# ФИЗИКО-МАТЕМАТИЧЕСКИЕ НАУКИ

## ON CONVECTION STABILITY OF GAS-VAPOR MIXTURE AT CLOSE TO CRITICAL TEMPERATURE

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**ABSTRACT.**

A new physical and mathematical model of Rayleigh-Benard convection in a gas-vapor mixture of oxygen and cyclohexane is proposed, where the dependence of density on temperature has its maximum. Linear analysis of stability is performed, two threshold cases are analyzed when the density inversion parameter is large or small enough. Formulae for the growth increment and attenuation, the neutral curve and the boundary of the instability domain on the wave plane have been obtained.

**АННОТАЦИЯ.**

Предложена новая физико-математическая модель конвекции Рэлея-Бенара в газо-паровой смеси кислорода и циклогексана, где зависимость плотности от температуры имеет максимум. Выполнен линейный анализ устойчивости, аналитически исследованы два предельных случая, когда параметр инверсии стремится к своим предельным значениям. Получены формулы для инкремента нарастания и затухания, нейтральной кривой и границы области неустойчивости на волновой плоскости.

**Keywords:** Rayleigh-Benard convection, Rayleigh number, Prandtl number, linear theory, increment.

**Ключевые слова**: конвекция Рэлея-Бенара, число Рэлея, число Прандтля, линейная теория, инкремент.

**Introduction**

Convection in the flows with monotonous dependence of density on temperature has been analyzed and described in details in [1,2]. However, in natural conditions and numerous important applications, such as transportation of hydrocarbon through pipelines, convection of cold water in the process of glacial melting [3-7], convection of inert and reactive gas-vapor mixture in chemical reactors and processing systems etc., the density of the convective flows is characterized by a non-monotonous, nonlinear function of the temperature with a clear maximum at its critical point. These applications explain the necessity of studying convection in such flows. The most complicated thing in terms of theory and practical application is to study a whole range of convection regimes in the vicinity of the maximum density point, where the buoyancy force changes its sign [6,7].

We would like to emphasize that a physically natural hypothesis about the cyclohexane condensation on the walls and, hence, formation of a more viscous liquid film on them allows us to consider the boundaries impermeable and free from viscous shear [5,9]. Our analysis of the dependence of the heat expansion coefficient on temperature shows that it is reasonable to consider the coefficient to be a piecewise constant temperature function as in [8].

We see that the problem of Rayleigh-Benard convection in such a gas-vapor mixture asymptotically turns into the classical Rayleigh problem of convective instability in incompressible fluids with impermeable horizontal boundaries in the absence of viscous shear [1], if the density maximum is reached at one of the horizontal boundaries.

We also see an evident analogy with the penetrating convection of cold water close to the point of maximum density, where the heat expansion coefficient goes through zero but is a linear temperature function [3,4], which allows us to compare and verify the model proposed [5].

In particular, the analogy with the penetrating convection of cold water suggests that if the critical temperature is higher than that of the cold boundary and lower than that of the heated one, the layer is divided into two sublayers, and the instability may develop only in the lower sublayer, whereas the upper one is always stable. The situation does not change depending on whether we heat the lower or the upper layer [4].

Similar to the convection of cold water, a decrease in the relative thickness of the lower, unstable layer is likely to cause flow stabilization [3,4].

In this article, we aim at describing a new physical and mathematical model of Rayleigh-Benard convection in a gas-vapor mixture of oxygen and cyclohexane taking into account the evaporation and condensation cyclohexane processes, and provide the results of the linear analysis of stability.

**Physical properties of a gas-vapor mixture**

Convection of a gas-vapor mixture with the oxygen $O_2$ and liquid-vapor cyclohexane $C_6H_{12}$ is considered in view of a possible condensation of the latter on the boundaries. To be specific, the total cyclohexane mass fraction, both in condensed and vapor forms, is set as $\beta_0 = 0.524$.

At the critical temperature $T_{cr}$, all the cyclohexane added to the system evaporates. Thus, when $T < T_{cr}$, the cyclohexane is present as a liquid vapor condensed on the boundaries and as a saturated vapor, with the rise of temperature leading to a rapid growth of the saturated cyclohexane pressure according to (1) and a corresponding growth of the density of the gas-vapor mixture. When $T > T_{cr}$, the cyclohexane is present only as an unsaturated vapor with a relatively small change in its partial pressure, and a further increase in temperature leads to a decrease in the density, according to the ideal gas-state equation.

Due to a small temperature change in the Boussinesq approximation used, the partial pressure of the oxygen is considered fixed and equal to *1 atm*. The total pressure $P$ includes the pressure of the oxygen and the vapor: $P = 1 + P_{sv}$.

When $T < T_{cr}$, the pressure of the saturated cyclohexane (atm) can be calculated according to the known absolute temperature:

$$P_{sv}(T) = 9.87 \cdot 10^{2.9764 - \frac{1206.5}{T - 273.15 + 223.14}}. \quad (1)$$

When $T > T_{cr}$, the cyclohexane is unsaturated and has a relatively week dependence on temperature, which can be neglected.

The molar weight of the gas mixture $\mu$ *(kg/kmol)* is determined by

$$\mu(T) = (1 \cdot 32 + P_{sv}(T) \cdot 84) / (1 + P_{sv}(T).$$

Here, *32kg/kmol* and *84kg/kmol* are molar weights of the oxygen $Q_2$ and cyclohexane $C_6H_{12}$, respectively. When $T > T_{cr}$, the molar weight of the gas-vapor mixture in the approximation described is constant.

$$\rho = (1 + P_{sv})\mu(T) / RT.$$

The ideal gas density can be determined according to the equation of state for the ideal gas (Fig.1):

Here $R$ is the is the universal gas constant. The temperatures of the cold and heated boundaries in Fig.1 are arbitrary, shown just to explain the formulation of the problem, and do not correspond to the real values.

The thermal expansion coefficient β can be calculated from

$$\beta = -\frac{1}{\rho}\frac{d\rho}{dT} = -\frac{1}{B}\frac{dB}{dT},\ B = (1 + P_{sv})(8 + 21 \cdot P_{sv}) / T.$$

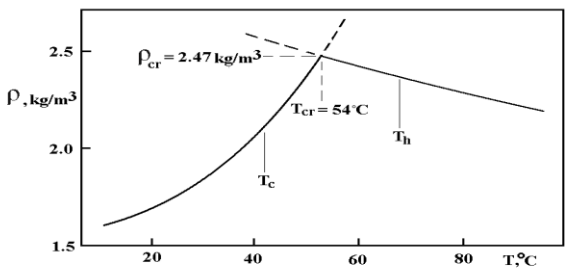


*Fig. 1. Density of the gas-vapor mixture.*

In our case, the density maximum is achieved inside the layer. The analysis of the thermal expansion coefficient dependence on the temperature proves that the piecewise-constant approximation with a discontinuity at the critical temperature is correct, with the coefficient ranging in the interval between *0.003054* in the lower unstable layer and *-0.01583* in the upper stable one. We should notice that at the point close to the critical temperature the kinematic viscosity $\nu$ and the thermal diffusivity $\chi$ are continuous functions and in terms of the Boussinesq approximation can be considered constant.

The critical (maximum) value of the saturated cyclohexane pressure is determined by its total mass fraction, both in condensed and vapor forms, $\beta_0$ and the corresponding critical temperature is found from reversed Eq. (1). Thus, the given $\beta_0 = 0.524$ corresponds to the critical values of the saturated gas pressure $P_{sv} = 0.4191atm$ and temperature $T_{cr} = 54.32°C\ (327.5K)$.

As it was stated above, the temperature of the upper cold horizontal boundary $T_c$ is lower than the critical temperature $T_{cr}$, and the temperature of the lower heated horizontal boundary $T_h$ is greater than that, hence $T_c < T_{cr} < T_h$. Similar to the case with cold water convection, the maximum density line where $T = T_{cr}$ divides the layer into two sublayers, the instability can develop only in the lower layer with the upper one being always stable [4]. Relative thicknesses of the sublayers are characterized by the inversion parameter $\tau = d/H = (T_h - T_c r)/(T_h - T_c)$, $0 < \tau < 1$, which shows the ratio of the height of the unstable sublayer $d$ to the thickness of the whole layer $H$. In the ultimate cases when $\tau = 1$ and $0$, the set of the problem turns to the classical Rayleigh problem with $\beta = 0.003054$ and $-0.01583$ respectively [1]. Similar to the case with cold water convection, the analysis of the density profile form (Fig.1) shows that the situation does not change at heating from above [4].

**Linear analysis**

By analogy with [1], in order to investigate convective stability in linear approximation, we obtain a set of equations:

$$\omega_t = \Delta\omega + C(\tau, y)\cdot Ra\cdot Q_x,\ \Delta\psi = -\omega,\ Q_t = (\Delta Q - \psi_x)/\Pr. \quad (2)$$

Here, factor $C(\tau,y)$ is determined as:

$$C(\tau, y) = 1,\ 0 \le y \le \tau \ \text{and}\ -5.183,\ \tau < y \le 1.$$

The stream function $\psi$ and vorticity $\omega$ were derived from $u = \psi_y$, $v = -\psi_x$ and $\omega = v_x - u_y$, and $Q$ is the deviation of temperature from the linear profile. Characteristic values are chosen as the layer thickness $H$ for the length, $\chi/H$ for velocity, $H^2/\nu$ for time, $\rho_0\nu\chi/H^2$ for pressure, $1/T_{cr}$ (in the unstable sublayer) for the coefficient of thermal expansion and $\delta T = T_h - T_c$ for temperature, where the density inversion parameter $\tau = d/H = (T_h - T_{cr})/(T_h - T_c)$, $0 < \tau < 1$ is the ratio of the height of the unstable sublayer $d$ to the height of the whole layer $H$. Here $Ra = gH^3\delta T/(\chi\nu T_{cr})$ and $Pr = \nu/\chi$ are the Rayleigh and Prandtl numbers.

In order to solve system (2), we use the Galerkin method provided that $\tau$ is not too small [1,10] and the orthogonalization method if $\tau$ is small.According to the Galerkin method, an approximated solution of (2) is considered as:

$$\omega(t,x,y) = \exp(-\lambda t)\sin(\alpha x)\sum_{m=1}^{N}\omega_m \sin(m\pi y),\ \psi(t,x,y) = \exp(-\lambda t)\sin(\alpha x)\sum_{m=1}^{N}\frac{\omega_m}{S_m}\sin(m\pi y),$$
$$Q(t,x,y) = \exp(-\lambda t)\cos(\alpha x)\sum_{m=1}^{N}Q_m \sin(m\pi y),\ S_m = \alpha^2 + m^2\pi^2. \quad (3)$$

Here $\lambda$ is the eigenvalue (increment), and $\alpha$ is a wave number. Negative λ values correspond to instability, while positive ones to the stable flow.

By substituting (3) into (2) and following the standard Galerkin method [1,10], we obtain the condition of non-trivial solution existence in the form of determinant:

$$det(A - D) = 0, \quad (4)$$

where $A$ is a square matrix with the components $A_{km}$

$$A_{mk} = A_{km} = \int_0^1 C(\tau, y)\sin(m\pi y)\sin(k\pi y)dy,\quad A_{mk} = -(a+1)\left(\frac{\sin(2m\pi\tau)}{2m\pi} - \tau\right)/2 - a/2,\ m = k,$$
$$A_{mk} = \frac{a+1}{\pi(k^2 - m^2)}(m\cos(m\pi\tau)\sin(k\pi\tau) - k\cos(k\pi\tau)\sin(m\pi\tau)),\ m \ne k.$$

and $D$ is a diagonal matrix with the components $d_{mm}$:

$$d_{mm} = \frac{S_m(S_m - \lambda)(S_m - \lambda\Pr)}{2Ra\cdot\alpha^2},\qquad 1 \le m \le N.$$

Consider now asymptotic ratios with $\tau$ close to $1$, when the density maximum is achieved at the upper horizontal boundary and the problem considered asymptotically turns into the classical Rayleigh problem [1].

From (5), one can find $A_{mk}$:

$$A_{mk} = \frac{1}{2} - \frac{1}{3}m^2\pi^2(1+a)(1-\tau)^3,\ m = k;\ \ A_{mk} = \frac{1}{3}mk\pi^2(1+a)(1-\tau)^3(-1)^{m+k+1},\ m \ne k.$$

The equations show that matrix $A$ at $\tau$ close to $1$ has a diagonal dominance, and the equations for dominant harmonic components can be considered separately. However, it is due to these harmonic components that the flow stability and other characteristics are determined [1,2,10].

Test calculations demonstrate that at $\tau$ close to $1$ the neutral curve and perturbation growth increments can be calculated accurately taking into account only one harmonic component $(N = 1)$.

When $N = 1$, the neutral curve has the form:

$$Ra = \frac{S^3}{2A_{11}\alpha^2},\ Ra = \frac{S^3}{\alpha^2}(1+\frac{2}{3}\pi^2(1+a)(1-\tau)^3) = \frac{S^3}{\alpha^2}(1+40.68\cdot(1-\tau)^3),\ S = \alpha^2+\pi^2. \quad (5)$$

Hence, one can obtain the critical value of the Rayleigh number (minimum of $Ra$ versus $\alpha$):

$$Ra_{cr} = 6.75\cdot\frac{\pi^4}{2A_{11}} = \frac{657.511}{2A_{11}} = 657.511\cdot(1+\frac{2}{3}\pi^2(1+a)(1-\tau)^3), \quad (6)$$
$$Ra_{cr} = 657.511\cdot(1+40.68(1-\tau)^3),\ \alpha_{cr} = 2.221.$$

For the growth increment $\lambda$, we find that

$$\lambda = \frac{S}{2}(\frac{1+\Pr}{\Pr}) - \sqrt{\frac{S^2}{4}(\frac{1-\Pr}{\Pr})^2 + \frac{2Ra\,\alpha^2 A_{1,1}}{S\Pr}}, \quad (7)$$
$$\lambda = \frac{S}{2}(\frac{1+\Pr}{\Pr}) - \sqrt{D} + \frac{\pi^2(a+1)Ra\,\alpha^2}{3\Pr S\sqrt{D}}\cdot(1-\tau)^3,\ D = \frac{S^2}{4}(\frac{1-\Pr}{\Pr})^2 + \frac{Ra\,\alpha^2}{S\Pr}.$$

According to (7), one can see that the increment values obtained differ from those for the classical Rayleigh problem by about $O(1\text{-}\tau)^3$ [1], and decrease in $\tau$ leads to a more stable flow.

Fig.2 shows the function of the wave number $\alpha$ as a growth increment $\lambda$, with numbers on the curves showing corresponding $\tau$ values when $Ra = 10\cdot Ra_{cr}$, $Ra_{cr} = 657.511$ and $Pr = 0.71$. Fig.3 shows the instability domain on the wave plane, with solid curves showing the boundary of the instability domain for Rayleigh problem $(\tau = 1)$, and dashed curves showing boundaries at $\tau = 0.8$ and $0.85$. Figures 2 and 3 show flow stabilization as $\tau$ decreases.

Consider now asymptotic ratios with $\tau$ close to $0$, when the density maximum is achieved at the lower horizontal boundary.

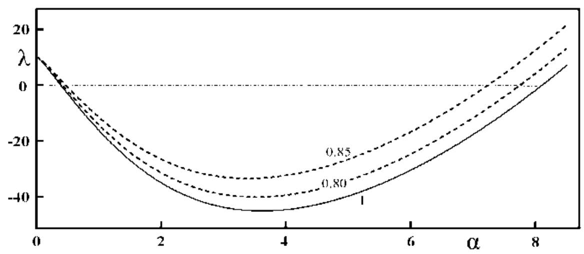


*Fig. 2. Growth increment.*

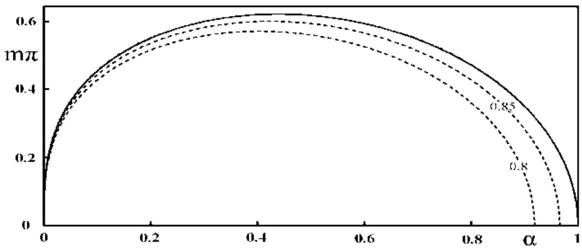


*Fig. 3. Instability domain.*

One can obtain for $A_{mk}$

$$A_{mk} = -\frac{1}{2}a + \frac{1}{3}m^2\pi^2(1+a)\tau^3,\ m=k;\ A_{mk} = \frac{1}{3}mk\pi^2(1+a)\tau^3,\ m\neq k.$$

These equations show that matrix $A$ at $\tau$ close to $0$ has a diagonal dominance, and the equations for dominant harmonic components can be considered separately. It follows from the first of the equations above for the neutral curve (7) that given $\tau$ close to $0$ all the disturbances attenuate.

We can show that attenuating disturbances at $\tau$ close to $0$ are typically oscillatory, which appears if Rayleigh number exceeds the threshold value $3.757$ [1].

Let us write down the equation for increment $\lambda$ at $\tau$ close to $0$ with $Pr = 1$ for simplicity:

$$\lambda = S - i\,\alpha\sqrt{a\,Ra/S}\cdot(1-\frac{\pi^2(a+1)}{3a}\tau^3),\ \lambda = S - 2.277\,i\,\alpha\sqrt{Ra/S}\cdot(1-3.925\tau^3).$$

The equation shows that the attenuation rate, which is determined by the real part of the increment $\lambda$, does not depend on inversion $\tau$, and its growth (or attenuation) leads to a small decrease (increase) in the damped oscillation frequency.

Let us now compare critical Rayleigh numbers and corresponding wave numbers obtained in the present paper for the convection in cold water [3-5] and for the classical Rayleigh problem [1]. Figures 4 and 5 show the scale of length as the height of the unstable sublayer, and our data were recalculated as $Ra_{cr}\cdot\tau^4$ and $\alpha_{cr}\cdot\tau$, respectively.

The solid curves in Fig. 4 and 5 show our results (line 1) and the data for convection in cold water ([4] - line 2 and [3] - dots), whereas the dotted line corresponds to the classical Rayleigh problem [1]. The dot-and-dash line in Figure 4 shows the result of the experiment with the convection of cold water poured on ice [5].

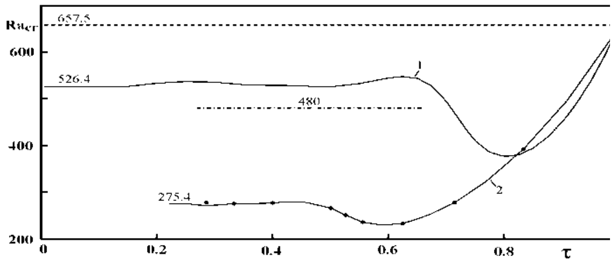


*Fig. 4. The critical value of the Rayleigh number.*

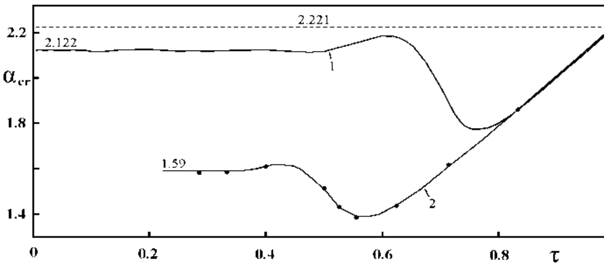


*Fig. 5. The critical value of the wave number.*

According to the data in Figures 4 and 5, we can see that at $0.8 < \tau \leq 1$ the results of our work are close to those of convection in cold water, and at $\tau = 1$ they coincide with the solution of the classical Rayleigh problem, while in the domain of infinitesimal $\tau$ they asymptotically tend to the constants corresponding to the asymptotes of $Ra_{cr} = 526.4/\tau^4$ and $\alpha_{cr} = 2.122/\tau$.

We should notice that according to the critical Rayleigh number value, the results of experimental studying for convection in cold water [5] are essentially close to our results (with a 9.6% deviation), especially if we compare the former with the theoretical results [3,4], which give a deviation of about 43%. A possible reason for this might be the fact that in both experiment [5] and our work the heat expansion coefficient $\beta$ was considered constant and equal to its average value on horizontal boundaries. Due to this, our results are also much closer to the results of solving the classical Rayleigh problem with constant $\beta$ [1] than the theoretical results for convection in cold water [3,4].

**Conclusion**

In present work we have described a new physical and mathematical model of Rayleigh-Benard convection in a gas-vapor mixture of oxygen and cyclohexane taking into account cyclohexane evaporation and condensation on the boundaries of the domain. It is shown that at a certain (critical) temperature, when all the added liquid cyclohexane has evaporated, the density of the gas-vapor mixture reaches a local maximum, where the heat expansion coefficient and the buoyancy force in the equation of motion change their sign going through zero.

We conducted linear analysis of stability and analyzed data obtained through calculating nonlinear steady conditions of Rayleigh-Benard convection.

A physically natural supposition about cyclohexane condensation on solid walls and, hence, a more viscous liquid film on the walls allows us to consider the boundaries of the domain horizontal and free from shear strength, which simplified the task.

We showed that the problem of Rayleigh-Benard convection in the gas-vapor mixture considered turns asymptotically into the classical Rayleigh problem if the density maximum is achieved on one of the horizontal boundaries. In both threshold cases we obtained analytical asymptotic formulae for stability characteristics. In terms of one mode approximation, expressions for the growth increment (or attenuation) of the first major mode were obtained. The boundary of the instability domain was also studied on the wave plane.

At $\tau$ close to $1$, a decrease in the inversion parameter $\tau$ leads to a growth in the flow stability, and at $\tau$ close to $0$ it leads to a growth in the damped oscillation frequency, but the rate of decay is constant.

Having solved the linear stability problem, we see that a decrease in the relative thickness of the lower, unstable sublayer or a corresponding growth in the relative thickness of the upper stable sublayer cause stabilization of the flow.

We draw a quality analogy between the convection in the gas-vapor mixture considered and the penetrating convection of cold water at close to maximum density point, where the heat expansion coefficient goes through zero but is a linear function of the temperature.

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