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Abstract. *The use of distribution entropy is discussed in this work as a tool to quantify diffusional processes in the context of thermophoresis. We use an in-house code to simulate the dynamical behavior of nanoparticles immersed in a Newtonian fluid subjected to a temperature gradient. The deterministic motion of the particles from hotter to colder regions due to the isotropic nature of Brownian motion is investigated in terms of statistical analysis. First we bring a historical discussion regarding the terminology of the word "entropy". Then we show that some classical statistical tools used in the context of Langevin dynamics fail to capture the effect of the temperature field from a statistical perspective. Then we propose the use of distribution entropy as a tool to quantify these effects. The methodology used for this purpose is discussed in details and the results are physically interpreted.*

Keywords: *thermophoresis, colloids, Brownian movement, distribution entropy, diffusion*

1. INTRODUCTION

Information entropy is a concept that defines the average information produced by a stochastic process. A diffusion like process is the result of the stochastic behavior of particles and molecules. Here, the information entropy is presented as a tool for the analysis of the distribution of particles during diffusion, under the effect of "phoresis" (that is, particle migration) and under the effect of some external force that disturbs the concentrations of the components of a mixture.

The information entropy can be used as a measurement for stratification and evolution of the process of diffusion. The relationship between the entropy of statistical mechanics and information theory is very debatable. There are many articles and discussions among which we can cite Ben-Naim (2008), Maroney (2009) and Thims (2012).

The entropy used by Shannon when he studied compression of information, is rather a mathematical concept than a generalization of Clausius entropy. A popular version of why Shannon called it entropy published by Holzinger (2010) says that John Neumann suggested Shannon this name for his logarithm function because of the similarity with the same function used in statistical mechanics to define Boltzmann entropy.

Most critics about the use of "information entropy" in relationship with thermodynamics is that it has no concept of energy and temperature in it. The pioneer of application of information theory to statistical mechanics was (Jaynes, 1957). His approach was criticized by several authors (Friedman and Shimony, 1971) , (Dias and Shimony, 1981). Later, (Jaynes, 1983) criticizes the use of the name *entropy* in information theory and wrote that it and the experimental thermodynamic entropy should be distinguished. Jaynes and Thims (2012) also criticizes the look for connection between thermodynamic and information entropies just because both use logarithm function of the form $\sum p_i \log P_i$ (mathematical isomorphism), as James exemplifies, the function $\cos\theta$ appears in many fields of physics and engineering with no profound connection between them. Ben-Naim (2008) arguments that indeed $\cos\theta$ appears in different fields like the swinging of a pendulum and electromagnetic wave, however it $\cos\theta$ indicates periodicity. As well the appearance of $\sum p_i \log P_i$ in many fields from linguistic to thermodynamics indicates measure of information.

The entropy in classical thermodynamics comes from Clausius inequality as consequence of the second law of thermodynamics, which arouse from Carnot Cycle. The analysis of the Carnot Cycle that led to the definition of entropy is the derivation of the Carnot-Clapeyron equation. Back from the caloric theory, the heat that must be added to change the temperature and volume of a body is (Ivory, 1827) equal to some heat absorbed by dilatation (called latent heat in that context) plus the internal heat (that depends on the mass and specific heat).

Clausius was interested in the internal heat as a state function, that is the energy absorbed that didn't become work. The difference of the absorbed energy and external work, $W = pdV$, would be equal to the increase in the latent heat by the change in volume. The doctrine of latent and specific heat was written as an amount of heat dQ equals the change in latent heat and specific heat with an amount of change in volume dV and temperature dT respectively. But Clausius criticized equating this way (Lavenda, 2009), because an infinitesimal amount of absorbed heat cannot be a function of temperature and volume, so equating dQ to the change in internal energy and latent heat yields a false derivative. So a integrability condition that says that the variation of the pressure with temperature is the difference of the variation of latent heat with the temperature and the variation of the heat capacity with volume.

By integrating the equation of heat over an enclosed area in the V, T-plane of the Carnot cycle gives off the work output. And dividing it by the latent heat yields the ratio that defines the Carnot efficiency $\eta_C = (T_h - T_c)/T_h$, where T_h and T_c are the temperatures of the hot and cold reservoir.

These last two paragraphs describe the ideas of the interconversion of heat and work and the maximal efficiency of a thermal engine. In a reversible cycle, all the work produced would be the difference between the heat taken from the hot reservoir and given to the cold reservoir. The entropy production is what makes the integration over a cycle $\oint \frac{dQ}{T}$ less than zero. An interpretation of the line integral $\oint \frac{dQ}{T}$ is a ratio of energy gained from the hot reservoir and average kinetic energy of the molecules in there dQ_h/T_h, less the ratio of energy transfered to the cold reservoir and the average kinetic energy of molecules in there dQ_c/T_c. If the ratio dQ_c/T_c is higher so that the integral is less than zero, some potential energy that could have become work was irreversible lost in the form of heat. The thermodynamic definition of entropy then comes from the inequality eq. 1.

$$\delta S = \oint \frac{dQ}{T} = \frac{Q_h}{T_h} - \frac{Q_c}{T_c} \leq 0 \tag{1}$$

Until now, there was no mention to atoms, probabilities and statistical mechanics. The caloric theory was falling apart during the time of Carnot and Clausius. Alto, the atomistic theory was not widely accepted. The papers about the second law of thermodynamics by Clausius with the study of dQ/T was published in 1854, but the concept of entropy only came in 1863. The statistical mechanics evaluation of entropy was developed in 1877. (Ben-Naim, 2008) suggests that if the concept of temperature as a measure of average kinetic energy of the molecules came earlier in the dawn of thermodynamics, events could have unfold differently. It would be more natural to identify temperature in units of energy when arriving at the identity

$$\frac{m \left\langle v^2 \right\rangle}{2} = \frac{3}{2}T, \tag{2}$$

instead of $3k_BT/2$ in the right hand side. It would have no effect on the expression of Carnot's engine or in the definition of Clausius entropy eq. 1. The difference would be the units of entropy, that would become dimensionless, what could make the acceptance of Boltzmann's statistical definition of entropy easier as well as the concept of entropy as some measure of information. It is important to note that even when saying that Shannon's and thermodynamic entropies are conceptually and formally the same measure of information, they are in different branches of science (communication and thermodynamics) with differences in orders of magnitude and serve for different finalities, as the compression of a gas in the engine of a car and of ZIP files are conceptually very different things.

The understanding of entropy as disorder is also problematic, and yet this explanation is commonly found in literature. "Disorder" is subjective, for example, in the fig. 1, which is more ordered? When (Ben-Naim, 2008) asked people that never heard about entropy the answers are diverse. "Disorder" isn't also quite extensive, why would two equally messy rooms be more messy than each room individually? That is similar to the assimilation of two gases fig. 2.

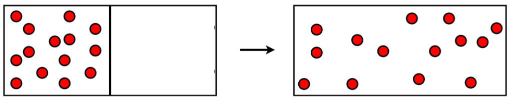


Figure 1: Expansion of gas.

Thermoforesis, growth of crystals and packing are phenomena that leads to spontaneous spatial ordering. The issue around situations where the order grows as entropy increases is presented in the article *Understanding shape entropy dense packing* by (G van Anders and Glotzer, 2012). Although the article doesn't quantify the entropy shape, it comments about shape entropy and how it leads to high packing of particles driven by directional entropic forces. The entropic force is the result of the entire thermodynamical system tending to increase entropy. The article also makes reference to the shape entropy role in the colloids regarding the (Asakura and Oosawa, 1954) model. In other context, Roux (1996) used shape entropy in an attempt to predict the settling velocity of non-spherical particles.

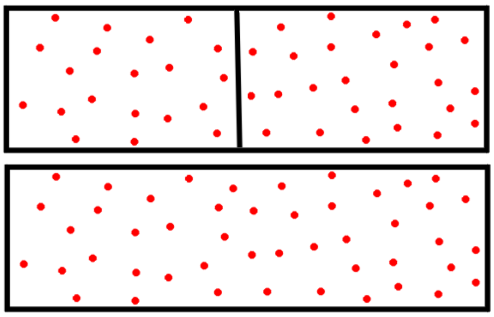


Figure 2: Assimilation of two gases of the same kind.

The information entropy is computed in this work using the probabilities of particles being in certain regions of our calculation domain in the context of thermophoresis. For that, we take the concentration distribution of the particles and use it as probability density functions to calculate continuous entropy. We show that a systematic increase of this relative continuous information entropy is associated with an increase in the temperature gradient.

2. CASE OF STUDY: THERMOPHORESIS

Thermophoresis is the migration of particles inside a fluid under a temperature gradient to colder regions. This phenomenon is illustrated in figure (3).

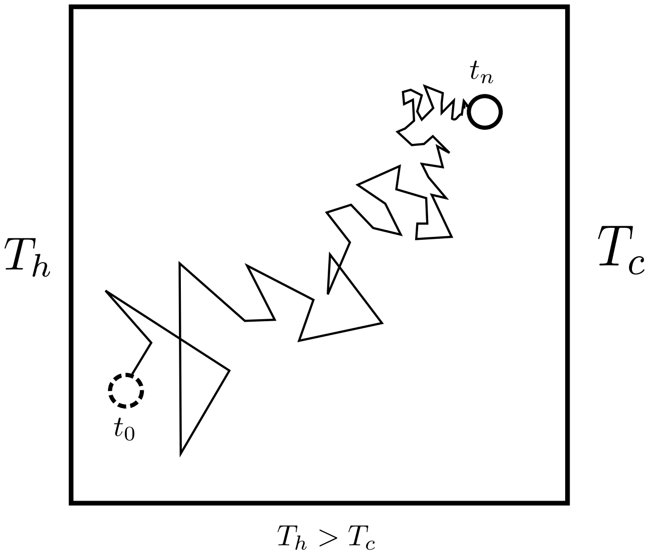


Figure 3: Illustration of the Brownian walk of a particle under a temperature gradient. The distance traveled by the particle after a time interval become shorter as it approaches the colder region. Here T_c denotes the temperature of the coolest wall and T_h represents the hottest wall temperature. We use t_0 to represent the time consider for the initial position of the particle and t_n denotes the time interval taken for the particle to leave its initial position and suffer n random walks due to Brownian motion.

The motion of each individual particle under Brownian motion (Gontijo and Cunha, 2015) is given by eq. 3:

$$m\frac{d\boldsymbol{u}}{dt} = -6\pi\eta\boldsymbol{u} + \frac{4}{3}\pi\Delta\rho g a^3\hat{g} + \boldsymbol{f}_{Th} + \boldsymbol{f}_B, \tag{3}$$

where m is the mass of the particle, u is its velocity, η the dynamic viscosity, $\Delta\rho$ the density difference between it and the fluid, a its radius, $\hat{g}$ the particle acceleration, $\delta\tau$ a typical time step and $\boldsymbol{\xi}$ a white noise unitary vector with uniform distribution in symmetric positive and negative intervals. At the term of diffusion coefficient D_t, which is function of the temperature $\mathrm{D}_t = \kappa_B T/(6\pi\eta a)$, is where the temperature gradient influences. The thermophoretic force $\boldsymbol{f}_{Th}$ is originated by different sources depending on the particle Knudsen number (McNab and Meisen, 1973). For the propose of this article, let's consider that the thermal gradient along the particle is negligible $\boldsymbol{f}_{Th} = 0$, therefore the migration of the particles depends only on the local temperature. The Brownian force $\boldsymbol{f_B}$ is given by

$$\boldsymbol{f_B} = 6\pi\eta a\left(\frac{6D_t}{\delta\tau}\right)^{1/2}\boldsymbol{\xi} \tag{4}$$

For the simulations realized in this work, an equation in terms of dimensionless quantities $Pe = D_t/(U_s a)$ (Brownian Peclet number) and $St = mU_s/(6\pi\eta a^2)$ (translational Stokes number) was used.

$$\mathrm{St}\frac{d\boldsymbol{u^*}}{dt^*} = -\boldsymbol{u^*} + \hat{g} + \left(\frac{6T^*}{\mathrm{Pe^o}\delta\tau^*}\right)^{1/2}\boldsymbol{\xi}. \tag{5}$$

In equation (5) the upper asterisks variables denote dimensionless variables. By neglecting the inertia of the particles due to their small masses ($St = 0$), equation 5 reduces to eq. 6:

$$\boldsymbol{u^*} = \hat{g} + \left(\frac{6T^*}{\mathrm{Pe^o}\delta\tau^*}\right)^{1/2}\boldsymbol{\xi}. \tag{6}$$

Here we define Pe_0 as the Peclet number for a fixed reference temperature T_0 and $T^* = T/T_0$ is the local dimensionless temperature in the space occupied by the Brownian particle.

The validation of the data produced by the code to solve eq. 6 is done by calculating the mean square deviation (MSD) of the particles. If correctly, it should correspond to Einstein equation $\langle x\rangle^2 = 2nD_t$, where n is the number of dimensions, D_t is the Stokes-Einstein diffusion coefficient. The dimensionless version of Einstein equation with the scales used in eq. 5 for the 2D case gives eq. 7.

$$\langle x^*\rangle^2 = 4\mathrm{Pet} \tag{7}$$

Figure 4 shows the MSD of the particles under influence of a temperature gradient. The case without temperature gradient followed Einstein's equation for the MSD, and the cases with thermophoresis produced a small deviation. These deviations are the result of the variation of the local Peclet number where the particles walked (just an average value was used). Although, the temperature gradient affects by a small amount the MSD, this alteration does not follow a systematic tendency capable of being physically interpreted directly as a consequence of the temperature gradient.

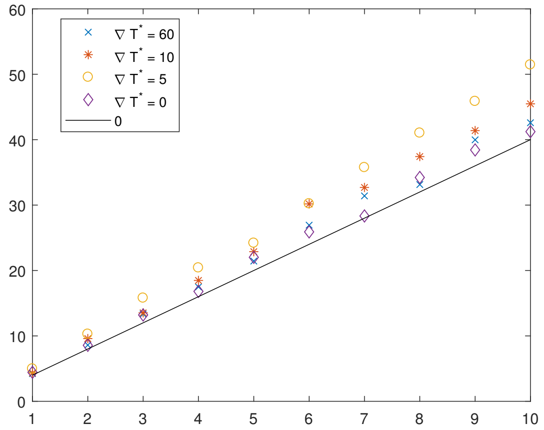
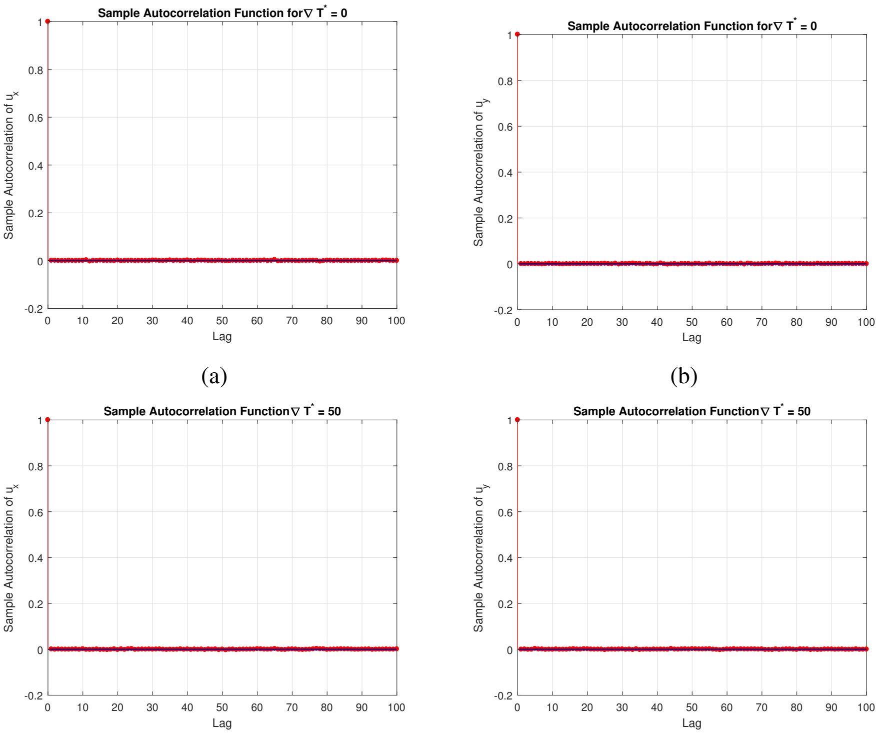


Figure 4: Mean square displacement of the particle under several temperature gradients. The Pe used was the average Pe in each computational domain.

Other important analysis in data that originates from stochastic behavior is the autocorrelation function. For the case without inertia (St = 0), there is no correlation of the particle's velocities parallel and perpendicular to the temperature gradient, when considering a Brownian suspension ($Pe \ll 1$). This shows that the time series of the particle's migration process (velocity fluctuations in time) is memoryless (Markovian process). So in this approach, the autocorrelation function is not much useful to quantify thermophoresis. Although the particles do migrate in a deterministic way from hotter to colder regions they do so following a Markovian path.

Since the tools presented here fail in capturing a systematic consistent behavior with the phenomenon of thermophoresis, the analysis of continuous entropy applied to the concentration distribution of phoretic particles is presented as an alternative tool to quantify such process.



(a) (b)

(c) (d)

Figure 5: Autocorrelation of the velocities of one realization with (fig. 5c , fig. 5d) and without temperature gradient (5a, fig. 5b).

3. DEFINITIONS

First, we present definition 3.1 of Shannon's entropy as a function of discrete probabilities .

Definition 3.1. (Shannon's entropy) The Shannon entropy $H(X)$ of a random variable X with the distribution $P(x)$ is defined as:

$$H(X) = \sum_{i} P(x_i) \log \frac{1}{P(x_i)} \tag{8}$$

In our context, instead of using discrete probabilities, we use a continuous normalized concentration (volume fraction of particles in each region) function. By substituting the sum term of definition 3.1 by an integral, we get definition 3.2.

Definition 3.2. (Continuous Entropy) The continuous entropy $h(X)$ of a random variable X with probability density of $f(x)$ is defined as:

$$h(X) = \int_{S} f(x) \log \frac{1}{f(x)} \tag{9}$$

But as discussed by (Marsh, 2013) in his article, definition 3.2 has its weaknesses. It is variant under change of variables. Which means that depending on the coordinates system a distribution might present different entropy values. The article further discusses that it is scale variant and presents negativity. A solution for this weakness was presented in definition 3.3, which remains non-negative in the continuous case:

Definition 3.3. (Relative Continuous Entropy (RCE) The continuous entropy $D(f||g)$ of two PDFs.

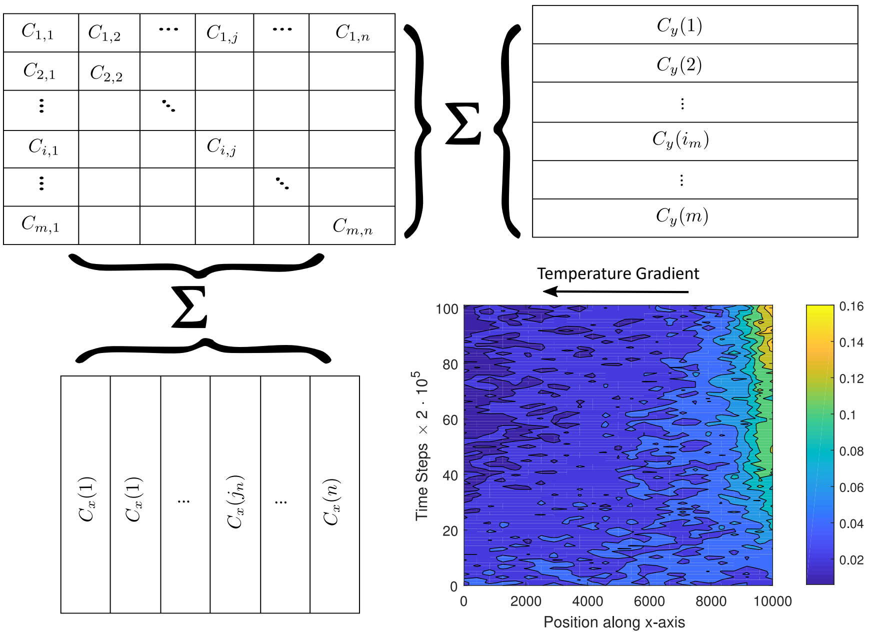
$$D(f||g) = \int_{S} f(x) \log \frac{f(x)}{g(x)} \tag{10}$$

Where S is the support set of f. Note that $D(f||g) = 0$ if $supp(g) \notin supp(f)$.

As he concluded, comparing entropy across continuous entropy distributions yielded good results through maximum entropy problems and central limit theorem. It should be noted that the uniform distribution is the one with maximum entropy in an interval. This means that the events described by an uniform PDF are more equiprobable (have the same weights in terms of recurrence) than random events described by any other PDF. We may say that the RCE works as a measure of the randomness degree of probabilistic events ruled by continuous PDFs.

4. METHODOLOGY

In order to calculate these PDFs, a concentration map can be used, that also gives a nice visualization of the progress of diffusion with time. In a 2D case, the region of interest is divided in an uniform mesh $n \times m$ and the number $N_{i,j}$ of particles in each element $a_{i,j}$ calculated. In a concentration map like fig. (6 b) the concentration C_y is calculated by summing the number of particles in the $i_n -$ th line element along y-axis, $C_y(i_m). = \sum_{i=1}^{n} N_{i,i_m}$. On the other hand C_x is calculated by summing the number of particles in the j_m-th column element, $C_x(j_m). = \sum_{i=1}^{m} N_{i,j_m}$, as shown in figure (6 a). The values C_x and C_y are then divided by the total number of particles, so that we have the fraction of particles along the axes (F_x and F_y) in each region and therefore, the probability of finding a certain number of particles in each column or line element.



a) b)

Figure 6: a) Way to divide the region in cells to calculate the concentration along the x and y directions. b) Concentration field for St$=0$, Pe$=20$ and $\nabla T^* = 20$.

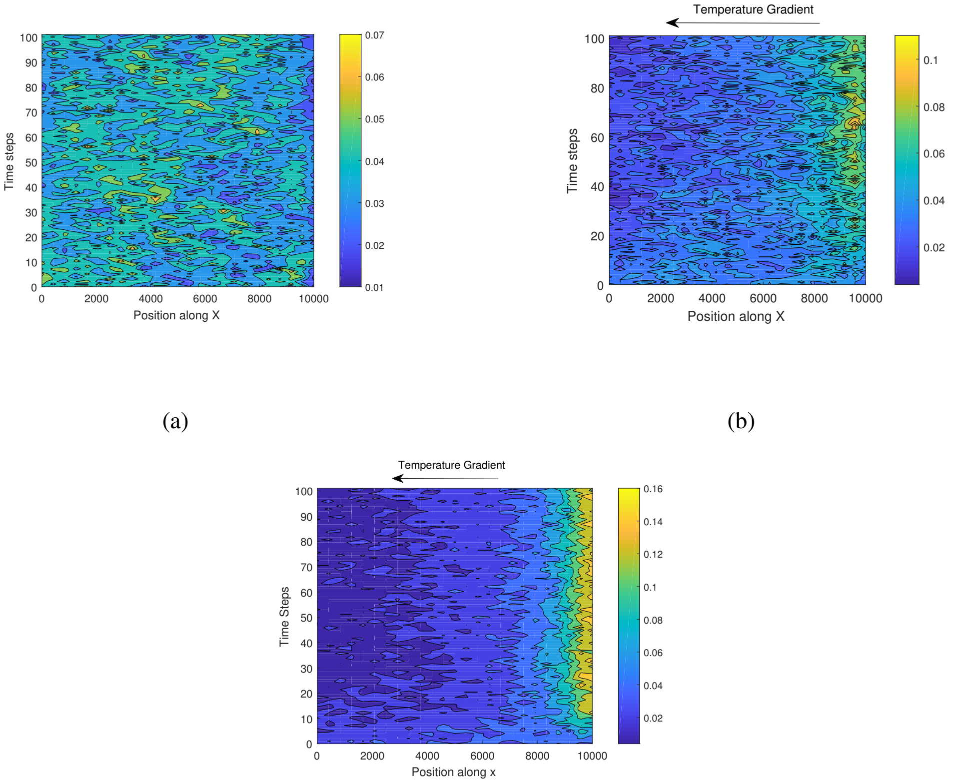
When the data of concentration C_x or C_y is plotted against the values obtained in several time steps in a color map, a figure like (6 b) is obtained.

In order to calculate the entropy of the distribution, the data of the distribution along the axes F_x e F_y are used to generate a function with some curve fitting technique that will describe a PDF of the particles distribution. The entropy is then calculated using eq. 33. Since the RCE compares the entropy of two distributions, some function must be chosen as a reference. Because the uniform distribution has the maximal entropy, it is logically the nicest reference.

For the 3D case, the same logic can be used to map the concentration, but using an uniform 3D mesh. Hence, we would have also the concentration C_z along the z-axis.

5. RESULTS AND DISCUSSION

First we present some images of typical concentration maps for both scenarios: with and without thermophoresis. Figure (7a) denotes the time evolution process of 500 particles in independent realizations without a temperature gradient. It is possible to observe that in the absence of thermophoresis the particles simply fluctuate through the calculation domain. It is also noticeable that the range of density number in the scale at the right side of figure (7a) varies in the interval $0.01 \leq n \leq 0.07$. Figure (7b) considers a dimensionless temperature gradient $\nabla^* T^* = 5$, here $\nabla^* T^* = (a/T_0)(T_h - T_c)/10.000a$, where T_0 denotes a reference temperature value, a is the radius of the particles and T_h and T_c represent the temperatures of the hot and cold wall respectively. It is possible to observe that as we increase the temperature gradients the particles tend to concentrate in the cold wall due to the thermophoretic effect. This effect is even more pronounceable for $\nabla^* T^* = 20$, as shown in figure (7c). Note also that the range in the density number on the right side of figures (7b) and (7c) lies in the range $0.01 \leq n \leq 0.12$ and $0.01 \leq n \leq 0.16$ respectively, meaning that indeed the numerical values of particle concentration through the calculation domain indicate a more heterogeneous distribution due to the thermophoretic mechanism.



(a)

(b)

(c)

Figure 7: Comparison of the particle's concentration under no temperature gradient (a), considering a temperature gradient $\nabla^* T^* = 5$ (b) and $\nabla^* T^* = 20$ (c).

The results shown in figure (7) indicate that the methodology proposed in this work seems to capture thermophoreses. The idea of using local values of the ordinary Stokes-Einstein Brownian diffusional coefficient based on local temperature distributions is capable of recovering at least qualitatively this systematic particle migration tendency due to the thermophoretic effect. It is important to mention that we have not considered explicitly a thermophoretic force, but its indirect calculation by weighting the intensity of the memoryless isotropic random walk according the local temperature values. In future works we intend to discuss this option in details and search for an appropriate model to compute this thermophoretic force in the particle scale.

Now we intend to avail how the RCE of these concentration (density number) maps is affected by an increase in the temperature gradient. The idea here is to test a new methodology that could be used to quantify the thermophoretic mechanism. The distribution entropy calculated with the methodology described in the previous section, gives a number that quantifies how uniformly or noisy a certain probabilistic data distribution is. By comparing different values of the distribution entropy under different conditions, it is possible to compare the influence level of each condition.

It is important to notice that in the way that the PDF function was calculated in the previous section, it identifies variations in the concentration in a single direction over the entire calculation domain, so it is suitable to use it when the boundary conditions of two opposite boundaries differ and all other are equal. If not, this can be still used, but is important to inspect the concentration along other directions.

Although in the way that the PDF of the particles distribution was calculated in the previous section accounts for unidirectional variations in the concentration, if the entropy is calculated for all directions (perpendicular to the boundaries), the effect of the different boundaries conditions or forces that act in different directions can be identified.

Here we consider the PDF functions generated in several simulations of 500 particles in Brownian regime under different temperature gradients. These PDFs are shown in fig. 8 and their relative distribution entropies calculated with eq. 32. In these simulations, there was no interaction between the particles. It is possible to see a sharp decline of relative entropy when the gradient of non-dimensional temperature is under 10, which means that the difference in the kinetic energy of the particles is not big enough to hold the particles in the colder places.

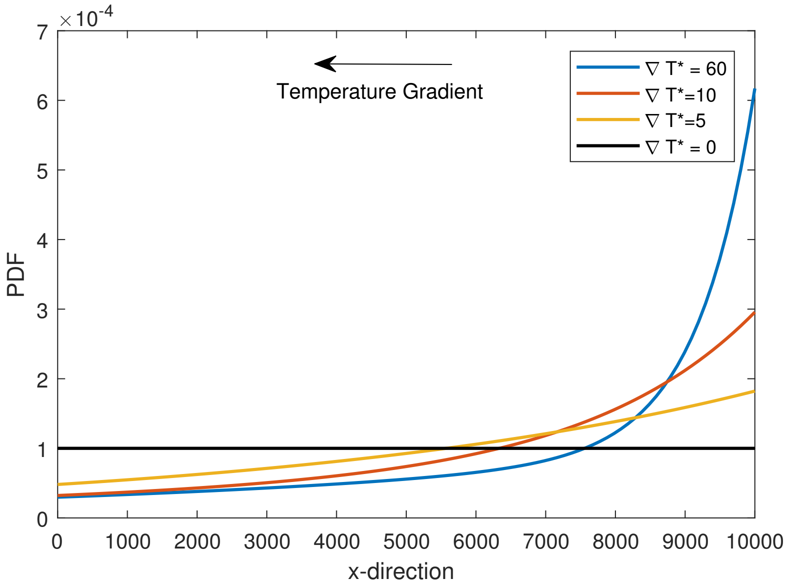


Figure 8: PDF functions of the position of particles under the effect of thermophoresis based on their concentration map.

Based on these PDF functions we calculate the relative entropy associated with each one of them, considering the uniform distribution (or the distribution obtained in the absence of a temperature gradient) as a reference. These calculations are summarized in table (1).

Table 1: Relative distribution entropies of particles under thermophoresis.

∇T^*	**Entropy [nats]**
100	0.4491
60	0.4096
40	0.3605
30	0.2765
20	0.2557
10	0.2059
5	0.0709

It is possible to observe a systematic tendency of increasing the relative continous entropy of the particles distribution as we increase the temperature gradients. From the preliminary results shown in this paper it is important to highlight that the capture of a systematic tendency in this probabilistic scenario by itself is a major result. We have shown that the self-correlation function of velocity fluctuations is not a suitable tool to avail the mechanism of thermophoreses since the particles migrate from hot to colder regions through a Markovian path. We have also shown that although some quantifiable difference may be noted in the MSD plots, this is not an interesting tool to avail the thermophoretic effect since the results do not follow a systematic tendency. However, the use of the relative continous entropy seems to indicate, or at least point a possible investigation direction towards the definition of a statistical tool capable of capturing the thermophoretic effect and hence quantify diffusion.

In terms of a physical interpretation of table (1) there are many points to mention here. First of all, the idea of increasing entropy with the increase of temperature gradients seems to be in accordance with our thermodynamic feeling of entropy. However, it is important to highlight here that the terminology of the word *entropy* is a major point in the topics explored in this paper.

We speculate that the thermophoretic effect leads to a higher deviation from the expected behavior described by a uniform distribution. In this sense we increase the continous relative entropy of particle distribution by increasing the temperature gradients. This affirmation is corroborated by the numerical values of table (1). We must have in mind that the entropy considered here is relative to a specific probability distribution function. A zero RCE means that the data fitted by a certain PDF could also be fitted by the reference PDF chosen to compute the RCE. This could be interpreted in terms with an analogy with the signal convention in the first law of thermodynamic for example. The distribution entropy here, being always defined in terms of a reference PDF, will indicate us wether the events described in a probabilistic scenario by a certain distribution will either approach or distance this reference distribution.

Indeed, considering that the absence of temperature gradients combined with the isotropic nature of Brownian motion will lead to uniform particle distribution it is expected to observe a systematic increase in the differences between the probabilistic particle distribution observed in higher temperature gradients from the isothermal case.

6. CONCLUDING REMARKS

In this work we have proposed a new model to capture the thermophoretic effect in colloidal liquids considering the local Brownian diffusional coefficient values. This methodology was able to capture qualitatively the process of particle migration from hotter to colder regions of the calculatio domain. We have checked the use of classical statistical tools used to quantify diffusion, such as the self-correlation function and the Mean Square Displacement (MSD) and concluded that they fail in the attempt to capture systematic tendencies related to the phoretic effect. Finally, we showed that the use of a relative continous entropy based on the probability distribution functions of the particles concentration maps seems to be an interesting statistical analysis tool to be used in order to quantify diffusion.

7. AKNOWLEDGMENTS

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