

Transient thermo-solutal convection in a tilted porous enclosure heated from below and salted from above

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Abstract

The confinement of CO₂ in deep geothermal reservoirs as a means of mitigation of greenhouse gas emissions is continuously motivating research on the retention capacity of these deep aquifers. An important physical containment mechanism is related with CO₂ dissolution and thermo-solutal convection. In this context, numerical simulations are performed in this work to assess the effect of inclination, Rayleigh number, and buoyancy ratio on the convective transport in a rectangular porous medium. The porous enclosure is heated from below and cooled from above, whereas a solute is dissolved through the upper boundary with a constant concentration condition and no mass loss through the other boundaries. A set of governing parameters is considered in this assessment: two buoyancy ratios with dominant solute buoyant forces (10 and 100), three Rayleigh numbers (10, 50, and 80), and three inclination angles plus the horizontal case for reference (5°, 10°, and 15°). The solution to the problem is based on a Finite Volume method along with fixed point iteration for the coupled differential equations, and a Conjugate Gradient algorithm for the algebraic system. The model is validated and tested under mesh analysis. The numerical results show that the inclination angle has a minor effect on the convective mixing properties of the porous medium in comparison with the Rayleigh number

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and the buoyancy ratio. Increasing the angle slightly decreases the mixing rate as a consequence of the formation of preferential flow paths associated with the inclination, these preferential flow paths make mixing less efficient and give rise to zonation of solute concentration.

Keywords: double-diffusive convection, porous medium, Boussinesq approximation, CO₂ dissolution.

1 **Nomenclature**

2 **Greek symbols**

3 α Thermal diffusivity

4 β Thermal expansion coefficient

5 ϵ Normalized porosity (ϕ/σ)

6 Γ Coefficient $-1/Ra$

7 μ Viscosity

8 ν Kinematic viscosity

9 ϕ Porosity

10 ψ Stream function

11 ρ Density

12 σ Ratio of solid-fluid heat capacities

13 θ Inclination angle

14 **Other symbols**

15 – Overbar denotes dimensional variables and operators

16 **Roman letters**

17 **e** Vector ($\sin \theta, \cos \theta$)

18	\mathbf{u}	Darcy's velocity
19	A	Area
20	B	Height of the porous enclosure
21	C	Width of the porous enclosure
22	D	Mass diffusivity
23	g	Gravitational constant
24	k	Permeability
25	Le	Lewis number
26	N	Buoyancy ratio
27	Nu	Nusselt number
28	P	Pressure
29	Ra	Rayleigh number
30	S	Solute concentration
31	S_a	Average concentration
32	Sh	Sherwood number
33	T	Temperature
34	t	Time
35	x, y	Cartesian coordinates
36	Subscripts	
37	$0, c, r$	Reference values
38	a	Average value
39	S	Solutal
40	T	Thermal

41 1. Introduction

42 Double-diffusive convection in porous media is a major process for the physi-
43 cal containment of CO₂ in deep aquifers [1, 2]. The problem arises from the fact
44 that supercritical CO₂ injected in a confined aquifer accumulates beneath the
45 cap rock, where dissolution of CO₂ in water takes place over time [3], this leads
46 to convective mixing due to a slightly higher density of the CO₂-Brine solution.
47 Moreover, the CO₂-Brine interface might not be horizontal, but present some
48 degree of inclination due to structural conditions and CO₂-front displacement.
49 We address the dissolution process of a solute in this situation.

50 It is worth mentioning that this topic is relevant in other scientific and
51 engineering fields, such as: evolution of hydrothermal systems [4], materials
52 manufacturing [5], and nuclear waste repository [6].

53 Fundamental aspects of double-diffusive convection have been stated by sev-
54 eral authors in the past. Early work was presented by Nield [7], who addressed
55 the onset of convection in a porous layer heated and salted from below. On the
56 fact that thermal diffusions occurs more rapidly than solute diffusion, he pointed
57 out that the onset of convention can be characterized either by monotonic or
58 oscillatory instability depending on whether the solute gradients enhance or
59 counteract the instability associated with thermal gradients. Taunton et al. [8]
60 extended the stability analysis of this problem and pointed out that concentra-
61 tion density differences are more effective in promoting instabilities. Trevisan
62 and Bejan [9] addressed the problem of steady-state solutions at high Rayleigh
63 numbers (up to 2000), their work revealed that the Sherwood number relates to
64 the governing parameters Ra and Le with three distinct scaling laws. Rosenberg
65 and Spera [10] conducted numerical simulations in order to find relations be-
66 tween Nu and Sh with the governing parameters Ra , Le , and N in steady-state
67 solutions. They also evaluated the effect of buoyancy ratio N on the dynamics
68 of transient convection at $Ra = 600$. The results showed that Nu follows an
69 oscillatory behavior as the flow develops and becomes more complex for greater
70 N .

71 Subsequent work was concerned with a variety of boundary conditions and
72 configurations. Lin [11] addressed this problem considering the lateral walls of
73 the porous enclosure as the source of thermal and concentration gradients, with
74 adiabatic and impervious top and bottom boundaries. In a similar configuration,
75 Mamou et al. [12] looked into the existence of multiple steady-state solutions
76 (a condition which is known from purely thermal free convection [13, 14]), they
77 obtained governing parameters that allow different convective modes. Assuming
78 the same kind of boundary conditions, Mamou et al. [15] presented the stabil-
79 ity analysis for the particular case of opposing buoyancy forces ($N = -1$), they
80 reported parametric relations ($Le, \bar{\epsilon}, A$) with the onset of stationary and oscilla-
81 tory convection. A three-dimensional version of this problem was presented by
82 Sezai and Mohamad [16] with the additional assumption of a Darcy-Brinkman
83 flow model. They identified steady-state solutions for a set of governing param-
84 eters (Ra, Le , and N) and found that in the case of opposing buoyancy forces
85 the convective mode is strictly 3D. Nithiarasu et al. [17], considered the case
86 of prescribed temperature and concentration in a lateral boundary, and con-
87 vective heat and mass transfer in the opposite, giving rise to the Biot number
88 as a governing parameter of the system. Zhao et al. [18] considered a porous
89 medium heated and salted from a segment of a lateral boundary keeping the
90 opposite boundary at constant temperature and concentration, and top and
91 bottom boundaries as adiabatic and impermeable. Their work is particularly
92 important to understand the transport behavior associated with localized heat
93 and mass sources. In a later model, the use of the Darcy-Brinkman equation
94 allowed the evaluation of the no-slip boundary condition on a solid wall [19].
95 Coupled porous medium and free fluid systems, as well as tilted porous enclo-
96 sures have also been studied in relation with these boundary conditions [20, 21].

97 With regard to dissolution processes from the top boundary, theoretical stud-
98 ies have determined the critical conditions for the onset convective mixing as
99 well as mixing evolution in absence of thermal gradients [22, 23, 24]. The prob-
100 lem is stated as the stability of a diffusive boundary layer in a semi-infinite
101 domain [22], which leads to the definition of a critical time and wave number

102 for the onset of convection, both proportional to the ratio between diffusive
103 and buoyancy forces. Further, the mixing has been described in terms of five
104 stages, comprising diffusion, onset of instability, onset of convection (fingering),
105 merging of convective fingers, and convective shutdown [3]. Concerning systems
106 heated from below and salted from above, Islam et al. [2] described the general
107 aspects of the convective transport, characterized by fingering and merging.
108 They use the average concentration of the solute in the porous medium as a
109 parameter to measure how the thermal and solute Rayleigh numbers enhance
110 solute transport with time. They also evaluated the aspect ratio of the porous
111 enclosure and found that wide is more advantageous for solute transport than
112 tall. Anisotropy effects, geochemical reactions, presence of impermeable layers,
113 and more recently, external forces due to fluid injection in the porous medium
114 have also been examined [25, 26, 27, 28].

115 Even though a variety of general aspects of this problem are now available
116 in the literature, we consider that the effect of inclination angles has received
117 less attention. In particular, the extent to which the tilt angle accelerates or
118 delays the convection and mixing has not been addressed in the context of this
119 problem. For this reason we present in this paper numerical simulations with
120 a focus on the inclination of the porous medium to provide answers to this
121 particular question.

122 **2. Problem formulation**

123 The problem concerns a rectangular porous enclosure of height B and length
124 C with impermeable walls and fully saturated with an incompressible fluid (Fig.
125 1). It is assumed a constant aspect ratio of the enclosure $C/B=3$. The medium
126 is heated from below at constant temperature, and salted from above at a con-
127 stant concentration. Initially the medium is at constant temperature \bar{T}_c and
128 in absence of dissolved solute ($\bar{S}=0$). The lateral boundaries are adiabatic and
129 the enclosure is inclined an angle θ with respect to the horizontal. The basic
130 assumptions for this problem include local thermal equilibrium, constant poros-

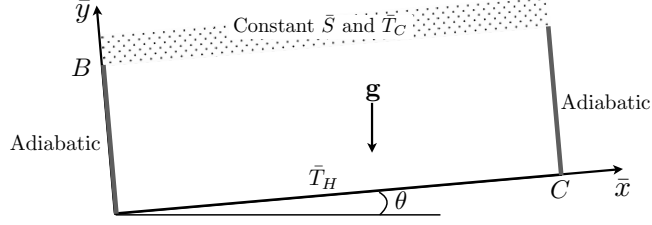


Figure 1: Schematic model of a rectangular porous enclosure tilted an angle θ . The enclosure is heated from its base and cooled from the top at constant temperatures, whereas dissolution occurs at constant concentration on the top boundary.

131 ity, fluid flow is described by Darcy's law, and the Boussinesq approximation
 132 can be applied. From these considerations the momentum equation can read as
 133 follows:

$$\bar{\mathbf{u}} = -\frac{k}{\mu} (\bar{\nabla} \bar{P} + \rho g \mathbf{e}) \quad (1)$$

where the vector $\mathbf{e} = (\sin \theta, \cos \theta)$ determines the components of the buoyancy force as a function of the inclination angle. The heat transfer equation takes the form [29]:

$$\sigma \frac{\partial \bar{T}}{\partial t} + \bar{\mathbf{u}} \cdot \bar{\nabla} \bar{T} = \bar{\nabla} \cdot (\alpha \bar{\nabla} \bar{T}), \quad (2)$$

likewise, the mass transfer equation reads:

$$\phi \frac{\partial \bar{S}}{\partial t} + \bar{\mathbf{u}} \cdot \bar{\nabla} \bar{S} = \bar{\nabla} \cdot (D \bar{\nabla} \bar{S}). \quad (3)$$

The Boussinesq approximation leads to the condition of incompressibility:

$$\bar{\nabla} \cdot \bar{\mathbf{u}} = 0, \quad (4)$$

furthermore, an equation of state for the buoyancy term is required [29]:

$$\rho = \rho_0 [1 - \beta_T (\bar{T} - \bar{T}_0) - \beta_S (\bar{S} - \bar{S}_0)] \quad (5)$$

134 The pressure \bar{P} (Eq. 1) is defined so that it is taken relative to the hydrostatic
 135 pressure $\rho_0 g z$, this leads to the common form to the Darcy momentum equation
 136 with the buoyancy term:

$$\bar{\mathbf{u}} = -\frac{k}{\mu}(\bar{\nabla}\bar{P} - (\beta_T(\bar{T} - \bar{T}_0) + \beta_S(\bar{S} - \bar{S}_0))\rho_0 g \mathbf{e}) \quad (6)$$

137 The following dimensionless parameters and operators are introduced for the
138 formulation of the mathematical problem:

$$x = \frac{\bar{x}}{B}, \quad y = \frac{\bar{y}}{B}, \quad t = \frac{\bar{t}\alpha}{\sigma B^2}, \quad \mathbf{u} = \frac{B}{\alpha}(\bar{u}, \bar{v}), \quad P = \frac{k}{\mu\alpha}\bar{P},$$

$$T = \frac{\bar{T} - \bar{T}_0}{\Delta\bar{T}}, \quad S = \frac{\bar{S} - \bar{S}_0}{\Delta\bar{S}}, \quad \Delta\bar{T} = \bar{T}_0 - \bar{T}_c, \quad \Delta\bar{S} = \bar{S}_0 - \bar{S}_r,$$

$$Ra = \frac{gBk\beta_T\Delta\bar{T}}{\alpha\nu}, \quad Le = \frac{\alpha}{D}, \quad N = \frac{\beta_S\Delta\bar{S}}{\beta_T\Delta\bar{T}}, \quad \epsilon = \frac{\phi}{\sigma}, \quad \bar{\nabla} = B\bar{\nabla}. \quad (7)$$

139 Next, we define the Nusselt number, Sherwood number, and the average
140 concentration of the solute as the physical parameters for the analysis of the
141 numerical results:

$$Nu = \int \left| \frac{\partial T}{\partial y} \right|_{y=1} dx, \quad Sh = \int \left| \frac{\partial S}{\partial y} \right|_{y=1} dx, \quad S_a = \frac{\int S dA}{A}. \quad (8)$$

142 The dimensionless equations read:

$$\frac{\partial T}{\partial t} - \nabla^2 T + \mathbf{u} \cdot \nabla T = 0, \quad (9)$$

$$\epsilon \frac{\partial S}{\partial t} - \frac{1}{Le} \nabla^2 S + \mathbf{u} \cdot \nabla S = 0, \quad (10)$$

$$\mathbf{u} + \nabla P = Ra(T + NS)\mathbf{e}. \quad (11)$$

143 In what follows, a constant $\epsilon = 1$ was assumed in our mathematical model.
144 Next, the momentum equation (Eq. 11) is written in terms of the stream
145 function, ψ : $u = \partial\psi/\partial y$, $v = -\partial\psi/\partial x$ [30, 31]. This leads to a Poisson equation
146 of the form:

$$\Gamma \nabla^2 \psi = \left(\frac{\partial T}{\partial x} + N \frac{\partial S}{\partial x} \right) \cos \theta - \left(\frac{\partial T}{\partial y} + N \frac{\partial S}{\partial y} \right) \sin \theta, \quad (12)$$

147 with $\Gamma = -1/Ra$. Equations 9, 10, and 12 represent the mathematical
 148 problem to be solved subject to the following boundary conditions:

For the heat transfer equation, the temperature field satisfies

$$\frac{\partial T}{\partial x} = 0, \quad \text{for } x = 0 \quad \text{and} \quad x = 3, \quad (13)$$

$$T = 1, \quad \text{for } y = 0 \quad \text{and} \quad t > 0, \quad (14)$$

$$T = 0, \quad \text{for } y = 1 \quad \text{and} \quad t > 0. \quad (15)$$

The mass transfer equation is subject to

$$\frac{\partial S}{\partial x} = 0, \quad \text{for } x = 0 \quad \text{and} \quad x = 3, \quad (16)$$

$$\frac{\partial S}{\partial y} = 0, \quad \text{for } y = 0, \quad (17)$$

$$S = 1, \quad \text{for } y = 1 \quad \text{and} \quad t > 0. \quad (18)$$

With regard to the momentum equation (Eq. 12), $\psi = 0$ is imposed at the boundaries to achieve no fluid flow through the walls:

$$\psi = 0, \quad \text{for } x = 0 \quad \text{and} \quad x = C, \quad (19)$$

$$\psi = 0, \quad \text{for } y = 0 \quad \text{and} \quad y = 1. \quad (20)$$

149 **3. Methods and solution**

150 The time-dependent mathematical problem was discretized with the Finite
 151 Volume numerical method [32]. The discretization of the convective terms of the
 152 heat and mass transfer equations (Eqs. 9 and 10) was done under the upwind
 153 scheme, and a first-order fully implicit scheme was used for the temporal term
 154 of both equations. A fixed point algorithm was implemented for the solution
 155 of the coupled differential equations [33]. Further, the algebraic systems were
 156 solved with a Conjugate Gradient algorithm. A convergence criterion of 1×10^{-6}
 157 was used in both iterative algorithms. The numerical model was implemented

158 in Fortran 90 with multithread libraries (OpenMP by Intel[®]). The simulations
 159 were performed in processors Xeon[®] E5-2630 v3, 2.40 GHz.

160 A time step $\Delta t = 1 \times 10^{-6}$ was selected for the simulations after a calibration
 161 process. Likewise, a mesh sensitivity analysis (Sec. 3.1) permitted us to choose
 162 a mesh given by $\Delta x = \Delta y = 1/250$. This mesh turned out a sufficiently high
 163 resolution mesh to capture small scale flow features.

164 3.1. Validation

165 The model was validated by comparison with the results reported by Islam
 166 et al. [2]. In this case, the aspect ratio of the porous enclosure is 1, and $\theta = 0$.
 167 Additionally, a perturbation of the concentration gradient is introduced as a
 168 boundary condition, in order to promote the onset of ‘fingering’ in the solute
 169 transport: $S(x, 1) = 1 + 0.01 \sin(48\pi x)$. Based on these conditions, three simu-
 170 lations were performed considering fixed $N = -100$, $Le = 1$, a simulation time
 171 $t = 6.3 \times 10^{-3}$ and three Rayleigh numbers. Figure 2 presents the comparison
 172 between our results and the reference. There is consistency in the results despite
 173 some differences as the system becomes more convective ($Ra = 100$), this slight
 174 deviation in the models can be attributed to the different approaches to solve
 175 both the system of differential equations and the algebraic systems.

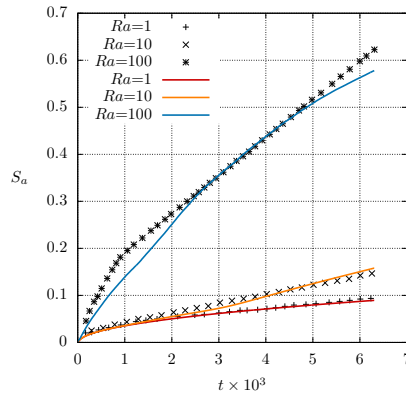


Figure 2: Comparison between the present results (curves) and the reference [2] (points).

176 The simulation result corresponding to $Ra = 100$ was further tested under

177 two finer meshes: $\Delta x = \Delta y = 1/500$ and $\Delta x = \Delta y = 1/1000$, a smaller time
 178 step was required, however, to maintain the stability of the solution ($\Delta t =$
 179 1×10^{-7}). A comparison of S_a after a simulation time $t = 6.3 \times 10^{-3}$ show that
 180 a small difference is obtained ($\sim 6\%$) with the finer meshes with a considerable
 181 increase in computing time (Table 1). On this basis $\Delta x = \Delta y = 1/250$ was
 182 preferred as a suitable discretization for the cases studied here, involving a
 183 larger aspect ratio and longer simulation times.

Table 1: Mesh sensitivity analysis.

Mesh	S_a	cpu time (h)
1/250	0.58	0.35
1/500	0.61	5.21
1/1000	0.63	33.6

184 4. Numerical results and discussion

185 4.1. Average concentration

186 The average concentration S_a is a measure of the degree of saturation of
 187 the solute in the fluid. Since the porous medium does not allow mass loss
 188 through the boundaries, the average concentration S_a tends to 1 in the porous
 189 enclosure, which is a saturation condition. This section is intended to quantify
 190 the extent to which the governing parameters N , Ra , and θ enhance (or delay)
 191 mixing whereas Le is set constant at 10. Figure 3 presents the relation of S_a
 192 for the three parameters examined, each buoyancy ratio is presented in separate
 193 graphs (Fig. 3-A and B). The most effective means to enhance mixing turned
 194 out to be the magnitude of the buoyancy ratio ($|N|$). Regardless of Ra and θ ,
 195 for $N = -100$, S_a reaches about 50% or more at $t = 0.05$, while the curves
 196 for $N = -1$ present more moderate slopes. At the end of the simulation time
 197 ($t = 0.3$) the cases $N = -1$ have all of them evolved up to $S_a = 0.9$ or above.

198 With regard to Ra , as expected the curves display a faster mixing for higher
 199 Ra . The buoyant forces of both thermal and solute gradients are increased with

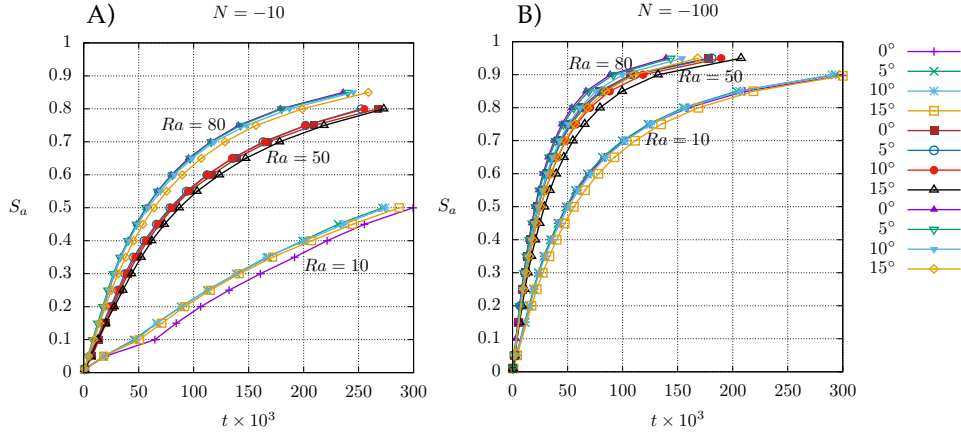


Figure 3: Average concentration S_a as a function of the governing parameters N , Ra , and θ .

200 Ra leading to a highly convective system. An interesting feature is that the
 201 curves corresponding to $Ra = 50$ and $Ra = 80$ for $N = -100$ form almost a
 202 single set of curves and difficult to distinguish between each other. Likewise,
 203 the set $Ra = 10$ appears less distant from $Ra = 50$ and $Ra = 80$ than in the
 204 case $N = -10$. This behavior is associated with the large magnitude of N ,
 205 that exerts a major control of the convection in the system. This feature is
 206 not evident for $N = -10$, where the three sets of curves corresponding with
 207 the three Rayleigh numbers are still clearly identified. Consequently, it can be
 208 concluded that Ra becomes a controlling parameter for moderate and small N .
 209 This is also expected from the inspection of Equation 12, where the buoyant
 210 force for the solute gradient is a multiple of Ra . Therefore, Ra and N are
 211 equally important to govern the flow system only when $N = -1$.

212 Figure 3 further shows that the effect of varying θ on the evolution of S_a is
 213 moderate for a given Ra and N . This effect appears to be negligible in some
 214 cases, for instance $N = -100$ and $Ra = 10$, where the curves $\theta=0^\circ$, 5° , and
 215 10° are almost equivalent. Moreover, the largest inclination angle, 15° , does
 216 not imply the most convective system. With a single exception ($N = -10$,
 217 $Ra = 10$), $\theta = 15^\circ$ delays mixing, even though moderately. Unlike purely ther-

mal convection, in which the increase of the slope angle (in the same space of parameters) leads to more convection-dominated transport with well defined multicellular convection [14], in this case solutal convection governs the convective transport with a strong dependency on N and more complex convective cell configurations. An inspection of S_a at long times shows the weak dependence of this parameter with regard to θ (Table 2). Even in the most diffusive case ($N = -10$, $Ra = 10$), S_a behaves similarly for every angle with differences no greater than 6%.

Table 2: Average concentration S_a at the end of the simulation time $t = 0.3$.

θ	S_a					
	$N = -10$			$N = -100$		
	$Ra = 10$	$Ra = 50$	$Ra = 80$	$Ra = 10$	$Ra = 50$	$Ra = 80$
0°	0.50	0.82	0.88	0.90	0.98	0.98
5°	0.53	0.83	0.88	0.90	0.98	0.98
10°	0.53	0.83	0.88	0.90	0.97	0.98
15°	0.51	0.82	0.87	0.90	0.97	0.98

4.2. Sherwood and Nusselt numbers

The behavior of the Sherwood number is presented in Figure 4 for limit angles (0° and 15°). There is a monotonic and steep decrease at the beginning when the transport is diffusive. This decrease lasts until the onset of convective mass transport characterized by fingering, the rebound on Sh is then followed by an oscillatory decrease characterized by merging of convective fingers. The decrease will continue until the porous medium is fully saturated. As expected, the time scale of the evolution of Sh varies considerably with N . For $N = -10$, the development from diffusion to merging of convective fingers takes about $t = 0.006$, whereas the same transition occurs for $t < 0.001$ in $N = -100$. Concerning the slope angle, it is evident from Figure 4-A and B that the inclination becomes a source of instability for the onset of convective fingers since $\theta = 0^\circ$ displays a slightly longer diffusive stage. With regard to $N = -100$ the inclination of the porous medium is accompanied by a more complex transition from

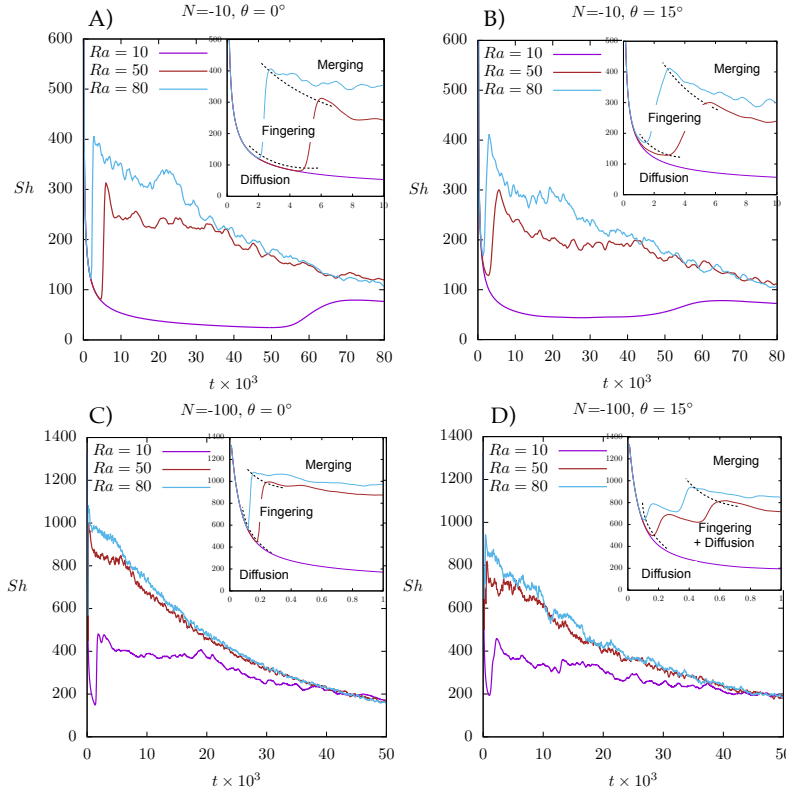


Figure 4: Sherwood number as a function of time for selected angles. The insets show the phases that characterize mass transport.

240 diffusion to convection, in which part of the upper boundary starts developing
 241 convection while the rest of it remains diffusive (Fig. 12), this region is labeled
 242 as a Fingering + Diffusion regime in the inset of Figure 4-D.

243 The evolution of the Nusselt number is in general oscillatory (Figure 5),
 244 with the exception of $N = -10$, $Ra = 10$, that displays a smoother trend for
 245 every θ . The curves present peaks of maximum Nu that appear considerably
 246 sooner in $N = -100$ (Fig. 5-C and D) in consistency with earlier convective
 247 effects associated with high N . It is important to highlight that the amplitude
 248 of the oscillations of Nu decreases over time, which indicates its relation with
 249 the mass flux through the boundary. As the fluid approaches saturation the

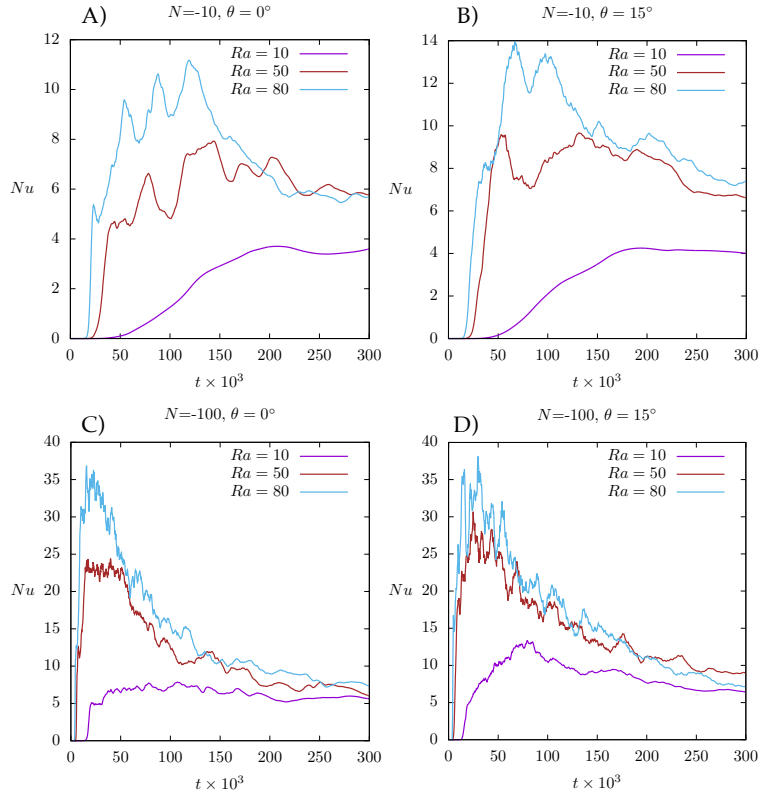


Figure 5: Nusselt number as a function of time for selected angles.

250 Nusselt number recovers a smoother trend (typical in pure thermal convection)
 251 because the mass flux starts vanishing and so do the fingering and merging that
 252 strongly and dynamically control transport at the upper boundary. A further
 253 evidence that the oscillations of Nu in the porous cavity are associated with the
 254 convective mass transport resides on the fact that Nu is smooth when Sh is so
 255 (4-A and B).

256 An alternative way to examine the behavior of Sh and Nu is integrating them
 257 over the simulation time in order to have a measure of the mass and heat fluxes
 258 over the entire period (from $t = 0$ to $t = 0.3$). These integrated Sh and Nu are
 259 presented in Figure 6. Two additional Rayleigh numbers ($Ra=50$ and $Ra=100$)
 260 and a buoyancy ratio ($N = -50$) are included in order to have wider view of

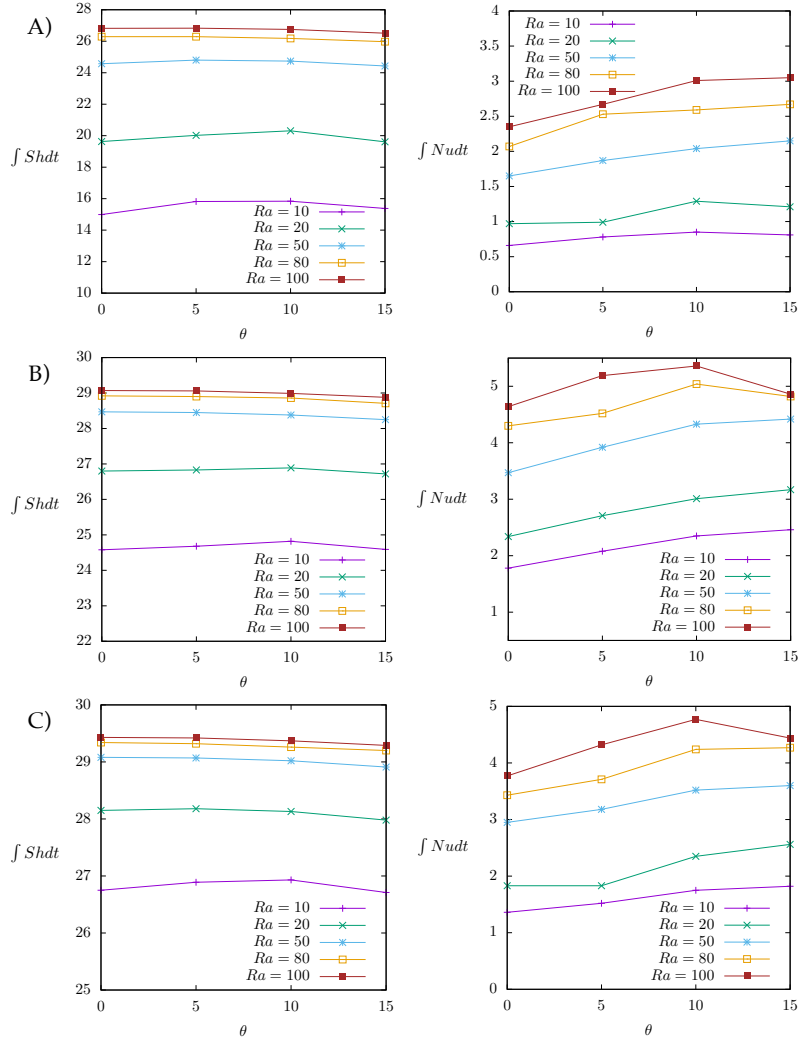


Figure 6: Integrated Sherwood and Nusselt numbers over the entire simulation time as a function of θ and for $Le = 10$: A) $N = -10$, B) $N = -50$, C) $N = -100$.

261 this parameter analysis. With respect to $\int Shdt$, there is a small sensitivity
 262 to θ , regardless of the buoyancy ratio (as expected from Figure 3). This is
 263 particularly evident for high Ra which curves display a small decrease with
 264 θ , associated with the formation of a preferential flow direction that decreases
 265 mass diffusion (Sec. 4.3). Additionally, as Ra increases the integrated Sherwood

266 number tends to constant value (see for example the small difference between
 267 $Ra = 80$ and $Ra = 100$). The vigorous convection associated with high Ra
 268 leads the system to saturation condition, so that the mass flux turns out almost
 269 equivalent.

270 As regards the Nusselt number (Fig. 6), there is a general trend of favoring
 271 the heat transfer increasing θ , in agreement with the case of purely thermal
 272 convection. This can be observed in Figure 7 that illustrates the effect of in-
 273 creasing the angle for a given set of governing parameters and the same t . For
 274 $\theta = 15^\circ$ the flow pattern tends to adopt a three-cell convective mode with two
 275 large upwelling thermal plumes, whereas $\theta = 0^\circ$ displays a lower magnitude
 276 multicellular convection highly conditioned by the convective mass transport.

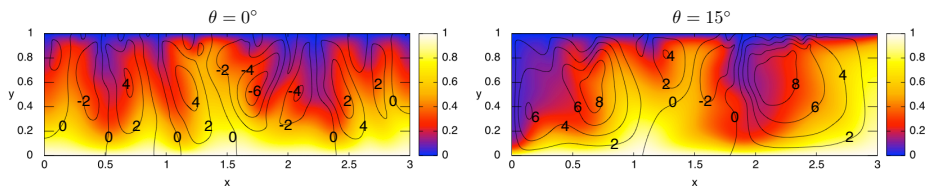


Figure 7: Temperature fields and streamlines for $N = 10$, $Ra = 50$ at $t = 0.15$.

277 It is convenient to compare this behavior with a different Lewis number. A
 278 more moderate case in which mass and thermal diffusivities are of the same order
 279 of magnitude ($Le = 2$) is presented in Figure 8. The mass transport ($\int Shdt$)
 280 presents a consistent behavior with Figure 6, however, smaller values of $\int Shdt$
 281 indicate smaller concentration gradients at the top boundary as a compensation
 282 for a higher mass diffusivity ($1/Le$, Eq. 10). An important exception is the most
 283 diffusive case ($Ra = 10$, $N = 10$) that displays a monotonic increase with θ , so
 284 that as the systems becomes more diffusive the inclination at these moderate
 285 angles can enhance mass transport. The integrated Nusselt number, on the
 286 other hand, strongly depends on the history of the convective regime, number
 287 and intensity of convective cells. Unlike the case $Le = 10$ (Fig. 6), it is observed
 288 less dependence of $\int Nudt$ on θ being essentially dependent on Ra and N .

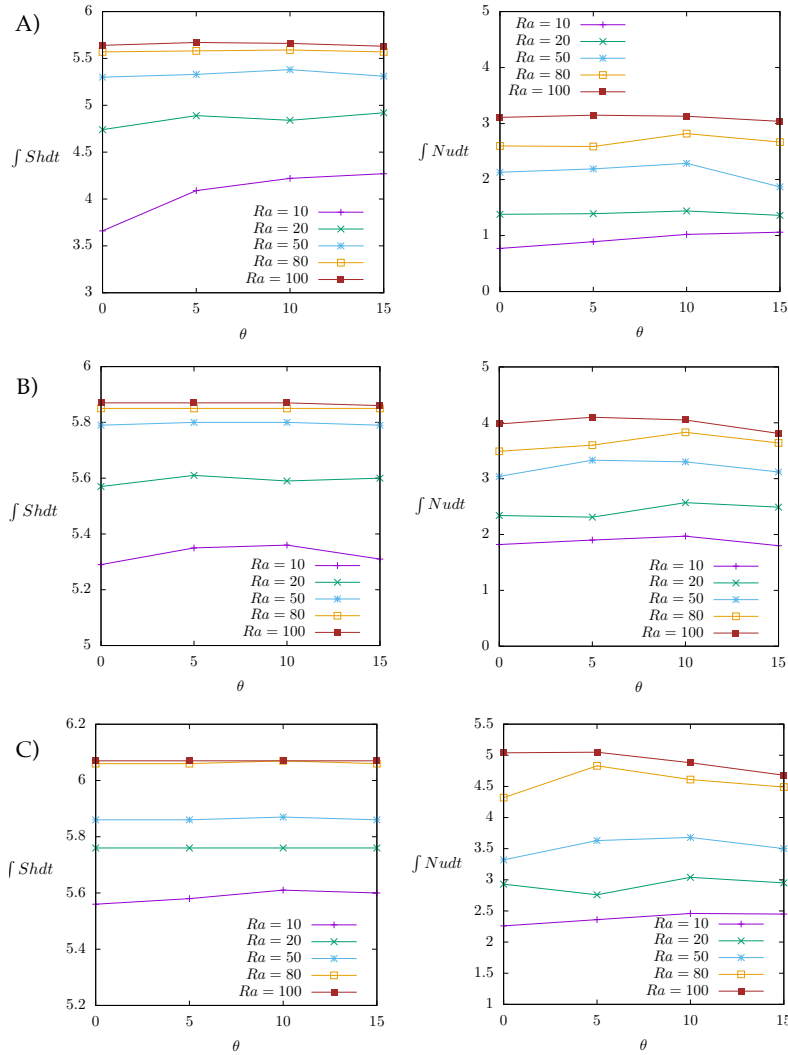


Figure 8: Integrated Sherwood and Nusselt numbers over the entire simulation time as a function of θ and for $Le = 2$: A) $N = -10$, B) $N = -50$, C) $N = -100$.

289 *4.3. Transient mass and heat transport*

290 The most representative convective features are described in this section.
 291 Firstly, the case with the lowest buoyancy forces is shown in Figure 9. Diffusive
 292 transport progresses faster for the temperature and subsequently the onset of
 293 convection takes place on the upper boundary. At the end of the simulation

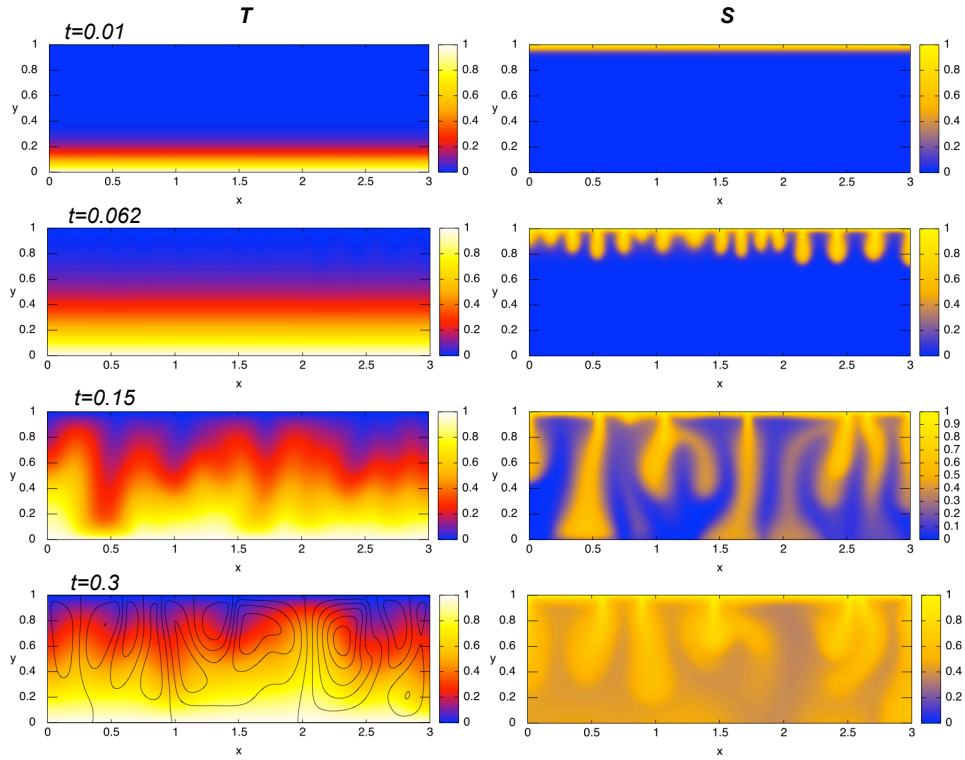


Figure 9: Concentration and temperature fields for $N = -10$, $Ra = 10$, and $\theta = 0^\circ$. Streamlines are included in the most developed stage.

294 time ($t = 0.3$) the average concentration in the cavity reaches about 50% with
 295 a multicellular convective pattern controlled by the downwelling mass flow.

296 As the Rayleigh number and the inclination angle are increased the onset of
 297 convection occurs earlier. This onset is characterize by smaller scale fingering
 298 and by the development of a large downwelling at the left boundary ($x = 0$).
 299 This preferred flow path remains throughout the entire simulation time giving
 300 rise to zonation of mass concentration in two opposite corners of the porous
 301 enclosure. This convective pattern is accompanied by the formation of two
 302 large thermal plumes ($t = 0.05$) unlike the horizontal case.

303 With regard to high buoyancy forces ($N = -100$), for the horizontal porous
 304 medium (Figure 11, video format available as supplementary material), the

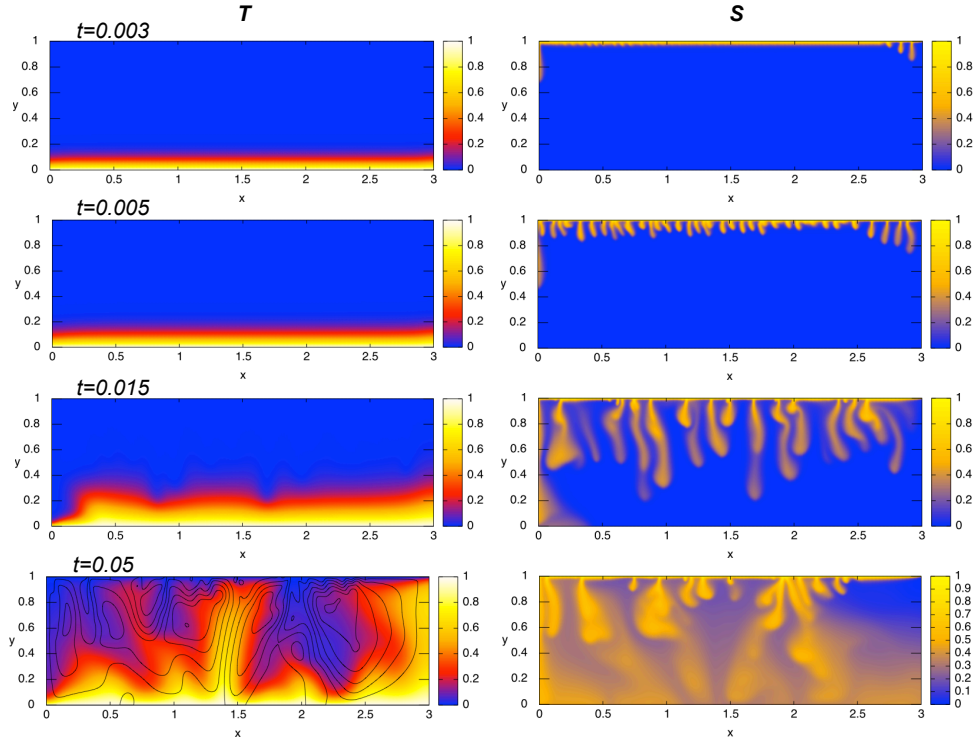


Figure 10: Concentration and temperature fields for $N = -10$, $Ra = 50$, and $\theta = 15^\circ$.

305 onset of convection is characterized by small scale fingering with a short diffusive
 306 stage. Downwelling mass flow is characterized by multiple fingers that remain
 307 throughout the simulation time. In this case about 50% of average concentration
 308 has been attained at $t = 0.02$. The flow pattern at this time consists of a complex
 309 arrangement of convective cells. On the basis that this flow pattern is highly
 310 dynamic, it explains the strong oscillatory character of Nu at these stages of
 311 the simulation.

312 Finally, in a combination of high N with an inclination angle $\theta = 15^\circ$ (Figure
 313 12, video format available as supplementary material), the onset of convection
 314 at the upper boundary is characterized by an uneven distribution, with the
 315 formation and development of downwelling mass flow for small and moderate
 316 values of x , whereas the remaining section of the boundary is still in a diffusive

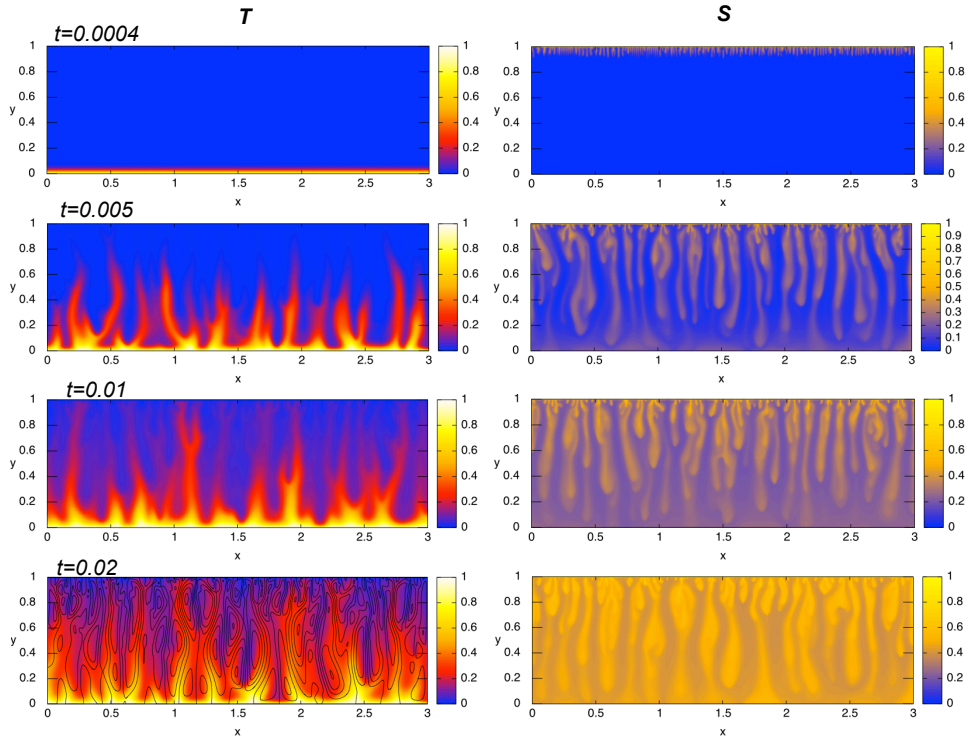


Figure 11: Concentration and temperature fields for $N = -100$, $Ra = 50$, and $\theta = 0^\circ$. A video format of this figure is available online as supplementary material.

317 stage or in an early stage of convective transport. The preferential direction of
 318 flow controlled by θ leads to an accelerated saturation in a zone of small x and
 319 y with high temperature gradients (heating zone), whereas the opposite corner
 320 of the cavity becomes a low saturation zone accompanied by a strong thermal
 321 upwelling (cooling zone).

322 5. Conclusion

323 To gain further physical understanding of the problem of underground CO_2
 324 dissolution, we conducted transient numerical simulations to assess the convec-
 325 tive mass and heat transport in a porous medium heated from below, salted
 326 from above, and subject to an inclination angle. We focused in a set of gov-

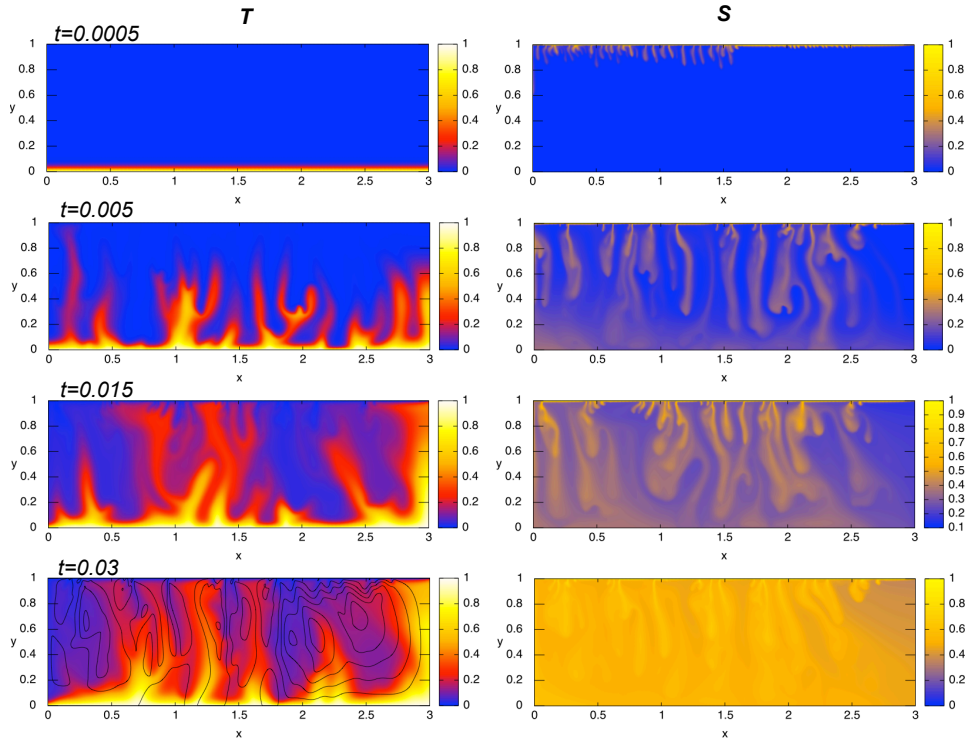


Figure 12: Concentration and temperature fields for $N = -100$, $Ra = 50$, and $\theta = 15^\circ$. A video format of this figure is available online as supplementary material.

327 erning parameters given by two buoyancy ratios: $N = -10$, $N = -100$; three
 328 Rayleigh numbers: $Ra=10$, $Ra=50$, and $Ra=80$; and four inclination angles:
 329 $\theta = 0^\circ, 5^\circ, 10^\circ$, and 15° . Our results provided a quantitative insight on how
 330 mixing is enhanced as the buoyancy forces are increased. We also described the
 331 potential consequences that θ can have on the rate of solute mixing and in the
 332 form that onset of convection (fingering) takes place. Finally, the implications
 333 that θ can have on the flow patterns and heat transfer properties were identified.
 334 The concluding remarks are summarized in the following key points:

- 335 • Even though the Rayleigh number and the buoyancy ratio considerably
 336 speed up the saturation of the fluid with the solute, the inclination angle
 337 has a minor effect. Furthermore, a slight decrease in the mixing rate can

338 occur when increasing θ as a consequence of preferential flow paths that
339 force the solute flow through high saturation zones in the porous medium.

340 • For high buoyancy ratios, the inclination of the porous medium has the
341 consequence of an uneven onset of convection at the boundary, being the
342 most stable part that in the opposite direction to the gravitational force
343 component.

344 • On the basis that thermal diffusivity is higher than mass diffusivity ($Le =$
345 10 in all the simulations), convective heat transport is controlled by con-
346 vective mass transport in early stages of dissolution. Thermal upwellings
347 adopt the shape of low solute concentration plumes. With regard to θ , the
348 heat transfer properties of the cavity (Nu) tend to increase with the incli-
349 nation, since the convective patterns allow less but more intense thermal
350 upwellings.

351 These concluding remarks help improve our understanding of fundamental
352 behavior of double diffusive convection in porous media in the context of
353 solute dissolution. We envisage further work that takes into account other
354 important effects such as surface tension in the upper boundary (which is
355 important for the case of miscible fluids) and heterogeneities.

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- 361 [1] S. Bouzgarrou, H. S. Harzallah, K. Slimi, Unsteady Double Diffusive Natu-
362 ral Convection in Porous Media-Application to CO₂ Storage in Deep Saline
363 Aquifer Reservoirs, in: Salame, C and Khoury, G and Aillerie, M (Ed.),
364 Terragreen 13 International Conference 2013 - Advancements in Renew-
365 able Energy and Clean Environment, volume 36 of *Energy Procedia*, Ter-
366 raGreen, 2013, pp. 756–765. doi:{10.1016/j.egypro.2013.07.088}, Ter-
367 raGreen International Conference on Advancements in Renewable Energy
368 and Clean Environment, Beirut, LEBANON, FEB 15-17, 2013.
- 369 [2] A. W. Islam, M. A. R. Sharif, E. Carlson, Numerical investigation of
370 double diffusive natural convection of CO₂ in a brine saturated geothermal
371 reservoir, *Geothermics* 48 (2013) 101 – 111.
- 372 [3] H. Emami-Meybodi, H. Hassanzadeh, C. P. Green, J. Ennis-King, Con-
373 vective dissolution of CO₂ in saline aquifers: Progress in modeling and
374 experiments, *International Journal of Greenhouse Gas Control* 40 (2015)
375 238 – 266. Special Issue commemorating the 10th year anniversary of the
376 publication of the Intergovernmental Panel on Climate Change Special Re-
377 port on CO₂ Capture and Storage.
- 378 [4] J. L. Bischoff, R. J. Rosenbauer, Salinity variations in submarine hydrother-
379 mal systems by layered double-diffusive convection, *Journal of Geology* 97
380 (1989) 613–623.
- 381 [5] M. J. Colçao, G. S. Dulikravich, Solidification of double-diffusive flows
382 using thermo-magneto-hydrodynamics and optimization, *Materials and*
383 *Manufacturing Processes* 22 (2007) 594–606.
- 384 [6] Y. Hao, J. J. Nitao, T. A. Buscheck, Y. Sun, Double-diffusive natural con-
385 vection in a nuclear waste repository, *Nuclear Technology* 163 (2008) 38–46.
386 International High-Level Radioactive Waste Management Conference, Las
387 Vegas, NV, APR 30-MAY 04, 2006.
- 388 [7] D. A. Nield, Onset of thermohaline convection in a porous medium, *Water*
389 *Resources Research* 4 (1968) 101 – 111.

- 390 [8] J. W. Taunton, T. Green, E. N. Lightfoot, Thermohaline instability and
391 salt fingers in a porous medium, *Physics of Fluids* 15 (1972) 748 – 753.
- 392 [9] O. V. Trevisan, A. Bejan, Mass and heat-transfer by high rayleigh number
393 convection in a porous-medium heated from below, *International Journal*
394 *of Heat and Mass Transfer* 30 (1987) 2341–2356.
- 395 [10] N. D. Rosenberg, F. Spera, Thermohaline convection in a porous-medium
396 heated from below, *International Journal of Heat and Mass Transfer* 35
397 (1992) 1261–1273.
- 398 [11] D. K. Lin, Unsteady natural-convection heat and mass-transfer in a satu-
399 rated porous enclosure, *Warme und Stoffubertragung - Thermo and Fluid*
400 *Dynamics* 28 (1993) 49–56.
- 401 [12] M. Mamou, P. Vasseur, E. Bilgen, Multiple solutions for double-diffusive
402 convection in a vertical porous enclosure, *International Journal of Heat*
403 *and Mass Transfer* 38 (1995) 1787–1798.
- 404 [13] M. Sen, P. Vasseur, L. Robillard, Multiple steady-states for unicellular
405 natural-convection in an inclined porous layer, *International Journal of*
406 *Heat and Mass Transfer* 30 (1987) 2097–2113.
- 407 [14] F. J. Guerrero-Martínez, N. Karimi, E. Ramos, Numerical modeling of
408 multiple steady-state convective modes in a tilted porous medium heated
409 from below, *International Communications in Heat and Mass Transfer* 92
410 (2018) 64 – 72.
- 411 [15] M. Mamou, P. Vasseur, E. Bilgen, Double-diffusive convection instability in
412 a vertical porous enclosure, *Journal of Fluid Mechanics* 368 (1998) 263–289.
- 413 [16] I. Sezai, A. A. Mohamad, Three-dimensional double-diffusive convection
414 in a porous cubic enclosure due to opposing gradients of temperature and
415 concentration, *Journal of Fluid Mechanics* 400 (1999) 333–353.

- 416 [17] P. Nithiarasu, T. Sundararajan, K. N. Seetharamu, Double-diffusive nat-
417 ural convection in a fluid saturated porous cavity with a freely convecting
418 wall, *International Communications in Heat and Mass Transfer* 24 (1997)
419 1121–1130.
- 420 [18] F. Y. Zhao, D. Liu, G. F. Tang, Free convection from one thermal and
421 solute source in a confined porous medium, *Transport in Porous Media* 70
422 (2007) 407–425.
- 423 [19] F. Y. Zhao, D. Liu, G. F. Tang, Natural convection in a porous enclo-
424 sure with a partial heating and salting element, *International Journal of*
425 *Thermal Sciences* 47 (2008) 569 – 583.
- 426 [20] N. Hadidi, R. Bennacer, Three-dimensional double diffusive natural con-
427 vection across a cubical enclosure partially filled by vertical porous layer,
428 *International Journal of Thermal Sciences* 101 (2016) 143–157.
- 429 [21] R. D. Jagadeesha, B. M. R. Prasanna, M. Sankar, Double diffusive convec-
430 tion in an inclined parallelogrammic porous enclosure, *Procedia Engineer-*
431 *ing* 127 (2015) 1346 – 1353. *International Conference on Computational*
432 *Heat and Mass Transfer (ICCHMT) - 2015.*
- 433 [22] A. Riaz, M. Hesse, H. Tchelepi, F. Orr, Onset of convection in a gravi-
434 tationally unstable diffusive boundary layer in porous media, *Journal of*
435 *Fluid Mechanics* 548 (2006) 87–111.
- 436 [23] A. C. Slim, T. Ramakrishnan, Onset and cessation of time-dependent,
437 dissolution-driven convection in porous media, *Physics of Fluids* 22 (2010)
438 124103.
- 439 [24] M. Szulczewski, R. Juanes, The evolution of miscible gravity currents in
440 horizontal porous layers, *Journal of Fluid Mechanics* 719 (2013) 82–96.
- 441 [25] A. W. Islam, H. R. Lashgari, K. Sephernoori, Double diffusive natural
442 convection of CO₂ in a brine saturated geothermal reservoir: Study of non-

- 443 modal growth of perturbations and heterogeneity effects, *Geothermics* 51
444 (2014) 325–336.
- 445 [26] A. Islam, A. K. N. Korrani, K. Sepehrnoori, T. Patzek, Effects of geo-
446 chemical reaction on double diffusive natural convection of CO₂ in brine
447 saturated geothermal reservoir, *International Journal of Heat and Mass*
448 *Transfer* 77 (2014) 519 – 528.
- 449 [27] E. B. Soboleva, Density-driven convection in an inhomogeneous geothermal
450 reservoir, *International Journal of Heat and Mass Transfer* 127 (2018) 784
451 – 798.
- 452 [28] S. Mrityunjay, A. Chaudhuri, P. H. Stauffer, R. J. Pawar, Simulation of
453 gravitational instability and thermo-solutal convection during the dissolu-
454 tion of CO₂ in deep storage reservoirs, *Water Resources Research* 56 (2020)
455 e2019WR026126.
- 456 [29] D. A. Nield, A. Bejan, *Convection in Porous Media*, 4th ed., Springer, New
457 York, 2013.
- 458 [30] F. J. Guerrero-Martínez, P. L. Younger, N. Karimi, Three-dimensional
459 numerical modeling of free convection in sloping porous enclosures, *Inter-*
460 *national Journal of Heat and Mass Transfer* 98 (2016) 257–267.
- 461 [31] F. J. Guerrero-Martínez, P. L. Younger, N. Karimi, S. Kyriakis, Three-
462 dimensional numerical simulations of free convection in a layered porous
463 enclosure, *International Journal of Heat and Mass Transfer* 106 (2017)
464 1005–1013.
- 465 [32] H. K. Versteeg, W. Malalasekera, *An Introduction to Computational Fluid*
466 *Dynamics, The Finite Volume Method*, Prentice Hall, 1995.
- 467 [33] E. Báez, A. Nicolás, 2D natural convection flows in tilted cavities: Porous
468 media and homogeneous fluids, *International Journal of Heat and Mass*
469 *Transfer* 49 (2006) 4773–4785.