



# A thermodynamic model of nanofluid viscosity based on a generalized Maxwell-type constitutive equation

Georgy Lebon, Hatim Machrafi\*

*Thermodynamics of Irreversible Processes, Liège University, Allée du 6-Août, 19, 4000 Liège, Belgium*

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## ABSTRACT

An original study of nanofluid viscosity is proposed. The carrier fluid is assumed Newtonian but the two-phase nanofluid displays properties abiding by a generalized Maxwell constitutive law. Our approach is based on an extension of Einstein's model describing suspensions of solid particles in fluids by the introduction of the following elements: presence of a layer around the nanoparticles and a thermodynamic description of the role of size effects. The theoretical formalism is applied to liquid argon with lithium nanoparticles and to alumina nanoparticles in water. Good agreement with experimental data and molecular dynamics simulation is observed.

## 1. Introduction

Nanofluids are binary mixtures consisting of nanoparticles dispersed in a host fluid. These systems have met an increasing interest in several industrial applications, like in biotechnology, nanotechnology, electromechanical systems, they have proved to be relevant in the developments of new drugs, paints, lubricants among others. It is well known that the presence of nanoparticles influences considerably the thermomechanical properties of the basic fluid like, in particular, thermal conductivity and viscosity.

In this paper, focus is put on the role of nanoparticles on the shear viscosity of the system, an impressive lot of works (e.g. [1–10]) have been published on the subject. Viscosity depends essentially on the temperature, the nature of the particles and the fluid, the volume fraction of particles and their size. Subsequently, the analysis is focused on the role of volume fraction and particles size, the temperature is assumed uniform.

It has been observed that the viscosity of nanofluids is much larger than that of the host fluid. Several theoretical and ad hoc expressions for the viscosity  $\eta$  in terms of the particle's volume fractions  $\varphi$  have been proposed, among which the celebrated Einstein formula [11]

$$\eta = \eta_f(1 + 2.5\varphi), \quad (1)$$

with  $\eta_f$  denoting the viscosity of the host fluid, this expression is valid for dilute mixtures

( $\varphi < 0.05$ ) with spherical particles. Other more sophisticated ad hoc relations have been formulated like a quadratic dependence in the viscosity [12]

$$\eta = \eta_f(1 + a_1\varphi + a_2\varphi^2), \quad (2)$$

wherein  $a_1$  and  $a_2$  are parameters taking different values according to the nature of the nanofluid. Other models like those of Chen et al. [5] and Mahbubul et al. [8] have also been exploited.

The dependence of viscosity on the size of nanoparticles has been a subject of debate. For some authors [13,14], viscosity increases with increasing dimensions of nanoparticles while others [15–17] assert that viscosity diminishes with increasing size. For sufficiently large particles, the dependence with respect to size becomes negligible. It is however worth to mention that no valuable theoretical considerations, outside molecular dynamic simulations [18] are able to explain such behaviors.

The gap will be filled in the forthcoming: a formalism based on Extended Irreversible Thermodynamics (EIT) [19–21] is presented wherein the nanofluid is viewed as a generalized Maxwell model. In addition, our analysis will emphasize the role of interfacial layers surrounding the solid particle, acting as a possible mechanism for momentum transfer. This concept was introduced by Choi [22] and exploited by Yu and Choi [23] and Xie et al. [24] to interpret the enhancement of thermal conductivity in nanofluids.

The working hypotheses of our model are the following:

- the dispersed particles are solid spheres of radius  $r$ ,
- the host fluid is homogeneous, incompressible, isotropic and Newtonian,
- the particles are homogeneously distributed in the fluid,
- no formation of aggregates,
- the effects of the Brownian motion of nanoparticles is neglected,

\* Corresponding author.

E-mail address: [h.machrafi@ulg.ac.be](mailto:h.machrafi@ulg.ac.be) (H. Machrafi).

- thermal effects are not taken into account and temperature is uniform.

The paper is organized as follows. In Section 2, the main ingredients underlying EIT are recalled and the evolution equations governing the behavior of the state equations, essentially the internal energy and the viscous pressure tensor, are formulated. In Section 3, an expression of the effective shear viscosity of nanofluids in presence of spherical rigid nanoparticles is derived. In Section 4, the model is applied to lithium particles dispersed in Argon and to aluminate particles in water, respectively. Conclusions are drawn in Section 5.

## 2. Extended Irreversible Thermodynamics

The description of systems at subscales, such as nanoparticles, requires to go beyond the classical theory of irreversible processes [25]. A good candidate for treating these classes of problems is Extended Irreversible Thermodynamics (EIT). The principal idea behind EIT is to elevate the fluxes, as the fluxes of mass, energy and momentum to the status of independent variables at the same level as the classical conserved variables like mass, energy or momentum.

As a case-study, let us consider the flow of a viscous incompressible fluid at uniform temperature. The generalization to more complicated systems as fluids mixtures [26], polymer solutions [27], suspensions [28], porous media [29] and others have been dealt with in detail in numerous publications and books. In the case of an incompressible fluid flow, the only relevant conserved variable is the specific internal energy  $e$  (per unit mass) whereas the flux variable is the viscous pressure tensor  $\mathbf{P}$ , it is a second order symmetric traceless tensor, in contrast with  $e$ , it is not a conserved quantity. The corner stone of EIT is to assume the existence of a specific non-equilibrium entropy function  $s$  depending on both  $e$  and  $\mathbf{P}$  so that  $s = s(e, \mathbf{P})$  or, in terms of time derivatives,

$$d_t s = \frac{\partial s}{\partial e} d_t e + \frac{\partial s}{\partial \mathbf{P}} \otimes d_t \mathbf{P}, \quad (3)$$

wherein  $\otimes$  stands for the inner product of the corresponding tensors, the symbol  $d_t$  designates the time derivative which is indifferently the material or the partial time derivative as the system is, respectively, in motion or at rest. It is assumed that  $s$  is a concave function of the variables and that it obeys a general time-evolution equation which can be written in the form

$$\sigma^s = \rho d_t s + \nabla \cdot \mathbf{J}^s \geq 0, \quad (4)$$

with  $\sigma^s$  its rate of production per unit volume (in short, the entropy production) to be positive definite in order to satisfy the second principle of thermodynamics,  $\rho$  is the mass density of the nanofluid and the vector  $\mathbf{J}^s$  is the entropy flux, the dot between  $\nabla$  and  $\mathbf{J}^s$  denotes the scalar product. Let us define the temperature by  $T^{-1} = \partial s / \partial e$  assumed to be independent of the dissipative flux  $\mathbf{P}$ , next, we select the constitutive equation for  $\partial \eta / \partial \mathbf{P}$  as assumed to be given by the linear relation  $\partial s / \partial \mathbf{P} = -(\gamma_1 / \rho) \mathbf{P}$ , where  $\gamma_1$  is a material coefficient depending generally on  $\rho$  and  $T$ ,  $\gamma$  is positive definite in order to meet the property that  $s$  is maximum at local equilibrium, the minus sign in front of  $\gamma(T)\mathbf{P}$  has been introduced for convenience. Under these conditions, expression (3), can be written as

$$\rho d_t s = -T^{-1} \mathbf{P} \otimes \mathbf{D} - \gamma_1 \mathbf{P} \otimes d_t \mathbf{P} \quad (5)$$

after use has been made of the energy conservation law

$$\rho d_t e = -\mathbf{P} \otimes \mathbf{D}, \quad (6)$$

wherein  $\mathbf{D}$  is the symmetric traceless velocity gradient tensor. In absence of heat flux, the entropy flux is zero and the entropy production takes the form

$$\sigma^s = -\mathbf{P} \otimes (T^{-1} \mathbf{D} + \gamma_1 d_t \mathbf{P}) \geq 0. \quad (7)$$

It is a bilinear relation in the flux  $\mathbf{P}$  and the quantity represented by the two terms between the parentheses that is usually called the thermodynamic force  $\mathbf{X}$ . The simplest way to guarantee the positiveness of the entropy production  $\sigma^s$  is to assume a linear flux-force relation of the form  $\mathbf{X} = -\mu_1 \mathbf{P}$  with  $\mu_1$  a positive phenomenological coefficient, this procedure leads to the well-known Maxwell model

$$\tau_1 d_t \mathbf{P} = -\mathbf{P} - 2\eta \mathbf{D}, \quad (8)$$

after one has put  $\gamma_1 / \mu_1 = \tau_1$  (relaxation time) and  $1/T\mu_1 = 2\eta$  (shear viscosity) and wherein  $\tau_1$  and  $\eta$  are positive quantities as  $\mu_1$  and  $\gamma_1$  have been proven to be positive coefficients. Letting  $\tau_1$  vanish, one finds back Newton's law  $\mathbf{P} = -2\eta \mathbf{D}$ . Although Maxwell's relation is useful at short time scales (high frequencies), it is not satisfactory with the purpose to describe short length scales wherein non-localities play a preponderant role, for instance fluids in presence of nanoparticles.

In more complex materials like in nanofluids, fluxes of higher order should be introduced as extra states variables. Non-local effects, which are important in presence of nanoparticles, are elegantly accounted for, in the framework of EIT, by appealing to a hierarchy of fluxes  $\mathbf{P}^{(2)}$  ( $\equiv \mathbf{P}$ ),  $\mathbf{P}^{(3)}$ , ...  $\mathbf{P}^{(N)}$  where the second order tensor  $\mathbf{P}^{(2)}$  is identified with the usual viscous pressure tensor  $\mathbf{P}$ ,  $\mathbf{P}^{(3)}$  (a tensor of rank three) is the flux of the pressure tensor, ...etc. Here for simplicity, we limit our developments to the use of  $\mathbf{P}$  and  $\mathbf{P}^{(3)}$  as flux state variables but there will be no difficulty to include higher order tensors as  $\mathbf{P}^{(4)}$ , ...  $\mathbf{P}^{(N)}$  as done previously in the problem of non-local heat conduction wherein an infinite number of extra fluxes have been introduced [30–32]. From the kinetic theory point of view,  $\mathbf{P}^{(2)}$  and  $\mathbf{P}^{(3)}$  represent the second and third order moments of the velocity distribution. Written in Cartesian coordinates and designating by  $f$  the distribution function, the fluxes  $\mathbf{P}^{(2)}$  and  $\mathbf{P}^{(3)}$  read as

$$P_{ij}^{(2)} \equiv P_{ij} = \int C_i C_j f dc, \quad P_{ijk}^{(3)} = \int C_i C_j C_k f dc, \quad (9)$$

with  $\mathbf{C} = \mathbf{c} - \mathbf{v}$  the relative velocity of particles with respect to their mean velocity  $\mathbf{v}$ .

Up to the third-order moment approximation, which is sufficient for the present purpose, the Gibbs equation generalizing expression (3) takes the form

$$d_t s(e, \mathbf{P}, \mathbf{P}^{(3)}, \dots) = T^{-1} d_t e - (\gamma_1 / \rho) \mathbf{P} \otimes d_t \mathbf{P} - (\gamma_2 / \rho) \mathbf{P}^{(3)} \otimes d_t \mathbf{P}^{(3)}, \quad (10)$$

while the entropy flux is no longer equal to zero but is given by

$$\mathbf{J}^s = \beta \mathbf{P}^{(3)} \otimes \mathbf{P}, \quad (11)$$

with  $\beta$ , a phenomenological coefficient allowed to depend on  $e$  and the volume fraction of the particles but not on the flux variables. The entropy production (4) is obtained by substitution of  $d_t s$  and  $\mathbf{J}^s$  from (10) and (11) respectively and elimination of  $d_t e$  via the energy balance (4), the result is

$$\sigma^s = -\mathbf{P} \otimes (T^{-1} \mathbf{D} + \gamma_1 d_t \mathbf{P} - \beta \nabla \cdot \mathbf{P}^{(3)}) - \mathbf{P}^{(3)} \otimes (\gamma_2 d_t \mathbf{P}^{(3)} - \beta \nabla \mathbf{P}) \geq 0. \quad (12)$$

The above bilinear expression in fluxes and forces (the quantities between parentheses) suggests the following linear flux-force equations

$$\gamma_1 d_t \mathbf{P} + \mu_1 \mathbf{P} = -T^{-1} \mathbf{D} + \beta \nabla \cdot \mathbf{P}^{(3)}, \quad (13)$$

$$\gamma_2 d_t \mathbf{P}^{(3)} + \mu_2 \mathbf{P}^{(3)} = \beta \nabla \mathbf{P}, \quad (14)$$

wherein  $\gamma_n \beta$  and  $\mu_n$  ( $n = 1, 2$ ) are phenomenological coefficients allowed to depend in particular on the temperature and/or the relative volume fraction of the constituents. Relations (13) and (14) can also be viewed as time evolution equations for the fluxes  $\mathbf{P}$  and  $\mathbf{P}^{(3)}$ . Making use of (13) and (14), expression (12) of the entropy production reads as

$$\sigma^s = \mu_1 \mathbf{P} \otimes \mathbf{P} + \mu_2 \mathbf{P}^{(3)} \otimes \mathbf{P}^{(3)} \geq 0, \quad (15)$$

from which follows that  $\mu_1 \geq 0$  and  $\mu_2 \geq 0$  to satisfy the positiveness of

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