Different convection dynamics in mixtures with the same separation ratio

Kristina Lerman, Guenter Ahlers and David S. Cannell

[†]*Department of Physics and Center for Nonlinear Science*

University of California at Santa Barbara

Santa Barbara, CA 93106.

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Abstract

We report a study of convection in circular geometry using ethanol–water mixtures with two very different ethanol concentrations, but with the same negative separation ratio. The two mixtures show very different convective behavior. In the high concentration mixture, convection begins with a sequence of transients that always results in a cell filling state. In the low concentration mixture, there is a range of control parameter where a novel "wall" state is the apparently stable state. In other runs at the same value of the control parameter, repeated transients and aperiodically fluctuating localized regions of disordered convection persist for days.

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Studies of thermal convection in binary mixtures [1–5] have revealed a wealth of pattern–forming phenomena and enhanced theoretical efforts to study the behavior of closely related complex Ginzburg–Landau equations [6,7]. To date it has been generally assumed that liquid mixtures are fully described by three dimensionless parameters: the separation ratio ψ, and the Lewis and the Prandtl numbers, with the latter two being relatively unimportant. Mixtures with sufficiently negative ψ exhibit an initial Hopf bifurcation to traveling wave convection. This bifurcation is backward for most negative ψ values, and can result in novel phenomena including stable localized pulses in one dimension [1], unstable pulses in two dimensions [3], periodic and aperiodic "blinking" states [2,4] and "dispersive chaos" [5], corresponding to apparently random formation and collapse of convective pulses, observed in an annular convection cell. Most of the behavior observed in one dimension has been captured, at least qualitatively, by the complex Ginzburg–Landau equation [8,6,9].

In the course of experimental studies of ethanol–water mixtures in two–dimensional cylindrical cells, we used mixtures of 1.1wt.% and 25.0wt.% ethanol to achieve separation ratios of -0.078 and -0.082 respectively. Despite the similar values of ψ, the behavior was qualitatively different. In both cases convection began with linear traveling waves (TW) which propagated radially. These waves eventually focused azimuthally and then collapsed radially to form a localized region of convection surrounded by quiescent fluid. For the 25.0wt.% mixture, this region was often a long–lived localized pulse, similar in appearance to those studied in one–dimensional cells [1], but sometimes it was a region of disorganized TW convection. When the applied temperature difference ΔT was less than 0.1% above the critical temperature difference ΔT_c, the localized convective region died out, convection started in the region around it and grew to fill the cell with stationary convection rolls, a steady state of the system. The convective behavior in the 1.1wt.% mixture at similar value of the control parameter was different. In one run, an apparently stable state consisting of a narrow ring of convection rolls next to the cell wall, was observed to form. During other runs in the 1.1wt.% mixture at the same control parameter, convection began at irregular time intervals, collapsed to form a localized region of convection which died away, returning the cell to the conduction state for a variable period of time, after which convection began again. This behavior persisted apparently indefinitely, while for the 25.0wt.% mixture, the cell always filled ultimately.

The convection cell consisted of a single crystal sapphire top plate and a polished silver bottom plate separated by circular Delrin sidewalls. The upper surface of the sapphire was controlled to ± 1 mK by circulating water, and the bottom plate temperature was controlled to ± 0.1 mK. All experiments were carried out at fixed temperature difference ΔT, not at fixed heat current. The cell height was uniform to $\pm 0.05\%$, as determined interferometrically, and the cell diameter was 7.90 cm. The 25.0wt.% mixture was studied in a cell of height $d = 0.343$ cm ($\Gamma \equiv$ radius/height $= 11.53$), and the 1.1wt.% mixture in a cell of height $d = 0.345$ cm ($\Gamma = 11.44$). The 25.0wt.% mixture had $\psi = -0.082$, Prandtl number $=$ 23.7, Lewis number $= 0.007$, and $\tau_v \equiv d^2/\kappa = 114.6$ s, where κ is the thermal diffusivity of the mixture. The 1.1wt.% mixture had $\psi = -0.078$, Prandtl number $= 7.2$, Lewis number $= 0.008$, and $\tau_v = 85.5$ s. The ψ values were determined from measured values of the linear TW frequency [10]. The convective threshold was crossed quasistatically making steps of $0.13\%\Delta T_c$ every three hours. The threshold ΔT_c ($\epsilon \equiv \Delta T/\Delta T_c - 1 = 0$) was defined to be midway between the last point in the conduction regime and the first point for which convection occurred. For the 25.0wt.% and the 1.1wt.% mixtures, the critical temperature differences were $\Delta T_c = 2.244$ K and $\Delta T_c = 3.183$ K, respectively. The patterns were visualized using the shadowgraph technique.

Immediately above onset, convection consisted of a superposition of radially inward and outward traveling waves for both mixtures (Figure 1a and Figures 2b&h). The only observable difference was occasional azimuthal modulation of the roll pair nearest the cell wall (Figure 2b) in the 1.1wt.% mixture. In both mixtures convection localized azimuthally onto a line or lines along the diameter of the container, where the amplitude of convection became very strong, while it was weaker elsewhere in the cell (Fig. 1b&c and Fig. 2a&e). For $\epsilon < 10^{-3}$, this focused line(s) of convection always collapsed radially to form a localized convective pulse surrounded by quiescent fluid (Fig. 1e and Fig. 2d). For the 25.0wt.% mixture at this value of the control parameter ϵ, the localized pulse always decayed in amplitude, leaving a "hole" (Fig. 1f). Convection then grew around the "hole," leading to a steady state in which the entire container was filled with stationary convection rolls some 25 hours later.

Whereas for the 25.0wt.% mixture convection grew to a steady cell–filling state, the steady state that formed at a similar value of the control parameter, $\epsilon = 6 \times 10^{-4}$, was differentin the 1.1wt.% mixture. This apparently stable "wall" state was an approximately one roll wide ring of TW convection along the cell periphery with the convection rolls perpendicular to the cell wall (Fig. 3). There was no convection observable in the interior of the cell. The dimensionless angular TW frequency was 1.52 at $\epsilon = 2.5 \times 10^{-3}$, and fell to 0.96 at $\epsilon = 1.79 \times 10^{-2}$. The "wall" state was observed to form only once. In other runs in the 1.1wt.% mixture at $\epsilon = 6 \times 10^{-4}$, erratically repeated transients persisted for days. During this regime, convection grew at irregular time intervals, collapsed to form localized regions of convection, which died away, returning the cell to the conduction state for a variable amount of time, after which convection grew again. Images in Figure 2 show the typical states seen at different times at this value of ϵ.

Figure 4 shows the evolution of the Nusselt number for the two mixtures during three runs at similar values of ϵ. The Nusselt number N is the ratio of the total heat transport through the cell to the heat transport due to conduction. The record of N for the 25% mixture at $\epsilon = 7 \times 10^{-4}$ (upper curve) began immediately after image Fig. 1f was taken. In that run, at $950\tau_v$, the system reached a steady state in which the cell was filled with stationary convection rolls. The dashed line in Fig. 4 shows the time series of the Nusselt number for the 1.1% mixture at $\epsilon = 6 \times 10^{-4}$, that resulted in formation of the "wall" state (Fig. 3). During another run in the 1.1% mixture (solid lower curve), repeated transients persisted over a period of five and a half days ($\approx 5,500\tau_v$) at the same value of ϵ, until the run was terminated.

Figure 5 compares N vs. ϵ diagrams for the two mixtures. As shown in Fig. 5a, we observed only two stable branches for the 25.0wt.% mixture. The upper branch corresponds to the stationary rolls filling the cell, and the lower branch to pure conduction. Open symbols show data obtained while increasing ϵ, and solid symbols show data taken while decreasing ϵ. For $0 < \epsilon < 10^{-3}$ the upper branch was reached by decay of a pulse, leading to a "hole" as described above. For runs in which ϵ was increased beyond 10^{-3} during the initial linear transient, the behavior was different. There an azimuthally focused line of convection resulted in either a long–lived ($\geq 40h = 1256\tau_v$) pulse, which eventually spread, or a localized but somewhat larger region of disordered convection, which also spread to fill the cell.

Figure 5b shows N vs. ϵ for the 1.1wt.% mixture. Again, open symbols show data for increasing ϵ, and solid symbols show data for decreasing ϵ. In addition to the conducting and the cell filling branches shown by squares, two other branches are present. The triangles correspond to the "wall" state but do not indicate its full stability range, which we have not determined. The open circles correspond to repeated transients for which N is erratic, with the symbols giving the average value and the error bars the standard deviation of N.

Steinberg *et al.* reported [11] the observation of spatio–temporal chaos in a wide rectangular cell with an aspect ratio of 1:9:20. The mixture they studied was 25wt.% ethanol [15] and had separation ratio $\psi = -0.1$. The chaotic state was observed under conditions of constant applied heat flux, 0.4% to 1.4% above threshold. For these parameters, the chaotic state was stable, and it was characterized by alternating localization of a line of convection near either of the long walls. Occasionally, the line of convection split into one or two pulses near the long wall. We have observed an aperiodically fluctuating state in the 25.0wt.% ethanol mixture in a cylindrical container but only under conditions of constant applied heat flux. Under these conditions the chaotic state persisted for days, and it was characterized by a repeated azimuthal focusing of convection rolls along alternating diameters of the container. The line of enhanced convection broke up into weak pulses, which died, while another line of convection, roughly perpendicular to the first, was growing stronger. Such persistent, aperiodically fluctuating states were observed for a small value of heat flux just above threshold, and also when the heat flux was reduced to a slightly subcritical value.

While the chaotic dynamics were somewhat reminiscent of the repeated transient regime seen in the 1.1wt.% ethanol mixture, there are significant differences. In the 25.0% mixture, persistent aperiodic states were never observed at fixed ϵ. Nusselt number fluctuations were about ten times smaller in the constant heat flux experiments than in the low concentration mixture at a fixed ϵ. Finally, large amplitude localized regions of disordered convection never died out at a fixed value of heat flux, as they were observed to do in the 1.1wt.% mixture at constant ϵ.

We believe that the oscillations observed by Steinberg and ourselves at a fixed value of heat flux are related to those predicted by Busse and are fundamentally different from the chaotic states seen in the 1.1wt.% ethanol mixture at fixed ϵ. Busse suggested [12] that a system near an unstable backward branch can show periodic oscillations when the applied heat flux is kept fixed. Under these conditions, the system, in a sense, repeatedly falls off and is pushed back onto the unstable convection branch. However, unlike the theoretical prediction, the oscillations we observe are aperiodic.

The erratic states we observe bear close resemblance to the "dispersive chaos" seen in an annular cell with a 0.40wt.% ethanol mixture [5], which was attributed to strong nonlinear dispersion. However, our failure to observe chaotic dynamics in the 25.0wt.% mixture with a very similar separation ratio, leads us to believe that some other effect is responsible for the behavior we observe. The most obvious and striking difference between the two mixtures is the separation ratio's dependence on the concentration: for the 25.0wt.% mixture $\delta\psi/\delta c = 1.8$, and for the 1.1wt.% mixture $\delta\psi/\delta c = -6.5$. As far as we know, this "non–Boussinesq" effect has not been considered before. It is known that traveling waves induce a large scale concentration current [14,13]. Though this current does not change the mean ethanol concentration in one–dimensional systems, it affects the local stability properties of the fluid by changing the vertical concentration gradient. This mechanism contributes to the stability of one–dimensional localized pulses, because the vertical concentration gradient built up ahead of the pulse stabilizes the quiescent fluid and prevents the invasion of this region by convection rolls. It is not known how this picture changes when an extra degree of spatial freedom is added to the problem. We speculate that the difference in the separation ratio's dependence on concentration, coupled with concentration transport by convection may lead to the differences in the dynamics of the two mixtures.

In addition, a state similar to the "wall" state we observe, was found in numerical simulations of binary fluid convection [16]. In that study fluid parameters appropriate for the 25.0wt.% mixture convection were used.

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FIG. 2. Images of the repeated transients at $\epsilon = 6 \times 10^{-4}$ in the 1.1wt.% mixture. Images shown here are in most cases 5.3 hours apart. Convection never reached a steady state, but grew and died erratically for the length of the run (5 days). The images correspond to the Nusselt numbers in Fig. 4 (black dotted line) at times (a) $3620\tau_v$ (b) $3863\tau_v$ (c) $4062\tau_v$ (d) $4285\tau_v$ (e) $4358\tau_v$ (f) $4731\tau_v$ (g) $4954\tau_v$ (h) $5177\tau_v$ (i) $5400\tau_v$.

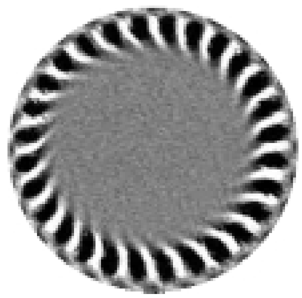

FIG. 3. An apparently stable "wall" state that formed in the 1.1wt.% ethanol mixture at $\epsilon = 6 \times 10^{-4}$. Convection rolls are traveling clockwise.

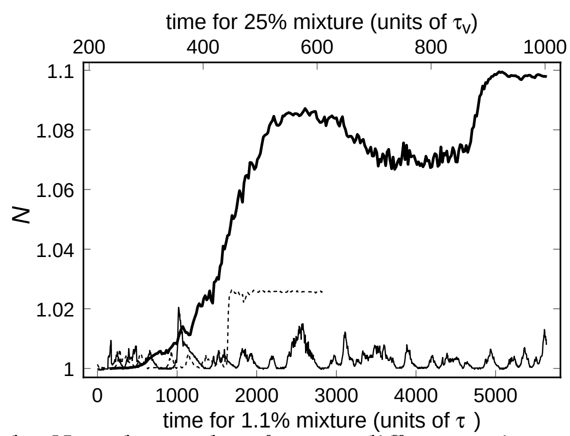

FIG. 4. Time series of the Nusselt number for two different mixtures. In the 25wt.% mixture (upper curve) at $\epsilon = 7 \times 10^{-4}$, convection grew to a cell filling state in about $800\tau_v$ (upper time axis). The Nusselt number record started after image Fig. 1f was taken. For the 1.1wt.% mixture at $\epsilon = 6 \times 10^{-4}$, a seemingly stable "wall" state (Fig. 3) was observed to form once (dashed line). During other runs at the same ϵ, irregularly repeated transients (lower solid curve) were seen over a period of $5500\tau_v$ (lower time axis). Images in Fig. 2 illustrate the patterns observed in this run.

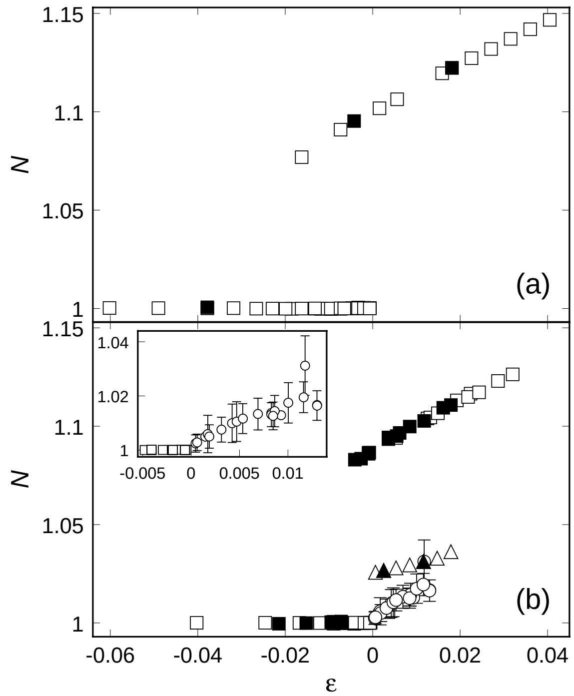

FIG. 5. (a) Nusselt number vs ϵ for the 25wt.% ethanol mixture in the $\Gamma = 11.53$ cell, that shows the conduction ($N = 1$) branch and the upper branch corresponding to the stationary cell–filling state. (b) Nusselt number vs ϵ for the 1.1wt.% ethanol mixture in the $\Gamma = 11.44$ cell, showing the conduction and stationary convection branches (squares), an apparently continuous branch corresponding to erratically fluctuating convection (circles), and the "wall" state (triangles). Open symbols show data obtained while increasing ϵ and solid symbols show data taken while decreasing ϵ, for both figures. Inset in (b) shows in detail the region in the neighborhood of the repeating transients branch.

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