

DIFFUSIVE INSTABILITY AND CONCENTRATION CONVECTION IN A THREE-COMPONENT GAS SYSTEM $0.360\text{C}_3\text{H}_8 + 0.640\text{N}_2\text{O} - \text{CO}_2$

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Abstract. Isothermal mixing in multicomponent gas media is a strongly nonlinear process in which molecular transport may give way to solutal gravitational convection, changing the intensity of mass transfer substantially. This transition is investigated using the system $0.360\text{C}_3\text{H}_8 + 0.640\text{N}_2\text{O} - \text{CO}_2$, in which the molar masses of all components differ by less than 0.2%, so that stratification driven by molecular-weight differences is excluded and the role of diffusive instability can be isolated. Three-dimensional unsteady modelling was performed with the Species Transport formulation (ANSYS Fluent) and compared with two-flask experimental data over the pressure range 0.2–0.85 MPa at $T = 298$ K. The agreement is characterised by a mean absolute error of 0.0106 mole fraction, a root-mean-square error of 0.0109, a mean absolute percentage error of 9.1% and a mean signed relative deviation of -1.3% , the last indicating the absence of systematic bias. It is shown that the upper mixture is denser than the underlying carbon dioxide over the entire pressure range examined, the relative density excess growing from about 1.0% at 0.2 MPa to about 6.0% at 1.0 MPa; this excess originates entirely from the difference in compressibility factors rather than from molar masses, and is confirmed by an independent Peng–Robinson calculation. Convection therefore sets in not through a change in the sign of the density difference but through the Rayleigh number crossing its critical range, which occurs between 0.3 and 0.4 MPa and coincides with the steep experimental rise of propane transport.

Keywords: multicomponent gas mixture; diffusive instability; solutal gravitational convection; density excess; Rayleigh number; Peng–Robinson equation of state; CFD modelling; ANSYS Fluent; mass transfer.

1. INTRODUCTION

Among the diverse types of motion in liquid and gaseous media, a special place is held by convection driven by the instability of the mechanical equilibrium of a mixture [1, 2]. Practical interest in this phenomenon stems from a wide range of applications, spanning gas separation, combined heat and mass transfer in chemical reactors, and atmospheric phenomena [3, 4]. In multicomponent media the situation becomes considerably more complex: differences in the mutual diffusion coefficients of the components give rise to strongly nonlinear concentration distributions, which alter the expected rate of composition homogenisation [5, 6].

A fundamental feature of multicomponent systems is that convective motion arises in them not only under the conditions of the classical Rayleigh–Bénard scheme [1, 2] but also in the presence of initially stable stratification, where the mixture density decreases with height. In this case the instability is governed by the relationship between several concentration gradients, and convective mass transport can exceed diffusive and thermodiffusive contributions by an order of magnitude or more [7]. Methodologies for predicting instability zones have been proposed to support accurate analysis [8, 9, 10].

Under isothermal conditions, effects associated with the diffusive mobility of the components, the pressure and the mixture composition become apparent. A review [11] and a number of experimental and theoretical studies [12, 13, 14] have shown that convective flows can arise in isothermal

multicomponent gas mixtures as early as the initial stage of mixing; moreover, under certain conditions preferential transport of the component with the highest molecular mass is observed. It has been established that the transition between regimes is determined not only by differences in diffusion coefficients but also by pressure and initial composition [15, 16, 17].

At the same time, the boundary between the diffusion and convection regimes in multicomponent gas mixtures has not been sufficiently studied. The case in which the component densities are close to one another remains virtually unexplored; here the stratification mechanism driven by differences in molecular masses is absent, and instability can arise solely from differences in diffusion fluxes and from deviations from ideal-gas behaviour. The $\text{C}_3\text{H}_8\text{--N}_2\text{O--CO}_2$ system is a convenient model for this type of study: the molar masses of its components differ by less than 0.2%, whereas their compressibility factors diverge significantly at the same pressure.

The aim of this study is to determine the mechanism of the transition from molecular diffusion to solutal gravitational convection in a system whose component molar masses differ by less than 0.2%, using the $0.360\text{C}_3\text{H}_8 + 0.640\text{N}_2\text{O} - \text{CO}_2$ composition as an example. The objectives are to perform three-dimensional unsteady modelling of mass transfer within the geometry of an actual two-flask apparatus, to verify the calculations against experimental data [18] over the range 0.2–0.85 MPa, to quantify the density excess and the corresponding Rayleigh number, and on this basis to explain the non-monotonic pressure dependence of mass transfer.

2. MATERIALS AND METHODS

2.1. Problem statement and experimental basis

The experimental data used to verify the calculations were obtained by the two-flask method [18]; a schematic of the setup is shown in Figure 1. Initially the internal volumes of the rig—the upper flask (1), the lower flask (2) and the connecting channel (3)—were evacuated with the shut-off valve (4) closed. The upper flask was then filled with a binary mixture of propane and nitrous oxide from cylinder (5) through valve (9), the pressure being adjusted to the experimental value and monitored with gauge (7). Similarly, the lower flask was filled with carbon dioxide from cylinder (6) through valve (10), with the pressure monitored by gauge (8). After the pressures in the flasks had equalised, valve (4) was opened and mixing began, with simultaneous recording of the time. After the set interval the channel was closed and the actual mixing time recorded; samples were withdrawn through valves (12) and (13) and analysed chromatographically (14).

Concentrations were determined by gas chromatography with an uncertainty of no more than 3%. Direct measurements were performed for propane and nitrous oxide, whereas the carbon dioxide concentration was derived from the condition of particle number conservation:

$$\sum_{i=1}^3 c_i = 1, \quad (1)$$

where c_i is the concentration of the i -th component. To improve reliability, the results were averaged over several series of measurements performed under identical conditions. In all experiments the upper flask contained the binary mixture of propane and nitrous oxide, while the lower flask contained carbon dioxide.

The geometric parameters of the cell were as follows: the volumes of the upper and lower flasks were $226.8 \cdot 10^{-6}$ and $214.5 \cdot 10^{-6} \text{ m}^3$ respectively; the connecting channel was a narrow slit with a cross-section of $0.005 \times 0.0006 \text{ m}^2$ and a height of $17 \cdot 10^{-3} \text{ m}$. The temperature was maintained constant at $T = 298 \text{ K}$ in all experiments, so that the system was strictly isothermal; the duration of

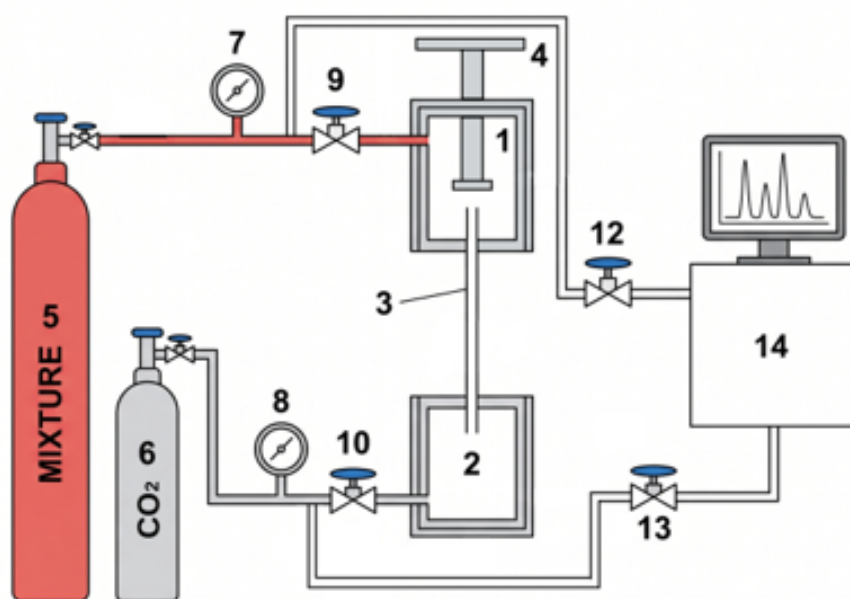


Figure 1 – Schematic of the experimental setup

each experiment was 20 min. The present study examines a single initial composition of the upper mixture, $0.360\text{C}_3\text{H}_8 + 0.640\text{N}_2\text{O}$, which makes it possible to follow the entire sequence of regimes on one object without averaging across compositions.

2.2. Thermophysical characteristics of the components

The input data for the calculation—density ρ , dynamic viscosity η and compressibility factor Z of the components at $T = 298\text{ K}$ —are listed in Table 1 from reference sources [19, 20].

Table 1. Density and compressibility factor of the components at $T = 298\text{ K}$ [19, 20]

p, MPa	$\rho(\text{C}_3\text{H}_8), \text{kg/m}^3$	$\rho(\text{N}_2\text{O}), \text{kg/m}^3$	$\rho(\text{CO}_2), \text{kg/m}^3$	$Z(\text{C}_3\text{H}_8)$	$Z(\text{N}_2\text{O})$	$Z(\text{CO}_2)$
0.2	3.74	3.65	3.64	0.966	0.988	0.989
0.3	5.64	5.52	5.50	0.948	0.981	0.983
0.4	7.58	7.42	7.38	0.929	0.974	0.977
0.5	9.56	9.36	9.29	0.910	0.966	0.970
0.6	11.60	11.35	11.23	0.890	0.958	0.963
0.7	13.70	13.39	13.21	0.870	0.950	0.956
0.8	15.87	15.48	15.23	0.849	0.942	0.949
0.9	18.12	17.63	17.29	0.827	0.933	0.941
1.0	20.46	19.84	19.40	0.804	0.924	0.933

The molar masses of the components are close to one another: $M(\text{C}_3\text{H}_8) = 44.097 \cdot 10^{-3}$, $M(\text{N}_2\text{O}) = 44.013 \cdot 10^{-3}$ and $M(\text{CO}_2) = 44.009 \cdot 10^{-3}\text{ kg/mol}$. The dynamic viscosity varies only slightly within the pressure range considered— $\eta(\text{C}_3\text{H}_8) = 8.31\text{--}8.62$, $\eta(\text{N}_2\text{O}) = 14.93\text{--}15.17$ and $\eta(\text{CO}_2) = 15.02\text{--}15.26$ (in units of $10^{-6}\text{ N} \cdot \text{s/m}^2$)—in agreement with the kinetic theory of gases, according to which the viscosity of a rarefied gas is determined primarily by temperature. The compressibility factors, in contrast, differ significantly: at 1.0 MPa , $Z(\text{C}_3\text{H}_8) = 0.804$ whereas $Z(\text{CO}_2) = 0.933$. It is this dis-

crepancy, rather than the difference in molar masses, that governs the formation of the density excess (Section 4.1).

The mutual diffusion coefficients of the components [19, 20] decrease with increasing pressure according to $D \propto 1/p$, reflecting the reduction of the molecular mean free path as the gas is compressed. At $p = 0.2$ MPa they are $D(\text{C}_3\text{H}_8\text{--CO}_2) = 0.218 \cdot 10^{-5}$, $D(\text{C}_3\text{H}_8\text{--N}_2\text{O}) = 0.215 \cdot 10^{-5}$ and $D(\text{CO}_2\text{--N}_2\text{O}) = 0.298 \cdot 10^{-5} \text{ m}^2/\text{s}$, and at $p = 0.5$ MPa $0.087 \cdot 10^{-5}$, $0.086 \cdot 10^{-5}$ and $0.119 \cdot 10^{-5} \text{ m}^2/\text{s}$ respectively. Two features of this data set are relevant to what follows. First, the $\text{CO}_2\text{--N}_2\text{O}$ pair diffuses faster than either pair involving propane, the ratio $D(\text{CO}_2\text{--N}_2\text{O})/D(\text{C}_3\text{H}_8\text{--CO}_2)$ remaining close to 1.37 over the whole pressure range; this asymmetry of diffusive mobilities creates local non-equilibrium density distributions in the transition zone of the channel, which act as the initial perturbation for the instability. Second, the two binary coefficients that involve propane differ from one another by only 0.9–2.3%, a point returned to in Section 4.4.

2.3. Computational domain and mesh generation

The problem was solved in a three-dimensional formulation: only a 3D model permits an accurate description of spatially non-uniform concentration fields, the formation of convective cells, and possible flow asymmetry in the region of the connecting channel. The computational domain reproduced the geometry of the actual two-flask apparatus (Figure 2).

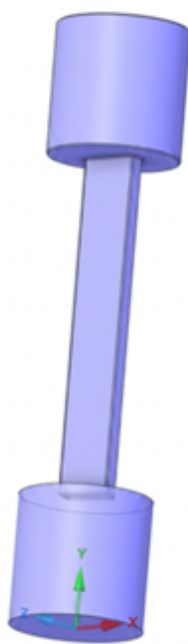


Figure 2 – Three-dimensional model of the two-flask diffusion cell

Discretisation was performed on a three-dimensional unstructured tetrahedral mesh with local refinement in the diffusion channel and in the adjacent parts of the flasks, where the concentration and velocity gradients are largest and where the first signs of instability appear. The element size in the bulk volumes of the flasks was $1.5 \cdot 10^{-3} \text{ m}$, while a refined size of 10^{-4} m was used in the connecting channel. The total number of cells in the computational domain was 1,038,742. The refinement level in the channel was selected so that its narrowest transverse dimension ($0.6 \cdot 10^{-3} \text{ m}$) was resolved by six cells across, which is the minimum required to represent the transverse velocity profile of the developing exchange flow. A formal grid-convergence study is discussed in Section 4.4.

2.4. Physical models and governing equations

Modelling was performed in a transient three-dimensional formulation using the Species Transport module of ANSYS Fluent. Mass transport was treated as a coupled process involving molecular diffusion and transport driven by the motion of the mixture as a whole. The mass conservation equation for each species was written in integral form for an arbitrary control volume V bounded by a surface S :

$$\frac{d}{dt} \int_V \rho Y_i dV + \int_S \rho Y_i \mathbf{v} \cdot d\mathbf{S} = - \int_S \mathbf{j}_i \cdot d\mathbf{S}, \quad (2)$$

where Y_i is the mass fraction of the i -th component, ρ is the mixture density, $\mathbf{v}$ is the velocity vector and $\mathbf{j}_i$ is the diffusion flux. This form directly expresses the mass balance within each cell and is convenient for the finite-volume implementation [22].

The hydrodynamic behaviour of the mixture was described by the momentum conservation equations, with the density treated as a function of the local composition. The concentration and velocity fields are therefore coupled; redistribution of the components alters the density, which in turn influences the flow structure. This feedback constitutes the physical basis of solutal gravitational convection.

The SST $k-\omega$ model [23] was employed for closure. The convective velocities developing in the connecting channel are small and the flow remains laminar throughout the process, so that the turbulent viscosity produced by the model is negligible relative to the molecular viscosity and the formulation reduces in practice to a laminar one. The model was retained for robustness in the presence of steep gradients within the confined channel geometry; the associated limitation is discussed in Section 4.4.

Binary mutual diffusion coefficients were determined from the kinetic theory of gases using Chapman–Enskog-type relations [24]:

$$D_{ij} \sim \frac{T^{3/2}}{p \sigma_{ij}^2 \Omega_{ij}}, \quad (3)$$

where σ_{ij} is the effective collision diameter, Ω_{ij} the collision integral, T the temperature and p the pressure. This relation accounts for the decrease of D_{ij} with pressure noted in Section 2.2.

2.5. Numerical solution procedure

The choice of numerical schemes was dictated by the need to describe unsteady mass transfer accurately in the presence of pronounced concentration gradients while minimising numerical diffusion, which could distort the transport characteristics within the channel. A second-order approximation was used for the momentum and turbulence transport equations in order to improve the representation of velocity and concentration gradients, which is critical when analysing the transition between regimes [21]. The PRESTO! scheme was used for pressure, ensuring a correct pressure distribution in problems involving gravitational effects; gradients were evaluated by the least-squares method, which improves stability on unstructured meshes. The nonlinear system was solved iteratively, the velocity, pressure and concentration fields being refined successively at each time step until convergence.

3. RESULTS

3.1. Pressure dependence of propane transport

The mole fraction of propane in the lower flask was selected as the representative parameter characterising the intensity of inter-flask mass transfer. This choice follows from the fact that the

component concentrations of a ternary system are linked by the material balance condition (1), so that analysing the remaining components yields no fundamentally new information. A comparison of the calculated results with the experimental data [18] is presented in Table 2.

Table 2. Mole fraction of propane in the lower flask for the composition $0.360 \text{ C}_3\text{H}_8 + 0.640 \text{ N}_2\text{O} - \text{CO}_2$

p , MPa	Experiment [18]	ANSYS Fluent
0.20	0.092	0.086
0.30	0.105	0.118
0.45	0.138	0.127
0.55	0.140	0.150
0.85	0.115	0.102

The agreement was assessed using several complementary measures, since a single figure of merit can be misleading for a data set of this size. The mean absolute error is 0.0106 and the root-mean-square error 0.0109 mole fraction; in relative terms the mean absolute percentage error is 9.1% and the largest single deviation 12.4%. These quantities characterise the magnitude of the scatter. Its direction is characterised separately by the mean signed relative deviation, which amounts to only -1.3% ; the residuals alternate in sign and very nearly cancel, so that the model exhibits no systematic bias with respect to the experiment and the discrepancies are random in character. In absolute terms the residuals lie between 0.006 and 0.013 mole fraction. For comparison, the 3% uncertainty of the chromatographic determination corresponds to 0.003–0.004 mole fraction at the concentration levels considered, that is, roughly one third of the observed residual; the remainder is attributable to the model and to discretisation.

The largest relative deviation occurs at 0.30 MPa, that is, precisely at the regime-transition boundary identified in Section 4.2, where the system is most sensitive to initial perturbations and where the predicted onset of instability depends most strongly on the mesh resolution in the zone of maximum gradients. Outside the vicinity of this boundary, the agreement is noticeably better. Significantly, the model reproduces not only the absolute values but also the qualitative structure of the pressure dependence: propane transport increases with pressure, reaches a maximum at $p \approx 0.55$ MPa (0.140 in the experiment against 0.150 in the calculation) and then decreases, falling to 0.115 at 0.85 MPa. The non-monotonic character of the dependence is reproduced without any additional tuning of the model, which indicates that its origin is physical rather than numerical.

3.2. Time evolution of the propane concentration

The evolution of the propane mole fraction in the lower flask at $p = 0.583$ MPa, corresponding to the region of maximum transfer, is shown in Figure 3.

The curve divides clearly into three sections. In the initial interval ($t \lesssim 100$ s) the concentration rises rapidly to about 0.05, corresponding to the establishment of the diffusive flux immediately after the channel is opened. Subsequently, over $t \approx 100$ –400 s, the rate of increase falls noticeably and the dependence becomes quasi-diffusive; the concentration grows smoothly, without pronounced nonlinearities.

A qualitative change occurs over $t \approx 400$ –700 s. The rate of increase rises again and appreciably exceeds the values characteristic of the preceding quasi-diffusive stage, the concentration growing from ≈ 0.07 to ≈ 0.125 . Such an acceleration of transport under unchanged external conditions cannot be explained within a purely diffusive framework and indicates the onset of convective motion

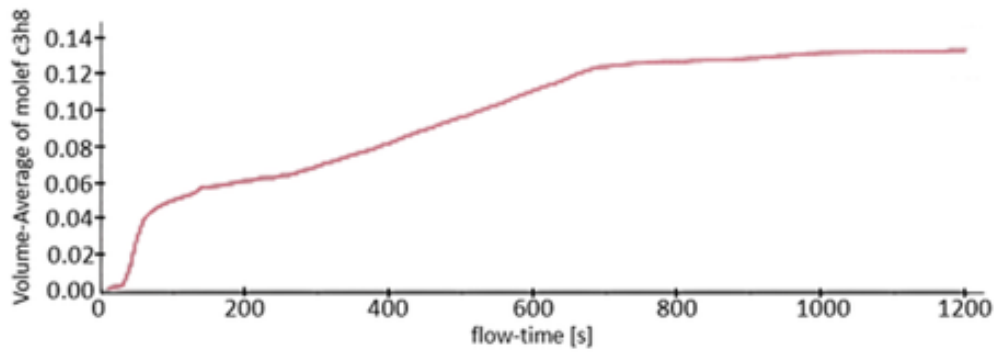


Figure 3 – Time evolution of the volume-averaged mole fraction of propane in the lower flask for the composition $0.360\text{C}_3\text{H}_8 + 0.640\text{N}_2\text{O} - \text{CO}_2$ at $p = 0.583\text{ MPa}$

in the channel. The interval $t^* \approx 400\text{--}600\text{ s}$ should be regarded as the delay time for the development of the instability, that is, the period required for the diffusive redistribution of the components to generate a sufficient density excess in the transition zone.

After $t \approx 700\text{ s}$ the curve enters a quasi-stationary regime, rising slowly to 0.135 by $t = 1200\text{ s}$. This saturation reflects the depletion of the driving concentration difference between the flasks as the compositions equalise.

4. DISCUSSION

4.1. Density excess as the source of the instability

The molar mass of the upper mixture, calculated from its initial composition, is $M = 0.360 \cdot 44.097 + 0.640 \cdot 44.013 = 44.043 \cdot 10^{-3}\text{ kg/mol}$, exceeding that of carbon dioxide by only 0.078% [cite: 9]. Under strict adherence to the ideal-gas equation of state such a difference would produce a relative density excess of the same order, a value far too small to drive convection in a channel of the dimensions considered. The instability mechanism must therefore be sought in the departure of the gases from ideality.

Writing the density of a real gas as $\rho = pM/(ZRT)$ and applying the additive rule $Z_{\text{mix}} = \sum x_i Z_i$ to the upper mixture, the relative density excess with respect to pure carbon dioxide at the same pressure and temperature takes the compact form:

$$\frac{\Delta\rho}{\rho} = \frac{\rho_{\text{mix}} - \rho_{\text{CO}_2}}{\rho_{\text{CO}_2}} = \frac{M_{\text{mix}}}{M_{\text{CO}_2}} \cdot \frac{Z_{\text{CO}_2}}{Z_{\text{mix}}} - 1. \quad (4)$$

Written in this form the result is immediate. Both factors exceed unity at every pressure: $M_{\text{mix}} > M_{\text{CO}_2}$ by construction, and $Z_{\text{mix}} < Z_{\text{CO}_2}$ because propane is markedly less ideal than carbon dioxide. The density excess is therefore positive throughout the pressure range, and no change of sign occurs. Its magnitude, however, is strongly pressure dependent, since the ratio $Z_{\text{CO}_2}/Z_{\text{mix}}$ grows as the gases are compressed. The computed values are given in Table 3.

Because the density excess is central to the proposed mechanism, the additive- Z approximation was verified independently against the Peng–Robinson equation of state with van der Waals mixing rules and zero binary interaction parameters, using standard critical constants for the three gases [26, 27]. This route gives a density excess that is likewise positive at every pressure, growing from +0.81% at 0.2 MPa to +2.02% at 0.5 MPa and +4.40% at 1.0 MPa. The two calculations therefore agree on every essential point: the sign of the excess, its monotonic growth with pressure, and its order of magnitude. The additive rule overestimates the excess by 0.2–1.6 percentage points, the

Table 3. Relative density excess of the upper mixture over carbon dioxide at $T = 298$ K

p , MPa	Z_{mix}	ρ_{mix} , kg/m ³	$\Delta\rho/\rho$, %
0.2	0.980	3.627	+0.99
0.3	0.969	5.503	+1.51
0.4	0.958	7.424	+2.08
0.5	0.946	9.397	+2.63
0.6	0.934	11.425	+3.24
0.7	0.921	13.507	+3.86
0.8	0.909	15.653	+4.53
0.9	0.895	17.878	+5.24
1.0	0.881	20.181	+6.01

discrepancy increasing with pressure as the mixture departs further from ideality—the expected behaviour, since additivity of compressibility factors is an Amagat-type approximation that neglects cross-interaction between unlike molecules. For the purposes of the present analysis this level of agreement is sufficient, and the conclusions drawn below are unchanged if the Peng–Robinson values are used instead.

The physical origin of the effect is the following. All three components have almost identical molar masses, so that under ideal behaviour their densities at a given pressure would coincide to within 0.08% at any pressure. In reality the density is governed by the ratio M/Z , and the behaviour of the compressibility factor becomes decisive. Of the gases considered, propane lies closest to its condensation region (the saturation pressure is about 0.9 MPa at 298 K), and its compressibility factor therefore falls much more rapidly with pressure—from 0.966 at 0.2 MPa to 0.804 at 1.0 MPa—whereas for carbon dioxide it falls only from 0.989 to 0.933[cite: 9]. A decrease in Z corresponds to an increase in density at the same pressure, so the presence of 36% propane in the upper flask makes that mixture heavier than pure CO₂, and increasingly so as the pressure rises. It is the non-ideality of propane, and not any difference in molecular masses, that maintains the heavy-over-light configuration.

A positive density excess is a necessary but not a sufficient condition for convection. The configuration is potentially unstable at every pressure examined, yet the experiment shows only weak transport at 0.2 MPa and a steep rise thereafter. The selection of the actual transition therefore rests with the balance between the destabilising buoyancy and the stabilising action of viscosity and diffusion, which is quantified by the Rayleigh number in the next section.

4.2. Rayleigh number

The stability of the configuration is characterised by the solutal Rayleigh number

$$\text{Ra} = \frac{g \left(\frac{\Delta\rho}{\rho} \right) L^3}{\nu D}, \quad \nu = \frac{\eta_{\text{mix}}}{\rho_{\text{mix}}}, \quad (5)$$

in which the choice of the characteristic length L requires comment, since the connecting channel has two very different transverse dimensions and a third, much larger, longitudinal one. Two scales are physically meaningful. The channel height $H = 17 \cdot 10^{-3}$ m is the distance over which the density difference between the flasks is supported and therefore sets the total buoyant driving force available to the exchange flow. The narrower transverse dimension of the slit, $d = 0.6 \cdot 10^{-3}$ m, is the scale that controls the onset of the instability, because it limits the size of the disturbance that can develop across the channel and it is over this distance that viscous damping acts. Rayleigh numbers based on the two

scales differ by the factor $(H/d)^3 \approx 2.3 \cdot 10^4$ and must be compared with different critical values. The kinematic viscosity was evaluated as $\nu = \eta/\rho$, the mixture viscosity $\eta \approx 12.5 \cdot 10^{-6} \text{ N} \cdot \text{s}/\text{m}^2$ being obtained from the additive rule, and D was taken as the $\text{C}_3\text{H}_8\text{--CO}_2$ coefficient at the corresponding pressure (Section 2.2).

Evaluated on the channel height, Ra grows from $6.3 \cdot 10^4$ at 0.2 MPa to $1.1 \cdot 10^6$ at 0.5 MPa and $1.1 \cdot 10^7$ at 1.0 MPa. Evaluated on the slit width, which is the scale relevant to the onset of the disturbance, it grows from 2.8 at 0.2 MPa to 9.7 at 0.3 MPa, 24 at 0.4 MPa and 48 at 0.5 MPa, reaching $4.7 \cdot 10^2$ at 1.0 MPa. This growth is driven by the combined effect of an increasing density excess and a diffusion coefficient falling as $1/p$. Since the product $\rho\nu = \eta$ is practically independent of pressure, equation (5) implies the approximate relation $Ra \propto (\Delta\rho/\rho) \cdot p$, which accounts for the roughly quadratic growth with pressure.

The essential point is that the Rayleigh number based on the slit width passes through values of order 10 between 0.3 and 0.4 MPa. For confined vertical geometries with an axial density gradient the critical Rayleigh number is of order $10^1\text{--}10^2$ depending on the aspect ratio and the boundary conditions [1, 2], so this crossing brackets precisely the interval over which the measured propane transport rises steeply, from 0.105 at 0.30 MPa to 0.138 at 0.45 MPa (Table 2). The transition observed in the experiment is therefore identified with the crossing of the critical Rayleigh number rather than with any change in the sign of the density difference. It should be emphasised that a critical value specific to the present slit geometry would require a dedicated linear stability analysis, which lies outside the scope of this work; the comparison made here is an order-of-magnitude one. Independent estimates of the convective velocity in the channel confirm that the resulting flow remains laminar over the whole range of conditions examined.

4.3. Nature of the pressure maximum

The monotonic growth of the Rayleigh number apparently contradicts the transport maximum observed in Table 2 at $p \approx 0.55$ MPa. The contradiction is removed once it is recognised that the measured quantity is a mole fraction and not an absolute mass flux. At fixed volume the total number of moles of gas in the lower flask increases in proportion to the pressure, so that even for a rising absolute flux of propane the mole fraction, normalised by this total, can pass through a maximum. The pressure maximum thus reflects the competition between the intensification of convective transport, driven by the increase of Ra , and the simultaneous dilution of the transported material within an increasing total number of particles.

A second factor is hydrodynamic. As the pressure rises, the diffusion coefficient continues to fall as $1/p$ while the convective velocity grows, and the exchange flow in a slit of high aspect ratio may reorganise from a through-going counter-flow into a system of closed recirculation cells localised within the channel. Such cells would mix the fluid efficiently inside the channel while contributing little to the net inter-flask transport, so that the overall transfer would again become diffusion-limited. This interpretation is consistent with observations reported for related two-flask systems at elevated pressures [13, 17], but it is offered here as a hypothesis: the present calculations were post-processed for concentration fields only, and a dedicated analysis of velocity vectors and streamlines at several pressures would be required to establish the existence and the geometry of such cells directly. This is identified as the principal direction for the continuation of the work.

4.4. Model limitations

Several limitations of the present approach should be borne in mind when interpreting the results. Diffusive transport was described through effective binary coefficients, whereas a rigorous treat-

ment of a ternary system requires the full Maxwell–Stefan matrix [25]. The magnitude of this simplification can be bounded from the data of Section 2.2. The species that carries the density signal is propane, and its two binary coefficients, $D(\text{C}_3\text{H}_8\text{--CO}_2)$ and $D(\text{C}_3\text{H}_8\text{--N}_2\text{O})$, differ from each other by only 0.9–2.3% across the entire pressure range. The effective diffusivity of propane is therefore almost independent of the local ratio of CO_2 to N_2O in the surrounding medium, and the binary approximation is well justified for the component that governs the buoyancy. The substantial difference in mobilities, a factor of 1.37, is between CO_2 and N_2O , that is, within the pair that does not carry a significant density contrast. Cross-diffusion effects are consequently expected to redistribute these two components relative to one another rather than to alter the propane flux appreciably; their neglect is not expected to change the conclusions, although it may shift the predicted onset time.

The mutual diffusion coefficients were held constant throughout the computational domain and were not adjusted for the local composition, whereas in the transition zone of the channel the composition varies over a wide range. Including the concentration dependence of D_{ij} would refine the predicted onset of convection.

The SST $k\text{--}\omega$ closure was applied to a flow which remains laminar over the whole range of conditions examined. The turbulent viscosity generated by the model is small compared with the molecular value, so the computation is effectively laminar; nevertheless a residual eddy viscosity cannot be excluded near the sharp gradients at the channel entrance, where it would act as an additional effective diffusivity and could shift the calculated onset time. A direct comparison with a fully laminar computation at two or three representative pressures is planned and will quantify this effect.

Finally, the mesh was refined locally in the channel to the level described in Section 2.3 rather than through a formal grid-convergence study on a sequence of systematically refined meshes. Because the predicted onset time of the instability is the quantity most sensitive to resolution, a study of this kind—reporting both the final concentration and the onset time as functions of cell count—is required to establish the numerical uncertainty of the present results and is the subject of ongoing work.

5. CONCLUSION

An experimental and numerical study of isothermal mixing in a three-component gas system has been carried out for the composition $0.360 \text{ C}_3\text{H}_8 + 0.640 \text{ N}_2\text{O} - \text{CO}_2$ over the pressure range 0.2–0.85 MPa at a constant temperature of $T = 298 \text{ K}$. The main results are as follows:

1. A three-dimensional CFD model based on the Species Transport approach reproduces the propane mole fraction in the lower flask with a mean absolute error of 0.0106 mole fraction, a root-mean-square error of 0.0109 and a mean absolute percentage error of 9.1%, the maximum relative deviation being 12.4%. The mean signed relative deviation of -1.3% shows that the model carries no systematic bias. The largest deviation occurs at the regime-transition boundary, where the predicted onset of instability is most sensitive to mesh resolution. The model reproduces the non-monotonic pressure dependence with a maximum at $p \approx 0.55 \text{ MPa}$.
2. In a system whose component molar masses differ by less than 0.2%, the upper mixture is nevertheless denser than the underlying carbon dioxide at every pressure examined, the relative density excess growing from about 1.0% at 0.2 MPa to about 6.0% at 1.0 MPa. The excess arises entirely from the difference in compressibility factors—propane, being closest to its condensation region, departs from ideality far more strongly than carbon dioxide—and not from the difference in molar masses, which would account for only 0.08%. An independent Peng–Robinson calculation confirms the sign, the monotonic growth and the order of magnitude of this excess.
3. Since the density excess is positive throughout, the onset of convection is not governed by a

change in the sign of the density difference but by the Rayleigh number crossing its critical range. Evaluated on the slit width, Ra grows from 2.8 at 0.2 MPa to 48 at 0.5 MPa and passes through values of order 10 between 0.3 and 0.4 MPa, that is, precisely over the interval in which the measured transport rises steeply.

4. The time evolution of the concentration exhibits three characteristic stages—an initial diffusive stage, a quasi-diffusive stage and a convective stage—with a delay time for the development of the instability of $t^* \approx 400\text{--}600$ s, after which the transport rate increases markedly.
5. The non-monotonic pressure dependence is explained by the competition between the intensification of convective transport, which follows the growth of the Rayleigh number, and the dilution of the transported material within the total number of particles, which increases with pressure.

The results obtained may serve as a basis for formulating quantitative criteria for identifying mixing regimes in multicomponent gas systems with a negligible initial molar-mass contrast, and for selecting conditions for the accurate measurement of mutual diffusion coefficients, whose reliable determination is possible only in the strictly diffusive regime.

AUTHOR CONTRIBUTIONS

V. Mukamedenkyzy — conceptualisation, methodology, supervision, manuscript editing; B.E. Akberdiyev — numerical modelling, data processing and analysis, visualisation, preparation of the initial draft; N.G. Valko — analysis of results, methodological review, manuscript editing. All authors have read and approved the final version of the manuscript.

DATA AVAILABILITY

Data supporting the results of this study are available from the corresponding author upon reasonable request. Experimental data used for verification were taken from previously published work.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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