Wärmeleitfähigkeit von Nanofluiden

Internationaler Vergleichstest INPBE

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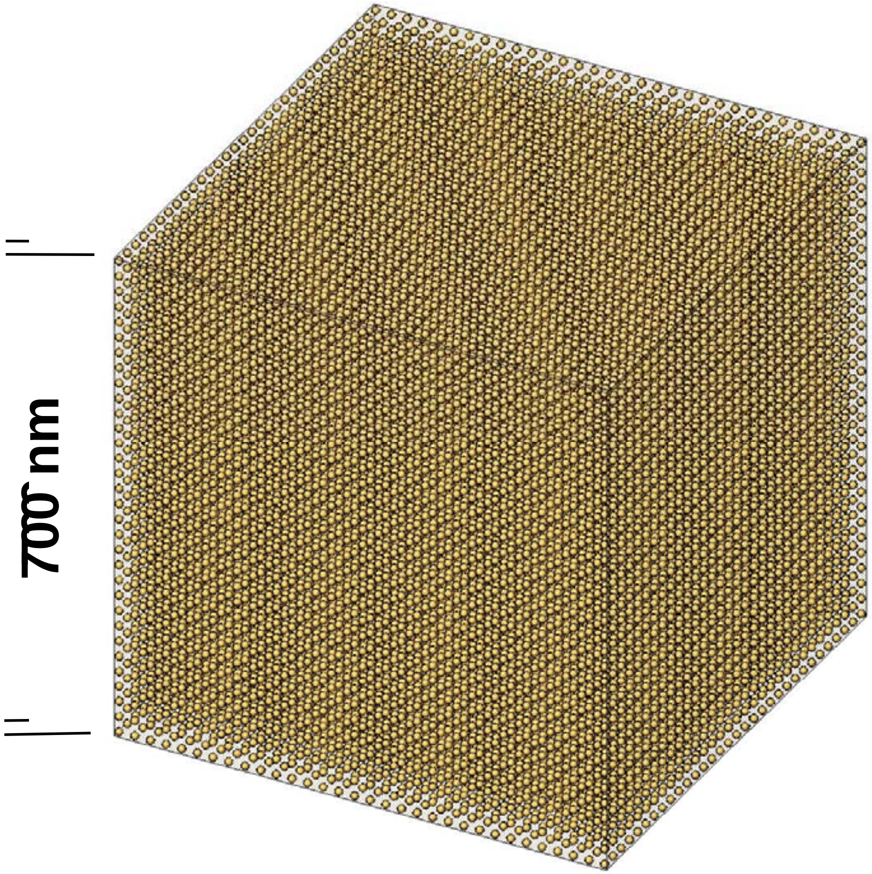
Nanofluid (Grundlagen)



uid

particle volume fraction:

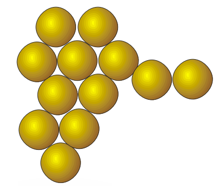
$\varepsilon_p = 5\%$

$d_p = 10$ nm

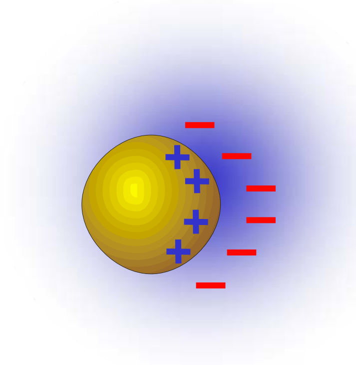
$\varepsilon_p = 5\%$

$d_p = 50$ nm

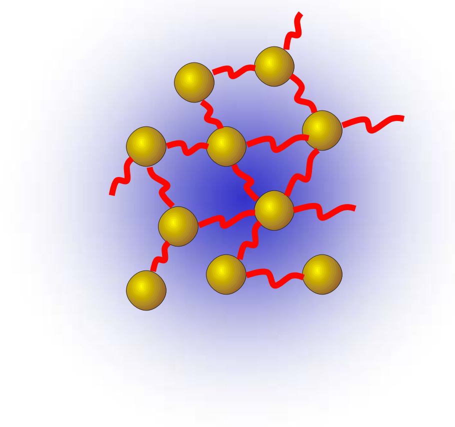
particle weight fraction 21 %



Agglomerate



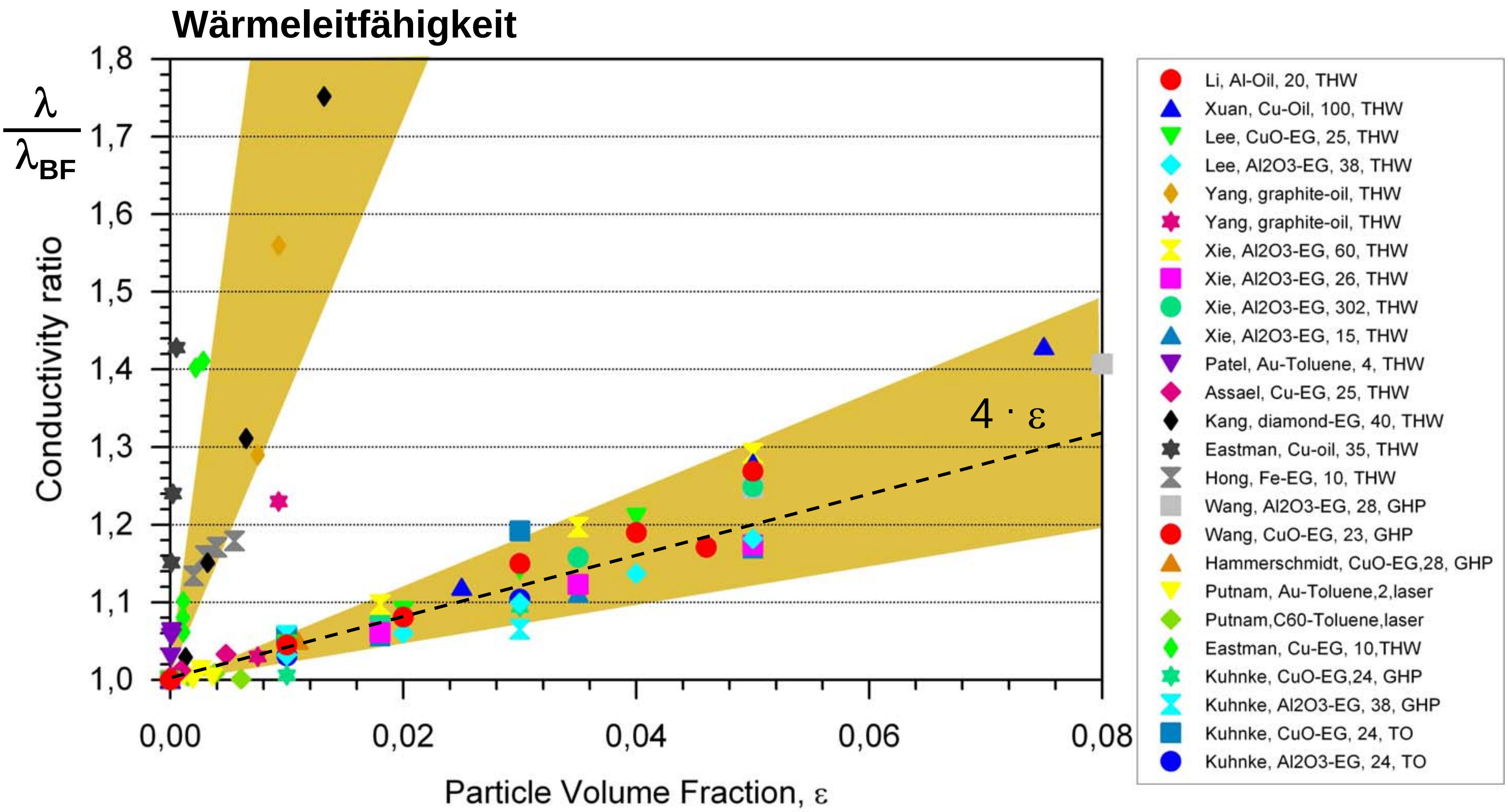
Elektrische Doppelschicht
Zeta Potential ζ



Stabilisatoren

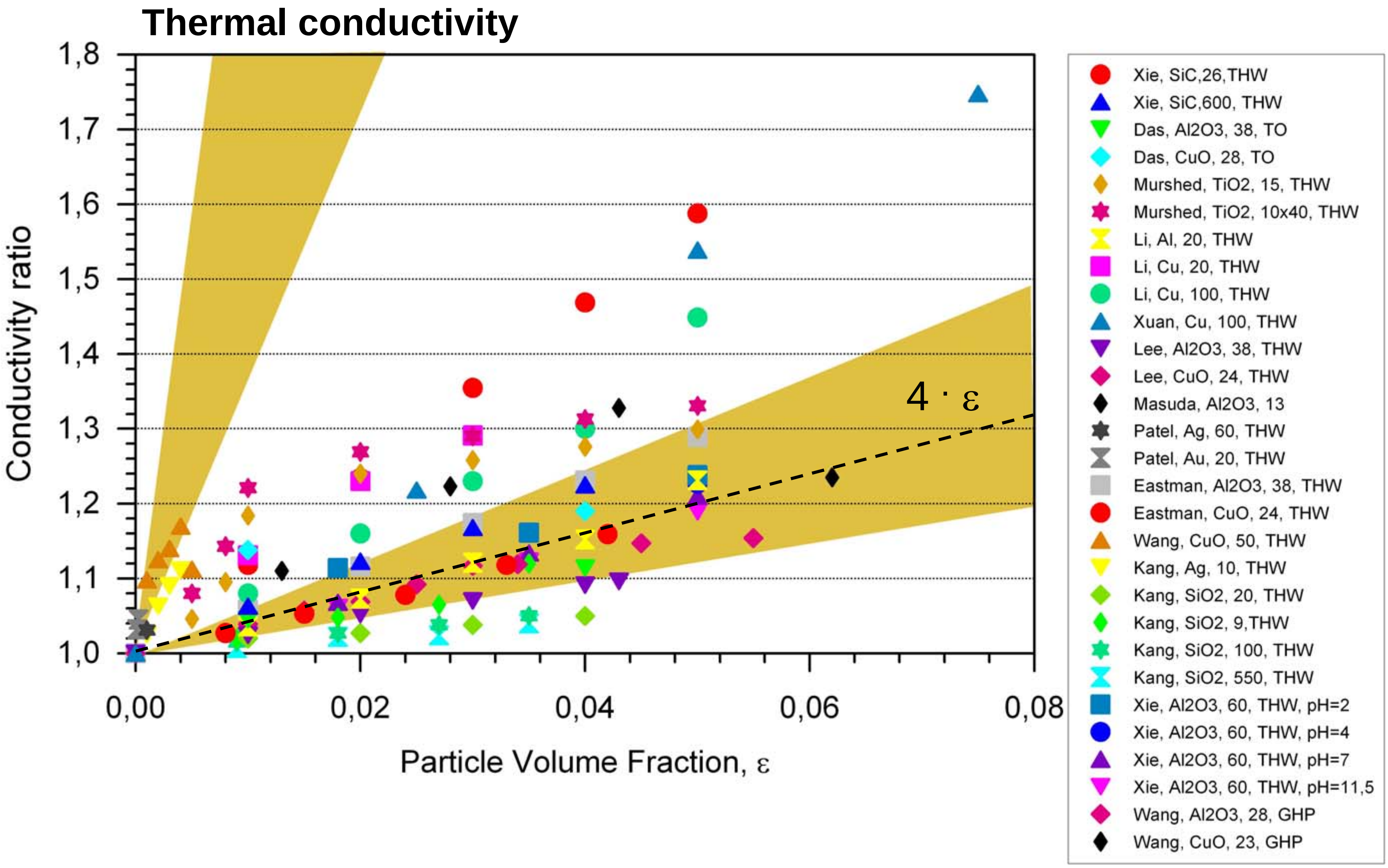
Organische Trägerfluide

alle Daten, T = 25°C



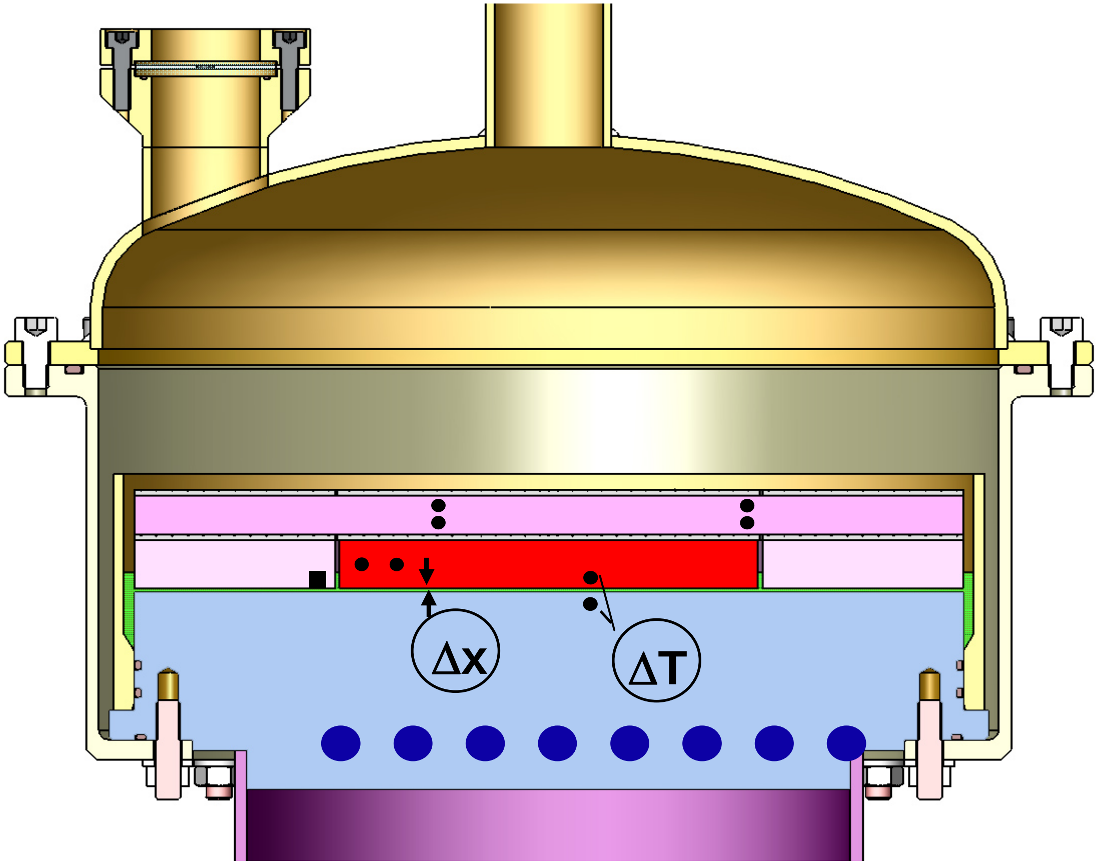
Wasser

alle Daten, T = 25°C

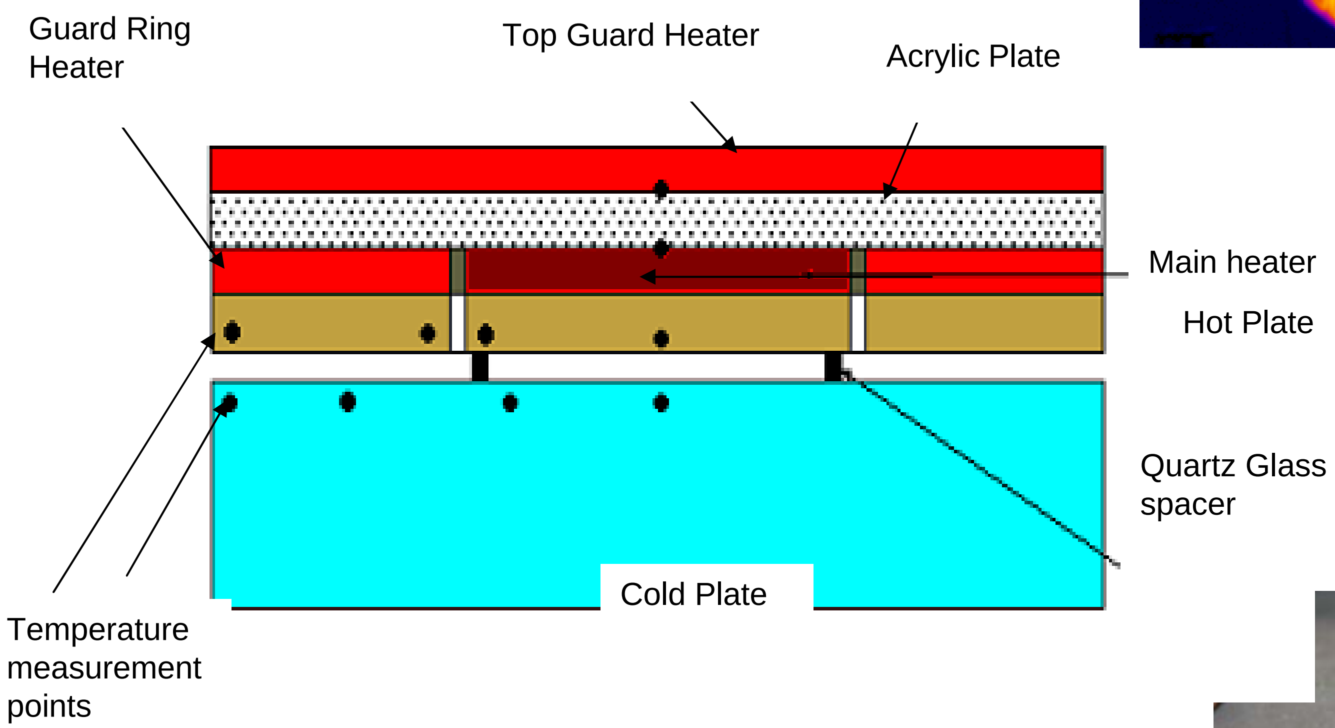


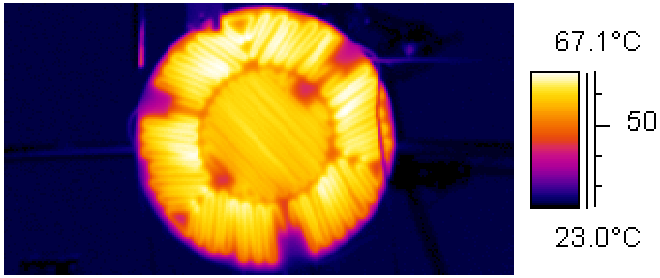
Parallelplatten - Apparatur an der HSU

$$\dot{q} = -\lambda \frac{\Delta T}{\Delta x}$$



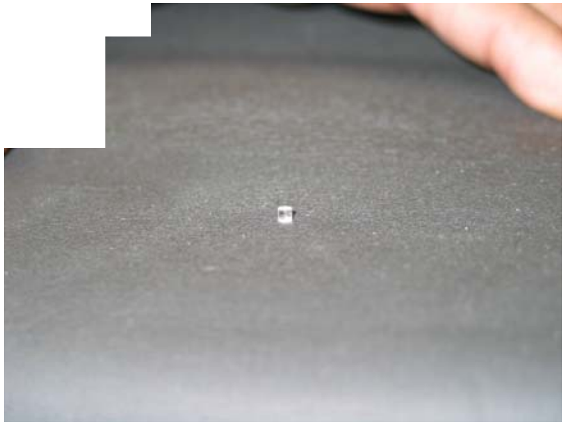
Einzelheiten





thermography check

spacer



Database (as of 2/2/09)

Institution / Contact persons	*Batch 1*	*Batch 2*	*Batch 3*	*Batch 4*
Argonne National Laboratory / Wenhua Yu	X	X	X	X
CEA-Saclay / Cecile Reynaud	X			
Chinese University of Hong Kong / Sheng-Qi Zhou	X	X	X	
DSO National Laboratories / Lim Geok Kieng				
ETH Zurich/ IBM Research / Werner Escher	X	X	X	X
Helmut-Schmidt University Armed Forces / Stephan Kabelac		X	X	X
Illinois Institute of Technology / David Venerus	X	X		
Indian Institute of Technology, Kharagpur / Indranil. Manna	X	X	X	X
Indian Institute of Technology, Madras / T. Sundararajan, S. K. Das	X	X		
Indira Gandhi Centre for Atomic Research / John Philip	X	X	X	
Kent State University / Yuriy Tolmachev	X	X	X	X
Korea Aerospace University / Seok Pil Jang			X	
Korea Univ. / Chongyoup Kim	X	X	X	
METSS Corp. / Frank Botz	X	X	X	
MIT / Jacopo Buongiorno, Lin-wen Hu, Tom McKrell	X	X	X	X
MIT / Gang Chen		X	X	
Nanyang Technological University / Kai Choong Leong	X			
NIST / Mark A Kedzierski	X			
Olin College of Engineering / Rebecca Christianson, Jessica Townsend	X	X		
Queen Mary University of London / Dongsheng Wen	X	X	X	X
Silesian University of Technology / Andrzej B. Jarzębski, Grzegorz Dzido,			X	X
South Dakota School of Mines and Technology / Haiping Hong	X	X	X	X
Stanford University / Patricia Gharagozloo, JooHyun Lee, Ken Goodson, John Eaton	X	X	X	
Texas A&M University / Jorge L. Alvarado	X	X	X	X
Tokyo Institute of Technology / In Cheol Bang	X	X	X	X
Universite Libre de Bruxelles / Carlo Saverio Iorio		X		
University of Leeds / Yulong Ding	X	X	X	X
Univ. of Missouri / Hongbin Ma				
Univ. of Nantes, Laboratoire de Thermocinétique / Ben-Abdallah				
Univ. of Pittsburgh, Minking Chyu	X	X	X	
University of Puerto Rico –Mayaguez / Jorge Gustavo Gutierrez				

Batch 1 Samples

Six samples (supplied by Sasol)

1. Alumina *nanorods* in de-ionized water
2. Alumina nanoparticles (first concentration) in Polyalphaolefins lubricant (PAO) + surfactant
3. Alumina nanoparticles (second concentration) in PAO + surfactant
4. Alumina *nanorods* (first concentration) in PAO + surfactant
5. Alumina *nanorods* (second concentration) in PAO + surfactant
6. PAO + surfactant

Sample #	Loading		Particle size		
	Sasol	MIT***	Sasol	MIT	IIT*****
1	1% vol.	1.2 to 1.3 % vol**	80x10 nm (nominal nanorod size),	60-64 nm****	131-134nm
2	1% vol.	0.7 to 0.8 % vol*	10 nm (nominal particle size)	na	75-88nm
3	3% vol.	1.9 to 2.2 % vol*	10 nm (nominal particle size)	na	99-112nm
4	1% vol.	0.7 to 0.8 % vol*	80x10 nm (nominal nanorod size)	na	70-110nm
5	3% vol.	2.0 to 2.3 % vol*	80x10 nm (nominal nanorod size)	na	100-115nm

* Measurements by neutron activation analysis (NAA)

** Measurements by inductive coupled plasma (ICP)

*** Range of values is given to account for expected hydration range of alumina (boehmite). Boehmite's chemical formula is Al2O3·nH2O, where n = 1 to 2. The hydrate is bound and cannot be dissolved in water. In most boehmites there is 70 to 82 wt% Al2O3 per gram of powder. Anhydrous boehmite density is 3.04 g/cm3.

**** Average size of dispersed phase, measured by dynamic light scattering (DLS). The range indicates the spread of multiple nominally-identical measurements. DLS systemic uncertainty is of the order of ±10 nm.

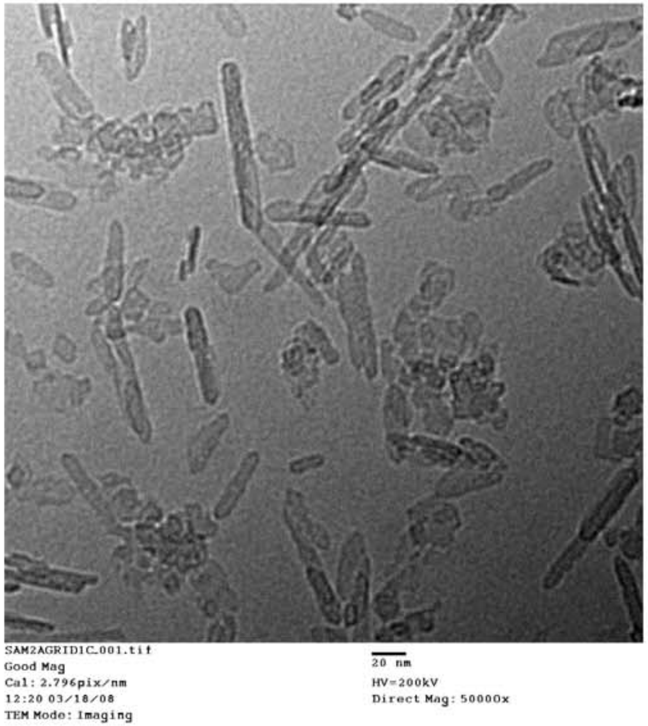
***** Average size of dispersed phase, measured by dynamic light scattering (DLS). The range indicates the spread of six nominally-identical measurements. DLS systemic uncertainty is of the order of ±10 nm. Malvern NanoS used to collect data.

na = not available due to unreliability of DLS analyzer with PAO-based samples

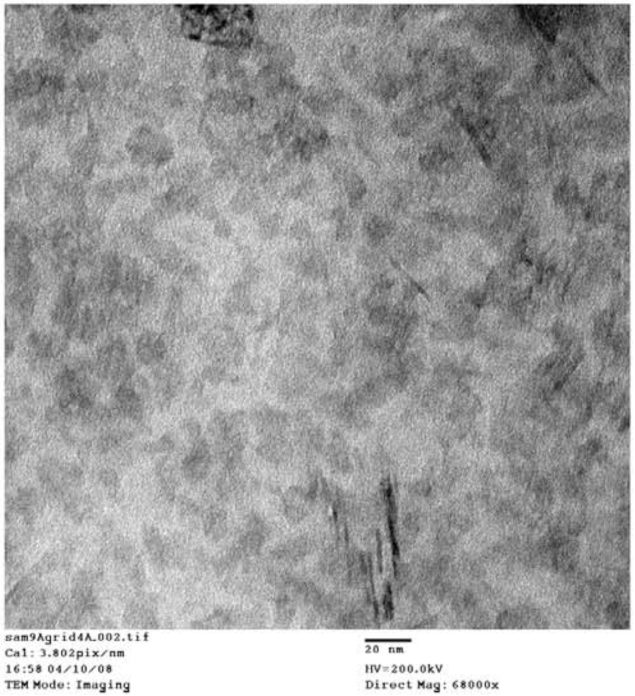
Batch 1 Samples

TEM images

Nanorods in water (Sample 1)



Nanoparticles in PAO (Sample 2)



TEM images of Samples 3-5 not available

Batch 2 Samples

Two samples (supplied by DSO National Labs, Singapore)

1. Gold nanoparticles in de-ionized water and trisodium citrate stabilizer.
2. De-ionized water + and sodium citrate stabilizer. (control sample)

Sample #	Au loading		Particle size			Stabilizer concentration (trisodium citrate)		pH
	DSO	MIT*	DSO	MIT***	IIT*****	DSO	MIT*	MIT
1	0.0010 vol%	0.0009 vol%**	20-30 nm	4-11 nm	14.8 nm ave (10-22 nm)	0.1 wt%	0.10 wt%	6.01
2	Zero	Zero ****	n/a	n/a	n/a	0.1 wt%	0.09 wt %	7.30

* Measurements by inductive coupled plasma (ICP). ICP has an accuracy of 0.6% of the reported value for gold in the concentration range of interest.

** Assumed density of gold is 19.32 g/cm3

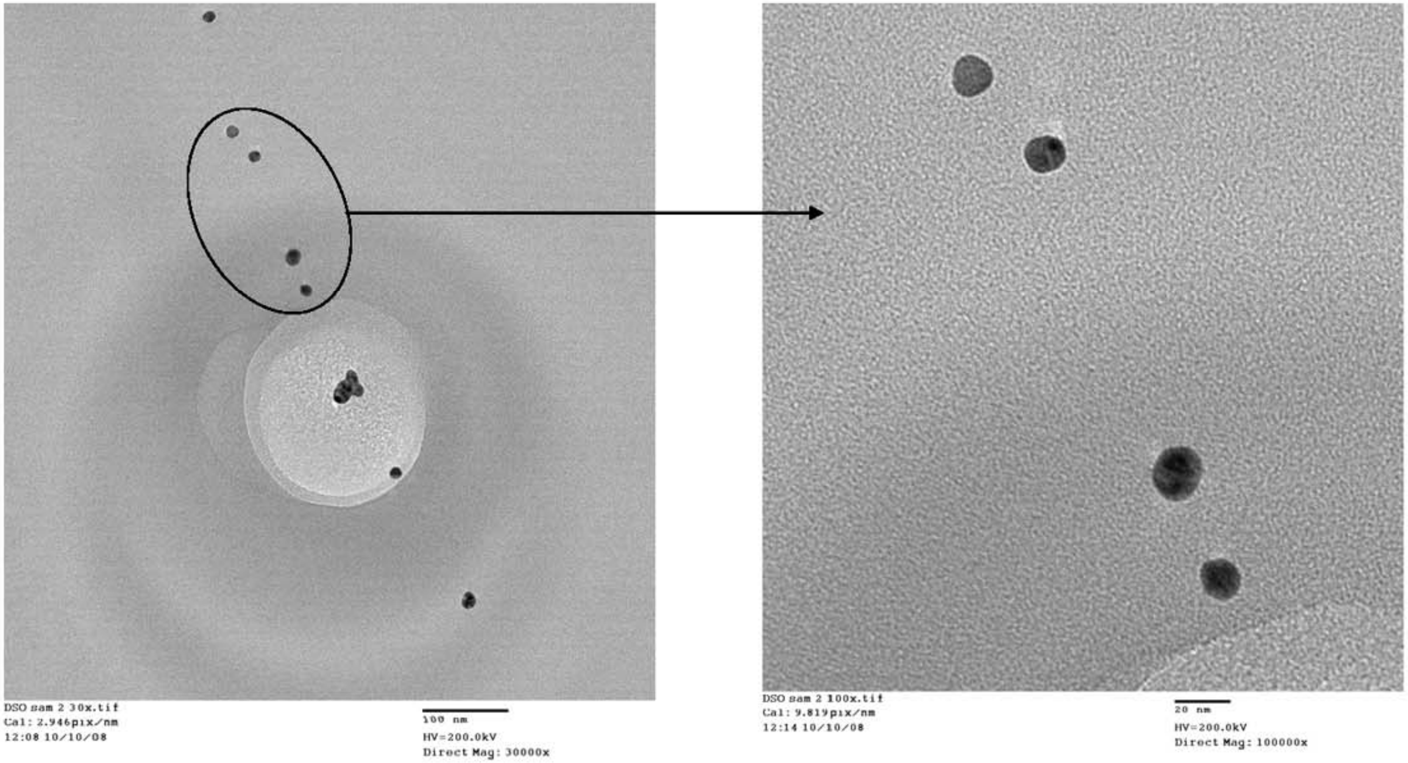
*** Number-weighted average size of particles, measured by dynamic light scattering (DLS). The range indicates the spread of two nominally-identical measurements. DLS systemic uncertainty is of the order of ±10 nm.

**** Within the detection limit of ICP.

***** Measurements by DLS. The values reported are the number-weighted average and the range at the full-width half maximum for six measurements.

Batch 2 Samples

TEM images



Batch 3 Sample

One sample (supplied by Grace Davison)

Silica spherical nanoparticles and stabilizer in de-ionized water (Ludox TM-50)

Comments

- Particles stabilized by pH control. The base is deprotonated silanol (SiO-) groups on the surface with Na ion as the counterion (0.1-0.2 wt% of Na ions).
- The dispersion contains also 500 ppm of a proprietary biocide.
- Grace Davison also stated that it is not possible to supply a control sample with only water and stabilizer "because of the way the particles are made".

Sample #	Silica (SiO_2) loading		Na_2SO_4 concentration		Particle size		pH	
	Grace Davison	MIT	Grace Davison	MIT*	Grace Davison	MIT***	Grace Davison	MIT
1	49.8 wt% 31.1 vol%**	43.6 wt%* 26.0 vol%**	0.1-0.2 wt% of Na	0.27 wt% of Na	22 nm	20-40 nm	8.9	9.03 (22.6°C)

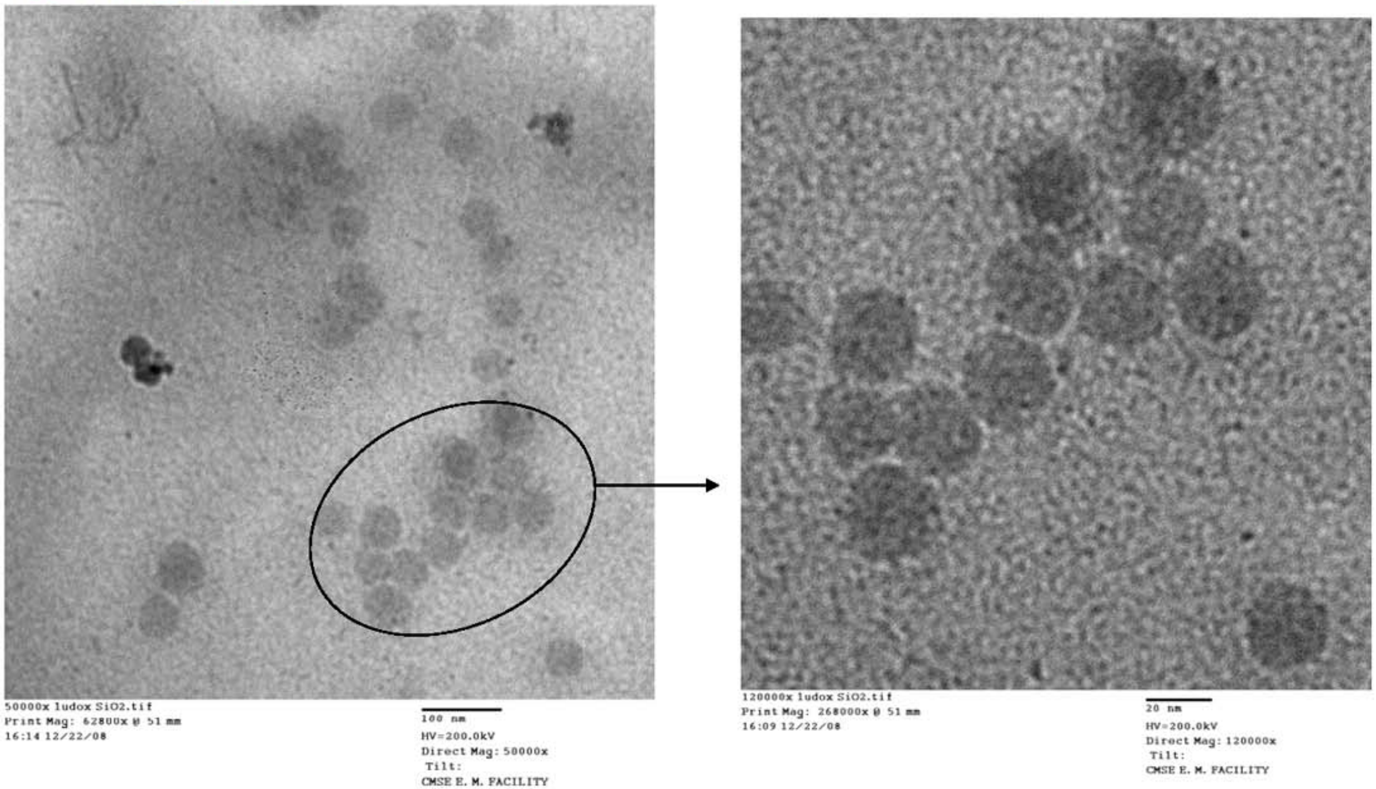
* Measured by inductive coupled plasma (ICP). ICP has an accuracy of 0.6% of the reported value.

** Assumed density of silica (SiO2) is 2.2 g/cm3

*** Number-weighted average size of particles, measured by dynamic light scattering (DLS). The range indicates the spread of three nominally-identical measurements. DLS systemic uncertainty is of the order of ±10 nm.

Batch 3 Sample

TEM images



Batch 4 Samples

Two samples (supplied by U-Puerto Rico at Mayaguez)

1. Mn-Zn ferrite ($Mn½-Zn½-Fe_2O_4$) particles in solution of stabilizer and water.
2. Solution of stabilizer (25 wt%) and water (75 wt%). (control sample)

Comments

- The stabilizer is Tetramethylammonium hydroxide, or (CH3)4NOH.

Sample #	Particle loading		Particle composition		Particle size		pH
	UPRM	MIT*	UPRM	MIT****	UPRM	MIT	MIT
1	0.86 vol%**	0.16 vol%	$Mn_{½}$-$Zn_{½}$-Fe_2***	Mn ~15 at%, Zn ~14 at%, Fe ~ 71 at%	12.9 nm*****	<20 nm******	15.2
2	N/A	N/A	N/A	N/A	N/A	N/A	15.1

* Measurements by inductive coupled plasma (ICP). Assumed a density of 4.8 g/cm^3 for Mn-Zn ferrite. ICP has an accuracy of 0.6% of the reported value.

** Determined from magnetic measurements

*** The molar fraction of Mn and Zinc was determined from stoichiometric balance.

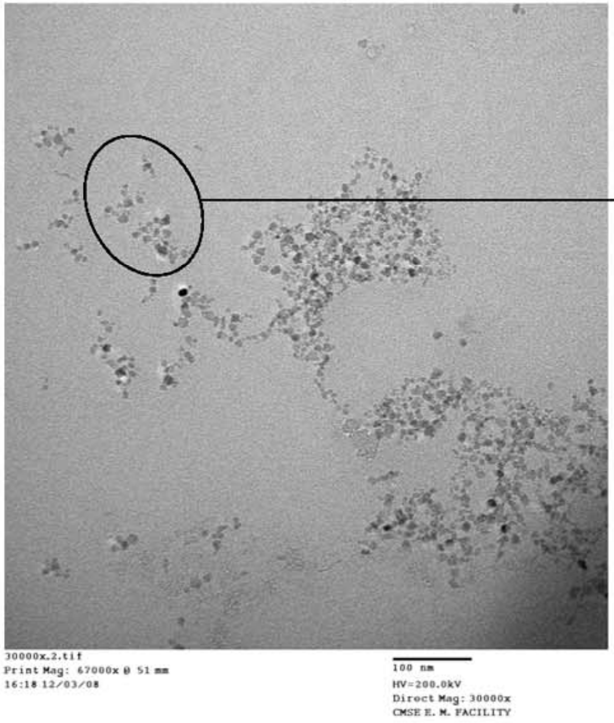
**** Atomic fraction measured by Energy Dispersive X-ray Spectroscopy (EDS).

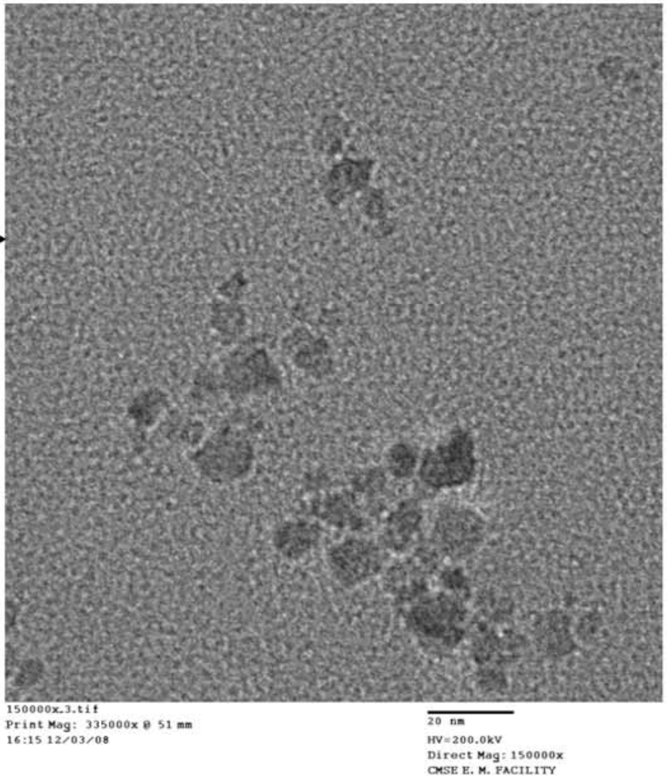
***** Average magnetic particle diameter

****** From TEM images

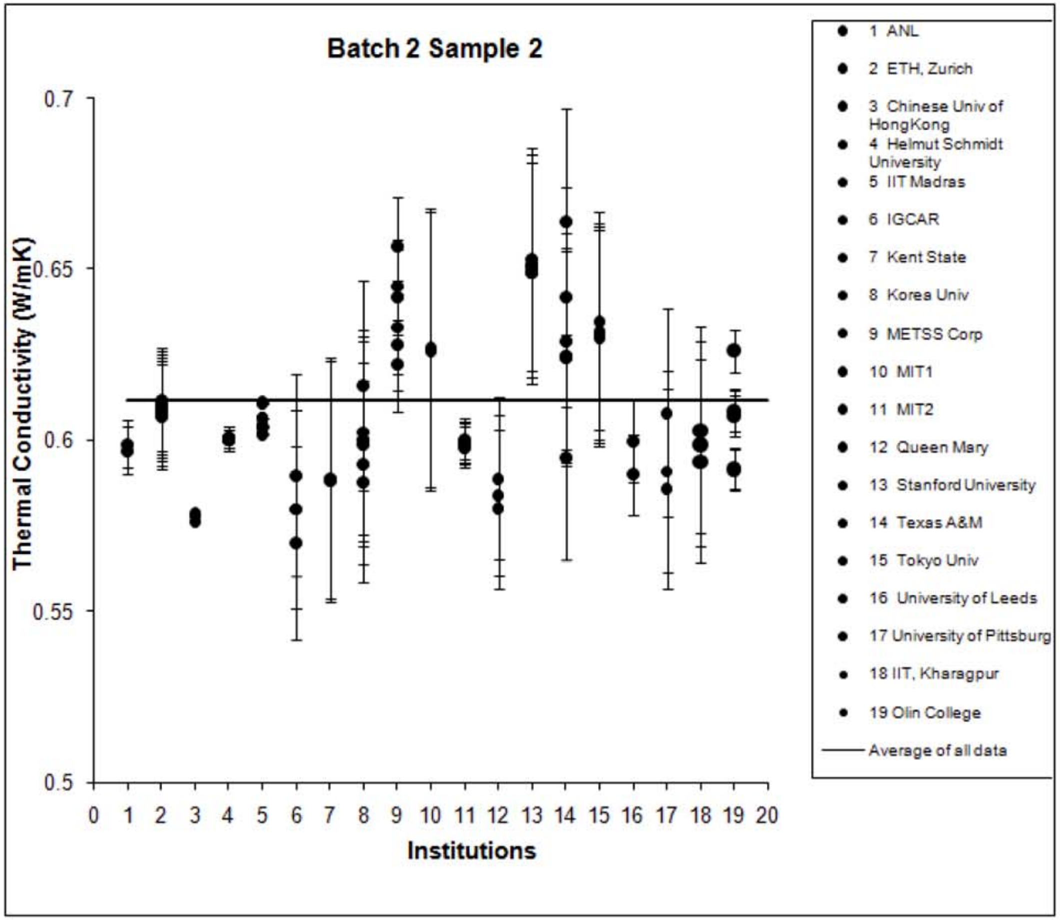
Batch 4 Samples

TEM images





BATCH II :- Sample 2 (Water + 0.1 wt% Trisodium Citrate)



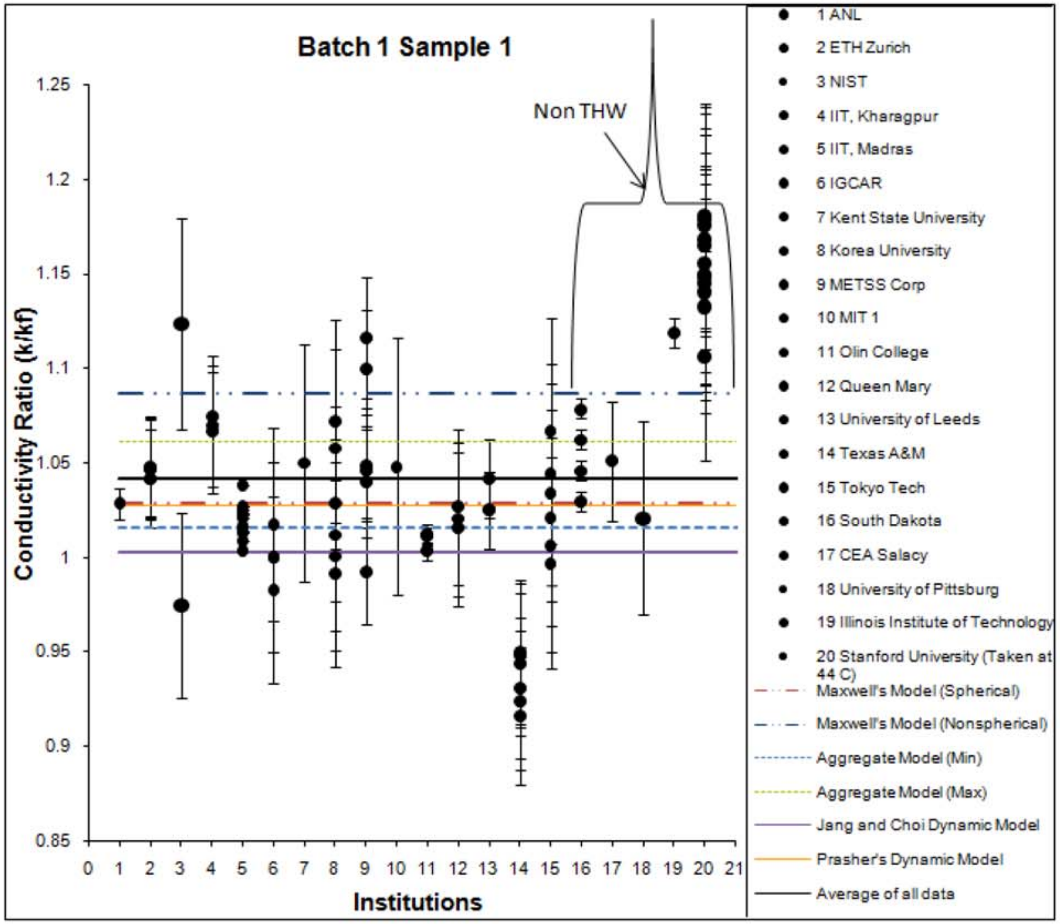
Observations

- Most data are within 5% of the mean value

- The average measured thermal conductivity was around 0.611 W/m-K

BATCH I :- Sample 1 (1 vol% Alumina Nanorods in Water)

Basefluid : Water



Observations :

- Most organizations report an enhancement in the range of 1% to 10 % with an average of around 4%

- Models predict an enhancement in the range 1% to 7%

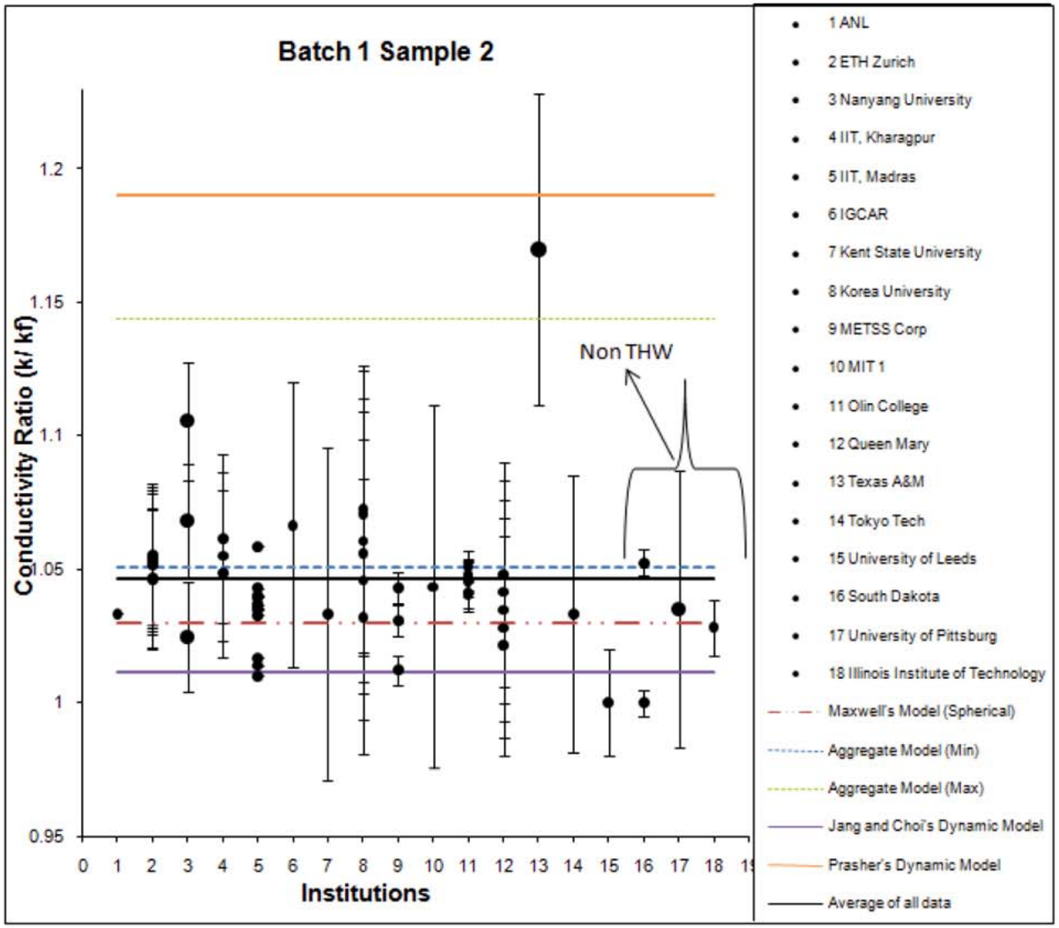
- No major differences observed between THW and non-THW data

- Prasher's Aggregate model and Maxwell's Static model give most accurate predictions

- Maxwell model for non-spherical nanoparticles over-predicts.

BATCH I :- Sample 2 (1 vol% Alumina Nanoparticles in PAO)

Basefluid : PAO + Surfactant

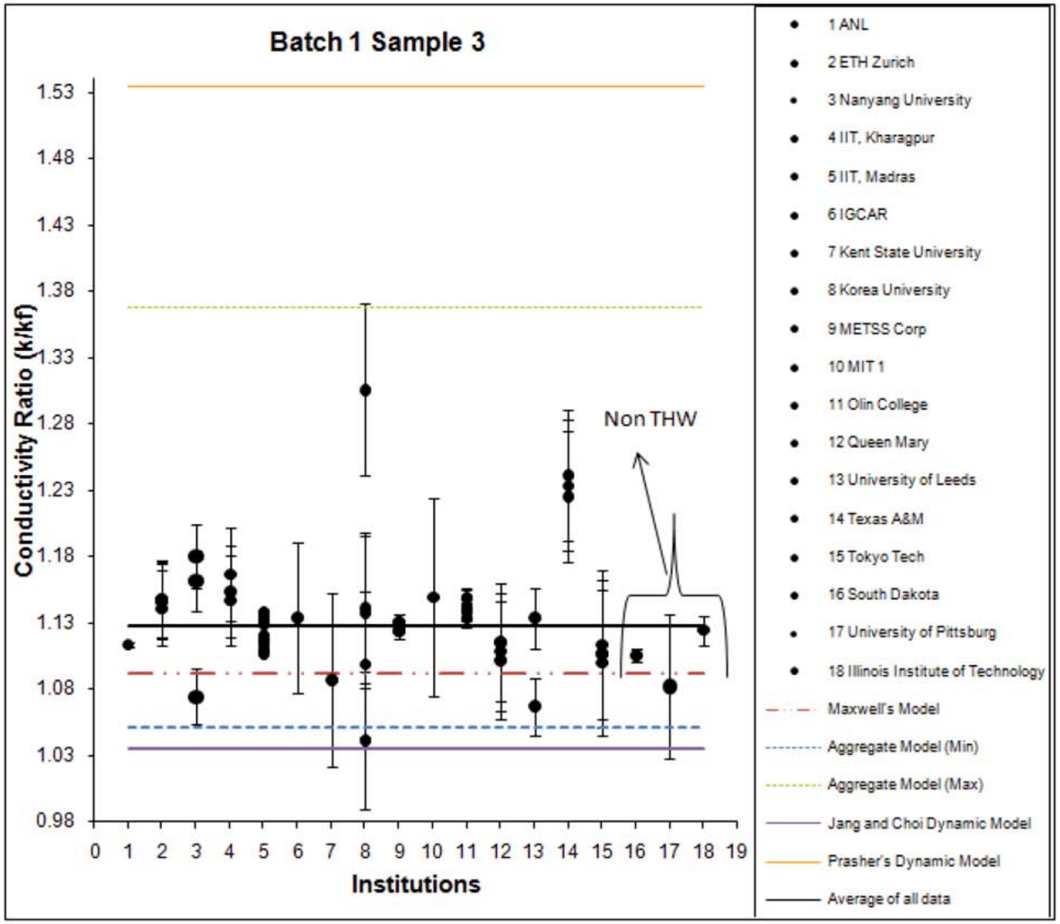


Observations :

- Enhancement of around 1% to 6% is observed with an average of about 4.5%
- Models predict an enhancement in the range of 1% to 18%
- Prasher's dynamic model seems to over-predict much in this case, while Jang and Choi's model under-predicts.
- The models predicting the observed behavior with a reasonable accuracy are Maxwell's Model and Prasher's Aggregate Model (at low aggregation state)

BATCH I :- Sample 3 (3 vol% Alumina Nanoparticles in PAO)

Basefluid : PAO + Surfactant



Observations :

- Most organizations observed enhancements in the range of 10% to 15% owing to higher concentration of nanoparticles

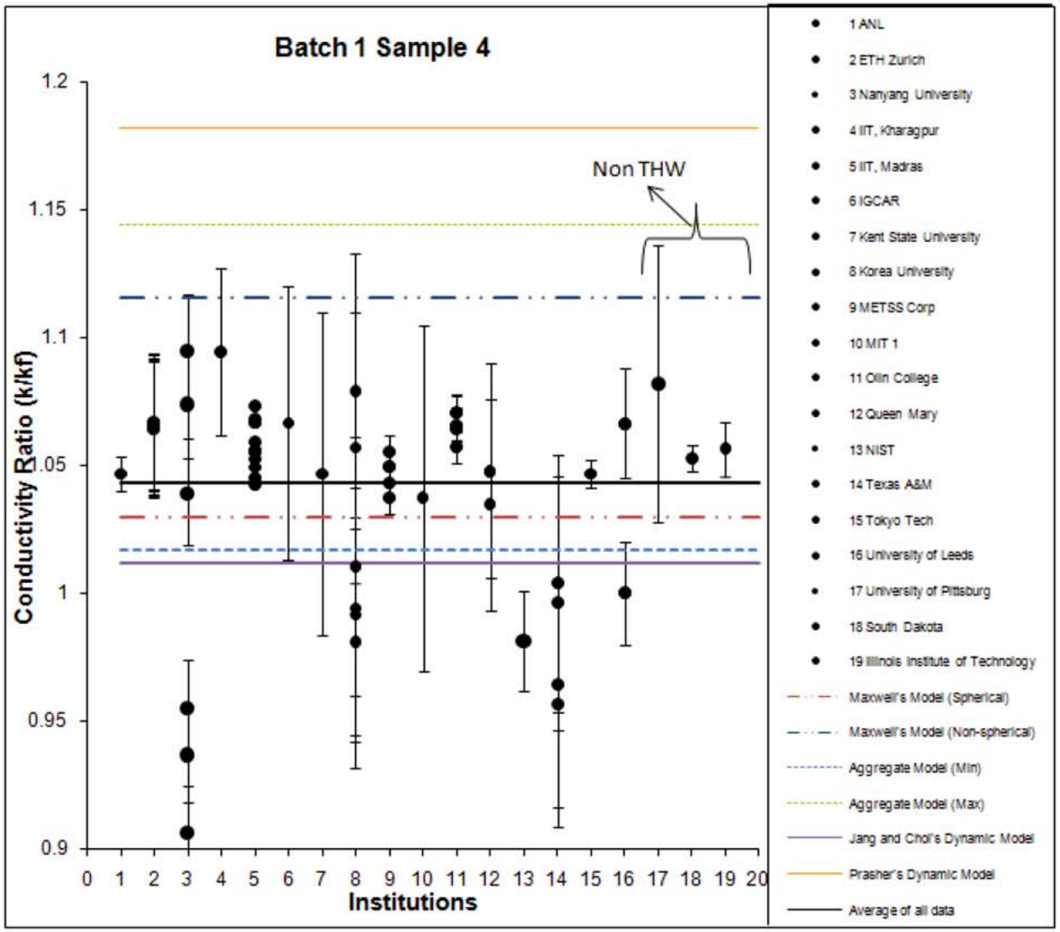
- The average enhancement observed is around 12.71%

- The best predictions are by the aggregate model (at intermediate aggregation states) and to some extent by the Maxwell's model (10%)

- Prasher's Dynamic models highly over predict the enhancement

BATCH I :- Sample 4 (1 vol% Alumina Nanorods in PAO)

Basefluid : PAO + Surfactant

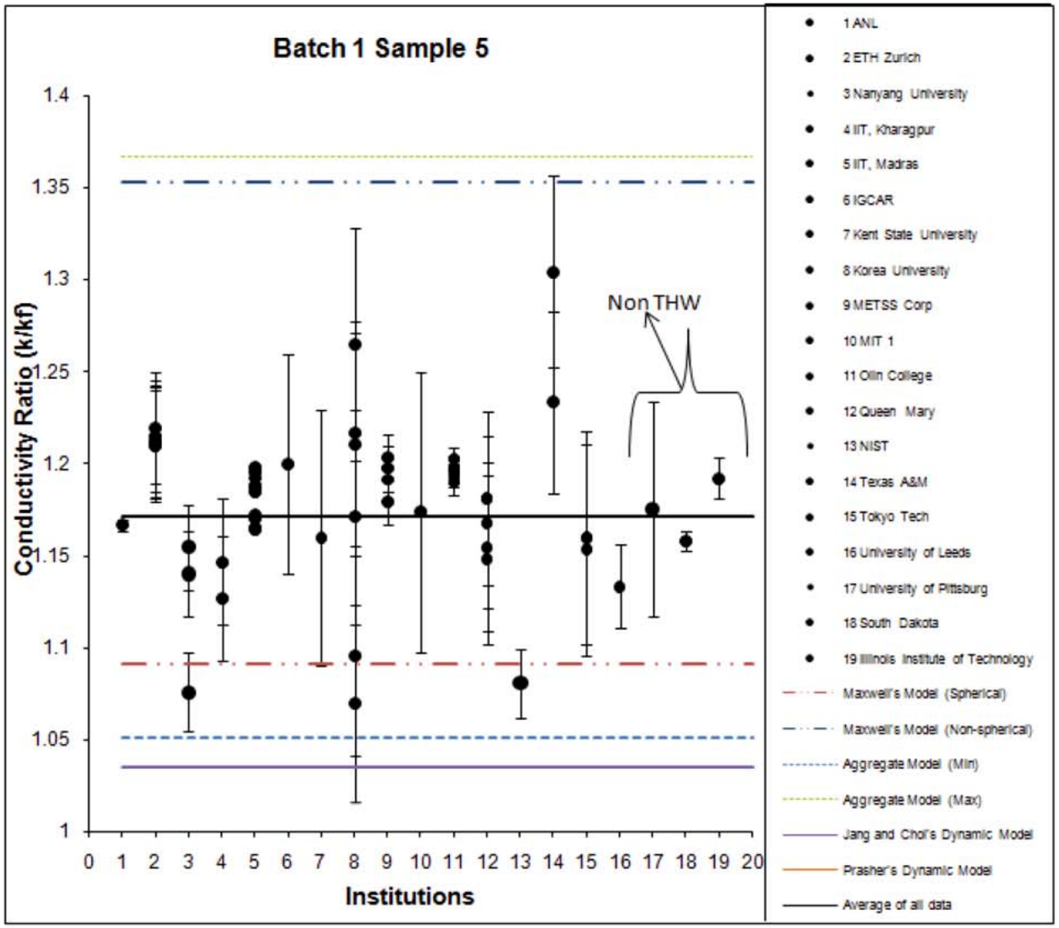


<u>Observations</u> :

- The reported enhancements are in the range of 2% to 8%
- The average enhancement observed is around 4.5%
- Prasher's aggregate model bounds most data.

BATCH I :- Sample 5 (3 vol% Alumina Nanorods in PAO)

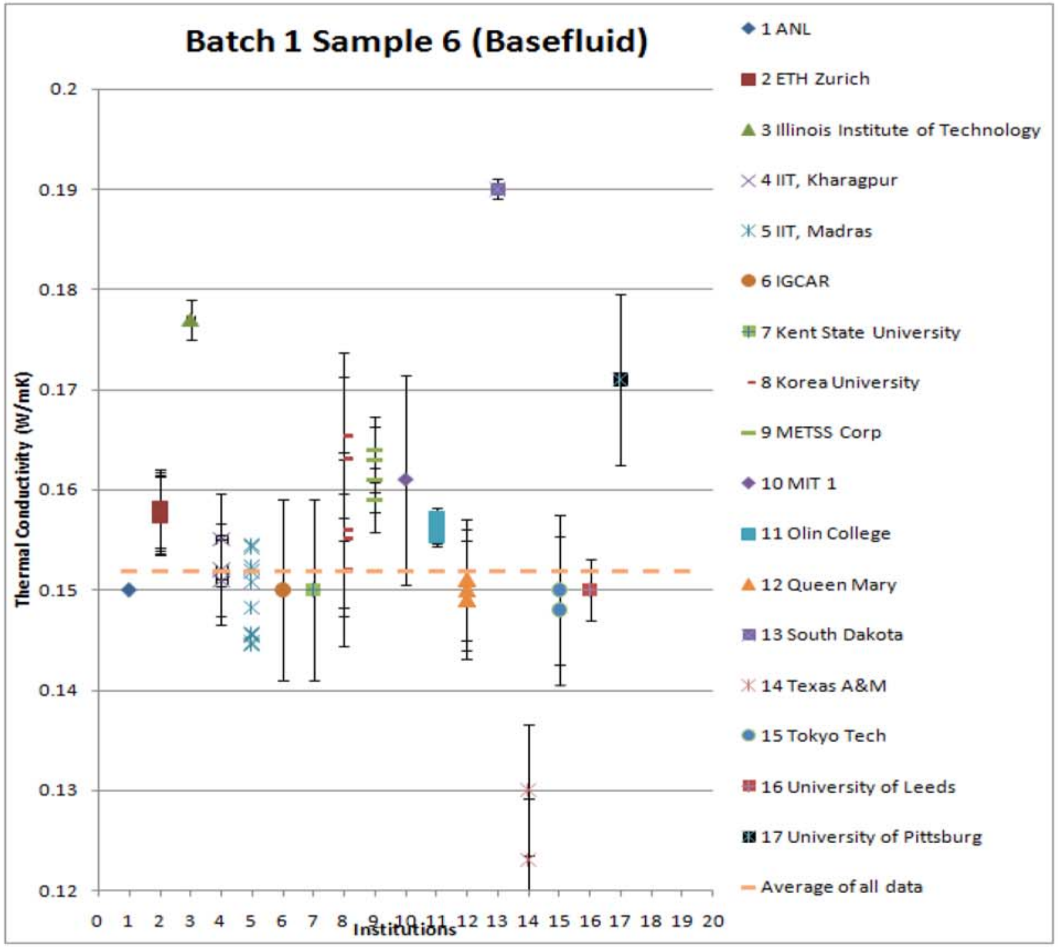
Basefluid : PAO + Surfactant



Observations :

- Most measured enhancement values are in the range of 12% to 22% with an average of around 17.5%
- Prasher's Aggregate Model bounds data
- We see a higher enhancement in case of nanorods wrt nanoparticles at the same concentration
- Maxwell's model for non-spherical nanoparticles seems to over-predict the data

BATCH I :- Sample 6 Basefluid(PAO + Surfactant)



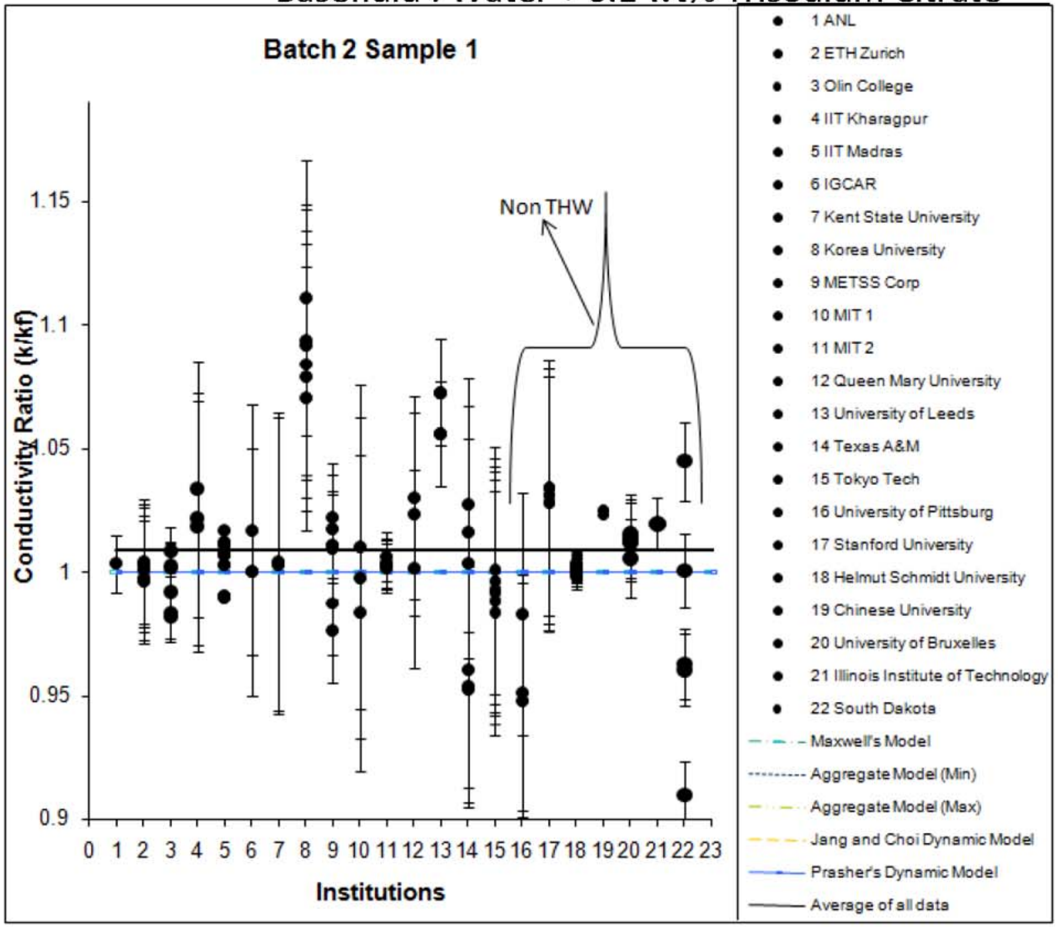
Observations

- Most data are within 10 % of the mean value

- The average measured thermal conductivity was around 0.152 W/m-K

BATCH II :- Sample 1 (0.001 vol% Gold Nanoparticles in Water)

Basefluid : Water + 0.1 wt% Trisodium Citrate

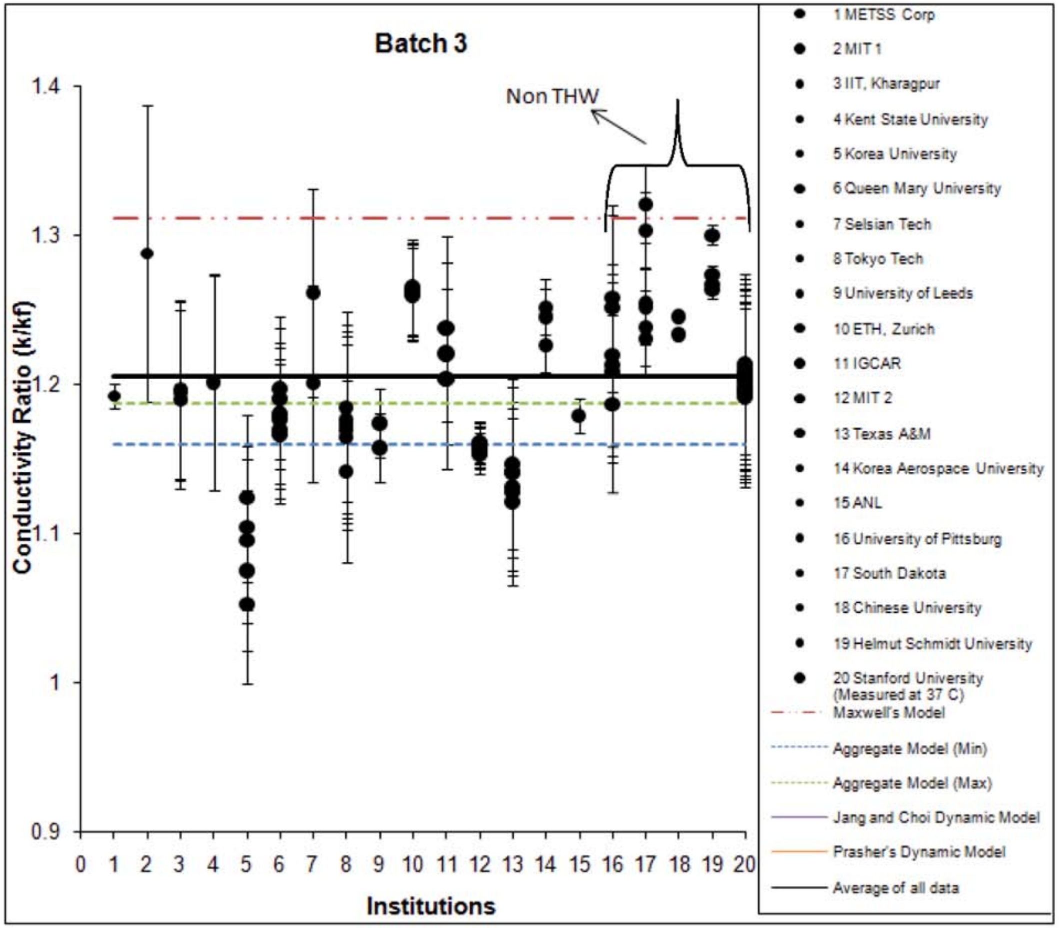


Observations :

- The enhancements reported are minute, consistently with the low nanoparticle concentration of this nanofluid
- In some cases a reduction in thermal conductivity wrt the base fluid is observed
- The average measured enhancement is around 0.9%
- All models predict no enhancement

BATCH III :- (50 wt% Silica in water)

Basefluid : Water + 0.2 wt% Sodium

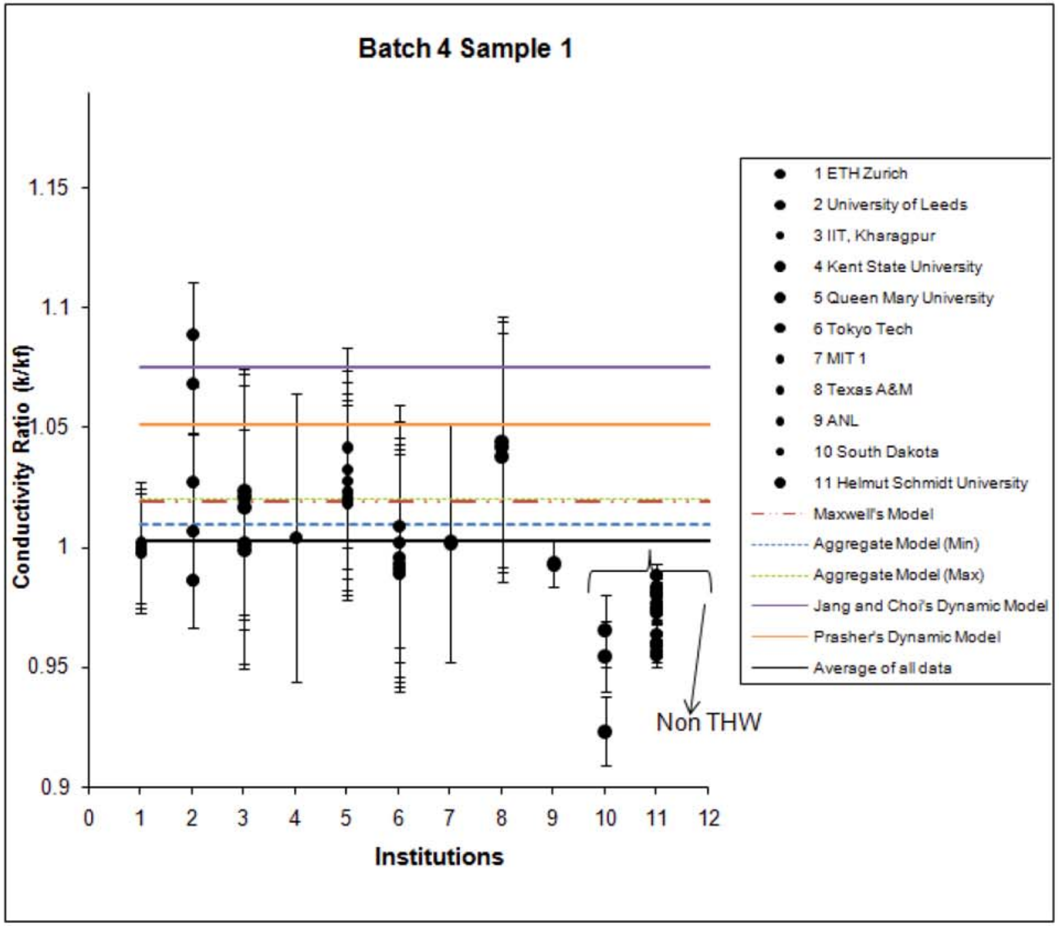


Observations :

- High enhancements of the order of 15% to 30% are observed owing to the high concentration of SiO_2 nanoparticles.
- The average enhancement observed was around 20.5 %
- Dynamic models greatly over-predict data. Values are off the chart
- Aggregate model bounds the data and Maxwell's model gives best predictions

BATCH IV :- Sample 1 (0.86 vol% Mn-Zn Ferrite in Water)

Basefluid : 75wt% Water + 25wt% Tetramethylammonium hydroxide

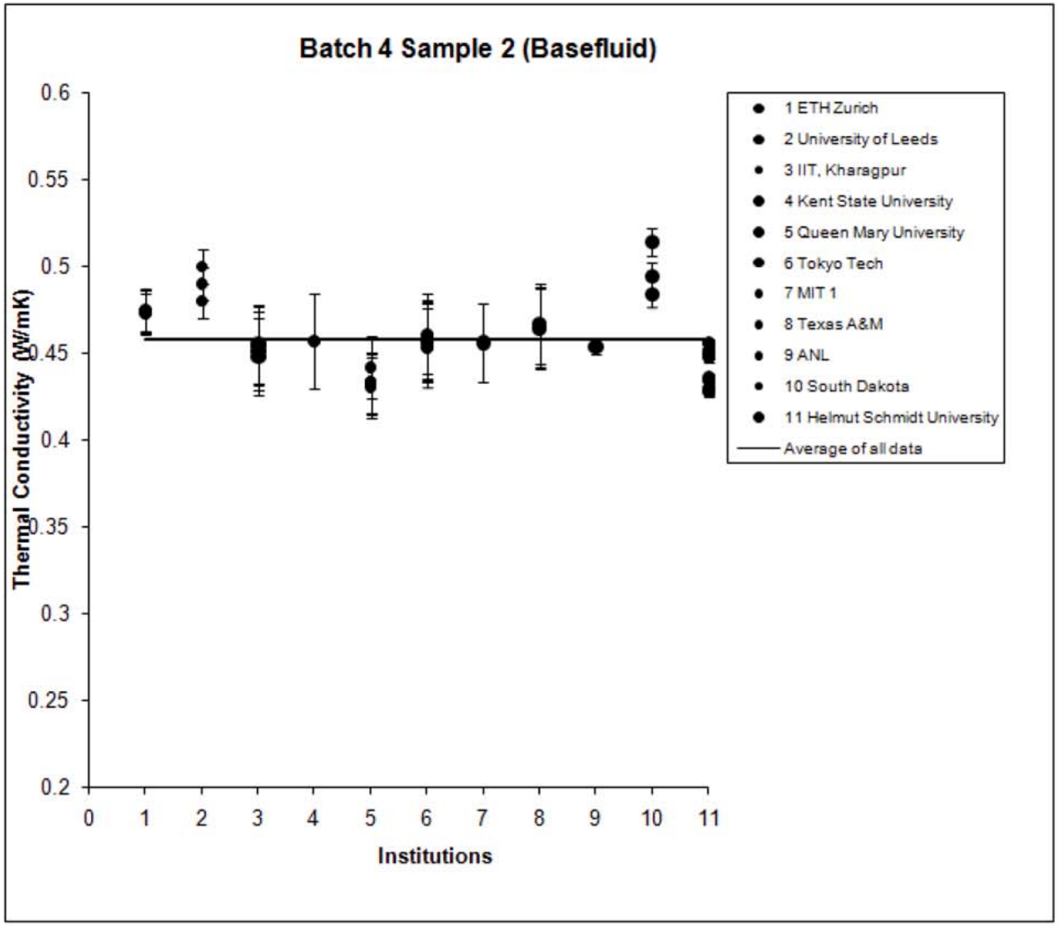


Observations :

- Most of the measurements show an enhancement ranging from 1% to 4%
- Some organizations measured a decrease in thermal conductivity
- On an average no enhancement is seen
- Prasher's Aggregate model is very close to the measured mean value (at low aggregation states)
- The dynamic models over-predict the enhancements

BATCH IV :- Sample 2 (Basefluid)

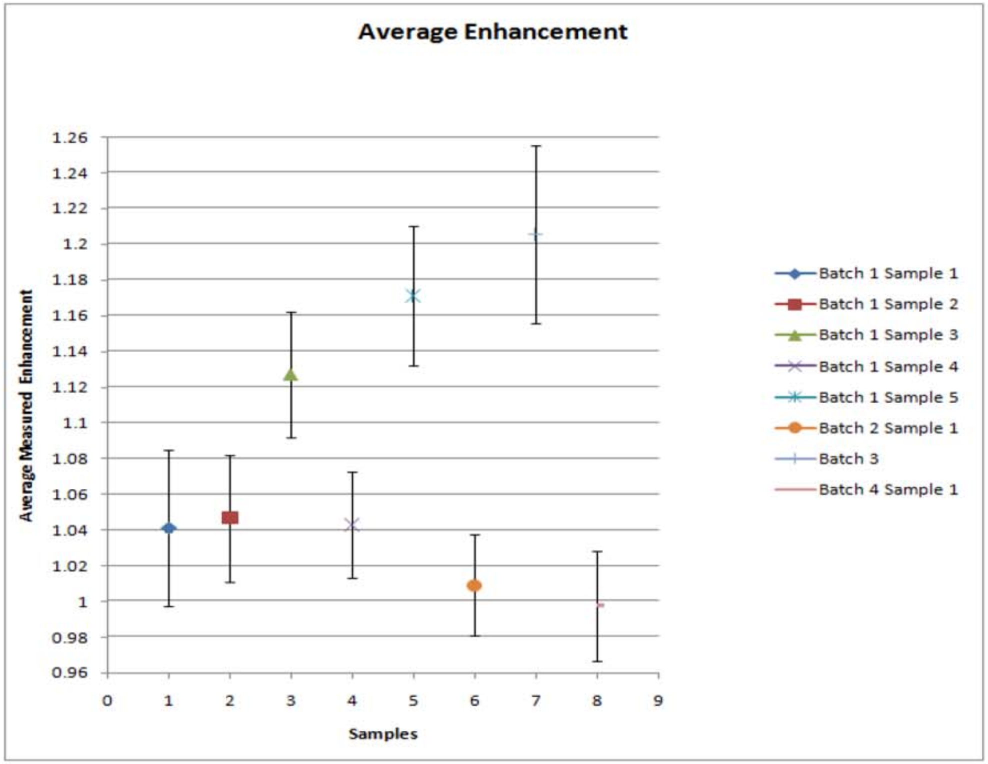
75wt% Water + 25wt% Tetramethylammonium hydroxide



Observations

- All organizations consistently report a decrease in the thermal conductivity (w.r.t water) owing to a high concentration of stabilizer
- Most data are within 10% of the mean value
- The average measured thermal conductivity was around 0.458 W/m-K

Average Enhancements Measured



Observations

Thermal conductivity is enhanced by using:

1)Elongated nanoparticles
2)Increasing concentration of nanoparticles

Static Theory: Effective Medium Approach

Overall Theory

(Particulate Composites)

Nan et al., 1997

thermal interface resistance

$$r_K \rightarrow 0$$

Maxwell, Hamilton – Crosser

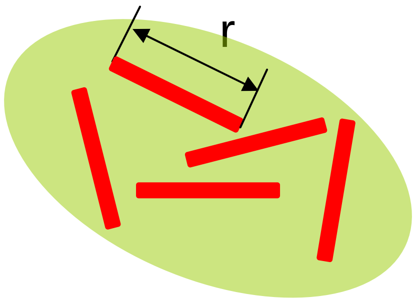
$$\frac{\lambda}{\lambda_{BF}} = \frac{\lambda_P + \lambda_{BF}(n-1) - \varepsilon(n-1)(\lambda_{BF} - \lambda_P)}{\lambda_P + \lambda_{BF}(n-1) + \varepsilon(\lambda_{BF} - \lambda_P)}$$

spheres....

(clusters)

$$\frac{\lambda}{\lambda_{BF}} \approx 3\varepsilon \rightarrow 5\varepsilon$$

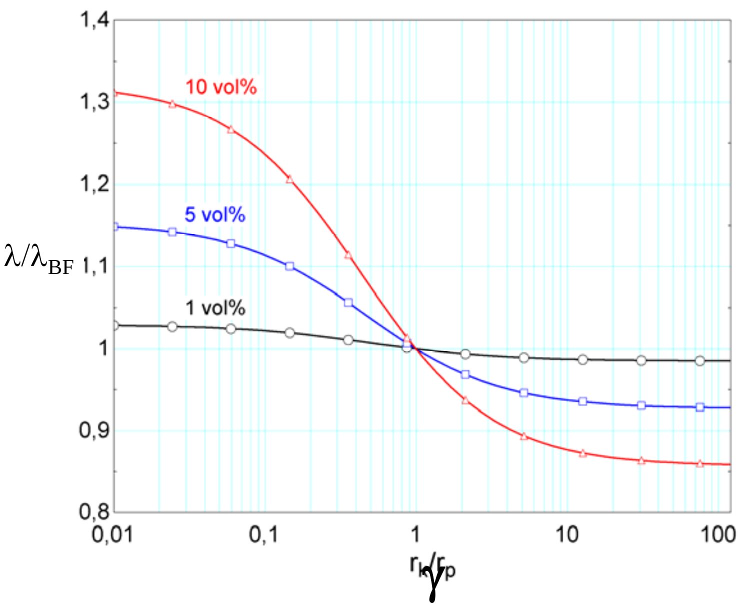
spheres:



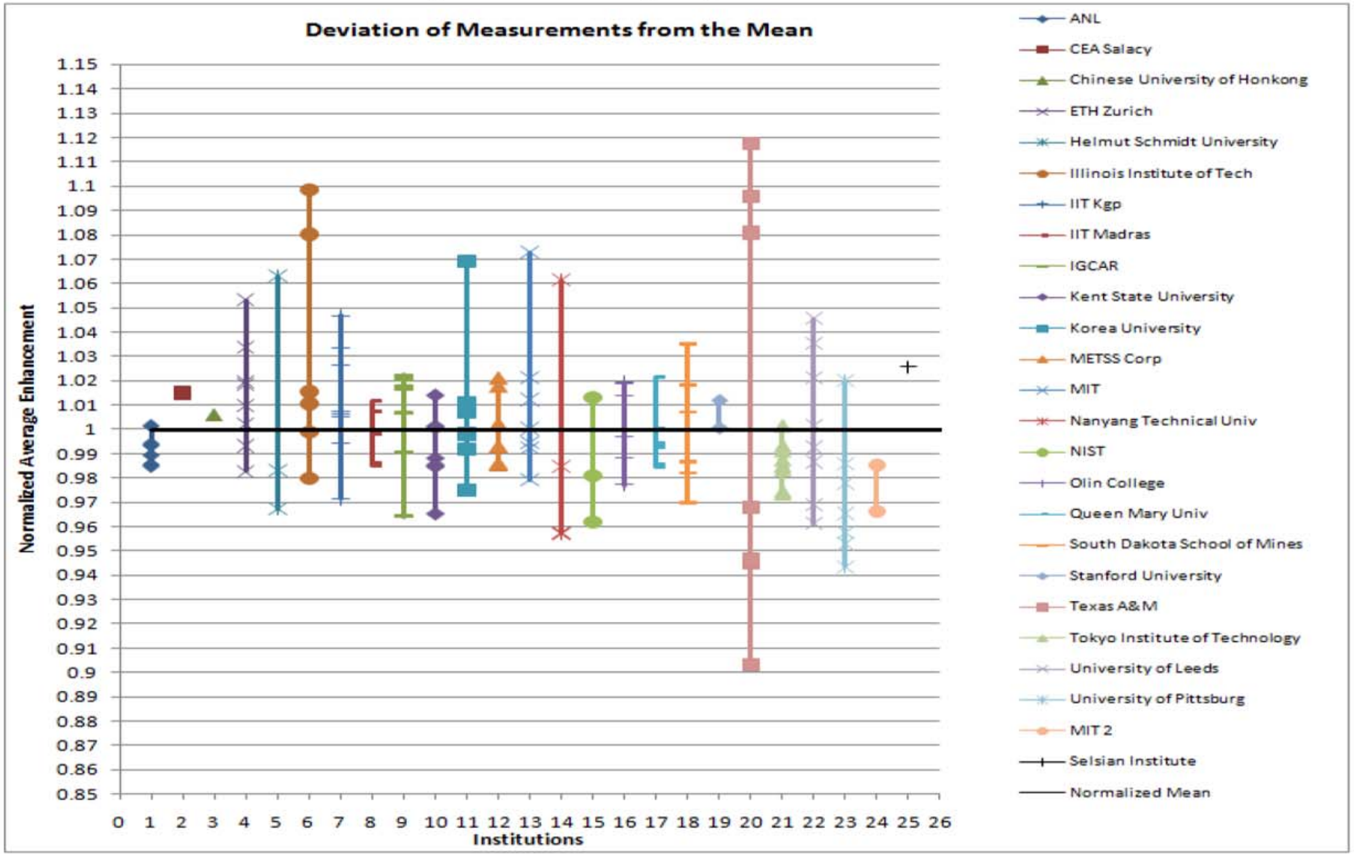
$$\frac{\lambda}{\lambda_{BF}} \approx 3\varepsilon \frac{\gamma - 1}{\gamma + 2} + 1$$

$\gamma = r_k / r$

$r_K = \lambda/G$ = Kapitza Radius

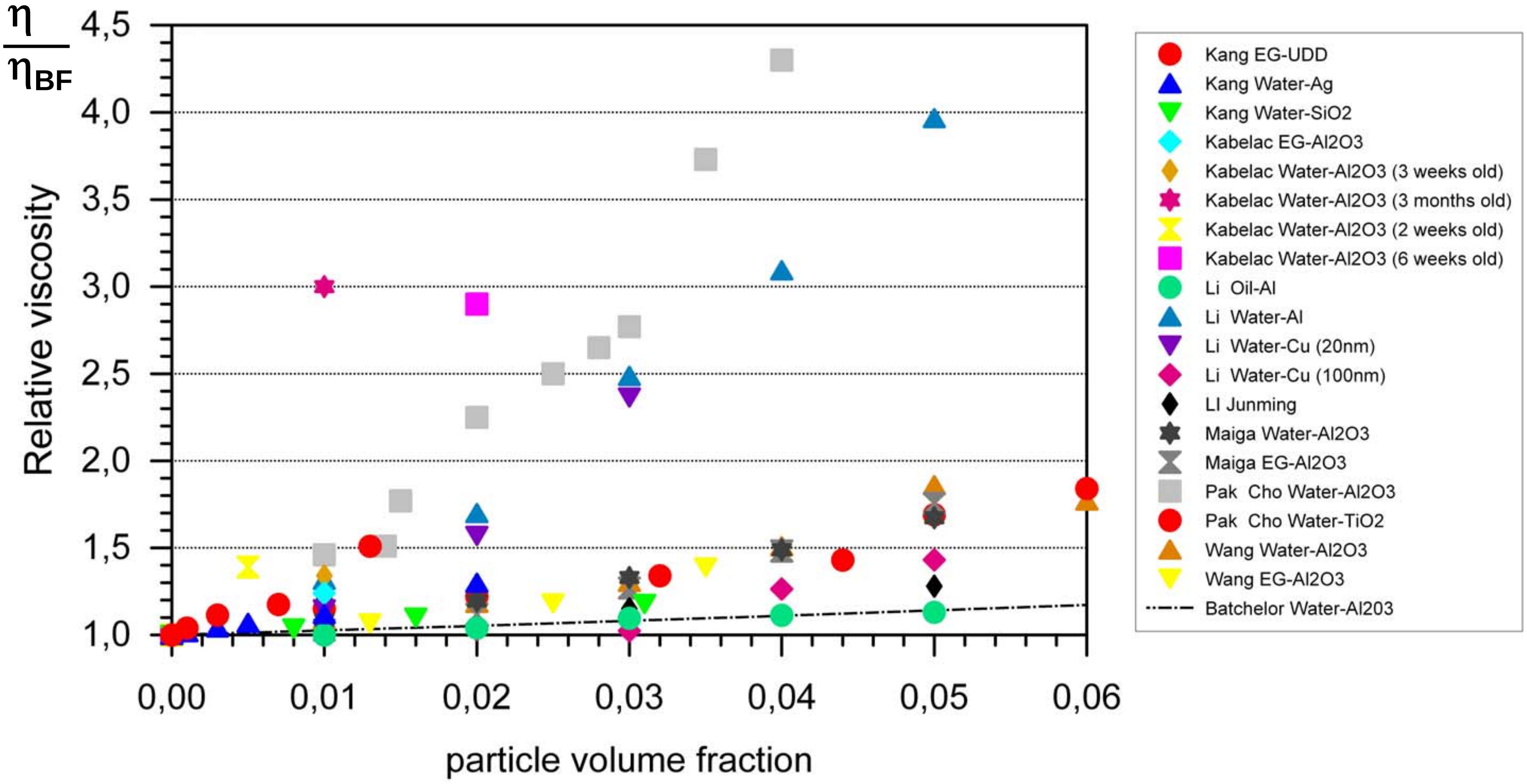


Analysis of any systematic errors



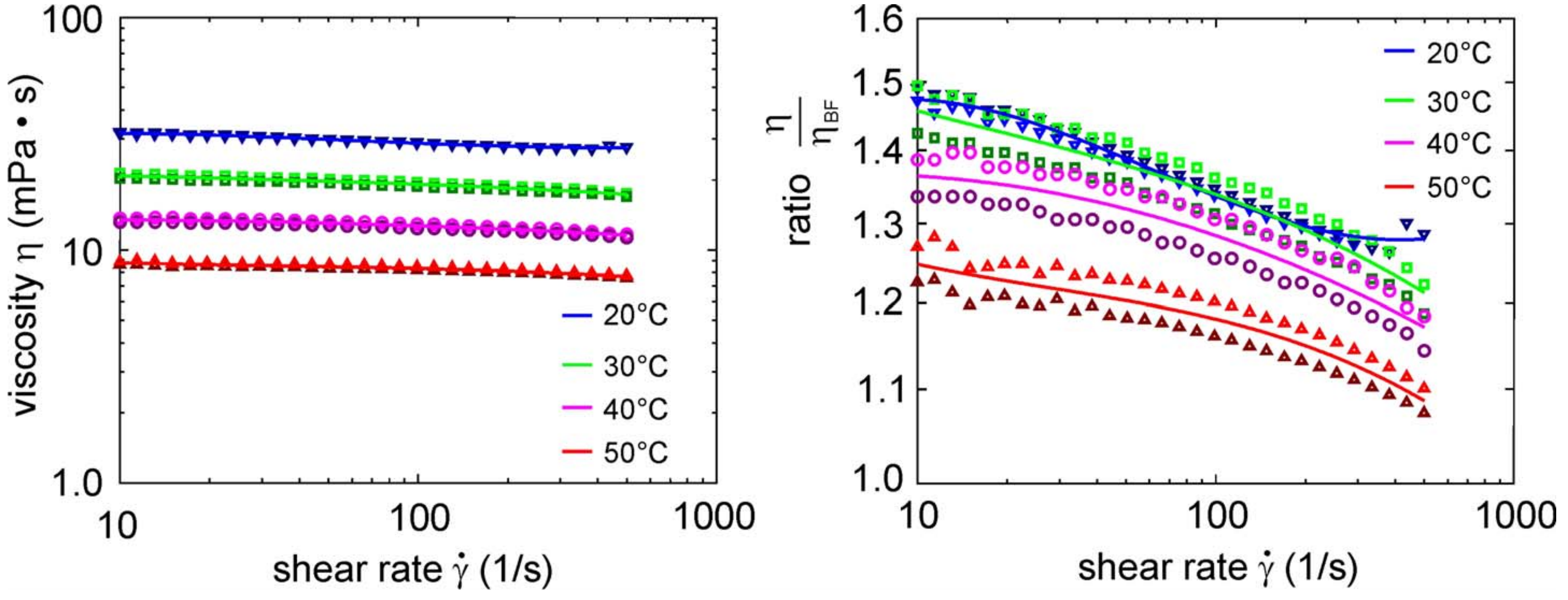
•Y-axis represents the ratio of mean value measured by an organization to the overall mean for a sample
•No participating organization reports measured values of thermal conductivity that are systematically above or below the average of all data.
•Also, no systematic differences are observed between THW and non-THW data

Viscosity data



Viscosity: Own Measurements

Al_2O_3 – EG **ε = I vol-%**



Zusammenfassung

Bei den untersuchten 8 Nanofluiden konnte von den insges. 34 Teilnehmern des Ringversuches **kein** anormales Verhalten der Wärmeleitfähigkeit festgestellt werden.

Die Daten zur Wärmeleitfähigkeit dieser Suspensionen werden innerhalb der Messunsicherheit durch die „effective medium“ Theorie (erweiterter Maxwell-Ansatz) wiedergegeben.

J. Buongiorno et al.
A benchmark study on the thermal conductivity of nanofluids.

Journal of Applied Physics **106**, 094312 (2009)

Validity of Fouriers Law (Transient)

diffusive:
(enough scattering events for excited heat carriers)

$$\dot{q} = -\lambda \bullet \text{grad } T \qquad \textbf{(Fourier)}$$

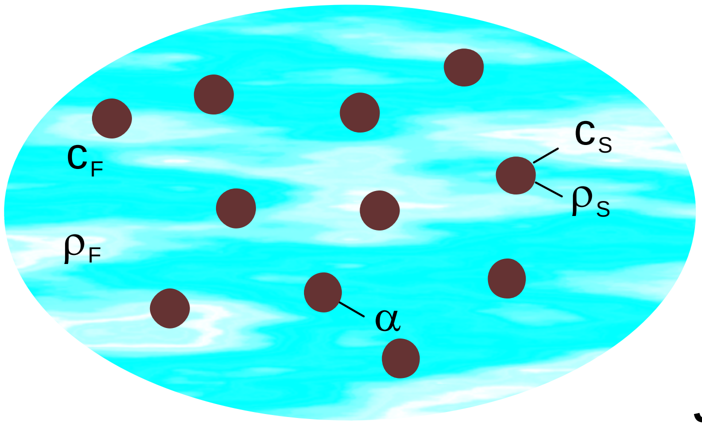
$$\tau \frac{\partial \dot{q}}{\partial t} + \dot{q} = -\lambda \bullet \text{grad } T \qquad \textbf{(hyperbolic)}$$

ballistic?
(phonon radiative transfer...)

non-homogeneous inner structure

$$\tau_q \frac{\partial \dot{q}}{\partial t} + \dot{q} = -\lambda \left[\text{grad } T + \tau_t \frac{\partial (\text{grad } T)}{\partial t} \right]$$

(dual phase lagging)



$$\tau_q = \frac{(c_S \bullet \rho_S)(c_F \bullet \rho_F)}{\alpha (c_S \bullet \rho_S + c_F \bullet \rho_F)} \;; \quad \tau_T = \frac{c_S \bullet \rho_S}{\alpha}$$

J. Vadasz **et al.** Int. J. Heat Mass Transfer 48 (2005)

Theory: Electro Thermal Approach I

(*J. Wang, G. Chen, Z. Zhang* Proc. Heat Transf. Conf. San Francisco July 2005)

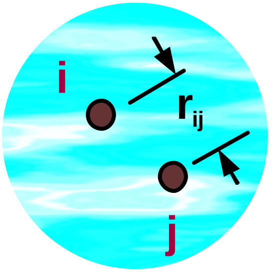
→ **in a stable nanofluid there <u>must</u> be a repulsive force between particles**

→ **Analogy: electroviscous effect on the rheology of colloid systems**

<u>basis:</u> **DLVO theory:** (Derjaguin – Landau – Verwey – Overbeek)

$$\Delta G = \frac{\varepsilon_o \zeta^2 d_p^2}{r_{ij}} \exp\left[-\kappa\,(r_{ij} - d_p)\right] - \underbrace{\frac{A_H}{12\,\pi\, r_{ij}^2}}_{\text{neglected}}$$

A_H: **Hamaker constant**



$$\frac{\partial(\Delta G)}{\partial r_{ij}}$$ **particle interaction force**

ζ: **Zeta – potential**

ε_o: **permittivity base fluid**

κ^{-1}: **Debye screening length**

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Background

nanofluids:

2000-2001 Sarit K. Das
2002-2006 DaimlerChrysler
2003-2007 F. Kuhnke Ph-D
2008 Anoop Kanjirakat
thermal conductivity
convective heat transfer

overall:

heat transfer (experimental)
fuel cells (thermal management)
radiation heat transfer
nonequilibrium thermodynamics

Abschätzung der Messunsicherheit

$$\lambda(T_m) = \frac{P}{A}\left(\frac{\Delta T}{d}\right)^{-1}$$

$$P = U^{el} I^{el} - \Sigma P_v + P_x$$

$$\Delta T = \Delta T_{ex} - \Delta T_m$$

P_{v1}	heater → guarded ring	0.1 %
P_{v2}	heater → top plate	0.2 %
P_{vs}	heat flow spacer	0.1 %
P_{vr}	radiation	0.15%
P_x	temperature jump interface	0.01 %