Applications of Viscoelastic Fluids Involving Hydrodynamic Stability and Heat Transfer

Ildebrando Pérez-Reyes,
René Osvaldo Vargas-Aguilar,
Samuel Bernardo Pérez-Vega and
Alejandro Sebastián Ortiz-Pérez

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Abstract

Rayleigh and Marangoni convection and rheology are linked in the thermal convection of viscoelastic fluids to some recent technological applications. Such technology developments as the ones presented here undoubtedly shall be based on interdisciplinary projects involving not only rheology or fluid mechanics but several other disciplines. Three practical applications which use Rayleigh or Marangoni convection in their working principle are presented along with some technical details. This contribution focus mainly on the physical mechanism and the involved hydrodynamics of some lab and industrial applications. Finally, a short discussion on the role play by the convective mechanisms is given in order to provide integration of the exposed ideas.

Keywords: viscoelastic fluid, convection, hydrodynamics

1. Introduction

Viscoelastic fluids are common in very important applications [1, 2]. Many of these are so common that people usually do not pay attention to its importance. Paints are a very good example since almost all buildings are painted with a combination of a polymer and a solvent, for different reasons. More complex applications involving polymeric solutions and some kind of process may exist.

On the other hand, not all viscoelastic fluids are paints and some of these may have biological nature or even may be intended for some optical application or the like. Also, viscoelastic fluids may be so different to each other that a reasonable number of constitutive equations have been developed to model their behavior. At the same time, some applications involving viscoelastic fluids may require physical mechanisms for proper working, like natural convection. Such is the case of deoxyribonucleic acid (DNA) replication and the fabrication of corrugated surfaces, for example. Viscoelasticity is then at the same level of importance than natural convection.

The present contribution is devoted to exposed the link between viscoelasticity and Rayleigh and/or Marangoni convection and how these fit in lab or industrial applications. In fact, this is also a different point of view for polymer rheology applications. In this case some examples involving viscoelastic fluids and hydrodynamic stability can be addressed to show another connection of rheology with fluid mechanics. These type of applications are only a few years old and remain relevant.

The author has been working in similar theoretical problems and is trying to make experimental contributions that would finally improve applications like these. Hopefully, this proposal could show the link between rheology and other subjects in new multidisciplinary research topics and developments. The aim of this contribution is to call the attention of researchers and graduate students to interdisciplinary subjects were important technological advances are awaiting to be discovered. Hydrodynamic stability and rheology are both matured topics with interesting theoretical and experimental findings.

The chapter is organized as follows. In Section 2, general comments on viscoelasticity are given so that a connection with thermal hydrodynamic stability in Section 3 can be made. In Section 3 a summary of the physics of thermal convection in Newtonian and viscoelastic fluids is presented too. A brief presentation of the methods used to study the thermal hydrodynamics in viscoelastic fluids is given in Section 4. Next in Section 5 interesting application examples that use thermoconvection are given. Finally, a short discussion on the role played by thermal convection in the example applications is given.

2. About viscoelastic fluids

Viscoelastic fluids are a type of non-Newtonian fluid formed by a viscous component and an elastic one. For short, viscoelastic fluids are the blend of a solvent and some polymer. Examples of these are paints, DNA suspensions, some biological fluids and others from the chemical industry. A number of features make the viscoelastic fluids very interesting and of industrial importance: polymers are almost everywhere. Take for example the case of paints whose annual production generates several USD millions. Proper understanding of viscoelasticity is key for industrial applications.

Polymeric suspensions show viscoelastic behavior but its stress - deformation relationship is not easily represented by a single model. In rheology these models are called constitutive equations and for which the books of Bird et al. [3] and Macosko [4] are two very good classic readings on this subject.

2.1. Some models

Modeling viscoelasticity is a complicated matter since viscoelastic fluids may show, for example, linear and non-linear behavior. On the other hand, some features of the polymeric component may have too much importance to be explicitly introduced in these models. Such is the case for thixotropic behavior in viscoelastic micellar solutions and liquid crystalline dispersions [5]. Some of the constitutive equations of common usage and interest in the academic and industrial community are

- the viscoelastic Maxwell fluid,
- the viscoelastic Jeffreys fluid,
- the viscoelastic Oldroyd fluid,
- the viscoelastic Carreau fluid,
- the viscoelastic “ordered” fluids
- and the viscoelastic KBZ fluid,
- among others.

The readers may find more details on the viscoelastic fluids in the papers of Larson [6, 7] and the textbook of Phan-Thien [8]. For thermal convective instabilities problems the constitutive equations for the viscoelastic Maxwell [9, 10], Jeffreys [11, 12] and second order [13, 14] fluids has some popularity. Several investigations has used these models to study the hydrodynamics of viscoelastic fluids heated from below. Also, these results are the foundation of a number of applications [15–17] like those presented in this contribution.

As a common and widely constitutive equation consider the case of Jeffreys viscoelastic fluids represented by the following expression

$$\left(\frac{\partial}{\partial t}+\lambda_1\right)\tau=-\eta_0\left(\frac{\partial}{\partial t}+\lambda_2\right)\dot{\gamma} \tag{1}$$

where τ is the stress tensor, $\dot{\gamma}$ is the rate of strain tensor, η_0 is the zero shear rate viscosity, λ_1 is the stress relaxation time and λ_2 is the stress retardation time. An important feature of Eq. (1) is that it reduces to the model for viscoelastic Maxwell fluids when $\lambda_2 = 0$, and to the constitutive equation for Newtonian fluids when $\lambda_1 = \lambda_2 = 0$. Several investigations dealing with Rayleigh and Marangoni convection has to do with this type of constitutive equations because of the previously mentioned feature.

3. Thermoconvection hydrodynamics

Thermal convection is of interest for the applications described below but mainly Rayleigh and Marangoni convection. A particular feature of these is the secondary flow generated solely by a thermal gradient. Also, Rayleigh and Marangoni convection form regular patterns which is related to the heat transfer across the fluid.

The problem of thermal convection in incompressible fluids is not new and several geometrical configurations have been considered. Also, the orientation of the thermal gradient and nature of the thermal source has been subjected to different arrangements. This has been driven mainly for potential industrial and/or lab applications [18, 19]. the following cases are of interest for the present contribution: Rayleigh and Marangoni convection in horizontal fluid layers and in vertical cylinders. Pattern formation and heat transfer are key for the proper understanding of the technological developments described below.

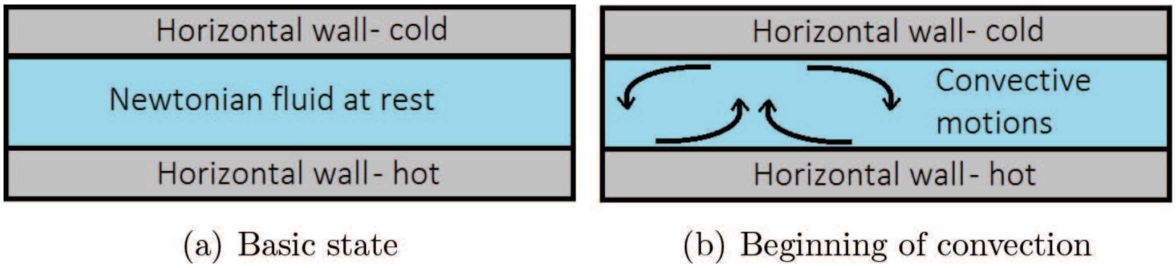
3.1. Convection in Newtonian fluids

For Newtonian fluids the physics behind these two classical problems of fluid mechanics is as follows. In horizontal fluid layers heated from below and cooled from above (see **Figure 1a**), near the bottom where the heating source is located the fluid changes its density by becoming lighter. At the same time the fluid near the top is heavy since because of the top cooling. This is an unstable arrangement of the fluid since portions of fluid with higher density tend to fall pushing portions of lower density fluid to the top (see **Figure 1b**). Next, the movement of the fluid occurs only if a critical temperature is achieved. This is called Rayleigh convection and investigation of the critical conditions at which the convective motions are set is key [20].

For the case of Marangoni convection the physical mechanism is quite different since the surface tension variations with temperature, at the surface, trigger fluid motions. This type of thermal convection occurs in very thin fluid layers or in low gravity conditions. Briefly, as the fluid layer is heated from below the energy is transferred by diffusion to the fluid surface. As the fluid surface tension depends on temperature, in hot surface spots the fluid moves away to cooler surface regions. Next, the convective motions take place.

Both, Rayleigh and Marangoni convection are connected and this has been demonstrated theoretical and experimentally. Most important is that the physical mechanisms has been studied and can be identified not only in the examples shown here but in other engineering areas.

Figure 1 only shows the beginning of the convective motion in the core of the fluid layer. As the process is reinforced, the motions become ordered in a periodical fashion. These convective



(a) Basic state (b) Beginning of convection

Figure 1. Schematics of the physical mechanism of thermal convection in a Newtonian fluid layer heated from below. (a) Basic state (b) Beginning of convection.

Rayleigh number	**Wavenumber**	**Walls**
1708.6	3.12	Conductors [9, 20]
720	0	Insulators [9, 21]
Marangoni number	**Wavenumber**	**Walls**
48	0	Insulators [22]

Table 1. List of critical values for the onset of convection of Newtonian fluids in horizontal fluid layers heated from below. These data correspond to perfect thermal conducting or insulating bounding horizontal walls. For viscoelastic fluids these critical numbers change.

motions are called convective cells. One important fact is that the problem of Rayleigh convection has been widely studied so that for certain cases the critical conditions at which convective motions set are well known as it is shown in **Table 1**. For the case convection of Newtonian fluids in horizontal fluid layers heated from below the critical conditions of perfect thermal conducting and thermal insulators are a common result. From these two cases, that of bounding perfect thermal conductors has been widely studied because it fits better with lab and industrial applications.

The Rayleigh number and the wavenumber are dimensionless parameters featuring the hydrodynamic stability of the fluid layer. As these two parameters achieve critical values thermal convection sets in. Otherwise the fluid motions eventually stop.

3.2. Convection in viscoelastic fluids

Convection in viscoelastic fluids has been widely studied too [9–12, 14, 16]. The discussion in this section shall be restricted to viscoelastic Maxwell and Jeffreys fluids. Then, the physical mechanisms is completely changed due to the introduction of two parameters featuring viscoelasticity as shown in Eq. (1)

- stress relaxation time λ_1,
- stress retardation time λ_2.

Also, convection may be set starting as oscillatory motions. Fortunately, a number of investigators have been working on this subject and the hydrodynamics is well known for some cases. For viscoelastic Maxwell and Jeffreys fluids physics is known to some extent with both the relaxation and the retardation time featuring its behavior. Rayleigh and Marangoni convection in viscoelastic fluids appear in industrial applications and are of interest because the industry of polymers generates millions of USD per year.

4. Methods

Research on hydrodynamics of viscoelastic fluids involves two different approaches. Theoretical and experimental studies are to be linked in order to improve practical applications, which are explained later in this chapter. The aim in hydrodynamic stability studies is to find the critical conditions that defined the onset of convection and later the formed patterns. In Section 5 the previously mentioned critical conditions make sense through the brief explanation of the physical mechanisms of each application.

4.1. Theoretical approach

The theoretical approach uses the common mathematical techniques of hydrodynamic stability for linear and non-linear problems. In either, Rayleigh or Marangoni convection these techniques are used since both are eigenvalue problems. In linear Rayleigh convection the analysis in made to find critical values of the Rayleigh number (*Ra*), and those of the Marangoni number (*Ma*) in linear Marangoni convection. As the mathematical procedure for different geometrical and heat source orientation, for example, are different only that for the convection in a fluid layer heated from below shall be presented.

Consider a horizontal Maxwell viscoelastic fluid layer heated from below and bounded by two horizontal solid walls which are very good thermal conductors. The physical arrangement is very similar to that shown in **Figure 1** with a Maxwell viscoelastic fluid instead of a Newtonian fluid. If the thermal convection in this system is to be studied then the momentum, the continuity, the heat conduction and a constitutive equations should be considered. These are,

$$(1 + Fi\omega)\left[\frac{i\omega}{\mathrm{Pr}}\left(\frac{d^2}{dz^2} - k^2\right)W - Rk^2\theta\right] = \left(\frac{d^2}{dz^2} - k^2\right)^2 W \tag{2}$$

$$\left[i\omega - \left(\frac{d^2}{dz^2} - k^2\right)W\right]\theta = W \tag{3}$$

subjected to the following boundary conditions

$$W = \frac{dW}{dz} = \theta = 0 \ \text{ at } \ z = 0, 1 \tag{4}$$

The system of differential Eqs. (2-4) is an eigenvalue problem for the Rayleigh number *Ra*. In Eqs. (2)–(4) Pr is the Prandtl number, ω is the frequency of oscillation, W is the vertical fluid velocity, θ is the temperature and k is the wavenumber. This eigenvalue problem can be analytically approached with the Galerkin method without fully solving for W and θ.

For short, as inputs, the Galerkin method needs approximated W and θ which are obtained as functions satisfying the corresponding boundary conditions. Then as the approximated functions are used to calculate the residual and find an analytical expression or numerical value of the Rayleigh number. This is a brief explanation of the solution process and further details can be found in Refs. [9, 10], for example. Then, the Rayleigh number *Ra*, the wavenumber k and the frequency of oscillation ω are obtained as outputs of the Galerkin method, for fixed values of Pr and F, as shown in the following **Table 2**

4.2. Experimental techniques

As the working fluids are viscoelastic, these should be characterized. In the case of Maxwell viscoelastic fluids a rheological study is necessary in order to find the corresponding relaxation

Ra	*k*	*ω*	Pr	*F*
226.7151	7.26	76.2593	10	0.1
0.04623	3.44	1.9625	10	100

Table 2. List of critical values for the onset of convection of viscoelastic Maxwell fluids in horizontal fluid layers heated from below. These data correspond to perfect thermal conducting horizontal walls. These are shown here as representative values [9].

time. Certain polymeric suspensions may fit the Maxwell viscoelastic fluid model. With the working fluid relaxation time *F* the theoretical methodology may help to find the corresponding critical conditions for the onset of convection.

Also, experiments in thermal hydrodynamics of convection in fluids are mainly based on visual techniques like Schlieren and shadowgraph. Some authors have also used particle image velocimetry to study the flow field of convective motions. Here, the shadowgraph techniques is considered because the evolution of the convective patterns is key. Besides, the temperature difference and the geometrical dimensions are sufficient for a discussion on the physics of this phenomena.

The experimental setup considered is sketched in **Figure 2**. The shadowgraph technique is very suitable for this type of investigations because it outputs important results at very low costs and time. It is based in the fact that fluid density changes also modify how the light is reflected by it. Then, an optical arrangement is built in ordered to detect light reflexion variations.

The aim of the experimental tests is to help find the real critical conditions at which a given convective pattern is formed. This is of paramount importance for improvement of existent and potential applications.

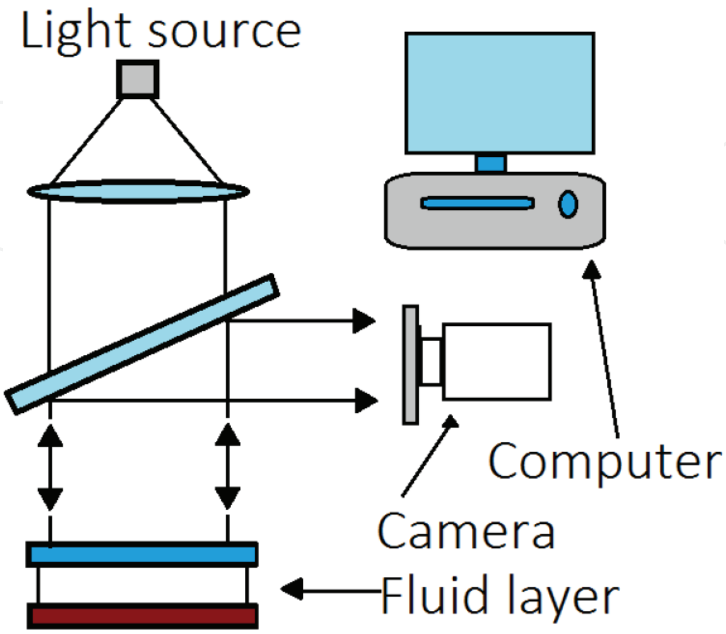


Figure 2. Experimental setup for the shadowgraph technique applied on the convection in a viscoelastic Maxwell fluid layer heated from below.

The theoretical results for *Ra*, *k* and ω are linked to the experimental observations as follows. The critical value of the Rayleigh number predicts the critical temperature difference at which the convective motions set and the critical value of the wavenumber indicates the number of formed convective cells. Then the theoretical results help for tuning the lab experiments and to control the formation of patterns in the fluid.

At this point both, theory and experiments are connected for the formation of convective patterns in viscoelastic fluids. As explain in the next section, this background is the foundation that makes it possible.

5. Some applications

The purpose of this section is to introduce two key example applications of thermal hydrodynamic instabilities in viscoelastic fluids. It is important to mention that these applications were selected because thermal convection is at the core of its working principles. In other words, proper understanding of the hydrodynamic stability in the convection of viscoelastic fluids would improve those applications.

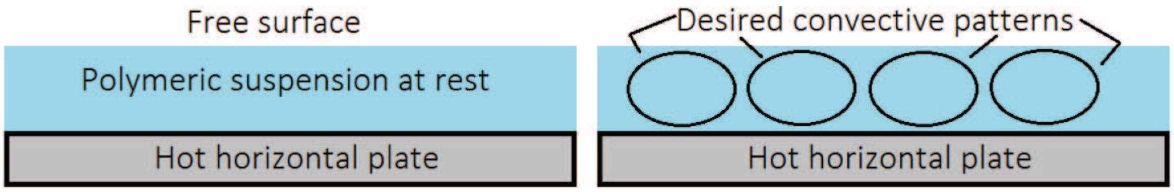
A remarkable fact of the applications described here is that these are placed at the frontier of at least two research topics. The fabrication of corrugated surfaces involved knowledge of hydrodynamics, rheology, heat transfer, optics and polymer chemistry. On the other hand, appliances for DNA replication may need knowledge of genetics, hydrodynamics, heat transfer and rheology.

5.1. Corrugated surfaces

In optics and some electronic appliances [1, 2, 23], it is necessary to print regular patterns over a surface. In fact, there are already technologies that print the mentioned patterns. However, redundant techniques are important for a number of reasons: for reduction of costs and production time, for special cases for which traditional techniques do not work, etc. [1, 2] (**Figure 3**).

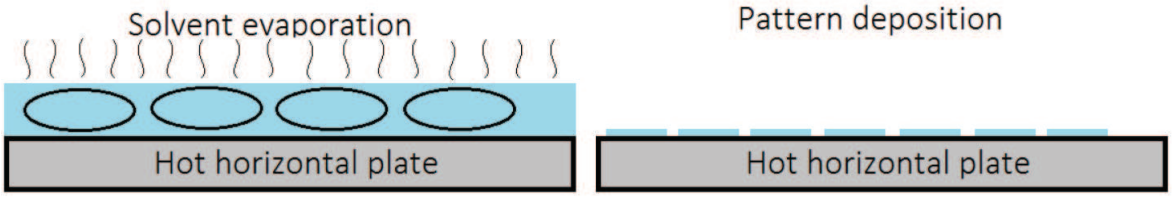
The pattern printed over a surface is in fact a corrugated surface. Precisely, natural convection in thin films of viscoelastic suspensions is used to produced this type of corrugated surfaces. The fabrication process of these corrugated surfaces briefly includes

1. preparation of the polymeric suspension,
2. application of the suspension in a plate surface,
3. heating from below of the plate
4. thermal control of the convection process in order to achieve and maintain the critical conditions,
5. formation of the desired pattern in the polymeric suspension,
6. evaporation of the solvent and deposition of the polymeric pattern.



(a) The suspension is at rest

(b) Convective pattern are formed in he fluid layer



(c) The solvent starts evaporating and the patterns reduce its size

(d) The formed patterns sediment on the dryed surface

Figure 3. Brief schematics for the fabrication of corrugated surfaces based on thermal convection. Notice that at the beginning Rayleigh convection dominates the process but at certain fluid layer thickness the Marangoni convection rules the process until the solvent evaporates completely. (a) The suspension is at rest (b) Convective pattern are formed in the fluid layer (c) The solvent starts evaporating and the patterns reduce its size (d) The formed patterns sediment on the dried surface.

Steps 4 to 6 in the above are tightly related to thermal convection. As the viscoelastic fluid is heated from below the critical conditions for the onset of convection may be achieved or not. It all depend on the control of the heating source, on the plate thermal and geometrical properties, on the fluid properties [9] and on the desired pattern. This is not an easy task and may resemble a craft work. However very good improvements on this technique has been made so that good quality can be achieved.

Several researchers have made advances on this matter which become hot topic with the paper by Nie and Kumacheva [2]. In fact, in 1998 Mitov and Kumacheva [17] reported instability induced patterning as printing technique for the fabrication of corrugated surfaces. More recently, other researchers have made findings on how the theory of Marangoni convection is coupled with the deposition of regular patterns on a surface.

Further work is needed for the improvement of the experimental technique. For example, it is known that thickness of the fluid layer decreases in time so that both Rayleigh and Marangoni convection may appear in the fluid. Also, as the fluid layer decreases may be the viscoelastic properties may change and in turn modify the formed patterns. In the literature can be found a few papers dealing with the evaporation rate. On the other hand, thermal control should be robust in order that the fluid layer may sustain the formed patterns. Also, several authors have reported new findings for the proportional and proportional integral control of Rayleigh convection. Although more isolated advances can be done in different fields interesting integrating work remains for this type of applications.

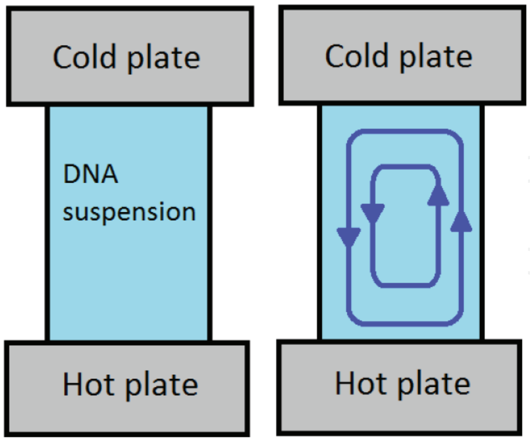
5.2. DNA replication

DNA technologies have attracted special attention, first because DNA suspensions are considered viscoelastic fluids because these are a combination of an aqueous solvent and a kind of polymer in the form of DNA chains. Second, hydrodynamics of biological fluids has become a subject of growing interest in recent decades because of its link to a biochemical reaction. For short, DNA replication needs a temperature gradient and an enzyme to be done so that possibly convective motions are involved. DNA replication is common task for geneticists and biotechnologists which also is an important tool for research and development in biotechnology, medicine, molecular biology and other important subjects.

In DNA replication a very expensive apparatus is involved. The thermocycler is small specialized piece of equipment that has become an indispensable tool for research laboratories for industry and academy. However, thermocyclers work is based on the physical mechanism of thermal convection. This is, the equipment creates a temperature gradient by heating from below small tubes and mixes the fluid by producing soft horizontal motions. Several years ago the process followed by the thermocycler was identified as thermal convection in a vertical cylinder [24] and a number of more simple and cheaper thermocyclers were prototyped [25].

The complicated DNA replication process can now be very briefly outlined. The steps given below should be taken as a general picture and the reader is encouraged to search specialized literature for more details on this matter. **Figure 4** shows a very simplified scheme of the thermoconvective motions that take place in this type of biological viscoelastic fluids. Thus, the process is as follows

1. an aqueous dilute suspension of a DNA fragments and the polymerase enzyme is prepared,
2. the suspension is placed in nearly cylindrical containers called vials,



(a) DNA suspension at rest (b) Convective motions in the DNA suspension

Figure 4. Brief schematics for thermal convection in a DNA suspension confined in a vertical cylinder. Notice that convection cannot be set if a proper critical Rayleigh number is not achieved. (a) DNA suspension at rest (b) Convective motions in the DNA suspension.

3. a thermal gradient is set vertically in the vial,
4. a temperature of 95°C is maintain in order that the polymerase chain reaction (PCR) takes place,
5. DNA replication.

The above given steps lack of some biochemical details but the aim is to show that a physical phenomenon involving thermal gradients and fluid motions is implied. As before, thermal convection is at the core of this appliance and it has been demonstrated the feasibility of cheaper and not too complicated thermocyclers [26]. The lab experiments conducted by Muddu et al. [26] with a very simple cycler prototype based on thermal convection are very important since these are based on the Rayleigh convection.

DNA replication is not the only application involving Rayleigh convection. Some other uses in molecular biology have been found like trapping of DNA with help of thermophoresis or the Soret effect [24]. The Soret effect and the Rayleigh and/or Marangoni convection are related, and for short it is mass transfer assisted by heat transfer and vice versa. This is, DNA material can be concentrated in suitable spot for further recovery.

6. Discussion

Some applications mainly based on Rayleigh and Marangoni thermal convection were briefly presented. These technologies were developed by researchers working on the frontier of various subjects which, from the authors point of view, is the perfect spot for human advances. Several complicated theories have a similar way finding unexpected common technological applications for most humans.

Thermal convection in its general sense has found practical applications in a number of other fields. For example, geology and volcanology use the theory of Rayleigh convection in fluids confined between insulating walls and many industrial equipments for heat transfer use the theory of convection too. On the other hand and to the best knowledge of the authors there are very few new technologies using Rayleigh or Marangoni convection as the ones presented here. It is remarkable that the physical mechanisms of these phenomena are at the core of the above presented technologies and what makes them work is the vast results reported since several decades.

Hopefully, researchers working on interdisciplinary projects could take advantage of the large amount of reported results on Rayleigh and Marangoni convection.

7. Conclusions

Here, an interesting connection between rheology and thermal hydrodynamics is presented through some applications. The theoretical approach is linked to an experimental setup by using dimensionless parameters like the Rayleigh number and the wavenumber. Already existent applications are then presented as the result a strong understanding of the physics involved. It can be concluded that for applications involving viscoelastic fluids rheology is intrinsically related to the thermal hydrodynamics. This is, pattern formation and heat transfer cannot be controlled if adequate constitutive model are not used. The rheological behavior of the fluid is key for these applications since relaxation time actually favors the heat transfer. It can be also concluded that robust control strategies, on the fluid rheology and heat transfer, may be needed for some applications as such is the fabrication of corrugated surfaces case. In this case the solvent is evaporated, so that viscoelasticity and then heat transfer change, which is not taken into account. Finally, it is shown that further investigation in this branch of rheology is needed.

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Author details

Ildebrando Pérez-Reyes[1]*, René Osvaldo Vargas-Aguilar[2], Samuel Bernardo Pérez-Vega[1] and Alejandro Sebastián Ortiz-Pérez[3]

*Address all correspondence to: ildebrando3@gmail.com

1 Chemical Sciences Faculty, Autonomous University of Chihuahua, New University Campus, University Circuit, Chihuahua, Chih., Mexico

2 SEPI ESIME Azcapotzalco, National Polytechnic Institute, Santa Catarina, Azcapotzalco, Mexico City, Mexico

3 Engineering Faculty, Autonomous University of Baja California, Mexicali, Mexico

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