Pattern formation in binary mixture convection in cylindrical three-dimensional cells

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Abstract. We present numerical results of pattern selection near the onset of convection for a water-ethanol mixture in a cylindrical container heated from below. Parameter values and boundary conditions relevant to experiments are used. The separation ratio of the mixture we consider is $S = -0.09$ and the radial aspect ratio of the cell is $\Gamma = 11$. The onset of convection occurs via a subcritical Hopf bifurcation. Slightly above the onset, in the linear regime, a $m = 1$ azimuthal mode consisting of radially travelling waves grows in amplitude. As convection evolves, the pattern focuses into one or several diameters of the cell. We do not observe a direct transition to a stable nonlinear state. Instead, collapses in the convection amplitude followed by subsequent growths take place. This behaviour resembles some experimental observations. The numerical results are obtained with an efficient time-evolution spectral code that solves the full convection equations in cylindrical coordinates.

1. Introduction
Convection in vertically heated binary-liquid mixtures is an excellent system for the study of pattern formation, especially for negative separation ratio mixtures, $S < 0$. In such mixtures, the primary bifurcation is subcritical and gives rise to a state of oscillatory convection. In large annular and rectangular containers, the linearly unstable state at the onset usually evolves either to a travelling wave state or to stationary rolls that are called states of stationary overturning convection. Nevertheless, experiments on these geometries, usually performed on water-ethanol mixtures, show that a great variety of states can arise near the onset of convection, including states of localised travelling-wave convection (pulses of travelling waves coexist with regions of quiescent fluid), states characterised by repeated bursts of amplitude or regimes exhibiting spatiotemporal chaos [1]-[5]. If the container is sufficiently narrow the resulting system is approximately two-dimensional and can be modelled neglecting the effect of the thickness of the cell. The numerical work dealing with the study of several aspects of the dynamics in two-dimensional containers is abundant (i.e. [6]-[9]).

With the aim of showing if the same types of states are found in truly three-dimensional geometries, a few experiments on cylindrical cells have been done [10]-[13]. The results of these experimental works indicate that new behaviour prevails. Waves that travel in the radial direction are present and travelling-wave convection patterns typically consist of several competing domains of travelling waves propagating in different directions. Transient localised pulses of travelling-wave convection similar to the states found in annular cells were observed, but these pulses either decayed back to pure conduction or grew to fill the cell.
In the experimental observations of Lerman et al. [10, 11, 12], $S \approx -0.08$ mixtures are used in cylindrical containers of aspect ratio $\Gamma \approx 11$ ($\Gamma = 10.91, 11.53$). Convection immediately above the onset consists of a superposition of radially inwards and outwards travelling waves filling the cell. The wave amplitude presents a sinusoidal azimuthal modulation, which is very sensitive to the actual value of the aspect ratio. While for the $\Gamma = 10.91$ cell the dominant azimuthal modes are odd, a small variation of 0.5 in the value of $\Gamma$ causes the even modes to be favoured at the onset. As the amplitude of convection grows, higher azimuthal modes become important and convection gets localised along one or more diameters of the cell. The focused lines of convection then collapse and can result in radially localised pulses, very similar to the ones observed in long annular cells, or in localised but disordered regions of convection. However, the localised states are not stable and always lead to a state in which the entire container is filled with convection rolls or they decay back to the conduction state and the process begins again. Apart from these confined states, in one occasion a wall state consisting of a narrow ring of azimuthal travelling waves very close to the wall and pure conduction in the interior has been observed [11].

The other available experimental study on binary mixtures in cylindrical containers is the work of La Porta and Surko [13]. They consider a mixture with a much stronger Soret coupling, $S = -0.24$, and survey the patterns arising in a $\Gamma = 26$ aspect ratio cell. The larger aspect ratio cell decreases the influence of the boundaries on the dynamics. Disordered states consisting of many small domains of travelling waves in which a significant part of the pattern does not interact with the boundaries are observed. Over time, these domains increase their size and the boundaries again become important. The resulting patterns typically consist of three or four domains of travelling waves whose dynamics seems to be controlled by the domain boundaries. The character of the patterns changes as the value of the control parameter increases, and rotating patterns are observed. Above a certain value, stationary overturning convection consisting of regions of straight rolls is observed.

The numerical work on this system is scarce. As far as we know, there are no available three-dimensional direct numerical simulations of the Navier-Stokes equations on binary-fluid convection on extended systems, since these types of three-dimensional computations are costly. The only previous numerical work on this system deals with the stability analysis of the conduction state in a cylindrical container [14], and shows that the eigenfunctions take the form of different types of right-handed or left-handed rigidly rotating spirals, or of convection rolls which travel radially inwards and outwards.

The present work is concerned with the direct numerical simulation of binary convection in shallow three-dimensional cells. We have developed an efficient three-dimensional time-evolution spectral code that solves the full convection equations in cylindrical coordinates. We present the first results we have obtained, showing the patterns arising in a $\Gamma = 11$ cylindrical cell. The parameters we consider are those used in one of the stability analysis presented in [14], which are very similar to the experimental values used in [10]-[12].

With our work we want to contribute to the understanding of the dynamics in three-dimensional binary mixtures convective layers, since many features remain unclear. Confined convection similar to that observed in two-dimensional systems has been obtained experimentally, but unlike in those systems, the pulses of convection seem not to persist indefinitely in cylindrical cells. A more detailed exploration, varying the value of the separation ratio, which is known to influence strongly the dynamics, is needed to confirm whether stable confined convection is possible or not and to provide an explanation. Another fundamental question is to what extent the arising dynamics is influenced by the lateral boundaries (shape and size) or it is determined mainly by the intrinsic features of the system. This problem is addressed in a very recent experimental work [15], where the global dynamics of travelling-wave patterns in circular, rectangular and stadium-shaped cells is analyzed.
2. Mathematical model
We consider Boussinesq binary fluid convection in a cylinder of height $d$ and radius $R$. The radial aspect ratio of the cylinder is defined as $\Gamma = R/d$. The cylinder is heated from below, being $\Delta T$ the temperature difference between the lids, and the mixture is in the presence of a vertical gravity $g = -g\hat{e}_z$. The nondimensional equations that describe the dynamics are

$$\nabla \cdot \mathbf{u} = 0,$$
$$\partial_t \mathbf{u} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla p + \sigma \nabla^2 \mathbf{u} + Ra\sigma [(1 + S)\Theta + S\eta] \hat{e}_z,$$
$$\partial_t \Theta + (\mathbf{u} \cdot \nabla) \Theta = w + \nabla^2 \Theta,$$
$$\partial_t \eta + (\mathbf{u} \cdot \nabla) \eta = -\nabla^2 \Theta + \tau \nabla^2 \eta.$$

Here, $\mathbf{u} = (u, v, w)$ is the velocity field in cylindrical coordinates $(r, \phi, z)$, $\Theta$ and $\eta$ denote the departure of temperature and concentration from the conduction profile, $\Theta = (T - T_c)/\Delta T$ and $\eta = -(C - C_c)/(C_0(1 - C_0)S_T\Delta T) - \Theta$, where $T$ and $C$ are the fields of temperature and concentration of the denser component, $T_0$ and $C_0$ are their mean values and $S_T$ is the Soret coefficient. Binary fluid convection is described by four dimensionless numbers, the Rayleigh number $Ra$, the Prandtl number $\sigma$, the Lewis number $\tau$ and the separation ratio $S$, defined as

$$Ra = \frac{\alpha \Delta T gd^3}{\kappa \nu}, \quad \sigma = \frac{\nu}{\kappa}, \quad \tau = \frac{D}{\kappa}, \quad S = C_0(1 - C_0)\frac{\beta}{\alpha}S_T,$$

where $\alpha$ and $\beta$ are the thermal and concentration expansion coefficients, $\kappa$ and $D$ are the thermal and mass diffusivities and $\nu$ is the kinematic viscosity. The Rayleigh number is the control parameter of the system and measures the strength of the imposed temperature gradient. The Prandtl number relates momentum diffusion to heat diffusion, while the Lewis number relates concentration diffusion to heat diffusion. The separation ratio gives the coupling between the thermal and concentration density gradients. A negative value of $S$ indicates that the concentration density gradient opposes the thermal density gradient, and tends to stabilise the fluid layer against thermal convection. For small $\tau$ and sufficiently negative values of $S$, the onset of convection is a Hopf bifurcation to a state of oscillatory convection.

For the boundary conditions, we consider a no-slip, no-flux, fixed temperature boundary at the top and bottom plates, and a no-slip, no-flux, insulating boundary on the lateral wall

$$\mathbf{u} = \Theta = \partial_z \eta = 0 \quad \text{on} \quad z = 0, 1,$$
$$\mathbf{u} = \partial_r \Theta = \partial_z \eta = 0 \quad \text{on} \quad r = \Gamma.$$

As a measure of the heat transport by convection, we use the Nusselt number $Nu$, defined as the ratio of heat flux through the top plate to that of the corresponding conductive solution:

$$Nu = 1 - A^{-1} \int_A \partial_z \Theta dA,$$

where $A$ is the area of the cylinder lids.

To integrate the equations in time we have used the second order time-splitting method proposed in [16] combined with a pseudo-spectral method for the spatial discretization, Galerkin-Fourier in the azimuthal coordinate $\phi$ and Chebyshev collocation in $r$ and $z$. The radial dependence of the functions is approximated by a Chebyshev expansion between $-R$ and $R$, but forcing the proper parity of the variables at the origin [17]. For instance, the scalar field $\Theta$ has an even parity $\Theta(-r, \phi) = \Theta(r, \phi + \pi)$, the vertical velocity $w$ and $\eta$ obey the same even parity condition, whereas $u$ and $v$ are odd functions. To avoid including the origin in the mesh grid, we have used an odd number of Gauss-Lobatto points in $r$ and we have enforced
the equations only in the interval $(0, R]$. We have used the standard combination $u_+ = u + iv$ and $u_- = u - iv$ in order to obtain, as a result of the splitting, Helmholtz equations for all the variables $\Theta, \eta, w, u_+$ and $u_-$. For each Fourier mode, these equations have been solved using a diagonalisation technique in the two coordinates $r$ and $z$. The imposed parity of the functions guarantees the regularity conditions at the origin needed to solve the Helmholtz equations [18].

3. Results

In this section we present results for a water-ethanol mixture with parameters $S = -0.09$, $\sigma = 24$ and $\tau = 0.008$. We analyse pattern formation near the onset of convection in a cylinder of aspect ratio $\Gamma = 11$. This choice of parameters is motivated by the experiments of Lerman et al [10]-[12], and are the same than those used in the linear analysis of Mercader et al [14]. We tested our time evolution code with linear stability results and we reproduce the critical Rayleigh number $Ra_c = 1917$ and frequency $\omega_c = 6.16$ reported in [14] for the parameters we consider.

As an indication of the arising patterns close to the convection threshold, we present here the results for $Ra = 1924$, a slightly supercritical Rayleigh number. To obtain the results we have used a mesh grid of $N_r = 54$, $N_z = 20$ and $N_\theta = 128$ points in the radial, vertical and azimuthal directions, respectively. The time step we have considered is $\Delta t = 5 \cdot 10^{-4}$. The same results are obtained with $N_\theta = 256$ and $\Delta t = 4 \cdot 10^{-4}$. Fig. 1 shows the time series obtained when the system is allowed to evolve at $Ra = 1924$, by using as initial condition a profile having a small Gaussian noise in the temperature field of the $m = 1$ azimuthal Fourier mode, which is the critical mode, and zero in the rest of modes and fields. The first series shows the variation of the Nusselt number, while the other three correspond to the time-dependence of the real part of the $m = 1$, $m = 2$ and $m = 3$ azimuthal modes of the vertical velocity at a given $(r, z)$ point.

To visualise the solutions, Fig. 2 shows the contour plots of the temperature field for several
time instants. In order to have an indication of the amplitude of convection, each figure includes the value of the Nusselt number. The time appearing in the plots is written in units of vertical thermal diffusion time, $d^2/\kappa$.

The first outstanding feature of the dynamics is that there is not a direct continuous evolution towards a stable nonlinear regime. The time series of the Nusselt number shows that repeated bursts of amplitude take place. When the amplitude of the Nusselt number is growing a sudden collapse that brings the system back to a small-amplitude state is produced. Then, the amplitude begins to grow again and the process repeats aperiodically. As can be seen in Fig. 1, during this regime the odd azimuthal modes are much larger in amplitude than the even modes. Although the $m = 1$ mode is the dominant one, other odd modes become important from a very early stage of the evolution. The presence of several azimuthal modes of the same order indicates that the critical Rayleigh number is nearly independent of the azimuthal mode. This is in agreement with the linear stability results of Mercader et al [14]. They obtained that the difference between

![Contour plots of the temperature field in $z = 0.5d$ showing the evolution of the pattern for a Rayleigh number $Ra = 1924$, slightly above the onset of convection $Ra_c = 1916$.](image)

Figure 2.
the critical Rayleigh numbers for the \( m = 1 \) and \( m = 3 \) modes is 0.02\% and found that the critical Rayleigh numbers for low-order odd modes are smaller than those for the low-order even modes.

During the linear transient, a \( m = 1 \) azimuthal mode begins to grow. The first three plots in Fig. 1 show the spatial structure of this solution. For \( t = 106 \) the amplitude of convection is really small, \( \text{Nu} - 1 \approx 10^{-6} \), and it grows up to \( \text{Nu} - 1 \approx 10^{-4} \) and \( \text{Nu} - 1 \approx 10^{-3} \) for \( t = 148 \) and \( t = 188 \). The pattern consists of radially inwards and outwards travelling waves and it is a nearly standing wave in the azimuthal direction. This structure results from the superposition of two counter propagating spiral modes, corresponding to the spiral eigenfunctions obtained in the linear stability analysis [14]. The eigenfunctions consist of right-handed and left-handed spirals travelling in the azimuthal direction in opposite senses. In \( t = 188 \) the influence of modes different from \( m = 1 \) can already be noticed, since a convection line along a diameter of the cell begins to develop.

As convection evolves, the pattern focuses into one or more diameters of the cell. This effect can be observed in the fourth plot in Fig. 2, which corresponds to \( t = 215 \). For this time instant, the Nusselt number is still growing, \( \text{Nu} - 1 \approx 0.5 \times 10^{-2} \), and the amplitude of convection is clearly larger along two diameters of the cell. For a time instant of about \( t = 220 \), a collapse in the amplitude of convection takes place. The plot in Fig. 2 corresponding to \( t = 228 \) shows how the convection pattern looks like shortly after the collapse. The Nusselt number has decreased an order of magnitude, and convection seems to persist only along three diameters, being its amplitude stronger in the center of the cell. In \( t = 248 \) and \( t = 268 \), during the second growth of the Nusselt number, we observe that convection is enhanced along only one diameter of the cell. But again a collapse of the convection pattern, followed by a new growth, takes place. In this occasion, the pattern fills a larger region of the cell during the growth of amplitude, as can be seen in Fig. 2 for \( t = 308 \). Finally, the last plot in Fig. 2 shows the solution for \( t = 348 \), after the third collapse of amplitude.

It is important to notice that during these processes of growth and decay of convection, the spatial structure of the patterns varies due to the different contribution of the azimuthal modes. For instance, we observe that the \( m = 2 \) mode, whose amplitude had remained rather small, begins to grow during the third collapse (for \( t > 325 \) its size is significant, as can be seen in Fig. 1). Nevertheless, more calculations are needed to see whether the system finally evolves to a stable nonlinear state or it remains indefinitely in these types of repeated transients and small-amplitude states.

The behaviour we obtain numerically is quite similar to the experimental observations reported in [12] for a \( S = -0.08 \) mixture in a \( \Gamma = 10.91 \) cylinder. For this geometry, the dominant modes near the threshold of convection were also odd azimuthal modes, and the exact value of the aspect ratio was found to influence strongly the selected mode. Apart from that, the azimuthal focusing along one or more diameters of the cell is also observed in the experiments. However, experiments report very confined small-amplitude states that we have not observed so clearly yet (the solution we show for \( t = 228 \) is quite confined in the center, but the structures along the diameter are still visible). In the experiment, the repeated transients and localised regions of convection persisted for many days, but at the end the system evolved towards a steady state in which the container was filled with stationary convection rolls.

4. Discussion

In this paper we present and analyse numerical simulations of binary convection in a cylinder slightly above the threshold of convection. We have considered a \( S = -0.09 \) binary mixture in a cell of aspect ratio \( \Gamma = 11 \). To obtain the results, we have developed an efficient time-evolution spectral code that solves the non linear equations in primitive variables. There is full agreement between the results obtained with our code and the linear stability results presented in [14]. We
reproduce the critical values of the Rayleigh number and frequency, and the dynamics we observe during the nearly linear transient growth is consistent with the dominant eigenfunctions. In this regime, we observe that a $m = 1$ azimuthal mode, travelling radially and drifting very slowly in the azimuthal direction, grows in amplitude. These types of modes result from the superposition of two counter propagating spiral eigenfunctions. Moreover, the nonlinear evolution of the arising patterns that we obtain agrees with the experimental observations reported in [10]-[12]. The relatively fast growth of spatial modes different from the dominant one produces a confinement of convection along one or more diameters of the cell during the nonlinear regime. Nevertheless, the system does not settles in a stable nonlinear state. Instead, collapses of convection and subsequent growths of amplitude take place aperiodically.

The repeated transients we obtain in the cylinder resemble the complex low-amplitude dynamics observed in very large aspect ratio annular containers ($\Gamma \approx 80$) for mixtures with a small value of $|S|$ (i.e.[5]). In that geometry, the system remains in small-amplitude states, and sudden bursts and collapses of amplitude are observed. It is believed that nonlinear dispersion, which is important in small $|S|$ mixtures, prevents the system from reaching a stable uniform nonlinear state. To confirm that the small $|S|$ value of the mixture plays a key role in the dynamics we observe in the cylinder, numerical simulations in larger $|S|$ mixtures should be performed.

For the time being, we have not found the localised states observed in some experiments for mixtures with parameters similar to ours [10]-[12]. In these experiments, unlike in convection in annular and rectangular containers, the highly confined states do not seem to remain stable.

Finally, we plan to compare the dynamics arising in the confined cylindrical cell, where the laterals walls play in principle an important role, with that of a periodic cell, where the dynamics is governed by intrinsic features of the system. To that aim, we have extended the two-dimensional time-evolution code used in [8] to three-dimensional rectangular periodic cells.

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