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#### Author for correspondence:

M. Avila e-mail: marc.avila@zarm.uni-bremen.de

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# Statistical analysis of thermal conductivity experimentally measured in water-based nanofluids

J. Tielke<sup>1</sup>, M. Maas<sup>2,3</sup>, M. Castillo<sup>1</sup>, K. Rezwan<sup>2,3</sup> and M. Avila<sup>1,3</sup>

<sup>1</sup>University of Bremen, Center of Applied Space Technology and Microgravity (ZARM), Am Fallturm, 2, 28359 Bremen, Germany <sup>2</sup>University of Bremen, Advanced Ceramics, Am Biologischen Garten, 2, 28359 Bremen, Germany

<sup>3</sup>MAPEX Center for Materials and Processes, University of Bremen, 28359 Bremen, Germany

(D) JT, 0000-0001-7119-5333

Nanofluids are suspensions of nanoparticles in a base heat-transfer liquid. They have been widely investigated to boost heat transfer since they were proposed in the 1990s. We present a statistical correlation analysis of experimentally measured thermal conductivity of water-based nanofluids available in the literature. The influences of particle concentration, particle size, temperature and surfactants are investigated. For specific particle materials (alumina, titania, copper oxide, copper, silica and silicon carbide), separate analyses are performed. The conductivity increases with the concentration in qualitative agreement with Maxwell's theory of homogeneous media. The conductivity also increases with the temperature (in addition to the improvement due to the increased conductivity of water). Surprisingly, only silica nanofluids exhibit a statistically significant effect of the particle size, whereby smaller particles lead to faster heat transfer. Overall, the large scatter in the experimental data prevents a compelling, unambiguous assessment of these effects. Taken together, the results of our analysis suggest that more comprehensive experimental characterizations of nanofluids are necessary to estimate their practical potential.

## 1. Introduction

The efficient removal of heat with circulating fluids is pivotal to applications in mechatronics, mechanical, aerospace and chemical engineering. The main limiting material property in heat transfer is the thermal conductivity (k) of the fluid, followed by the viscosity. Conventional liquids used for heat transfer, such as water or ethylene glycol, are inexpensive but exhibit low k-values. In an influential paper published in 1995, Choi & Eastman [1] proposed that 'an innovative class of heat transfer fluids can be engineered by suspending metallic nanoparticles in conventional heat transfer fluids. The resulting nanofluids are expected to exhibit high thermal conductivities'. Their experimental measurements with ethylene glycol exhibited an increase in thermal conductivity up to 20% at a volume fraction of 4% through the addition of copper oxide particles ( $d \approx$ 20 nm) and spurred many theoretical and experimental investigations [2]. Several models were proposed to explain what was historically termed as anomalous heat transfer, meaning that the effective material properties of the suspension (e.g. effective viscosity and thermal conductivity of the nanofluid) could not solely account for the enhanced heat transfer [3,4]. In an influential paper, Buongiorno [5] considered many possible physical mechanisms behind convective heat transfer enhancement in nanofluids and assessed their plausibility and relative importance. He argued that Brownian motion and thermophoresis (i.e. the motion of particles along gradients of temperature) were the two only plausible mechanisms for heat transfer enhancement. Keblinski et al. [6] critically analysed some experimental datasets and concluded that effective medium theories are capable of explaining the data.

In 2009, Buongiorno *et al.* [7] performed an experimental benchmark study to determine the influence of the measurement technique on the experimental thermal conductivity of nanofluids. This study showed that the data scattered in a range of at least  $\pm 5\%$  about the median, which implied that a total enhancement of a few per cent cannot be detected. Subsequently, Khanafer *et al.* [8] developed correlations for thermal conductivity and viscosity based on experimental data. There have been no additional benchmark studies or statistical analysis since their study. Characterization efforts have mainly focused on the analysis of the nanoparticle sizes and particle concentrations [9–13].

Recently, the state of the art in this field was reviewed by Buschmann *et al.* [14], who analysed several experiments on convective heat transfer with nanofluids. Their analysis supported the conclusion of Buongiorno [5] that there are no anomalies in the convective or conductive heat transfer of nanofluids. Buschmann *et al.* [14] argued that nanofluids can be treated as homogeneous fluids by considering their effective properties. Hence, their heat transfer can be correctly predicted by well-established correlations for pure fluids, provided that the effective thermal conductivity and the effective viscosity of the nanofluid are known. Finally, Buschmann *et al.* [14] pointed out that there is currently a lack of knowledge of how and why nanoparticles change the thermal conductivity of a fluid.

The observation that thermophysical properties are modified by additions of particles to a fluid dates back to the theoretical work of Maxwell in 1881 [15]. According to his Effective Medium Theory, the effective thermal conductivity of a nanofluid  $k_{\text{eff}}$  depends on the thermal conductivities of the nanoparticles  $k_{\text{p}}$ , thermal conductivity of the base fluid  $k_{\text{ff}}$ , and particle fraction  $\varphi$ ,

$$\frac{k_{\rm eff}}{k_{\rm f}} = 1 + \frac{3\varphi(k_{\rm p} - k_{\rm f})}{3k_{\rm f} + (1 - \varphi)(k_{\rm p} - k_{\rm f})}.$$
(1.1)

Eapen *et al.* [16] argued that there may be different dispersion states in nanofluids that influence thermal conductivity, which are not considered in Maxwell's theory. For example, particles can form percolating structures at moderately low concentrations [16,17]. Through the use of a theory developed by Hashin & Shtrikman [18], Eapen *et al.* [16] derived the following Hashin and Shtrikman (HS)-bounds for the effective thermal conductivity of nanofluids

$$k_{\rm f} \left[ 1 + \frac{3\varphi(k_{\rm p} - k_{\rm f})}{3k_{\rm f} + (1 - \varphi)(k_{\rm p} - k_{\rm f})} \right] \le k_{\rm eff} \le k_{\rm p} \left[ 1 - \frac{3(1 - \varphi)(k_{\rm p} - k_{\rm f})}{3k_{\rm p} - \varphi(k_{\rm p} - k_{\rm f})} \right].$$
(1.2)

The lower HS-bound represents the well-dispersed state, where the particles are the disperse phase and the base fluid is the continuous phase, which was already described by Maxwell, see equation (1.1). Hence, in the well-dispersed state, heat is mainly transferred through the fluid. The upper HS-bound represents a state in which heat is mainly transferred between particles [16,18,19]. Specifically, at high concentrations ( $\varphi \approx 1$ ), the base fluid becomes the dispersed phase and particles the continuous phase. Even if this limit is not realistic for e.g. spherical particles (due to close-packing), it is still useful in describing particle configurations (chains, percolation networks), which can arise even at low concentrations and result in strongly enhanced heat transfer [20].

Maxwell's and Hashin & Shtrikman's theories of suspensions do not account for the particle size, whereas the distinct feature of nanofluids is that the particles are less than 100 nm in one dimension. Vadasz [21] considered heat conduction in nanofluids and reviewed the transient-hot-wire method. He showed that by accounting for the dependence of the heat transfer coefficient on the particle size (through the particle's specific area *S*), the experimental data became consistent with classical theories of suspensions. However, he stated that his analysis was inconclusive and that more experimental data were required. Further experimental studies [11,22–25] showed that thermal conductivity increases linearly with increasing temperature and suggested an effect from the particle size.

Bouguerra *et al.* [20] recently showed that the effective thermal conductivity and the effective viscosity of water-based alumina nanofluids strongly depend on pH. They were able to distinguish between different dispersion states that included well-dispersed particles, the formation of percolation networks, and fully agglomerated particles, through measurements with volume concentrations between 0.2% and 2% at different pH levels. Variations of the pH modify the suspension stability through repulsive forces of electrostatic origin. An alternative to this is the use of surfactants. Depending on the choice and concentration of the surfactant, the thermal conductivity of the nanofluid may be increased or decreased [12,26–30], and to date there is no general, coherent picture as to what the effect of surfactants on the nanofluid is.

In this paper, we present a statistical correlation analysis of thermal conductivity measurements of water-based nanofluids available in the scientific literature. The aim of our analysis is to assess nanofluids for potential applications and to identify suitable nanomaterials for heat transfer. To this avail, we analyse whether the influences of particle concentration, size, temperature and surfactants on the thermal conductivity of nanofluids are statistically significant, and we quantify their relative importance.

## 2. Material and methods

We compiled a database with N = 1656 data points (experimental measurements of the thermal conductivity) from 73 publications concerned with water-based nanofluids. The data points were taken from publications in which temperature, volume concentration and particle size were fully specified. The database is given in the electronic supplementary materials and contains data for 17 different nanoparticle materials. In the first part of our analysis, all data points were considered. Subsequently, six individual materials were analysed for which there are at least four different publications available comprising  $N \ge 50$  data points altogether. This was done to avoid spurious results due to small samples. The six individual materials are

- alumina (Al<sub>2</sub>O<sub>3</sub>, N = 470): [2,9,12,13,20,22–24,31–57]
- titania (TiO<sub>2</sub>, N = 188) [44,50,57–63]
- copper oxide (CuO, N = 106) [12,22,24,44,52,56,64]
- copper (Cu, N = 94) [26,27,56,65,66]
- silica (SiO<sub>2</sub>, N = 86) [13,32,67–72]
- silicon carbide (SiC, N = 53) [73–77].

Descriptive statistics for these datasets are shown in electronic supplementary materials, table S1. We note that for the analysis of copper nanofluids we excluded the study of Liu [65]. This study

features results which are very different from those of all other works for the same material. The decision to exclude them is further justified later. Here it suffices to say that as a result of excluding this, the regression significantly improves. For completeness, a comparison to the regressions with all data points (without exclusion) can be found in the electronic supplementary materials.

For the materials below, there are fewer than four publications and/or fewer than 50 data points. Thus, we did not analyse them individually, because the analysis would have little statistical significance:

- iron(III)oxide (Fe<sub>2</sub>O<sub>3</sub>, N = 86) [78,79]
- iron(II,III)oxide (Fe<sub>3</sub>O<sub>4</sub>, N = 98) [80–82]
- zinc oxide (ZnO, N = 3) [50]
- graphene (G, N = 145) [28,83,84]
- graphene oxide (GO, N = 20) [85]
- carbon-nanotubes (CNT, N = 188) [28,29]
- nanodiamond (ND, N = 38) [86,87]
- silver (Ag, N = 34) [68,70,88,89]
- iron (Fe, N = 15) [90]
- aluminium (Al, N = 20) [56]
- gold (Au, N = 4) [91,92].

We caution that the published data for metals have to be interpreted critically because metallic particles easily oxidize in water [93,94].

#### (a) Linear statistical model

We employed a linear model to statistically quantify the effect of the volume concentration  $\varphi$ , the temperature *T* and the particle size (through the specific surface *S*) on the normalized thermal conductivity

$$k^*(\varphi, T, S) = \frac{k_{\text{eff}}(\varphi, T, S)}{k_f(T)},$$
(2.1)

where  $k_f(T)$  is the thermal conductivity of the base fluid (pure water) as a function of the temperature. Changes in the thermal conductivity of the base fluid  $k_f$  due to the addition of surfactants or changes in the pH were taken account of, if the data were specified in the respective studies. Nevertheless, most of the data are given normalized or are normalized on pure water.

Our linear statistical regression model reads

$$k^{*}(\varphi, T, S) = C_{0} + C_{\varphi} \varphi + C_{T} T^{*} + C_{S} S^{*}, \qquad (2.2)$$

where the coefficients  $C_i$  (with  $i = \{0, \varphi, T, S\}$ ) were determined from linear regressions of the datasets. These coefficients are dimensionless, due to the definitions of  $T^* = (T - T_{ref})/T_{ref}$  and  $S^* = (S - S_0)/S_{ref}$ . We chose  $T_{ref} = 293$  K because most measurements found in the literature were taken at room temperature. For large particles (with specific surface  $S \approx 0$ ), we do not expect an increase in thermal conductivity beyond Maxwell's theory, therefore we chose  $S_0 = 0$  as reference. Finally, we chose  $S_{ref} = 6/d_{ref}$  with  $d_{ref} = 1$  nm for simplicity (with these choices the last term simplifies to  $S^* = S/S_{ref} = d_{ref}/d$ ).

#### (b) Physical interpretation of the linear statistical model

Starting with Maxwell's equation (1.1) and linearizing about zero concentration ( $\varphi$ =0), one obtains a linear prediction of the normalized effective thermal conductivity for small concentrations

$$k^*(\varphi) = 1 + C_{\varphi,\text{Maxwell}}\varphi, \qquad (2.3)$$

where

$$C_{\varphi,\text{Maxwell}} = 3 \frac{(k_{\rm p} - k_{\rm f})}{(k_{\rm p} + 2k_{\rm f})}.$$
 (2.4)

**Table 1.** Results of the linear regressions fitted to the entire database and for each individual material, with the number of data points (*N*), corrected correlation coefficient ( $R^2$ ), model coefficients ( $C_{\varphi}$ ,  $C_T$ ,  $C_S$ ) and their corresponding standardized correlation coefficients ( $\beta_{\varphi}$ ,  $\beta_T$ ,  $\beta_S$ ).

material	N	<i>R</i> <sup>2</sup>	<i>C</i> <sub>0</sub>	$\mathcal{C}_{arphi}$	$eta_arphi$	ζ	$eta_{ extsf{T}}$	Cs	$eta_{ extsf{S}}$
general	1656	0.29	1.031	1.81	0.39	0.51	0.27	0.092	0.34
alumina	470	0.53	1.025	1.75	0.72	0.31	0.21	0.16	0.12
titania	188	0.75	1.018	1.69	0.58	0.63	0.51	—0.19	-0.097
copper oxide	106	0.23	1.051	1.45	0.54	0.27	0.20	-0.47	-0.057
copper	94	0.43	1.059	7.46	0.72	-0.24	-0.086	0.48	0.073
silica	86	0.27	0.994	0.61	0.35	0.10	0.31	0.42	0.47
silicon carbide	53	0.68	1.082	2.97	0.70	-0.018	-0.017	—1.25	-0.25

The coefficient  $C_{\varphi,\text{Maxwell}}$  increases monotonously as the thermal conductivity of the particles  $k_p$  increases. In particular, in the low conductivity limit ( $k_p \ll k_f$ ),  $C_{\varphi,\text{Maxwell}} = -1.5$ , whereas in the opposite limit ( $k_p \gg k_f$ ),  $C_{\varphi,\text{Maxwell}} = 3$ . Hence in the framework of the linear Maxwell's equation (2.3),  $C_{\varphi,\text{Maxwell}} \in [-1.5, 3]$ . This theoretical prediction can be compared with the coefficients obtained from the linear regression of (2.2) to the compiled datasets, whereby regression values with  $C_{\varphi} > 3$  indicate higher performance than admissible in Maxwell's theory. For example, in the aforementioned experiments of Lee *et al.* [2], the heat transfer rates increased up to 20% at a volume fraction of 4% ( $\varphi = 0.04$ ). This would imply  $C_{\varphi} = 5$ , which cannot be explained with Maxwell's theory. Furthermore, if there are no particles ( $\varphi = 0$ ), the thermal conductivity must remain unchanged and it would be expected that the linear correlation analysis yielded values of  $C_0 \approx 1$ . The deviation of  $C_0$  from unity serves as a proxy for the level of quality of the linear model and/or for the scatter of the data.

#### (c) Data processing and statistical methods

Analyses were performed with IBM SPSS Statistics v. 25 using regressions with the normalized thermal conductivity ( $k^*$ ) as a dependent variable and  $\varphi$ ,  $T^*$  and  $S^*$  as independent variables. All variables were entered in a single step and confidence intervals were calculated to 95%. We evaluated the corrected correlation coefficient  $R^2$ , the coefficients ( $C_0, C_{\varphi}, C_T, C_S$ ), the standard deviations of the coefficients and the standardized regression coefficients ( $\beta_{\varphi}, \beta_T, \beta_S$ ). The values of ( $\beta_{\varphi}, \beta_T, \beta_S$ ) were calculated by subtracting the mean from the variable and dividing by its standard deviation. The larger the standardized regression coefficient of a parameter, the higher the influence of that parameter on  $k^*$ . Our analysis demonstrated that when  $|\beta_i| < 0.1$  the influence of parameter *i* was insignificant.

Additional analyses were also carried out with subsets of the data or variations of the model, which allowed an assessment of the robustness of the data and model. For example, to verify the use of the linear regression in terms of the concentration, we restricted the data to  $\varphi \le 0.02$ . The influence of surfactants was tested by performing separate regressions for the data without surfactants. Furthermore, we fitted nonlinear regressions with quadratic and cubic terms on the concentration and also with  $C_0 = 1$  fixed. Detailed results for all the linear and nonlinear regressions can be found in the electronic supplementary materials.

## 3. Results

The result of using a linear regression to fit all data points of our database with model equation (2.2) is shown in the first row of table 1. In figure 1, the model prediction (vertical axis) is plotted against the experimentally measured normalized thermal conductivity (horizontal



**Figure 1.** Experimentally measured versus modelled normalized thermal conductivity ( $k^*$ ) for water-based nanofluids. The ideal values of  $k^*$  and a  $\pm 10\%$  interval are displayed as a solid line and a grey area, respectively. Colours are used to distinguish single materials (analysed separately) from other materials (see the legend). The model is  $k^* = 1.031 + 1.81 \varphi + 0.51 T^* + 0.092 S^*$  ( $R^2 = 0.29$ , N = 1656). (Online version in colour.)

axis). Data points lying along the black line exhibit perfect agreement with our linear model, equation (2.2), whereas the discrepancy is larger the further the data points are from the line. The shaded region depicts a  $\pm 10\%$  interval about the model prediction. The correlation coefficient is  $R^2 = 0.29$  and  $C_0 = 1.031$ . The most significant parameter is the concentration, with  $C_{\varphi} = 1.81$  ( $\beta_{\varphi} = 0.39$ ), which is well within the bounds from the linearized Maxwell equation,  $C_{\varphi,\text{Maxwell}} \in [-1.5, 3]$ . The thermal conductivity also increases with increasing temperature and specific surface, with  $C_T = 0.51$  ( $\beta_T = 0.27$ ) and  $C_S = 0.092$  ( $\beta_S = 0.34$ ), respectively. These results allow a first estimation of the performance of water-based nanofluids. For example, if d = 30 nm at a 4% concentration and T = 303 K is substituted into equation (2.2)

$$k^* = C_0 + C_{\varphi} \varphi + C_T T^* + C_S S^* = 1.031 + 0.072 + 0.017 + 0.003 = 1.124$$

a 12.4% increase in thermal conductivity is obtained (when compared to pure water at T = 303 K). Noteworthy, the contribution directly from the particle size (last term) is negligible.

The linear regressions to the data sets for single materials are shown in figure 2. Clearly, the scatter in the data and the goodness of the fit depend strongly on the material, which is quantified by the respective  $R^2$  (given in the fifth column of table 1). Overall, the values of  $\beta_{\varphi}$  given in the fifth column of table 1 confirm that increasing the particle concentration leads to a statistically significant enhancement of the thermal conductivity for all materials analysed. In addition, there is also a significant influence of the temperature and/or surface for specific materials (see the seventh and ninth columns of table 1). In what follows, we discuss the influence of each of these three factors separately.

#### (a) Effect of particle concentration

The computed values of  $C_{\varphi}$  are shown in figure 3*a* as a function of the thermal conductivity of the particle material. The corresponding 95% confidence intervals indicate that silicon carbide nanofluids are in excellent agreement with Maxwell's prediction (equation (2.3) and lower solid line in the figure), whereas silica, alumina, titania and copper oxide nanofluids are below it. Only copper nanofluids appear to exceed Maxwell's prediction. Overall, the discrepancy with



**Figure 2.** Experimentally measured versus modelled normalized thermal conductivity ( $k^*$ ). The ideal values of  $k^*$  and a ±10% interval are displayed as a solid line and a grey area, respectively. (*a*) Alumina with N = 470 and  $k^* = 1.025 + 1.75 \varphi + 0.31 T^* + 0.16 S^*$  ( $R^2 = 0.53$ ). (*b*) Titania with N = 188 and  $k^* = 1.018 + 1.69 \varphi + 0.63 T^* - 0.19 S^*$  ( $R^2 = 0.75$ ). (*c*) Copper oxide with N = 106 and  $k^* = 1.051 + 1.45 \varphi + 0.27 T - 0.47 S^*$  ( $R^2 = 0.23$ ). (*d*) Copper with N = 94 and  $k^* = 1.059 + 7.46 \varphi + -0.24 T + 0.48 S^*$  ( $R^2 = 0.43$ ). (*e*) Silica with N = 86 and  $k^* = 0.994 + 0.61 \varphi + 0.10 T^* + 0.42 S^*$  ( $R^2 = 0.27$ ). (*f*) Silicon carbide with N = 53 and  $k^* = 1.082 + 2.97 \varphi + -0.018 T^* - 1.25 S^*$  ( $R^2 = 0.68$ ). (Online version in colour.)

Maxwell's prediction is not large in view of the large scatter in the datasets. By linearizing the upper HS-bound of equation (1.2), we obtained an upper bound for  $C_{\varphi}$  (upper solid line in figure 3*a*). This upper bound embraces all possible dispersion states and is satisfied also by copper.



**Figure 3.** The coloured symbols show the values of the model coefficient  $C_i$  (with corresponding 95% confidence-interval, shown as error-bars) as a function of the thermal conductivity  $k_p$  of each material. (*a*) Concentration coefficients  $C_{\varphi}$ . The linearized HS-bounds are displayed as solid black line. The lower bound is given by the linearized Maxwell equation (2.3), whereas the upper HS-bound was calculated by linearizing the right-hand side of equation (1.2) (with  $\varphi k_p/k_f = 0$ ). (*b*) Temperature coefficients  $C_1$ . (*c*) Surface coefficients  $C_5$ . (Online version in colour.)

#### (b) Effect of nanofluid temperature

The computed values of  $C_{\rm T}$  for single materials are shown in figure 3*b*. Increasing the temperature in titania nanofluids enhances the thermal conductivity significantly and strongly. For alumina and copper oxide nanofluids, the increase in thermal conductivity is less pronounced and also less significant. For silica the effect is significant, but the performance increase is even weaker. Finally, there is no significant influence of temperature on the thermal conductivity for copper and silicon carbide nanofluids, which is due to a lack of experimental measurements for a sufficiently large range of temperatures (see electronic supplementary material, table S1, fig. S2).

#### (c) Effect of the particle size

The computed values of  $C_S$  are shown in figure 3*c*. A strong effect of the particle size is found only in silica nanofluids ( $C_S = 0.42$ ,  $\beta_S = 0.47$ ). For alumina nanofluids, the particle size has a mildly significant and weak effect. In both cases, reducing the particle size appears to enhance the thermal conductivity. By contrast, for silicon carbide nanofluids the computed coefficient is negative ( $C_S = -1.25$ ), suggesting that increasing the particle size leads to higher thermal conductivity. However, the large scatter in the data for this material (see the corresponding confidence intervals in figure 3*c*) does not allow a conclusive statement. For copper oxide and copper nanofluids, the effect is statistically insignificant, which may be attributed to the absence of variation in particle size in the experimental measurements available (see electronic supplementary material, table S1 and figure S2).

The case of titania is more complicated to interpret. Here the experimental measurements span a wide range of particle sizes, yet the particle-size effect is insignificant if all data are used. If only the measurements without surfactant are considered in the statistical analysis, the particle size appears to have a very strong and significant influence, with  $C_S = 1.931$  and  $\beta_S = 0.367$ . The descriptive statistics and boxplot diagram in the supplementary material (see figures S2 and S4 therein) show an irregular distribution of the measured particle sizes, with most measurements for d = 21 nm and a few for 100 nm. This may result in the significant regression coefficients.

#### (d) Effect of surfactants

Out of N = 1656 data points, 1038 data points (62%) were obtained in suspensions without surfactants, whereas 618 (38%) data points were obtained with surfactants. Half of these data points (309) were from surfactant concentrations of less than 1.67 wt%, the rest with concentrations higher than 10 wt% (225 data points) or provided no information about the concentration (84 data points). Descriptive statistics and corresponding boxplots can be found in the electronic supplementary material, §2. Depending on the type of the surfactant and concentration, the thermal conductivity of both, the base fluid and the nanofluid, may increase or decrease [12,26–29]. Because of the heterogeneity of the measurements with surfactants (in type and concentration), it would be meaningful to analyse data with similar surfactant configurations together. However, this would lead to small data subsets and would prevent statistically significant analyses. Hence, in this work, we statistically investigated the effect of surfactants by performing regressions of the data points obtained without surfactants (separately for each material), and then comparing the results to those presented in the previous sections (with and without surfactants).

The data for copper were all obtained with surfactants except for the study of Liu *et al.* [65]; this is the reason why their work was excluded from the analysis for copper. For SiO<sub>2</sub> and SiC nanofluids, no surfactants were used. Hence the effect of surfactants cannot be investigated for these materials, as no comparisons (with/without surfactants) are possible. Still, we stress that the results for copper reveal a  $C_{\varphi}$  beyond Maxwell's theory in contrast to the other materials and insignificant  $C_{\rm T}$  and  $C_{\rm S}$  coefficients with large error-bars.

Comparisons were possible for Al<sub>2</sub>O<sub>3</sub> and CuO, with (405/470) and (94/106) data points obtained without surfactants, respectively. The changes in the regressions of alumina and copper oxide without surfactants lie within the 95% intervals of the regression for the whole datasets. Major changes occurred in the case of TiO<sub>2</sub>. Out of N = 188 data points, 83 were obtained without surfactants. The concentration coefficient  $C_{\varphi}$  increases and nearly approaches Maxwell's prediction. The coefficients  $C_{\rm T}$  and  $C_{\rm S}$  increase beyond the 95% interval of the whole data set, whereas  $C_0$  decreases.

Overall it can be concluded that while surfactants are expected to change the stability and the thermal conductivity of nanofluids, no conclusive statements can be made with the data available in the literature.

#### 4. Discussion

Buongiorno et al. [7] pointed out already one decade ago that nanofluid data in the literature exhibit large scatter ( $\pm$ 5% about the median). A first critical statistical analysis of the scatter in the experimental data was done by Khanafer & Vafai [8]. Our statistical analysis extends and updates their analyses and confirms that the scatter in the different datasets is very large, as illustrated by the regression coefficients  $R^2 \in [0.23, 0.75]$ . In line with Buongiorno *et al.* [7], we attribute the large scatter in the data to two factors. First, the experimental determination of thermophysical properties (e.g. thermal conductivity) of fluids is known to be susceptible of errors (even for pure fluids). Chirico et al. [95] reviewed thermophysical data from five major journals and found errors in nearly one-third of all publications. The most common problems were data with omitted uncertainties and the use of volume-based units [95]. Furthermore, in many publications the particle parameters (e.g. the size) were directly taken from the manufacturers, which is another source of error. Second, the characterizations of the nanofluids performed in the literature are often insufficient and additional variables need to be taken into account. Nieto de Castro et al. [96] considered the different factors influencing the thermal conductivity of nanofluids and emphasized the need for accurate, comparable measurements including stability characterizations (see also [97,98]).

#### (a) Dispersion state

The stability of nanofluids depends on their preparation, i.e. on the use of surfactants and/or the pH value of the fluid. For example, the colloidal stabilization of titania, alumina and other oxidic particles highly depends on pH [20], whereas hydrophobic particles (e.g. silicon carbide) agglomerate without the addition of dispersing agents [6,99,100]. Hence the surfactant itself and the concentration must be selected very carefully to maximize the thermal conductivity. Owing to the heterogeneity in surfactant type and concentration (and particle material and size) used across studies, it is difficult to compare data and draw general conclusions from them. Hence, despite the widespread use of surfactants, the results are ambiguous at best. Many studies suggest improved thermal conductivity, whereas others suggest the opposite [12,26–29]. The copper nanofluids in the literature were synthesized with the use of surfactants. In most studies, the concentration of surfactants was above the critical micelle concentration, possibly generating networks of micelles in the nanofluid. Hence the high value of  $C_{\varphi}$  obtained for copper may arise from a dispersion state different than well-dispersed particles. We note however that micellar aggregates, wormlike micelles and network structures increase the viscosity of the nanofluid [30,101]. Thus, the use of high surfactant concentrations may lead to high thermal conductivities, but may be rather unfavourable for the application of nanofluids.

Our analysis employs a linear regression to assess the importance of concentration, temperature and specific surface. This precludes an investigation of combined effects, whereas surface (particle-size) effects may depend on the concentration. Unfortunately, the quality and quantity of the published data do not currently allow for a statistically significant analysis of such combined effects via the inclusion of nonlinear terms (e.g. of the form  $C_{\varphi,S} \varphi S^*$ ). We note

however that we did not find any significant cross-correlations between variables. The aging and stability of the nanofluid, which change the dispersion state, could not be included in our statistical analysis, because these factors are seldom described in sufficient detail in published data [7,102,103]. For the same reason, we could not analyse the dependence of the effective viscosity on concentration, temperature and size of the nanoparticles.

The viscosity is crucial in heat transfer applications and complex structures, such as particle clusters or percolation networks, dramatically increase the viscosity of suspensions. Bouguerra *et al.* [20] have shown that for alumina nanofluids the well-dispersed regime (when available), is the preferred one for heat transfer applications. Studies employing molecular dynamics simulations have also shown that clustering influences the thermal conductivity of nanofluids [104–106], and increases their viscosity [104,105]. Tahmooressi *et al.* [106] showed for nanoparticles at high concentrations ( $\varphi = 0.1$ ) that the agglomeration of nanoparticles into small clusters may be more efficient towards high thermal conductivities than well-dispersed particles or percolating networks. This is in line with the experimental observations of Bouguerra *et al.* [20] measuring higher thermal conductivities for percolation than for dispersed particles. Clearly, simultaneous measurements of thermal conductivity and viscosity, and direct measurements of the state of dispersion of the nanofluid would be very useful. Here the determination of the diffusion coefficient and the size of the hydration layer would help [19,107,108].

#### (b) Regressions with $C_0 = 1$

We also fitted all datasets discussed above, but imposing  $C_0 = 1$  in equation (2.2). The results are shown in electronic supplementary material, table S6a–g and figures S5–S7. For silica nanofluids, the results do not change much when  $C_0 = 1$  is imposed. For all other particle materials, imposing  $C_0 = 1$  leads to an increase in the values of  $C_{\varphi}$ ,  $C_T$  and  $C_S$ . This is because for all these materials  $C_0 > 1$  in the linear regressions. In fact, the larger the value of  $C_0$  in the linear regression, the larger the increase of the other coefficients when  $C_0 = 1$  is set. For silicon carbide, copper oxide and copper nanofluids, the value of  $C_S$  even changes from negative to positive. Hence nothing can be said about particle size for these nanofluids. In both linear and nonlinear analyses, the 95% confidence intervals for the surface coefficient  $C_S$  reflect the large scatter in the data regarding particle size effects. Overall, it can be concluded that setting  $C_0 = 1$  does not qualitatively change the results of our analysis, but rather fortifies our findings and our assessment of the data quality.

#### (c) Analysis for small concentrations $\varphi \leq 0.02$

We analysed the data for concentrations below 2% separately to assess the validity of the linear assumption for the concentration in our statistical regression model. We found statistically identical results for silica, silicon carbide and copper. For copper oxide, titania and alumina (see electronic supplementary material, §2.1 and figure S3) the (relative) performance is better at low concentrations. For alumina, there is then perfect agreement with Maxwell, whereas titania and copper oxide are above the Maxwell curve, but with confidence intervals nearly touching it. It can be concluded that higher concentrations decrease the (relative) performance, possibly due to unfavourable agglomeration effects (e.g. [20]).

### 5. Conclusion and outlook

Our statistical analysis shows that the experimentally measured thermal conductivity of waterbased nanofluids increases approximately linearly with the particle concentration. Within statistical uncertainty, this increase can be accounted for by Maxwell's Effective Medium Theory for all materials. A possible exception may be copper nanofluids, although more experiments are necessary to assess their performance and the role of surfactants. A linear increase of the conductivity with the temperature was observed for all materials in which measurements covering sufficiently large temperature ranges are available. Finally, only silica nanofluids exhibit statistically significant, strong particle-size effects, which makes this scarcely investigated material interesting for fundamental investigations. In particular, in silica nanofluids, reducing the particle size (at constant concentration) leads to higher conductivity. Their performance is however low (compared to other nanofluids) because of their low value of  $C_{\varphi}$ .

In the view of the detrimental effect of the increased viscosity and of stability issues, we suggest that the potential of nanofluids in engineering practice is limited. The main advantage of nanofluids, when compared to suspensions of larger particles, is their reduced sedimentation speed. We conclude that the large scatter found in the experimental measurements makes it difficult to test and compare theories for the effective thermal conductivity of water-based nanofluids. More comprehensive and precise characterizations, including the analysis of the dispersion state and of the stability on nanofluids (e.g. depending on the surfactant), are needed to quantify the sources of the data scatter. We believe that an improvement of the state of the art can only be achieved by ensuring the reproducibility of results with a priori identical conditions in different research groups. Silica nanoparticles are usually homogeneous in their size, are easy to handle and are sufficiently stable in dispersion. Hence, silica nanofluids are good candidates to precisely quantify the effects of particle size and dispersion stability on nanofluids. Copper nanofluids exhibit high thermal conductivities, but in order to compare their performance to other materials, it would be necessary to stabilize them with small concentrations of surfactant (below the critical micelle concentration). Unfortunately, this is challenging because metal nanoparticles rapidly oxidize and agglomerate without surfactants [109].

Finally, we suggest that more sophisticated statistical analyses [110,111] could be employed to shed light on the source of the variability in the measured thermal conductivity of nanofluids. We will investigate possible factors in future work.

Data accessibility. All data used in the paper is referenced to the published articles. Additionally electronic supplementary material and the data table are made accessible together with the article.

Authors' contributions. J.T.: Conceptualization; methodology, formal analysis, investigation, writing: original draft, visualization, writing: review and editing; M.M., M.C. and K.R.: conceptualization, writing: review and editing. M.A.: conceptualization, methodology, validation, writing: original draft; writing: review and editing. All authors gave final approval for publication and agree to be held accountable for the work performed therein.

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