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# Suppression of Buoyancy in Gaseous Media at High Temperatures

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# SUPPRESSION OF BUOYANCY IN GASEOUS MEDIA AT HIGH TEMPERATURES

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## Introduction

Natural convection, induced by buoyancy forces in the presence of a gravitational field, is driven by density gradients generated by either temperature or concentration gradients. If the gradients are perpendicular to the gravity vector, the fluid is unconditionally unstable. Above a horizontal surface, however, the transport of heat and mass in a quiescent fluid will be governed solely by conduction and diffusion until buoyancy forces become strong enough to initiate motion. In our discussions below, we limit our considerations only to *thermal gradient* cases where the heated surface is below the colder fluid and is horizontal with respect to the gravity vector. The onset and development of buoyant flow under such conditions depends quantitatively on Rayleigh number, defined as

$$Ra \equiv \frac{\Delta\rho}{\rho_o} \frac{gL^3}{\alpha\nu} \quad (1)$$

where  $\rho = \rho_o - \rho$ . If the Rayleigh number is smaller than a critical Rayleigh number  $Ra_c$ , then the fluid remains still (known as the Rayleigh-Bénard criterion).

The Rayleigh number can also be expressed as a product of the Grashof number and the Prandtl number, respectively defined as

$$Gr \equiv \frac{\Delta\rho}{\rho_o} \frac{gL^3}{\nu^2}, \quad Pr \equiv \frac{\nu}{\alpha} = \frac{\mu c_p}{\lambda} \quad (2)$$

such that  $Ra = Gr \cdot Pr$ . Under small normalized temperature differences at constant pressure, Boussinesq conditions apply (Boussinesq, 1903); i.e., it is commonly understood that the governing conservation equations consist of the following approximations: (1) density is assumed constant except when it directly causes buoyant forces, (2) all other fluid properties are assumed constant, and (3) viscous dissipation is assumed negligible, as was first used by Oberbeck (1879). The normalized density difference can then be approximated by the first term of its Taylor-series expansion; i.e.,

$$\frac{\Delta\rho}{\rho_o} \approx \beta_T (T - T_o) = \beta_T \Delta T \quad (3)$$

where  $\beta_T$ , defined as  $\beta_T \equiv - (1/\rho) \cdot (\partial\rho/\partial T)_p$ , is the coefficient of thermal expansion at constant pressure. If a temperature gradient (i.e., density stratification) is too large for diffusion to sustain, such that a  $Ra_c$  is exceeded, then finger-like intrusions of the lighter gas into the heavier gas, and the heavier gas into the lighter gas, result rapidly.

The validity of the Boussinesq approximation (Gray and Giorgini, 1976) and departures from the Boussinesq approximation (Paolucci and Chenoweth, 1987) have been studied in the literature. These studies led to the specification of explicit conditions under which the neglect of various terms in the traditional Boussinesq approximation can be justified.

Goldstein and Volino (1995) give an extensive description of various investigations in the literature for the onset of convection and flow development in horizontal layers. Many questions are raised due to discrepancies among different flow structures. Some studies had free surfaces, while others used rigid surfaces. The heated surface was applied either a step change or a linear increase in temperature, or a step change in heat flux. Some of the investigators report plunging sheets, while others observe isolated plumes and thermals, and still others see both. They use different criteria and different experimental observations for the basis of determining the onset of instability. Several authors report widely ranging values of  $Ra_c$ , easily from 1000 to 3000.

### **Temperature-Dependent Properties for Gases and Its Consequences on Buoyancy**

Albeit the uncertainty on  $Ra_c$ , even under simpler Boussinesq conditions, the problem of determining a critical Rayleigh number for *non*-Boussinesq conditions (*large* normalized temperature differences), using high-order eigenvalue formulations/solutions, is quite intriguing and remains to be thoroughly treated. Upon extension to large temperature differences, the temperature dependence of the thermodynamic and transport properties becomes a matter of significance and can no longer be neglected.

More recently, Fröhlich *et al.* (1992) investigated large departures from the Boussinesq approximation in the Rayleigh-Bénard problem. They determined that temperature-dependent properties affect the mid-plane symmetry and lead to quantitative and qualitative changes of the flow, in particular, in the bifurcation from the conduction state. Their most important finding states that the non-Boussinesq behavior becomes more confined to small regions in the flow, and hence, the relative importance of these effects is expected to diminish for higher Rayleigh numbers. However, their study fails to determine that gaseous flows tend to become *more* stable towards the conduction state at *larger* normalized temperature differences, due primarily to the dampening effect of gas viscosity at higher temperatures, as investigated in this paper.

Here, we limit our considerations only to *gaseous* media because the temperature dependence of thermodynamic and transport properties for gaseous systems leads to some interesting consequences in regards to flow stability and onset of buoyant convection. For ideal gases, it is readily shown that  $\beta_T = 1/T$ . The power-law approximations of the temperature dependence of other gaseous properties are typically taken as (Svehla, 1962):  $\rho \sim T^{-1}$ ,  $\mu \sim T^{0.7}$ ,  $\lambda \sim T^{0.85}$ ,  $c_p \sim T^{0.19}$ ,  $D \sim T^{1.7}$ . These powers are also consistent with the kinetic theory of gases (Kennard, 1938; Hirschfelder, Curtiss, and Bird, 1954), and are accurate descriptions for the properties of air in the 600 to 1600 K range (Brown and Donoughe, 1951; Kays and Crawford, 1980).

Interestingly, the dimensionless numbers based on the relative importance of momentum, heat, and species mass diffusion, namely the Prandtl (Pr), Schmidt ( $Sc = \nu/D = \mu/\rho D$ ), and Lewis numbers ( $Le = \alpha/D = \lambda/\rho c_p D$ ), become temperature *independent*; i.e., the power of temperature is smaller than 0.04. Hence, ignoring variable property effects for many convection problems is

justified. Since the temperature dependences of the Rayleigh and Grashof numbers become about the same, we focus our attention to Gr from here on.

For large normalized temperature differences, the variable (temperature-dependent) gas properties are typically accounted for by evaluating their values at a *reference* temperature  $T_*$ , which is customarily taken as the arithmetic mean of the hot and cold surface temperatures in the system (Kays and Crawford, 1980; Bird, Stewart, and Lightfoot, 1963); i.e.,

$$T_* = T_o + 0.5(T_h - T_o) = \frac{(T_h + T_o)}{2} \quad (4)$$

Paolucci and Chenoweth (1987) also found in their calculations that this reference temperature describes average properties more accurately than other alternatives. The temperature dependence of the Grashof number can now be expressed as

$$Gr = \beta_r \Delta T \frac{gL^3}{\nu^2} = \frac{\Delta T}{T_*} \frac{gL^3}{\nu^2} \sim \frac{\Delta T}{T_*} \frac{1}{T_*^{3.4}} = \Delta T T_*^{-4.4} \quad (5)$$

It is indeed noteworthy that the temperature dependence of  $\nu$ , the gas kinematic viscosity, is primarily responsible for making the Grashof number so strongly temperature dependent.

Typically, the Boussinesq approximation is valid if  $\frac{\Delta T}{T_*} \leq 0.1$  (6)

For cases where  $T_o$  is room temperature around 300 K,  $\Delta T$  is thus limited to smaller than 35 K. For most heat transfer applications,  $\Delta T$  is much larger than 35 K, and for combustion, it will routinely exceed 1000 K. Hence, one has to be cautious in the use of such things as the Rayleigh stability criteria or heat transfer correlations based on the Rayleigh (Grashof) number.

For quantifying the temperature dependence of the Grashof number, consider a constant-property Grashof number,  $Gr_o$ , for which the gas properties are evaluated at the cold-surface temperature,  $T_o$ , and the normalized temperature difference is taken as unity (or  $\Delta T = 1$  K); i.e.,

$$Gr_o = \frac{1}{T_o} \frac{gL^3}{\nu_o^2} \quad (7)$$

Then the variation of the Grashof number with temperature at larger normalized temperature differences can be quantified by normalizing the Grashof number with respect to  $Gr_o$ . Thus, the normalized Grashof number, defined as GR, has the following temperature dependence:

$$GR \equiv \frac{Gr}{Gr_o} = \Delta T \left( \frac{T_o}{T_*} \right)^{4.4} \quad (8)$$

Note that Eq. 8 is independent of the type of gas and the pressure of the system as long as the power-law descriptions for the temperature dependence of gas properties are adequate. Figure 1 below describes the behavior of GR for three different cold surface temperatures. The main

curve is the one labeled for cold-surface temperature of  $T_o = 300$  K and forms the basis of most of the discussions presented in this paper. Other curves labeled at lower cold-surface temperatures apply to arguments related to cryogenic applications.

Figure 1 has a number of interesting implications. It suggests that if a system becomes unstable when the hot-surface temperature is increased, then there are *two* distinct critical hot-surface temperatures for the onset of buoyant convection, one at a lower temperature and another at a higher temperature. Below and above these temperatures, the system is stable, i.e., conduction-dominated. It seems that, for a fixed cold-wall temperature, the lower the critical low-temperature is for a system, the higher the critical high-temperature is. The question of whether there is any significant difference in the critical Rayleigh numbers for the onset of convection at the lower and higher temperature regions will be explored below.

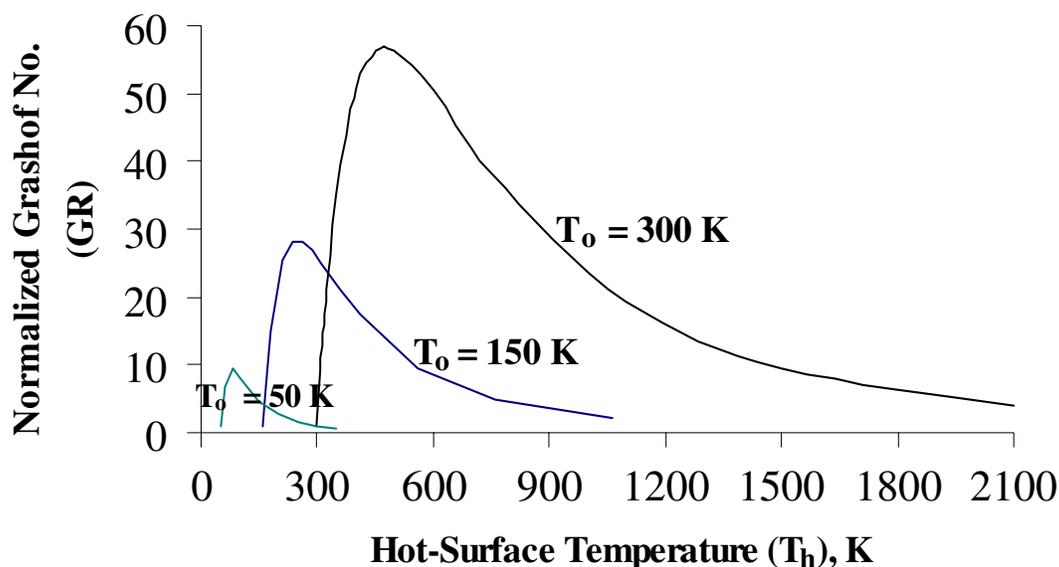


Figure 1.—Variation of normalized Grashof number with hot surface temperature (eq. 8) describing the effect of temperature-dependent gas properties.

### Numerical Modeling Results

In order to provide an initial check for the validity of fig. 1, we have built a 2-D model using the computational fluid dynamics code FLUENT. The rectangular geometry and the conditions are taken to represent the traditional Bénard problem, where the distance between the bottom hot and the top cold surfaces is fixed at 1.25 cm and the aspect ratio is chosen as 6. The vertical side-walls are insulated (adiabatic), the top cold-surface temperature is fixed at 300 K, and the bottom hot-surface temperature is varied parametrically at fixed levels. We use either neon or argon whose temperature-dependent properties are fully accounted for by using curve fits. Pressure is also fixed, but parametrically varied to explore, under *steady-state* conditions, whether the two different regimes are dominated either by conduction or buoyant convection.

Our numerical experiments for Ne under atmospheric conditions reveal that Ne is always stable for all hot-surface temperatures. This finding is not surprising since the *largest* Rayleigh number for Ne at the peak temperature of  $\sim 476$  K of figure 1 is calculated to be  $\sim 1500$ . This is

smaller than  $Ra_c$  values reported by Fröhlich *et al.* (1992) and Goldstein and Volino (1995), which are larger than 1800 and 2000, respectively. These results demonstrate that one cannot make the Ne system unstable to onset buoyant convection by simply raising the hot-surface temperature. However, when we switch the working gas to Ar, the system is unstable at the hot-surface temperature of 476 K, as expected and depicted in fig. 2. In all of our calculations where the gas becomes unstable, recirculating cells with aspect ratios of about unity are generated, creating warm and cool fingers. The stable, quiescent cases, however, result in almost linear temperature profiles, where the slight departures from linearity are due to temperature-dependent gas properties. Note that the color scale is linear and applies the same way to all figures.

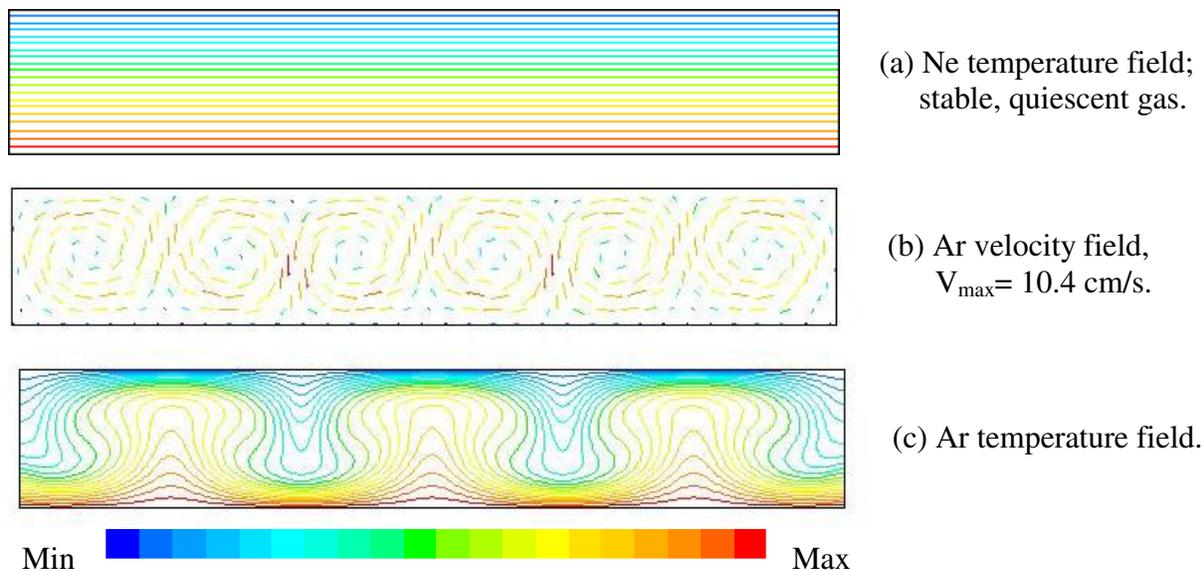


Figure 2.—Ne and Ar temperature and velocity fields for  $T_h = 476$  K (peak temperature for fig. 1) and  $p = 1$  atm. The color scale is linear.

Based on the molecular-weight ratio of about 2 between Ar and Ne, the dependence of the Grashof number on the square of density and that Ne has smaller viscosity than Ar, we expect Ne to be *unstable* for the same hot-surface temperature of 476 K, but at 5 atm. Indeed, figure 3 demonstrates the resulting slightly stronger velocity field with its associated temperature field.

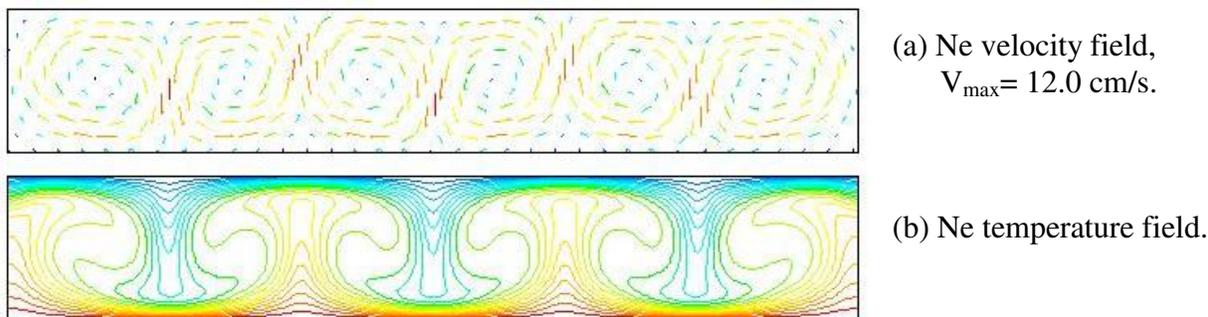


Figure 3.—Ne velocity and temperature fields for  $T_h = 476$  K and  $p = 5$  atm.

In the next set of calculations, we successively increase the pressure for Ne gas from atmospheric pressure, for which Ne stays stable for all hot-surface temperatures, to as high

as 5 atm. The results are schematically shown in fig. 4 below and reproduce fig. 1 in reversed shape. Indeed, the implications of fig. 1 are numerically demonstrated again. Furthermore, one can readily observe from fig. 4 that some systems remain stable with respect to the onset of convection regardless of how large the normalized temperature difference becomes. In fact, beyond a certain maximum hot-surface temperature, the tendency of systems to become more stable towards a conduction-dominated state *increases* as the hot-surface temperature increases.

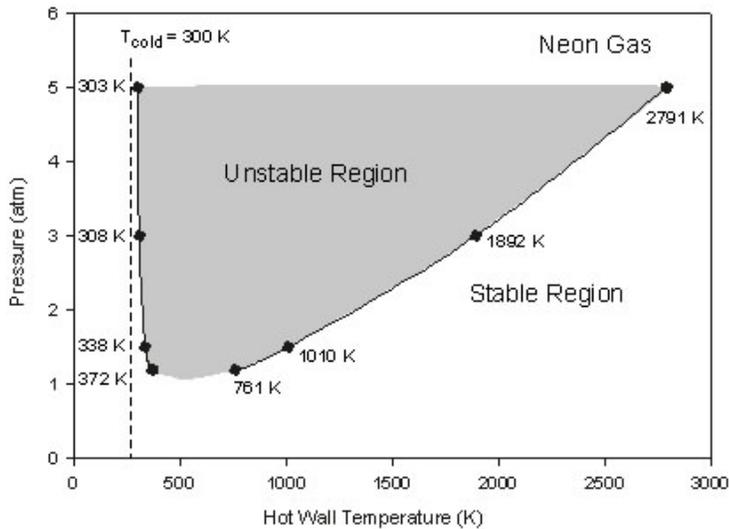


Figure 4.—A schematic of calculated stable and unstable regions, and hot-surface temperatures at which transition occurs for Ne at various pressure levels.

The proper determination of the critical Rayleigh numbers for the onset of convection at the lower and higher temperature regions requires *transient* analysis. Such calculations (subject of our current work) can also quantify the required heating rates of the hot-surface in order to pass through the unstable regime in the middle to reach the stable regime at higher temperatures. We tabulated below the calculated Rayleigh numbers corresponding to the transition temperatures depicted in fig. 4, cognizant of the limitations of using steady-state analysis.

| <b>P, atm</b> | <b>T<sub>h</sub>, K</b> | <b>Ra<sub>c</sub></b> |
|---------------|-------------------------|-----------------------|
| 1.2           | 372                     | 1683                  |
|               | 761                     | 1706                  |
| 1.5           | 338                     | 1812                  |
|               | 1010                    | 1672                  |
| 3.0           | 308                     | 1853                  |
|               | 1892                    | 1613                  |
| 5.0           | 303                     | 1996                  |
|               | 2791                    | 1570                  |

It is noteworthy that, for the same pressure pairs, the critical Rayleigh numbers depart more from one another as the pressure increases; i.e., as the T<sub>h</sub> differences increase. This may be due to the fact that there are larger inaccuracies in the calculation of Ra<sub>c</sub> at larger differences between hot- and cold-wall temperatures, since properties evaluated at the average temperature are used. In fact, the Boussinesq approximation is valid in only two of these cases (see eq. 6). It should also be noted, however, that all critical Rayleigh values are in the range of those most recently reported by Goldstein and Volino (1995), which vary depending on the measurement technique, and are given as 2673 on the basis of thermocouple, 2700 on the basis of liquid crystal, and 1265 on the basis of interferometry measurements.

Naturally, the level of convection is weaker at the hot-surface temperature where transition from stable to unstable occurs (determined numerically to within 1 K). An example of such small velocities is shown below in fig. 5 relative to those shown in figs. 2 and 3. We determined this transition temperature of 308 K for Ne at 3 atm, such that at  $T_h = 307$  K the temperature profile was completely conduction dominated.

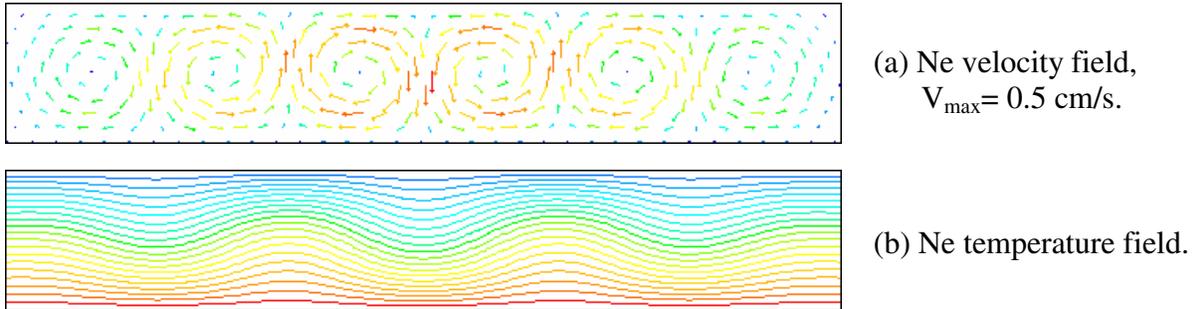


Figure 5.—Ne velocity and temperature fields for the transition  $T_h = 308$  K and  $p = 3$  atm.

### Some Practical Implications

Generalization of the Rayleigh-Bénard criterion for the onset of convective instability to non-Boussinesq conditions is long overdue in the heat transfer/fluid physics community. We list below a few examples where such information is not only relevant, but critical.

The application of existing heat transfer correlations involving the Rayleigh and Grashof numbers for engineering purposes is a routine practice, even under conditions where the Boussinesq approximation is no longer valid. Though for liquid systems there is little concern, we may not be so fortunate for gaseous systems, especially if the hot-surface temperature exceeds  $\sim 500$  K. According to fig. 1, we predict that the tendency for buoyant convection will be suppressed more at higher temperatures. If a practicing engineer uses recommended traditional methods to calculate the relevant Rayleigh (Grashof) numbers for hot-surface temperatures much *beyond* 500 K, he/she may be misled to a wrong conclusion, because these numbers will be larger at even lower temperatures through which the system will have to pass to reach the higher levels. One may conclude a stable, quiescent, conduction-dominated system behavior based on the calculated smaller numbers, whereas the system may become unstable and convection-governed while heating up to the higher temperature. Similarly, even if both predictions and reality may result in the unstable, convection-governed regime, the intensity of buoyant convection may be totally misjudged by not considering the history of the system during heat-up. The consequences of such misjudgments can be severe in both earth and space applications with partial levels of gravitational acceleration, such as Lunar or Martian.

For materials processing involving gaseous media, particularly chemical vapor deposition (CVD), buoyancy-driven convection is avoided to improve deposition rate uniformity and deposit quality. The effect of temperature-dependent gas properties on flow dynamics in the CVD process has long been recognized (Rosenberger, 1987). In horizontal CVD reactors, buoyancy leads to a helical flow structure. In vertical, impinging-jet or stagnation-flow CVD reactors, buoyancy is suppressed by either enhanced reactant flow rates or by lowering the pressure, both of which result in reduced efficiencies (Gokoglu, 1992). General perception in

CVD is that if the temperature of the hot-substrate increases, the tendency for buoyant convection, which is undesirable, also increases. Yet, our analysis above suggests that if the hot-substrate is heated to even higher temperatures (in excess of about 500 K), this may even be beneficial in suppressing buoyancy. The challenge would be to heat the substrate to these high temperatures rapidly enough to avoid the development of buoyant convective instabilities at lower temperatures during the ramp-up. This should be possible with currently available technology used in rapid thermal processing (RTP) of materials. Ironically, because high substrate temperatures are sometimes harmful to the desired deposit quality, especially for electronic CVD materials, the low-temperature CVD processes employed to circumvent this problem may be more prone to the buoyant convection problem. For applications of CVD to the growth or coating of structural materials, where the use of high substrate temperatures is not as critical an issue, the results presented here are directly relevant.

There are serious consequences of our proposed research for partial-gravity combustion applications in space and for the combustion community at large. Diffusive-transport-controlled flames with planar symmetry create environments with large normalized temperature differences where the traditional application of Rayleigh-Bénard criterion for the onset of convective instability is quite inadequate (Baumstein and Fendell, 1998; Fendell and Mitchell, 1996). Upon extension to non-Boussinesq conditions, it may prove necessary to regard the critical Rayleigh number as a function of the Prandtl and Lewis numbers, as well as the normalized temperature difference between the diffusion flame and the cold upper surface. The thermodynamic dependence of the transport properties becomes a matter of import.

Applying traditional scaling approaches by the use of dimensionless numbers to micro-scale combustion has been shown to be difficult (Sitzki *et al.*, 2001). Critical dimensions of such micro-combustors and the heat loss mechanisms associated for optimal operation require a more careful analysis. The results presented here suggest that designers of micro-combustors select the temperatures of operation and dimensions between hot and cold surfaces more judiciously in order to better control conduction versus convection-dominated heat transfer.

Figure 1 also suggests that the behavior of gaseous systems under cryogenic conditions is quite different for the onset of instability. For these systems, where room temperature may be the “hot” surface temperature and cryogenically cooled sensors or electronic components may constitute the cold surfaces, heat transfer mechanisms may be quite different than those expected from simpler, Boussinesq type analyses, because variable gas properties seem to dominate the flow behavior. Indeed, for the curve labeled as  $T_o = 50$  K, the system is drastically dampened by the higher viscosity of the gas when the hot surface temperature  $T_h$  is only 300 K! If the desired mode of heat transfer is conductive, then the designers of such devices may highly benefit from the results presented here by utilizing even larger temperature differences.

### **Concluding Remarks**

Our 2-D numerical work utilizing the computational fluid dynamics code FLUENT demonstrate that modeling can provide insightful information to understand Rayleigh instability at large temperature differences in gaseous media. The steady state analysis is believed to be sufficient for describing the phenomena, though not as accurate as transient calculations, which we will report on soon. We show here that under normalized temperature differences larger than justifiable for the Boussinesq approximation, there are terrestrial situations where a system

cannot *ever* be made unstable with respect to the onset on buoyant convection no matter how *large* the temperature (density) difference becomes at a given pressure! This behavior is due to temperature-dependent gas properties. Highly temperature-sensitive kinematic viscosity is primarily responsible for counteracting the tendency toward instability and dampens convection by making the gas more “viscous” at higher temperatures. This compensation of the buoyant force by the viscous force exhibits itself by the formation of a peak hot-surface temperature beyond which a system will tend to be *more* stable as the hot-surface temperature *increases*. We also show, for a given pressure, that there are *two* distinct critical hot-surface temperatures for the onset of buoyant convection, one at a lower temperature and another at a higher temperature, below and above which, respectively, the system is stable, i.e., conduction-dominated.

Our current work is also exploring the behavior in sealed *enclosures*, where the pressure is allowed to rise during the transient heating of the hot surface. The rate of pressure rise depends on the heat-loss rate of the system relative to the heat-up rate of the hot surface and the diffusion rate in the gas mixture. While the higher temperatures help dampen the buoyancy effect, the higher pressure oppose it. The effect of aspect ratio naturally becomes another parameter in this regard.

## Nomenclature

|           |  |
|-----------|--|
| $c_p$     | constant-pressure heat capacity  |
| $D$       | species mass diffusivity   |
| $g$       | acceleration of gravity  |
| $Gr$      | Grashof number, Eq. (2)  |
| $L$       | characteristic length (height) between hot and cold surfaces                 |
| $Le$      | Lewis number ( $\alpha/D =$ thermal diffusivity / species mass diffusivity)  |
| $M$       | molecular weight   |
| $p$       | pressure   |
| $Pr$      | Prandtl number, ( $\nu/\alpha =$ momentum diffusivity / thermal diffusivity) |
| $Ra$      | Rayleigh number, Eq. (1)   |
| $Sc$      | Schmidt number ( $\nu/D =$ momentum diffusivity / species mass diffusivity)  |
| $T$       | temperature  |
| $V$       | velocity   |
| $\alpha$  | thermal diffusivity, $\lambda/(\rho c_p)$                                    |
| $\beta_T$ | coefficient of thermal expansion at constant pressure                        |
| $\lambda$ | thermal conductivity   |
| $\mu$     | dynamic viscosity  |
| $\nu$     | kinematic viscosity ( $\nu = \mu/\rho$ )                                     |
| $\rho$    | density  |

## Subscripts

|     |  |
|-----|--|
| $c$ | cold surface condition                         |
| $h$ | heated, hot surface condition                  |
| $o$ | initial ambient or cold surface condition      |
| $p$ | constant pressure condition                    |
| *   | average, arithmetic mean temperature condition |

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| <b>13. ABSTRACT</b> ( <i>Maximum 200 words</i> )<br><br>Consider a rectangular box filled with a fluid having a heated bottom and a cold top surface, and insulated side-walls (Bénard problem). As the temperature difference between the horizontal top and bottom surfaces increases, a critical condition, defined quantitatively by the Rayleigh number, is reached beyond which density stratification can no longer be sustained by conduction and the fluid disrupts from its stable, quiescent state into an unstable, convective mode in which lighter and heavier gas mix. This paper suggests that such a statement is not necessarily true for gaseous media under normalized temperature differences that are much larger than justifiable for the Boussinesq approximation! In fact, there may be situations where a system cannot ever be made unstable with respect to the onset on buoyant convection no matter how large the temperature (density) difference becomes at a given pressure even under normal gravity! This "unexpected" behavior is primarily attributed to highly temperature-sensitive kinematic viscosity which counteracts the tendency toward instability and dampens convection by making the gas more "viscous" at higher temperatures. This compensation of the buoyant force by the viscous force exhibits itself by the formation of a peak hot-surface temperature beyond which a system will tend to be more stable as the hot-surface temperature increases. |   |  |                                   |
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