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Large time behaviour of the solution of a parabolic-hyperbolic system modelling the codiffusion of isotopes

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Abstract.

We study the asymptotic behaviour of the solution of a hyperbolic-parabolic problem in an isolated domain when $t \to \infty$. The problem models the diffusion of n species of radiative isotopes of the same element in a medium. The model is based on the assumption that, since the isotopes are chemically indistinguishable, the flux of each isotope depends on the gradient of the total concentration of the element weighted by the relative percentage of the isotope. We show that the asymptotic behaviour strongly depends on the radiative law, and, in some cases, on the pointwise distribution of the initial concentration

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1 Introduction

We consider the model proposed in [4] for the diffusion of n species of isotopes of the same element in a medium and based on the assumption that the flux of the i^{th} component J_i is given by

$$J_i = -D_i \frac{c_i}{c} \nabla c, \quad i = 1, ..., n, \qquad c = \sum_{i=1}^{n} c_i, \tag{1.1}$$

where the coefficients D_i are related to the interaction among the isotopes, which are chemically indistinguishable, so that each component feels the gradient of the total element concentration in a relative percentage $\frac{c_i}{c}$.

In the case of radioactive isotopes, we have to take into account the radioactive decay law, which for spatially homogeneous distributions is a linear ODE system

$$\dot{\mathbf{C}} = \Lambda \mathbf{C}, \quad \mathbf{C} = (c_1, ..., c_n) \in \mathbb{R}^n, \tag{1.2}$$

Λ suitable $n \times n$ constant matrix, so that the problem is

$$c_{it} = -\mathrm{div} J_i + \Lambda \mathbf{C}, \qquad i = 1, ..., n. \tag{1.3}$$

If we assume that $D_1 = \max D_j, D_n = \min D_j$, $c_n = c - \sum_{j=1}^{n} c_j$ and define $\tilde{\mathbf{C}} = (c_1, ..., c_{n-1}, c)$ then system (1.3) becomes

$$\begin{cases} c_{it} = \left(D_i \frac{c_i}{c} c_x \right)_x + (\tilde{\Lambda} \tilde{\mathbf{C}})_i \qquad i = 1, ..., n-1 \\ c_t = (a(\tilde{\mathbf{C}}) c_x)_x + (\tilde{\Lambda} \tilde{\mathbf{C}})_n, \end{cases} \tag{1.4}$$

where $a = D_n + \sum_{j=1}^{n-1} (D_j - D_n) \frac{c_i}{c}$ and $\tilde{\Lambda}$ is obviously obtained from Λ (see [4]). In the physical assumption that $0 \leq c_i \leq c$, $i = 1, ..., n$, we have that the total concentration c satisfies a uniformly parabolic quasilinear equation in divergence form, since $0 < D_n \leq a \leq D_1$, $\forall c_i, c \geq 0$. Hence $c(x,t)$ has a "parabolic" behaviour while, once c is given, the equations for the single species c_i are first order linear equations, so that we expect for c_i a "hyperbolic" behaviour. In particular c_i will have finite speed of propagation and will in general be non smooth for $t > 0$, showing the possibility of the onset of regions depleted of a component c_i and of strong oscillations also asymptotically, say a "asymptotic localization property".

This behaviour is in accordance with experimental data, as we have shown also numerically (see [6], where we considered the case in which all D_i are equal). The physical motivation for model (1.1)-(1.3) comes in fact from examples of discordance between predictions and experiments in the co-diffusion of isotopic molecules (see [9], [10]): many studies often assume that the isotopes ratios are constant, at the so called "secular equilibrium", however the analysis of isotopes in long-term experiments in bedrock and buffer materials (see [16]) evidentiates changes in the original isotope composition, called "fractionations". This phenomenon can be observed e.g. in the field of Uranium mobilization in geological media where there is, next to fractures surfaces, a narrow zone where the

U^{234}, U^{238} activity ratio (the ratio of the concentrations of the two isotopes) is significantly different from unity, i.e. from secular equilibrium. In some sense, the activity ratio does not seem to support a simple classical diffusion mechanism: there are jagged areas of enrichment and depletion of one isotope over the other, not as expected from classical diffusion theory, even if the profile of the total Uranium is diffusion-like.

The assumption (1.1) on the flux, which gives rise to the "hyperbolic problem", is an approximation of a more complete model ([21], [3]), denoted "parabolic problem" where the resulting problem consists in a parabolic system strongly coupled:

$$\frac{\partial c_i}{\partial t} = \mathrm{div}\left(\tilde{D}_i \nabla c_i + D_i \frac{c_i}{c} \nabla c\right) + \Lambda \mathbf{C}, \quad i = 1, ..., n, \quad c = \sum_{i=1}^{n} c_i, \tag{1.5}$$

where the additional term $\tilde{D}_i \nabla c_i$ in the flux is taken into account, being $\tilde{D}_i$ the usual parabolic diffusion coefficients, due to the interaction of the i^{th} component with the surrounding medium. The mathematical analysis of the above system in a bounded domain, with Dirichlet boundary conditions was the object of paper [7], in the physically relevant assumption that the total concentration of the element is positive and bounded, i.e.

$$K \geq c_i \geq k > 0, \quad i = 1, ..., n, \quad k, K \text{ constant}.$$

Aim of the present paper is the study of the asymptotic behaviour of the solution of the problem in an isolated domain when $t \to \infty$, in the assumption that all the D_i are equal. We will show in Sec.3 that the asymptotic behaviour as $t \to \infty$ strongly depends on the radiative law, i.e. on Λ.
Quite surprising it turns out that there are cases in which the asymptotic strongly depends on the pointwise distribution of the initial concentration, the simplest case being the one of a set of stable isotopes, e.g. (Cl^{37}, Cl^{35}), i.e. $\Lambda = 0$. But there are physical cases in which the asymptotic is uniform, that is a typically parabolic behaviour, see e.g. (U^{234}, U^{238}), or more generally Example 2 in Sect.4, where we will give some explicit examples of matrix Λ and discuss the corresponding behaviour.

2 Statement of the problem

After a suitable scaling on time, we can set $D_i = D = 1$, $\forall i$ in system (1.4), and consider the following Neumann problem in a bounded domain $\Omega = \{|x| < L\}$:

$$\begin{cases} c_{it} = \left(\dfrac{c_i}{c} c_x\right)_x + (\tilde{\Lambda}\tilde{\mathbf{C}})_i, & i = 1, ..., n-1 \\ c_t = c_{xx} + (\tilde{\Lambda}\tilde{\mathbf{C}})_n, & \\ c_i(x,0) = c_{i0}(x) \geq 0, \quad i = 1, ..., n-1, & x \in \Omega, \\ c_i(-L,t)\dfrac{c_x(-L,t)}{c(-L,t)} = c_i(L,t)\dfrac{c_x(L,t)}{c(L,t)} = 0, & t > 0. \end{cases} \tag{2.1}$$

As we mentioned before, in the physical assumptions

H1) $c_{i0} \in H^{2+l}(\overline{\Omega}), \quad l > 0, \quad i = 1, ..., n, \qquad 0 \leq c_{i0} \leq K, \qquad c_0 = \sum_{i=1}^{n} c_{i0} > 0,$

H2) positivity property for ODE: if $c_{i0} \geq 0$, then $c_i(t) \geq 0, \;\; i = 1, ..., n,$

there exists a unique classical solution of (2.1), moreover the total concentration c satisfies a uniformly parabolic equation, see Thm 5.3, 5.4 of [4]; here we use the notations of [18]. Then, as c is strictly positive, the solution constructed along the characteristics is the "viscosity solution" obtained as the limit of the complete physical model (1.5), with $\tilde{D}_i = \tilde{D} \neq 0, \; D_i = D = 1$ as $\tilde{D} \to 0$ (see [4]).

Remark that assumption **H2)** together with the uniqueness of the solution of the ODE (1.2) imply that if $\mathbf{C}_0 \neq \mathbf{0}$ and $c_{i0} \geq 0$, then $c = \sum_i c_i > 0$ for $t \geq 0$ and $c_i \leq c, \quad i = 1, ..., n.$

A common feature of this class of models (see also [2], [13], [14]) is that the ratio $r_i = \frac{c_i}{c}$ has an evolution law simpler than the one of c_i. In the case of isotopes, r_i is related to the activity ratio which e.g. for two species is $\frac{c_1}{c_2}$, but it is simpler to deal with mathematically, since it is bounded by 1:

$$r_{it} = r_{ix}\frac{c_x}{c} + P_i(r_1, ..r_{n-1}), \qquad i = 1, ..., n-1, \tag{2.2}$$

where P_i is a polinomial of second degree.
This means that each r_i evolves along the characteristics with the same law as the ratios $\dfrac{c_i(t)}{\sum_{i=1}^n c_i(t)}$, $c_i(t)$ being the solution of the ODE (1.2) $\dot{\mathbf{C}} = \Lambda\mathbf{C}$.

Since c is positive and regular, then we can define the characteristics starting at time $t = 0$ in $x = x_0$ by $X(t; x_0)$ as

$$\frac{dX(t;x_0)}{dt} = -\frac{c_x}{c}\bigg|_{x=X(t;x_0)}, \quad X(0;x_0) = x_0. \tag{2.3}$$

In the assumptions **H1)**, **H2)** the evolution of r_i on the characteristics satisfies a positivity property, i.e.

$$0 \leq r_{i0} = \frac{c_{i0}}{c_0} \leq 1 \text{ then } 0 \leq r_i(X(t), x_0) \leq 1$$

and (r_i, c) are solutions of the following problem:

$$\begin{cases} r_{it} = \dfrac{c_x}{c} r_{ix} + P_i(r_1, ...r_{n-1}), \qquad i = 1, ..., n-1 \\ c_t = c_{xx} + bc, \\ c_x(-L, t) = c_x(L, t) = 0, \qquad t > 0, \\ c(x, 0) = c_0(x), \qquad x \in \Omega, \\ r_i(x, 0) = \dfrac{c_{i0}(x)}{c_0(x)}, \qquad i = 1, ..., n-1, \end{cases} \tag{2.4}$$

where b depends on $r_1, ...r_{n-1}$ and is defined by

$$b = (\tilde{\Lambda}\tilde{\mathbf{r}})_n, \quad \tilde{\mathbf{r}} = (r_1, ..., r_{n-1}, 1). \tag{2.5}$$

A main information on the behaviour of the characteristics is obtained considering the total mass and the mass of a single specie among two characteristics (the same information

is used in Ref. [2]), in particular, since we deal with a Neumann homogeneous problem, $x = -L$ and $x = L$ are two characteristics, so that we will consider the characteristics $x = -L$ and $x = X(t; x_0)$, $x_0 \leq L$ to define:

$$\begin{aligned} &m_i(x,t) = \textstyle\int_{-L}^{x} c_i(\xi,t)\, d\xi, \quad i = 1, ..., n-1, \\ &m(x,t) = \textstyle\int_{-L}^{x} c(\xi,t)\, d\xi, \\ &\mathbf{m} = (m_1, ..., m_{n-1}, m), \end{aligned} \tag{2.6}$$

and the masses in the whole $\overline{\Omega}$ as

$$\mathbf{M}(t) = \mathbf{m}(L,t). \tag{2.7}$$

We proved in [4] that both $\mathbf{M}(t)$ and $\mathbf{m}(X(t;x_0),t)$ are the solutions of the ODE

$$\dot{\mathbf{Y}} = \tilde{\Lambda}\mathbf{Y}, \tag{2.8}$$

with initial datum $\mathbf{Y}_0$ derived from $\mathbf{C}_0(x)$, i.e.

$$\begin{aligned} &\mathbf{m}(X(0;x_0),0) = \mathbf{m}_0(x_0), \qquad x_0 \in \overline{\Omega}, \\ &m_{i0}(x) = \textstyle\int_{-L}^{x} c_{i0}(\xi)\, d\,\xi, \quad i = 1, ..., n-1, \\ &m_0(x) = \textstyle\int_{-L}^{x} c_0(\xi)\, d\,\xi. \end{aligned} \tag{2.9}$$

We pointed out that both the evolution in time along the characteristics of the r_i and of the masses m_i is the same as the one of the solution of the corresponding spatially homogeneous problem.

Since we want to study a set of isotopes which either decay or are stable, we will assume that all the eigenvalues of the matrix Λ in (1.2) are non positive, precisely:

H3) All the eigenvalues of Λ are real, nonpositive and if the maximum eigenvalue is zero, then it is semisimple (i.e. its algebraic multiplicity and geometric multiplicity coincide)

In assumption **H3)** any solution of $\dot{\mathbf{Y}} = \tilde{\Lambda}\mathbf{Y}$ with bounded initial datum $\mathbf{Y_0}$ is bounded for any time, remarking that the eigenvalues of $\tilde{\Lambda}$ coincide with the ones of Λ, so we can assume **H3)** directly on $\tilde{\Lambda}$.

From the ODE theory (see e.g. [1], [12]), we have a complete description of the solutions of the ODE. Assuming that $\tilde{\Lambda}$ has $s \leq n$ distinct eigenvalues $\lambda_s < ... < \lambda_1 \leq 0$, we denote, for $i = 1, ..., s$:

$\mu(\lambda_i)$ = algebraic multiplicity of λ_i,
$\nu(\lambda_i)$ = geometric multiplicity of λ_i,
$E(\lambda_i)$ = generalized autospace of λ_i,
$h(\lambda_i)$ = the least integer k such that $\mathrm{Ker}\,(\tilde{\Lambda} - \lambda_i I)^{k+1} = \mathrm{Ker}\,(\tilde{\Lambda} - \lambda_i I)^{k}$,
so that $E(\lambda_i) = \mathrm{Ker}\,(\tilde{\Lambda} - \lambda_i I)^{h(\lambda_i)}$, with $I = Id$ matrix $n \times n$.

Any solution is a linear combination of the product of exponential functions time polinomials ones. Quite precisely:

$$\mathbf{Y}(t) = \sum_{i=1}^{s} \left[\sum_{k=0}^{h(\lambda_i)-1} (\tilde{\Lambda} - \lambda_i I)^k \frac{t^k}{k!} \right] e^{\lambda_i t} \mathbf{Y}_{0,i}, \tag{2.10}$$

with $\mathbf{Y}_0 = \sum_{i=1}^{s} \mathbf{Y}_{0,i},\ \mathbf{Y}_{0,i} \in E(\lambda_i)$.
Assumption **H3)** implies that the leading term in (2.10) as $t \to +\infty$ is of the order $t^{h(\lambda_1)-1}e^{\lambda_1 t}$ and hence

$$\begin{aligned} &\lim_{t\to+\infty} t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}\mathbf{Y}(t;\mathbf{Y}_0) = \\ &\frac{1}{(h(\lambda_1)-1)!}(\tilde{\Lambda}-\lambda_1 I)^{h(\lambda_1)-1}\mathbf{Y}_{01} = \hat{B}\mathbf{Y}_0. \end{aligned} \tag{2.11}$$

Here $\hat{B}$ is a constant $n \times n$ matrix, determined by the autospaces $E(\lambda_i)$, $i = 1, ..., s$. To be precise, denoted by

$$\begin{aligned} E(\lambda_i) &= \{\mathbf{v}^{i,1}, ..., \mathbf{v}^{i,\mu(\lambda_i)}\}, \\ B^{n\times n} &= (\mathbf{v}^{1,1}, ..., \mathbf{v}^{s,\mu(\lambda_s)}\}, \\ \tilde{B} &= B\,\mathrm{diag}\,(I_{\mu(\lambda_1)}, 0)B^{-1}, \end{aligned} \tag{2.12}$$

we have $\mathbf{Y}_{01} = \tilde{B}\mathbf{Y}_0$ and $\hat{B} = \dfrac{1}{(h(\lambda_1)-1)!}(\tilde{\Lambda}-\lambda_1 I)^{h(\lambda_1)-1}\tilde{B}\mathbf{Y}_0$.

Concerning the mass $m(x,t)$ we have that it is a linear function of x and

$$\lim_{t\to+\infty} t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}m(x,t) = \frac{x+L}{2L}M_\infty \tag{2.13}$$

uniformly in $\overline{\Omega}$, where $M_\infty = (\hat{B}\mathbf{m}_0(L))_n > 0$, for any initial data $\tilde{\mathbf{C}}_0 \in \Gamma = \{\mathbf{y} \in \mathbb{R}^n : y_n > 0,\ 0 \le y_i \le y_n,\ i = 1, ..., n-1,\ (\hat{B}\mathbf{y})_n > 0\}$.
In the next Section we will prove a similar result for $m(x,t)$ for general not spatially homogeneous initial data, see Thm 3.1.
Finally for any $\mathbf{C}_0 \in \Gamma$ we have

$$\lim_{t\to+\infty} t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}\tilde{\mathbf{C}} = \hat{B}\tilde{\mathbf{C}}_0. \tag{2.14}$$

3 Asymptotic behaviour for $t \to \infty$

With the notations of Section 2, we define

$$F(x) = (\hat{B}\tilde{\mathbf{C}}_0(x))_n, \quad \mathbf{F}(x) = (F_1, ..., F_{n-1}, F) = \hat{B}\tilde{\mathbf{C}}_0(x). \tag{3.1}$$

Remark that the matrices involved are constant matrices, depending only on $\tilde{\Lambda}$. Moreover, let $\mathbf{M}(t)$ to be the solution of $\dot{\mathbf{Y}} = \tilde{\Lambda}\mathbf{Y}$ with initial datum $\mathbf{Y}_0 = \mathbf{m}_0(L)$ and, as in Sec.2,

$$M_\infty = (\hat{B}\mathbf{m}_0(L))_n = \int_{-L}^{L} F(\xi)\,d\xi, \tag{3.2}$$

then we have the following

Theorem 3.1 *In the assumptions* **H1)**, **H2)**, **H3)** *and for any* $\tilde{\mathbf{C}}_0(x)$ *such that* $F(x) \ge \delta > 0,\ \forall x \in \overline{\Omega}$ *we have*

$$\lim_{t\to+\infty} t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}m(x,t) = \frac{x+L}{2L}M_\infty, \tag{3.3}$$

uniformly in $\overline{\Omega}$*, with* $M_\infty > 0$.

Proof.

Defined $u = (1+t)^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}c(x,t)$, then u is solution of

$$\begin{cases} u_t = u_{xx} + \left(b - \lambda_1 - \dfrac{h(\lambda_1)-1}{1+t}\right)u = u_{xx} + \tilde{b}u, \quad x \in \Omega,\ t > 0, \\ u(x,0) = c_0(x), \quad x \in \Omega, \\ u_x(-L,t) = u_x(L,t) = 0, \quad t > 0. \end{cases} \tag{3.4}$$

Since b depends only on $(r_1, ..., r_{n-1})$ and the r_i evolve on each characteristic as the solution of the ODE problem, we have the following uniform estimate for the coefficient $\tilde{b}$ in (3.4):

Lemma 3.1

$$|\tilde{b}| \le k(\delta)\left[\frac{h(\lambda_1)-1}{t^2} + (s-1)e^{\frac{\lambda_2-\lambda_1}{2}t}\right] = k(\delta)g(t),\ \forall x \in \Omega,\ t \ge 1, \tag{3.5}$$

Then, by classical methods, we have that u is uniformly bounded for any time, precisely

$$0 \le u \le \max c_0\ e^{\int_0^t k(\delta)g(s)\,ds} \le k(\delta, \max c_0),\ \forall t \ge 0. \tag{3.6}$$

Let us define now

$$v(x,t) = (1+t)^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}m(x,t), \tag{3.7}$$

v is solution of

$$\begin{cases} v_t = v_{xx} + \int_{-L}^{x} \tilde{b}\,u\,d\xi = v_{xx} + f(x,t), \quad x \in \Omega,\ t > 0, \\ v(x,0) = m_0(x), \quad x \in \Omega, \\ v(-L,t) = 0, \qquad t > 0, \\ v(L,t) = (1+t)^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}m(L,t), \quad t > 0. \end{cases} \tag{3.8}$$

Since $m(L,t)$ evolves as the ODE, see (2.13), we have from the assumptions of the Theorem that

$$v(L,t) \to M_\infty \text{ as } t \to \infty.$$

Since u is uniformly bounded for any time, from Lemma 3.1 we have that $f(x,t)$ tends to zero uniformly in $\overline{\Omega}$ as $t \to \infty$.

From classical results, see [11], Cap.VI, Thm.1, (3.3) follows.

□

Proof. of Lemma 3.1

Fix any $x_0 \in \Omega$, then on the characteristic $X(t;x_0)$ for $t \ge 1$ we have, see (2.5):

$$b - \lambda_1 - \frac{h(\lambda_1)-1}{t} = \frac{(\tilde{\Lambda}\tilde{\mathbf{C}})_n}{c} - \lambda_1 - \frac{h(\lambda_1)-1}{t}\bigg|_{x=X(t;x_0)},$$

with $\tilde{\mathbf{C}}$ solution of the ODE $\dot{\mathbf{Y}} = \tilde{\Lambda}\mathbf{Y}$ and $\mathbf{Y}_0 = \tilde{\mathbf{C}}_0$.
Hence

$$b - \lambda_1 - \frac{h(\lambda_1)-1}{t}\bigg|_{x=X(t;x_0)} = \frac{1}{c}\left(\left(\tilde{\Lambda} - \lambda_1 I - \frac{h(\lambda_1)-1}{t} I\right)\tilde{\mathbf{C}}\right)_n. \tag{3.9}$$

From the explicit expression for $\tilde{\mathbf{C}}$ (see (2.10)) we have for the denominator c in previous expression:

$$c(X(t;x_0),t) = e^{\lambda_1 t} t^{h(\lambda_1)-1} F(x_0) + z_n(x_0,t), \tag{3.10}$$

with

$$|z_n e^{-\lambda_1 t} t^{-(h(\lambda_1)-1)}| \le k_1\left(\frac{h(\lambda_1)-1}{t} + (s-1)e^{\frac{\lambda_2-\lambda_1}{2}t}\right),$$

and for the numerator in (3.9):

$$\left(\tilde{\Lambda} - \lambda_1 I - \frac{h(\lambda_1)-1}{t} I\right)\tilde{\mathbf{C}} = \mathbf{a}(x_0,t) + \mathbf{z}_2(x_0,t), \tag{3.11}$$

with

$$||\mathbf{a}|| \le k_2(h(\lambda_1-1)t^{h(\lambda_1)-3}e^{\lambda_1 t}, \qquad ||\mathbf{z}_2|| \le k_3(s-1)t^{h(\lambda_1)-1}e^{\lambda_1 t}e^{\frac{\lambda_2-\lambda_1}{2}t},$$

for any $x \in \overline{\Omega}$, $t \ge 1$.
All the constants above, k_j, $j = 1,2,3$, depend on $\tilde{\Lambda}$ and on max $||\tilde{\mathbf{C}}_0(x_0)||$ in $\overline{\Omega}$.
The assumption $F(x_0) \ge \delta > 0$ together with the above estimates give the result of the Lemma.

□

The asymptotic behaviour of the characteristics follows from the previous Theorem:

Corollary 3.1 *In the hypotheses of Thm 3.1 we have that*

$$\lim_{t\to+\infty} X(t;x_0) = X_\infty(x_0) = \frac{2L}{M_\infty}\int_{-L}^{x_0} F(\xi)\,d\xi - L, \tag{3.12}$$

uniformly in $\overline{\Omega}$.

Proof. Recalling that $m(X(t;x_0),t)$ evolves in time like the n^{th} component of the solution of the ODE $\dot{\mathbf{Y}} = \tilde{\Lambda}\mathbf{Y}$ with initial datum $\mathbf{m}_0(x_0)$, we have

$$\lim_{t\to+\infty} t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}m(X(t;x_0),t) = (\hat{B}\mathbf{m}_0(x_0))_n = \int_{-L}^{x_0} F(\xi)\,d\xi = m^*(x_0) < M_\infty.$$

The uniform convergence of the mass $m(x,t)$ proves the result, remarking that, for any t, m is a monotone increasing function of x and $m^*(x_0)$ is increasing in x_0, so that there exists a unique X_∞ such that $\dfrac{X_\infty + L}{2L}M_\infty = m^*(x_0)$.

□

Let us denote by $X_\infty^{-1}(x)$ the inverse function of the monotone increasing function $X_\infty(x)$ defined in (3.12). From the previous Corollary and from the proof of Thm.3.1, we obtain the behaviour of the ratioes $r_i = \dfrac{c_i}{c}$, $i = 1,...,n-1$:

Corollary 3.2 *In the hypotheses of Thm 3.1 we have that*

$$\lim_{t\to+\infty} r_i(x,t) = r_{i\infty}(x) = \frac{F_i(X_\infty^{-1}(x))}{F(X_\infty^{-1}(x))}, \qquad i = 1, ..., n-1, \tag{3.13}$$

uniformly in $\overline{\Omega}$.

Proof.

Let us recall again that any r_i evolves along one fixed characteristic like $\frac{c_i}{c,}$, with $\tilde{\mathbf{C}}$ solution of the ODE $\dot{\mathbf{Y}} = \tilde{\Lambda}\mathbf{Y}$ with initial datum $\tilde{\mathbf{C}}_0$
Moreover from the explicit expressions for c and c_i we have (see (3.1))

$$r_i = \frac{F_i + z_i e^{-\lambda_1 t} t^{-(h(\lambda_1)-1)}}{F + z_n e^{-\lambda_1 t} t^{-(h(\lambda_1)-1)}}, \quad i = 1, ..., n-1,$$

where, as in Lemma 3.1, the following estimate holds:

$$||\mathbf{z}|| \le k_1 \left(\frac{h(\lambda_1)-1}{t} + (s-1) e^{\frac{\lambda_2-\lambda_1}{2} t} \right).$$

Therefore we have that (3.12) holds uniformly in $\overline{\Omega}$.

□

Remark 3.1

1. . If $\mu(\lambda_1) = 1$ then $h(\lambda_1) = 1$, and $E(\lambda_1) = \{\mathbf{v}^1\}$, where $\mathbf{v}^1$ is the eigenvector corresponding to λ_1, so that $\mathbf{F}(x) = \hat{B}\tilde{\mathbf{C}}_0(x) = \alpha_1(x)\mathbf{v}^1$, α_1 depending on the initial data.

   Therefore in this case, for any $x \in \overline{\Omega}$,

   $$r_{i\infty}(x) = \frac{v^{1,i}}{v^{1,n}}, \qquad i = 1, ..., n-1,$$

   so that we have a unique limit independent of x for any initial datum satisfying the assumptions of Thm 3.1. This is what is called "secular equilibrium for the set of isotopes", see e.g. the couple (U^{234}, U^{238}) and other examples in Sec.4.

2. . If $1 < \mu(\lambda_1) = \nu(\lambda_1)$ then again $h(\lambda_1) = 1$, but $E(\lambda_1) = \{v^{1,1}, ..., v^{1,\mu(\lambda_1)}\}$, $v^{1,i}$ being the $\mu(\lambda_1)$ independent eigenvectors of $\tilde{\Lambda}$ corresponding to λ_1. Therefore

   $$\mathbf{F}(x) = \hat{B}\tilde{\mathbf{C}}_0(x) = \alpha_1(x)v^{1,1} + ... + \alpha_{\mu(\lambda_1)}(x)v^{1,\mu(\lambda_1)},$$

   and $r_{i\infty}(x)$ strongly depend on the pointwise distribution of $\tilde{\mathbf{C}}_0$, this is e.g. the case of a set of n stable isotopes, i.e. $\Lambda = 0$.

3. . If $1 \le \nu(\lambda_1) < \mu(\lambda_1)$ then $h(\lambda_1) > 1$. Recalling the explicit limits (2.11), (2.12) and remarking that the vectors

   $w^{1,i} = (\tilde{\Lambda} - \lambda_1 I)^{h(\lambda_1)-1} v^{1,i}$, $i = 1, ..., \mu(\lambda_1)$

are either zeros or eigenvectors of $\tilde{\Lambda}$ corresponding to λ_1, we have that $r_{i\infty}$ either behaves as in case 1 or as in case 2. For example if $\nu(\lambda_1) = 1$, then all the $w^{1,i}$ not null are multiple of the unique eigenvector of $\tilde{\Lambda}$, $\mathbf{v}^1$, so that $\mathbf{F}(x) = \hat{B}\tilde{\mathbf{C}}_0(x) = \alpha_1(x)\mathbf{v}^1$ and hence we have, as in case 1, a unique spatially homogeneous limit for r_i, see Example 2 in Sec. 4.

As a conclusion we have that, depending on the radiative decay law, that is on Λ, we can have either a "parabolic behaviour" with a secular equilibrium as in case 1, or a "hyperbolic behaviour" possibly with strong oscillations in the limit as in case 2.

Remark 3.2

We have considered assumption **H3)** in order to describe the physical problem of decaying or stable isotopes, but clearly (see (2.10)), all the results of Section 3 hold without any restriction on the sign of λ_i, provided we still denote by λ_1 the maximum eigenvalue.

4 Examples and comments

Example 1
In the general case of n isotopes, assume that each one is either stable or decays out of the element, that is Λ is a diagonal matrix and the ODE is

$$\dot{c}_i = -\gamma_i c_i, \quad i = 1, ..., n, \quad \gamma_i \geq 0. \tag{4.1}$$

This is the case e.g. of the stable couple (Cl^{37}, Cl^{35}), i.e. $\Lambda = 0$, and of the radiative couple (U^{235}, U^{238}), for which $0 < \gamma_1 < \gamma_2$, very close one to the other so that as a first approximation they can be taken equal.
Without loss of generality we can order the diagonal element, setting
$-\lambda_1 = \gamma_1 \leq \gamma_2 \leq ... \leq \gamma_n$. In this case $h(\lambda_1) = 1$ and we have that:

1.a) if $\mu(\lambda_1) = 1$, asymptotically we have the unique isotope 1, with

$$F(x) = c_{10}(x), \quad M_\infty = m_{10}(L),$$

and the limits for the r_i are uniform for any initial datum such that $c_{10} \geq k > 0$, that is

$$\lim_{t\to\infty} r_i = \delta_1^i, \qquad i = 1, ..., n-1. \tag{4.2}$$

1.b) if $n \geq \mu(\lambda_1) > 1$ then the asymptotic distribution of the r_i strongly depends on the initial data. Precisely:

$$F(x) = \sum_{i=1}^{\mu(\lambda_1)} c_{i0}(x), \quad M_\infty = \sum_{i=1}^{\mu(\lambda_1)} m_{i0}(L),$$

and

$$\begin{array}{l} \lim_{t\to\infty} r_i(x,t) = \dfrac{c_{i0}(X_\infty^{-1}(x))}{\sum_{i=1}^{\mu(\lambda_1)} c_{i0}(X_\infty^{-1}(x))}, \qquad i = 1, ..., \mu(\lambda_1), \\ \lim_{t\to\infty} r_i(x,t) = 0, \qquad i = \mu(\lambda_1), ..., n. \end{array} \tag{4.3}$$

This can be observed in particular in the cases $\Lambda = 0$ and $\Lambda = -\gamma Id$ in which the r_i are constant along the characteristics.

Example 2
A chain of n isotopes of which the i^{th} decays into the $(i+1)^{th}$, for any $i = 1, ..., n-1$, and the n^{th} decays out of the element according to the following ODE:

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1, \\ \dot{c}_i = \gamma_{i-1} c_{i-1} - \gamma_i c_i, \quad i = 2, ..., n, \end{cases} \tag{4.4}$$

with $\gamma_i > 0,\ i = 1, ..., n$.
This is the case e.g. of U^{234}, U^{238}, for which $0 < \gamma_1 << \gamma_2$. In this example the asymptotic distribution is always uniform.
Infact let us analyze three possible situations:

2.a) Suppose that all the γ_i are distinct, as it usually happens in nature, then the matrix $\tilde{\Lambda}$ has n negative distinct eigenvalues $-\gamma_i$, $i = 1, ..., n$ and the solution of the ODE $\dot{\tilde{\mathbf{C}}} = \tilde{\mathbf{\Lambda}}\tilde{\mathbf{C}}$ with initial datum $\tilde{\mathbf{C}}_0$ can be expressed directly in the form:

$$\tilde{\mathbf{C}}(t, \tilde{\mathbf{C}}_0) = \Gamma(t)\Gamma(0)^{-1}\tilde{\mathbf{C}}_0, \tag{4.5}$$

with $\Gamma(t) = (e^{-\gamma_1 t}\mathbf{v}^1, ..., e^{-\gamma_n t}\mathbf{v}^n)$ fundamental matrix for $\tilde{\Lambda}$ (see [1]), and a direct computation of the eigenvectors gives for any $k = 1, ..., n$:

$$\begin{array}{l} v^{k,i} = 0, \qquad i = 1, ...k-1, \\ v^{k,i} = \prod_{j=i}^{n-1} \dfrac{\gamma_{j+1} - \gamma_k}{\gamma_j}, \qquad i = k, ..., n-1, \\ v^{k,n} = 1 + \sum_{i=1}^{n-1} \prod_{j=i}^{n-1} \dfrac{\gamma_{j+1} - \gamma_k}{\gamma_j}. \end{array} \tag{4.6}$$

Denoted by $\lambda_1 = -\min_{i=1,...,n} \gamma_i$ the maximum eigenvalue, say $-\gamma_k$, then

$$\tilde{\mathbf{C}}(t, \tilde{\mathbf{C}}_0)e^{\gamma_k t} \to \left(\Gamma(0)^{-1}\tilde{\mathbf{C}}_0\right)_k \mathbf{v}^k = \alpha_k \mathbf{v}^k = \mathbf{F},$$

as $t \to \infty$.

Let us remark that both $\Gamma(0)$ and $\Gamma(0)^{-1}$ are inferior triangular matrices and, if $\gamma_k = \min \gamma_i$, then the eigenvector $\mathbf{v}^k$ has the first $k-1$ components null and the other positive. Therefore we have that:

if $\gamma_1 = \min_{i=1,...,n} \gamma_i$, then all the components of $\mathbf{v}^1$ are positive and

$$F(x) = c_{10}(x)\frac{v^{1,n}}{v^{1,1}}, \qquad M_\infty = \frac{v^{1,n}}{v^{1,1}} \int_{-L}^{L} c_{10}(\xi)d\xi,$$

$$r_{i\infty} = \frac{v^{1,i}}{v^{1,n}} > 0, \qquad i = 1, ..., n-1. \tag{4.7}$$

Therefore we have "secular equilibrium" for all isotopes, each one in a proportion depending only on γ_i and independent of the initial data, provided the physical assumption $c_{10} \geq \delta > 0$, that guarantees that the isotope 1 is initially present, i.e. the start of the chain. Let us remark that also M_∞ depends on the initial mass of the isotope 1.

If $\gamma_k = \min \gamma_i$, $1 < k \leq n$, then $\mathbf{v}_k$ has the first $k-1$ components null, and the others positive.

Then $r_{i\infty} = 0$ for $i = 1, ..., k-1$ and positive for $i \geq k$, which means that the isotopes $1, ..., k-1$ vanish asymptotically, and the "secular equilibrium" holds only for the isotopes $k, ..., n$. However now $\alpha_k(x)$ and M_∞ depend on $c_{i0}(x)$, $i = 1, ..., k$.

In particular, when $k = n$ only the n^{th} isotope is present at ∞.

2.b) Again let $\lambda_1 = -\min \gamma_i$ be the maximum eigenvalue, with $\mu(\lambda_1) = 1$, the other eigenvalues possibly being non distinct.

Then the eigenspace $E(\lambda_1)$ again consists of the unique eigenvector corresponding to λ_1, so that the r_i behave like in case 2.a). However the matrix $\Gamma(0)$ depends now on the generalized eigenvectors, so that the dependence of $\mathbf{F}$ on the initial data is different from the previous one.

2.c) If $\mu(\lambda_1) > 1$, since in this example the geometric multiplicity $\nu(\lambda_i) = 1$ $\forall \lambda_i$, then, recalling Remark 3.1.3, we have that if $\gamma_k = \min \gamma_i$, then

$$\mathbf{F}(x) = \hat{B}\tilde{\mathbf{C}}_0(x) = \beta(x)\mathbf{v}^k,$$

with $\mathbf{v}^k$ given by (4.6) (remark that if there is an index $i > k$ such that $\gamma_k = \gamma_i$ then $v^{k,j} = 0$, for $j \leq i-1$, so that all the isotopes up to $i-1$ desappear asymptotically).

Therefore the behaviour of r_i as $t \to \infty$ is again uniform.

Let us remark that when all the eigenvalues coincide, that is $\gamma_i = \gamma > 0$, $i = 1, ..., n$, then since the matrix Λ is multiple of a Jordan normal form, we have $(\mathbf{F})_i(x) = \dfrac{c_{10}(x)}{(n-1)!}\delta_i^n$ and $h(\lambda_1) = n$. Therefore only the n-th isotope remain asymptotically, but $M_\infty = \dfrac{m_{10}(L)}{(n-1)!}$ depends only on the initial mass of the isotope 1.

Example 3

A chain of n isotopes such that the i^th decays into the $(i+1)^{th}$, for any $i = 1, ..., n-1$, and the n^{th} is stable. That is the radiative law is the same as in Example 2 (see (4.4)) but with $\gamma_n = 0$, $\gamma_i > 0$, $i = 1, ..., n-1$.
In this case the equation for the total concentration c is uncoupled and the ODE $\dot{\tilde{\mathbf{C}}} = \tilde{\mathbf{\Lambda}}\tilde{\mathbf{C}}$ becomes

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1, \\ \dot{c}_2 = \gamma_1 c_1 - \gamma_2 c_2, \\ \vdots \\ \dot{c}_{n-1} = \gamma_{n-2} c_{n-2} - \gamma_{n-1} c_{n-1}, \\ \dot{c} = 0. \end{cases} \tag{4.8}$$

The maximum eigenvalue is $\lambda_1 = 0$ and then $\mathbf{F}(x) = (0, ..., 0, c_0(x))$, $M_\infty = m_0(L)$ so that $r_i \to 0$, $i = 1, ..., n-1$ as $t \to \infty$, that is for any physical initial datum only the stable isotope remains asymptotically.

Remark 4.1

Many other examples combinations of the previous ones could be considered. Let us limit ourselves to one that is a mixture of Ex.1 and Ex.2, for which isotopes 1 and 3 decade out of the element and isotope 2 decades in isotope 3, that is:

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1, \\ \dot{c}_2 = -\gamma_2 c_2, \\ \dot{c}_3 = \gamma_2 c_2 - \gamma_3 c_3, \end{cases} \tag{4.9}$$

It can be shown that if the γ_i are all distinct, then the asymptotic distribution for r_i is uniform, while if e.g. $\gamma_1 = \gamma_2 < \gamma_3$ then

$$F(x) = c_{10}(x) + \frac{\gamma_3}{\gamma_3 - \gamma_1} c_{20}(x).$$

If moreover $F \geq \delta > 0$, then

$$r_1 \to \frac{c_{10}(\hat{x})}{F(\hat{x})}, \qquad r_2 \to \frac{c_{20}(\hat{x})}{F(\hat{x})},$$

with $\hat{x} = X_\infty^{-1}(x)$, that is we obtain an asymptotic non uniform distribution which depends on the pointwise initial concentrations, that is an example of Remark 3.1.2.

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