Rend. Istit. Mat. Univ. Trieste
Volume 44 (2012), 133–151

On the asymptotic behaviour of the characteristics in the codiffusion of radioactive isotopes with general initial data

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Dedicated to Fabio Zanolin on the occasion of his 60th birthday

Abstract. *The large-time behaviour of the solution of a hyperbolic-parabolic problem in an isolated domain, which models the diffusion of n species of radiative isotopes of the same element, is studied, assuming general hypotheses on the initial data.*
Depending on the radiative law and on the distribution of the initial concentration, either a uniform distribution for the concentration of each isotope or the presence of oscillations may be possible when $t \to \infty$.

Keywords: isotopes, diffusion, hyperbolic equations
MS Classification 2010: 35L50, 35K57, 35B05

1. Introduction

Let us consider the following problem in $\Omega = (-L, L)$:

$$\begin{cases} c_{it} = \left(\dfrac{c_i}{c} c_x\right)_x + \displaystyle\sum_{j=1}^{n} \Lambda_{ij} c_j, & x \in \Omega,\ t > 0, \\ c_i(x,0) = c_{i0}(x) \geq 0, & x \in \Omega, \\ c_i \dfrac{c_x}{c}(-L,t) = c_i \dfrac{c_x}{c}(L,t) = 0, & t > 0, \\ i = 1, \ldots, n, \ \ c = \sum_{k=1}^{n} c_k. \end{cases} \tag{1}$$

The problem comes from a model for the diffusion of n species of isotopes of the same element in a medium, in the assumption that the flux of the $i-th$ species, whose concentration is c_i, is

$$J_i = -\frac{c_i}{c} c_x,\ i = 1, \ldots, n,\ \ x \in \Omega,$$

where $c = \sum_{i=1}^{n} c_i$ is the total concentration.
This assumption means that any component varies with the total gradient of the element in a relative percentage $\frac{c_i}{c}$ (see [7, 20]).

Actually the above law for the flux is an approximation of a more complete model where the flux is $J_i = -(\tilde{D}_i c_{ix} + D_i \frac{c_i}{c} c_x)$. If one assumes $D_i = 0$ then the problem becomes a classical parabolic problem whose solution does not quite agree with the experimental data (see [20]). On the other hand there are physical situations, such as self-diffusion, in which it is sensible to try the model with $\tilde{D}_i = 0$, thus obtaining solutions more in agreement with experimental data, at least qualitatively.

Moreover, it would be reasonable, for solutes, that the coefficients D_i are practically the same for all isotopic molecules of the element, as they have the same partial molar volume and the same electronic configuration, especially for the heavier chemical elements. Although it would be interesting from a mathematical point of view to study the model in the general hypothesis that the diffusion coefficients are different (see [7]), numerical simulations evidentiate no significant difference in the qualitative behaviour of the solution in dependence on the diffusion coefficients D_i, here assumed to be all equal to 1 after rescaling (see [6]). For more details on the physical motivations of the model see [5].

The coefficients Λ_{ij} are the elements of a constant $n \times n$ matrix Λ which expresses the "radiative decay law" in the case of radiative isotopes. In the physically relevant hypothesis that $\mathbf{C} = (c_1, ..., c_n)$ is regular and satisfies

$$c_{i0}(x) \geq 0, \qquad c_0 = \sum_{i=1}^{n} c_{i0}(x) > 0, \tag{2}$$

there exists a unique classical non negative solution (see Section 2 for the precise assumptions, [7] for the complete model and [5] in the present case). We remark that it has been proved that the total concentration c satisfies a parabolic equation with data $c_x(\pm L) = 0$ and it is regular and strictly positive for any $t \geq 0$. Once c is given, the concentrations c_i for the single isotopes are solutions of linear hyperbolic first order equations and they can be derived by means of the method of the characteristics, defined by the total concentration. In this case, denoted by $X(t; x_0)$ the characteristic starting in x_0 at time 0, we have:

$$\frac{dX(t;x_0)}{dt} = -\frac{c_x}{c}\bigg|_{x=X(t;x_0)}, \qquad X(0;x_0) = x_0. \tag{3}$$

Let us remark that if the initial total concentration $c_0(x)$ has zeroes, there can be effects of "loss of regularity". Actually it can happen that, also if the data are regular, c_i has discontinuities for positive time. Although from a physical point of view it is more sensible to consider c_0 small rather than

$c_0 \equiv 0$, a mathematical approach to the hyperbolic problem was performed in [5], defining, also if the data are regular, a weak solution as in [2]. Let us stress that, since the total concentration satisfies a uniform parabolic equation, it will be strictly positive for any positive time also if it is initially zero on subintervals. The problem is that this initial "holes" may possibly cause the c_i to be discontinuous for positive time (for details see [5]). Since we wish to understand first the asymptotic behaviour for physically relevant initial data, possibly strongly oscillating but smooth, we need to assume $c_0 > 0$. In this assumption, one can use the results of [4] and show that the solution constructed along the characteristics is the "viscosity solution" obtained as the limit of the complete physical model, with $\tilde{D}_i = \tilde{D} \neq 0,\ D_i = D = 1$ as $\tilde{D} \to 0$. Numerical simulations confirm this result, also for the complete physical model, in very general situations, and they have been performed using a program for solving parabolic equations, with initial data possibly zero ([6]); however the proof of existence and uniqueness of the solution of the complete parabolic problem and its convergence to the hyperbolic problem in the possible presence of zeroes in the initial total concentration is still an open problem.

We remark that the asymptotic behaviour of the solutions for $t \to \infty$ strongly depends on the decay law, that is on Λ, and on the first significative term of the asymptotic expansion for $t \to \infty$ of the solutions of the ODE

$$\begin{cases} \dot{\mathbf{C}} = \Lambda \mathbf{C}, \qquad \mathbf{C} = (c_1, ..., c_n), \\ \mathbf{C}(0) = \mathbf{C}_0. \end{cases} \tag{4}$$

These results are evidentiated in [8], under strong assumptions on the positivity of the initial data in the whole Ω. However there are physically relevant initial data that do not satisfy such assumptions in the whole Ω but still the corresponding solution should have a similar asymptotic behaviour. In the present paper we will study the problem assuming the most general hypotheses.

2. Statement of the problem

Existence and uniqueness of a classical non negative solution of Problem (1) have been obtained in [5] under the following assumptions:

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

H2) positivity property for the ODE (4):
if $c_{i0} \ge 0$, then $c_i(t) \ge 0,\ \ i = 1, ..., n,$

Since we want to consider a set of isotopes which either decay or are stable, it is natural to assume that all the eigenvalues of the matrix Λ are real non positive, actually we can assume:

H3) all the eigenvalues of Λ are real.

Due to the structure of Problem (1) it is convenient to consider instead of $\mathbf{C}$, $\tilde{\mathbf{C}} = (c_1, ...c_{n-1}, c)$, $c = \sum_{i=1}^{n} c_i$, then (4) is transformed in the following:

$$\begin{cases} \dot{\tilde{\mathbf{C}}} = \tilde{\boldsymbol{\Lambda}}\tilde{\mathbf{C}}, \\ \tilde{\mathbf{C}}(0) = \tilde{\mathbf{C}}_0, \quad \tilde{\mathbf{C}}_0 = (c_{10}, ..., c_{(n-1)0}, c_0). \end{cases} \tag{5}$$

where $\tilde{\Lambda}$, for which **H3** holds too, is given by

$$\tilde{\Lambda} = \begin{pmatrix} \Lambda_{11} - \Lambda_{1n} & \dots & \Lambda_{1n} \\ \Lambda_{21} - \Lambda_{2n} & \dots & \Lambda_{2n} \\ \vdots & \ddots & \vdots \\ \sum_{m=1}^{n}(\Lambda_{m1} - \Lambda_{mn}) & \dots & \sum_{m=1}^{n} \Lambda_{mn} \end{pmatrix}.$$

Assuming that $\tilde{\Lambda}$ has $s \leq n$ distinct eigenvalues $\lambda_s < ... < \lambda_1$, for $i = 1, ..., s$, let us denote by (see [1, 12])

$\mu(\lambda_i) =$ 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 s.t. $\mathrm{Ker}\,(\tilde{\Lambda} - \lambda_i I)^{k+1} = \mathrm{Ker}\,(\tilde{\Lambda} - \lambda_i I)^k$,

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 polynomials. Quite precisely:

$$\tilde{\mathbf{C}}(t) = \sum_{i=1}^{s} \left[\sum_{k=0}^{h(\lambda_i)-1} (\tilde{\Lambda} - \lambda_i I)^k \frac{t^k}{k!} \right] e^{\lambda_i t} \tilde{\mathbf{C}}_{0,i}, \tag{6}$$

with $\tilde{\mathbf{C}}_0 = \sum_{i=1}^{s} \tilde{\mathbf{C}}_{0,i}$, $\tilde{\mathbf{C}}_{0,i} \in E(\lambda_i)$.

Therefore, since λ_1 is the highest eigenvalue, we have:

$$\begin{aligned} &\lim_{t \to +\infty} t^{-(h(\lambda_1)-1)} e^{-\lambda_1 t} \tilde{\mathbf{C}}(t; \tilde{\mathbf{C}}_0) \\ &\quad = \frac{1}{(h(\lambda_1) - 1)!} (\tilde{\Lambda} - \lambda_1 I)^{h(\lambda_1)-1} \tilde{\mathbf{C}}_{0,1} = \hat{B}\tilde{\mathbf{C}}_0. \end{aligned} \tag{7}$$

Here $\hat{B}$ is a constant $n \times n$ matrix, determined by the $E(\lambda_i)$ (see [8]).

Given $\tilde{\mathbf{C}}_0(x),\ x \in \overline{\Omega}$, let us define:

$$\mathbf{F}(x) = \hat{B}\tilde{\mathbf{C}}_0(x), \quad F(x) = (\hat{B}\tilde{\mathbf{C}}_0(x))_n. \tag{8}$$

Let us remark that the positivity hypothesis **H2** together with **H1** guarantees $F(x) \geq 0$, moreover, if for some x_0 $F(x_0) = 0$, then $\mathbf{F}(x_0) = \mathbf{0}$.

We proved in [8, Theorem 3.1], that, assuming **H1**, **H2**, **H3**, for any initial datum $\tilde{\mathbf{C}}_0$ such that

H4) $F(x) \geq \delta > 0$ in $\overline{\Omega}$,

we have

$$\lim_{t\to+\infty} t^{-(h(\lambda_1)-1)} e^{-\lambda_1 t} m(x,t) = \frac{x+L}{2L} M_\infty, \tag{9}$$

uniformly in Ω, where

$$m(x,t) = \int_{-L}^{x} c(\xi,t)\,d\xi, \quad M_\infty = \int_{-L}^{L} F(\xi)\,d\xi. \tag{10}$$

Then the first asymptotic term for the total concentration c is given by $t^{h(\lambda_1)-1} e^{\lambda_1 t} \dfrac{M_\infty}{2L}$, that is a uniform distribution of the total concentration, and this is in agreement with the physics of the problem.

Moreover, once the characteristics have been defined as in (3), it is possible to get their asymptotic behaviour, and precisely (see [8, Corollary 3.1]):

$$\lim_{t\to+\infty} X(t;x_0) = X_\infty(x_0) = \frac{2L}{M_\infty} \int_{-L}^{x_0} F(\xi)\,d\xi - L. \tag{11}$$

The hypothesis **H4** ensures that the function $X_\infty(x_0)$ is monotone increasing, and consequently it is possible to obtain the information on the ratioes $r_i = \dfrac{c_i}{c},\ i = 1, ...n-1,\ \dfrac{c_n}{c} = 1 - \sum_{i=1}^{n-1} r_i$, precisely:

$$\lim_{t\to+\infty} r_i(x,t) = \frac{F_i(X_\infty^{-1}(x))}{F(X_\infty^{-1}(x))}, \quad i = 1, ..., n-1 \tag{12}$$

uniformly in Ω (see [8, Corollary 3.2]).

Of course, if $M_\infty = 0$, that is $F \equiv 0$, the first significative term of the asymptotic expansion of m and c changes, but it is natural to investigate what happens if $F \not\equiv 0$ but e.g. it is null in a subset of Ω.

In order to better understand the question, let us consider the couple of isotopes (U^{238}, U^{234}) whose decay law is:

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1 \\ \dot{c}_2 = \gamma_1 c_1 - \gamma_2 c_2, \qquad 0 < \gamma_1 < \gamma_2, \end{cases} \tag{13}$$

that is the isotope 1, U^{238}, decays into the isotope 2, U^{234}, and the second one decays out of the element. In this example one can see that $F(x) = \dfrac{\gamma_2 - \gamma_1}{\gamma_2} c_{10}(x)$. If the isotope 1 is not present initially (i.e. $c_{10} \equiv 0$), then the solution is $c_1 \equiv 0$ and $c_2 \equiv c = e^{-\gamma_2 t} w(x,t)$, with $w(x,t)$ solution of

$$\begin{cases} w_t = w_{xx}(x), & x \in \Omega,\ t > 0, \\ w(x,0) = c_0(x), & x \in \Omega, \\ w_x(\pm L, t) = 0, & t > 0, \end{cases}$$

that is, for large time,

$$m(x,t) \simeq e^{-\gamma_2 t} \frac{x+L}{2L} \int_{-L}^{L} c_0(\xi)\, d\xi, \quad \text{and } r \equiv 0.$$

If on the contrary assumption **H4** holds, that is the isotope 1 is initially present everywhere in Ω, then from (9)-(12):

$$\begin{cases} m(x,t) \simeq e^{-\gamma_1 t} \dfrac{x+L}{2L} \left(1 - \dfrac{\gamma_1}{\gamma_2}\right) \displaystyle\int_{-L}^{L} c_{10}(\xi)\, d\xi, \\ r(x,t) \simeq r_E = 1 - \dfrac{\gamma_1}{\gamma_2}, \end{cases}$$

uniformly in Ω, with $0 < r_E < 1$. We have in this case the so called "secular equilibrium" of the two isotopes, that are both present in Ω for $t > 0$ and tend, for $t \to \infty$, respectively to r_E, $1 - r_E$. The question is what happens if the isotope 1 is absent only in a subset of Ω but $M_\infty > 0$. We will prove in the sequel that the asymptotic behaviour of m is still given by (9).

Other significant examples will be analyzed in Section 4.

3. Main result

Aim of this Section is to prove that the same result (9) holds if instead of **H4** we assume the following hypothesis:

H5) $\quad F(x) = (\hat{B}\tilde{\mathbf{C}}_0(x))_n \geq 0, \qquad F(x) \not\equiv 0$ in $\overline{\Omega}$.

We have the following:

THEOREM 3.1. *In the assumptions* **H1**, **H2**, **H3**, **H5**, *then*

$$\lim_{t \to +\infty} t^{-(h(\lambda_1)-1)} e^{-\lambda_1 t} m(x,t) = \frac{x+L}{2L} M_\infty, \tag{14}$$

uniformly in $\overline{\Omega}$, with m and M_∞ defined in (10).

Proof. Taking as an unknown $\tilde{\mathbf{C}} = (c_1, ..., c_{n-1}, c),\ c = \sum_{i=1}^n c_i$, the original problem (1) becomes:

$$\begin{cases} c_{it} = \left(\dfrac{c_i}{c} c_x\right)_x + (\tilde{\Lambda}\tilde{\mathbf{C}})_i,\ i = 1, ..., n-1, \quad x \in \Omega,\ t > 0, \\ c_t = c_{xx} + (\tilde{\Lambda}\tilde{\mathbf{C}})_n, \quad x \in \Omega,\ t > 0, \\ c_x(-L, t) = c_x(L, t) = 0, \quad t > 0, \\ \tilde{\mathbf{C}}(x, 0) = \tilde{\mathbf{C}}_0(x) = (c_{10}(x), ..., c_{(n-1)0}(x), c_0(x)), \\ c_0(x) = \sum_{i=1}^n c_{i0}(x), \quad x \in \Omega. \end{cases} \tag{15}$$

As in other problems of this kind, see [2, 5, 13, 14, 18], it is more convenient to consider, instead of (15), the problem for

$r_i = \dfrac{c_i}{c},\ i = 1, ..., n-1$:

$$\begin{cases} r_{it} = \dfrac{c_x}{c} r_{ix} + P_i(\mathbf{r}),\ i = 1, ..., n-1, \quad x \in \Omega,\ t > 0, \\ c_t = c_{xx} + b(r_1, ..., r_{n-1})c, \quad x \in \Omega,\ t > 0, \\ c_x(-L, t) = c_x(L, t) = 0, \qquad t > 0, \\ c(x, 0) = c_0(x), \qquad x \in \Omega, \\ r_i(x, 0) = \dfrac{c_{i0}(x)}{c_0(x)}, \quad i = 1, ..., n-1, \qquad x \in \Omega, \end{cases} \tag{16}$$

where P_i are polynomial expressions of degree ≤ 2 in $\mathbf{r} = (r_1, ..., r_{n-1})$, the coefficients depending on Λ, and b is defined by

$$b = (\tilde{\Lambda}\tilde{\mathbf{r}})_n, \quad \tilde{\mathbf{r}} = (r_1, ..., r_{n-1}, 1). \tag{17}$$

Let us remark that under hypotheses **H1** and **H2** we have proved in [5] the existence of a unique classical solution of problem (16).

Moreover, $c(x, t)$ is always positive, satisfying a linear parabolic equation with zero flux on the boundary and positive initial datum.
Once c is known, the characteristics depend only on c, see (3), but the r_i evolve along each characteristic, independently of c, like the solutions of the spatially omogeneous problem. Then, fixed x_0 and $\tilde{\mathbf{C}}_0(x_0)$, the r_i are given explicitely on the characteristic $X(t; x_0)$ by the ratioes c_i/c, with c_i, c given in (6) with initial datum $\tilde{\mathbf{C}}_0(x_0)$.

Moreover, we proved in [5] that the "masses" between two characteristics $X(t; x_1)$, $X(t; x_2)$ starting respectively in x_1, x_2, with $-L \leq x_1 < x_2 \leq L$, defined by

$$\tilde{\mathbf{M}}(t) = \int_{X(t;x_1)}^{X(t;x_2)} \tilde{\mathbf{C}}(\xi, t)\, d\xi, \tag{18}$$

are solutions of the ODE system:

$$\dot{\tilde{\mathbf{M}}} = \tilde{\mathbf{\Lambda}}\tilde{\mathbf{M}}, \qquad \tilde{\mathbf{M}}(\mathbf{0}) = \int_{\mathbf{x_1}}^{\mathbf{x_2}} \tilde{\mathbf{C}}_{\mathbf{0}}(\xi)\,\mathbf{d}\xi = \tilde{\mathbf{M}}_{\mathbf{0}}, \tag{19}$$

and hence are given explicitly by (6) with initial datum $\tilde{\mathbf{M}}_0$ instead of $\tilde{\mathbf{C}}_0$.

This means that, since $x = -L$ is the characteristic starting in $x_0 = -L$, we know the evolution in time of $m(x,t)$ on any characteristic $x = X(t; x_0)$ and in particular for $x = X(t; L) \equiv L$.

Then $v(x,t)$, defined by

$$v(x,t) = (1+t)^{-(h(\lambda_1)-1)} e^{-\lambda_1 t} m(x,t), \tag{20}$$

is solution of:

$$\begin{cases} v_t = v_{xx} + f(x,t), \quad x \in \Omega,\ t > 0, \\ v(x,0) = \int_{-L}^{x} c_0(\xi)\,d\xi, \quad x \in \Omega, \\ v(-L,t) = 0, \qquad t > 0, \\ v(L,t) = H(t), \quad t > 0. \end{cases} \tag{21}$$

with

$$\begin{aligned} & f(x,t) = \textstyle\int_{-L}^{x} \tilde{b}\, u\, d\xi, \\ & u = (1+t)^{-(h(\lambda_1)-1)} e^{-\lambda_1 t} c, \\ & \tilde{b} = b - \lambda_1 - \frac{h(\lambda_1)-1}{1+t}, \qquad b = (\tilde{\Lambda}\tilde{\mathbf{r}})_n, \\ & H(t) = (1+t)^{-(h(\lambda_1)-1)} e^{-\lambda_1 t} \times \\ & \quad \times \textstyle\sum_{i=1}^{s} \left\{ \left[\sum_{k=0}^{h(\lambda_i)-1} (\tilde{\Lambda} - \lambda_i I)^k \frac{t^k}{k!} \right] e^{\lambda_i t} \int_{-L}^{L} \tilde{\mathbf{C}}_{0,i}(\xi)\,d\xi \right\}_n . \end{aligned} \tag{22}$$

The expression of $H(t)$ comes from (19), recalling that $x \equiv \pm L$ are the characteristics starting at $x_0 = \pm L$, since there $c_x = 0$. Then, see (21), v is solution of a Dirichlet problem for the heat equation with source $f(x,t)$ and known boundary data.

Under the hypothesis **H5**, from (7) and the definition (8) of F, we have

$$\lim_{t \to +\infty} H(t) = \int_{-L}^{L} F(\xi)\,d\xi = M_\infty. \tag{23}$$

Using a classical result ([11, Theorem 1, Chapter V]) the proof of Theorem 3.1 follows, provided that

$$\lim_{t \to +\infty} f(x,t) = 0 \tag{24}$$

uniformly in Ω.

In order to prove (24), fixed an arbitrary $\sigma > 0$, let us divide the interval $\Omega = (-L, L)$ into the two subsets:

$$\begin{aligned} \Omega_- &= \{x \in \Omega : F(x) < \sigma\}, \\ \Omega_+ &= \{x \in \Omega : F(x) \geq \sigma\}. \end{aligned} \tag{25}$$

Let us remark that, for any σ sufficiently small, Ω_+ is not empty and, if $F(x_0) = 0$, there exists a neighborhood of x_0 where $F < \sigma$ and Ω_- is not empty.

For any fixed $t > 0$, let us divide Ω into

$$\begin{aligned} \Omega_-(t) &= \{x \in \Omega : x = X(t; x_0),\ x_0 \in \Omega_-\}, \\ \Omega_+(t) &= \{x \in \Omega : x = X(t; x_0),\ x_0 \in \Omega_+\}, \end{aligned} \tag{26}$$

that is $\Omega_-(t)$, $\Omega_+(t)$ are the set of the characteristics at time t starting from Ω_-, Ω_+ respectively.

Then

$$\begin{aligned} f(x,t) &= \textstyle\int_{[-L,x]\cap\Omega_-(t)} \tilde{b} u \, d\xi + \int_{[-L,x]\cap\Omega_+(t)} \tilde{b} u \, d\xi = \\ &= f_-(x,t) + f_+(x,t). \end{aligned} \tag{27}$$

Let us consider first f_+. In [8, Lemma 3.1], we proved that if for some x_0 $F(x_0) \geq \sigma > 0$, on the characteristic $X(t; x_0)$ starting in x_0, the following estimate on $\tilde{b}$ depending on σ holds:

$$|\tilde{b}| \leq \frac{k_1}{\sigma} \left[\frac{h(\lambda_1) - 1}{t^2} + (s-1) e^{\frac{\lambda_2 - \lambda_1}{2} t} \right] = \frac{k_1}{\sigma} g(t), \tag{28}$$

for $x = X(t; x_0)$ and $t \geq 1$, where k_1 is a constant depending on Λ and on $\max_\Omega ||\tilde{\mathbf{C}}_0(x)||$.

Then, being $u > 0$, recalling (21)-(23), we have:

$$\begin{aligned} |f_+(x,t)| &\leq \frac{k_1}{\sigma} g(t) \int_{[-L,x]\cap\Omega_+(t)} u(\xi,t) \, d\xi \\ &\leq \frac{k_1}{\sigma} g(t) \int_{-L}^{L} u(\xi,t) \, d\xi \\ &\leq \frac{k_1}{\sigma} g(t) H(t) \leq 2 \frac{k_1 M_\infty}{\sigma} g(t), \quad t \geq T_1. \end{aligned} \tag{29}$$

Let us consider now f_-. Notice that for any $x \in \overline{\Omega}$, $t > 0$, $\tilde{b}$ is uniformly bounded because the r_i are bounded between 0 and 1 (see (22)), that is $|\tilde{b}| \leq k_2$. Since $u > 0$ we have:

$$\begin{aligned} |f_-(x,t)| &\leq k_2 \textstyle\int_{[-L,x]\cap\Omega_-(t)} u(\xi,t) \, d\xi \\ &\leq k_2 \textstyle\int_{\Omega_-(t)} u(\xi,t) \, d\xi. \end{aligned} \tag{30}$$

From (18), (19), (6), (8), the last term in (30) can be written in the form

$$\begin{aligned} \textstyle\int_{\Omega_-(t)} u(\xi,t)\,d\xi &= (1+t)^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}\times \\ &\times \textstyle\sum_{i=1}^{s}\left\{\left[\sum_{k=0}^{h(\lambda_i)-1}(\tilde{\Lambda}-\lambda_i I)^k \dfrac{t^k}{k!}\right]e^{\lambda_i t}\displaystyle\int_{\Omega_-}\tilde{\mathbf{C}}_{0,i}(\xi)\,d\xi\right\}_n \\ &= \left(\hat{B}\textstyle\int_{\Omega_-}\tilde{\mathbf{C}}_{0,1}(\xi)\,d\xi\right)_n + \tilde{z} = \textstyle\int_{\Omega_-}F(\xi)\,d\xi + \tilde{z}, \end{aligned} \tag{31}$$

with $\tilde{z}$ bounded for any $x \in \overline{\Omega},\ t \geq 1$ by:

$$|\tilde{z}| \leq k_3\left(\frac{(h(\lambda_1)-1)}{t} + (s-1)e^{\frac{\lambda_2-\lambda_1}{2}t}\right) = k_3 g_1(t), \tag{32}$$

with k_3 depending on Λ and on $\max_\Omega ||\tilde{\mathbf{C}}_0||$.

Recalling that $F < \sigma$ in Ω_-, from (31), (32) it follows

$$|f_-| \leq k_4(\sigma + g_1(t)),\ x \in \overline{\Omega},\ t \geq 1. \tag{33}$$

From the estimates (29), (33) on f_+, f_- we have, for any $x \in \overline{\Omega},\ t \geq \max(1, T_1)$:

$$|f| \leq k_5\left(\sigma + \frac{g(t)}{\sigma} + g_1(t)\right). \tag{34}$$

Then, fixed an arbitrary $\epsilon > 0$, e.g. $\sigma = \dfrac{\epsilon}{3}$, recalling that $g(t)$ and $g_1(t)$ tend to zero as $t \to \infty$, from (34) we have that there exists a time $T(\epsilon)$ such that

$$|f| \leq \epsilon,\ \forall x \in \overline{\Omega},\ t > T(\epsilon),$$

that gives the proof of the theorem. □

From Theorem 3.1, as in [8], it is possible to obtain the asymptotic behaviour of the characteristics, precisely we have:

COROLLARY 3.2. *In the hypotheses of Theorem 3.1 we have that*

$$\lim_{t\to+\infty} X(t;x_0) = X_\infty(x_0) = \frac{2L}{M_\infty}\int_{-L}^{x_0} F(\xi)\,d\xi - L, \tag{35}$$

uniformly in $\overline{\Omega}$.

Proof. The proof is the same as the one of [8, Corollary 3.1], let us mention here that the idea of the proof is that we know the evolution in time of $m(X(t;x_0),t)$, since m is solution of the ODE (19). Therefore we have that, by the definition of $X_\infty(x_0)$ in (35) and by (6)-(8):

$$t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}m(X(t;x_0),t)$$
$$= \int_{-L}^{x_0} F(\xi)\,d\xi + \hat{z}(x_0,t) = \frac{X_\infty(x_0)+L}{2L}M_\infty + \hat{z},$$

where

$$|\hat{z}(x_0,t)| \le k_6 g_1(t),$$

with k_6 constant depending on Λ and on $\max_\Omega ||\tilde{\mathbf{C}}_0||$, and $g_1(t)$ defined in (32).

On the other hand, Theorem 3.1 implies that, for t sufficiently large and for any x_0 in $\overline{\Omega}$, $t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}m$ on the characteristic $X(t;x_0)$ is close to $\frac{X(t;x_0)+L}{2L}M_\infty$. □

Concerning the asymptotic behaviour of the $r_i = \dfrac{c_i}{c}$, $i=1,...,n-1$, as in [8] we have:

Corollary 3.3. *In the hypotheses of Theorem 3.1, and assuming that $F(x) \ge \delta > 0$ in $[x_1,x_2], \subset \overline{\Omega}$, we have:*

$$\lim_{t\to+\infty} r_i(x,t) = \frac{F_i(X_\infty^{-1}(x))}{F(X_\infty^{-1}(x))}, \tag{36}$$

uniformly in $[X_\infty(x_1), X_\infty(x_2)]$, and

$$\left| r_i\left(X(t;X_\infty^{-1}(x)),t\right) - \frac{F_i(X_\infty^{-1}(x))}{F(X_\infty^{-1}(x))}\right| \le k(\delta)g_1(t), \tag{37}$$

for $t > T(\delta) = g_1^{-1}\left(\dfrac{\delta}{2}\right)$, g_1 defined in (32).

Proof. From the hypothesis $F(x) \ge \delta > 0$, $x \in [x_1,x_2]$, it follows that the function $X_\infty(x)$ is monotone increasing in $[x_1,x_2]$, consequently the inverse function is monotone increasing in $[X_\infty(x_1), X_\infty(x_2)]$.

Moreover the characteristics are ordered so that $\forall \bar{t} > 0$ and $\forall \bar{x} \in [X(t;x_1), X(t;x_2)]$ there exists a unique $\hat{x} \in [x_1,x_2]$ such that $\bar{x} = X(t;\hat{x})$ and $F(\hat{x}) \ge \delta > 0$. Then we can repeat the arguments of [8, Corollary 3.2]. The estimate (37) on r_i comes from the explicit expression of $\tilde{\mathbf{C}}(t)$ in (6). □

From the explicit expression of $X_\infty(x)$, see (35), we have the following

REMARK 3.4. **i)** *If $F(x) \equiv 0$ for $x \in [x_1, x_2] \subset \Omega$, then $X_\infty(x_1) = X_\infty(x_2)$. That is, if F is identically zero in a subinterval of Ω, all the subinterval asymptotically reduces to the point*

$$X^* = X_\infty(x_1) = \frac{2L}{M_\infty} \int_{-L}^{x_1} F(\xi) - L.$$

ii) *If $0 \le F(x) \le \beta,\ \beta > 0$ for $x \in [x_1, x_2] \subset \Omega$, then*

$$X_\infty(x_2) - X_\infty(x_1) = \frac{2L}{M_\infty} \int_{x_1}^{x_2} F(\xi)\, d\xi \le \frac{2L}{M_\infty}(x_2 - x_1)\beta.$$

That is the asymptotic measure of the subinterval is of the order β.

In the next Section we will consider some examples in order to make clearer the above observations concerning the asymptotic behaviour of $\mathbf{r} = (r_1, ..., r_{n-1})$.

4. Examples and comments

Let us consider the example described in Section 2, for the couple (U^{238}, U^{234}), where the matrix Λ is given by (13). If we assume in this example that $F(x) = \dfrac{\gamma_2 - \gamma_1}{\gamma_2} c_{10}(x)$ is null in a subinterval $[x_1, x_2] \subset \overline{\Omega}$ and positive out of this interval, then (see Remark 3.4), the whole interval $[x_1, x_2]$ reduces, for $t \to \infty$ to the unique point

$$X^* = X_\infty(x_1) = \frac{2L}{M_\infty} \int_{-L}^{x_1} F(\xi) - L.$$

In this case there does not exist the $\lim_{x \to X^*,\, t \to \infty} r(x,t)$, because in any neighborhood of X^* there are characteristics on which $r \equiv 0$ (precisely $X(t; x_0)$, $\forall x_0 \in [x_1, x_2]$) and characteristics on which

$$r \to r_E = \frac{\gamma_2 - \gamma_1}{\gamma_2},\ 0 < r_E < 1,$$

precisely the ones starting at a point out of $[x_1, x_2]$.

However, fixed a neighborhood of X^*, out of it r tends uniformly to r_E for $t \to \infty$, because of Corollary 3.3. From a physical point of view in this case $(0 < \gamma_1 < \gamma_2)$ there is not a uniform asymptotic distribution for c_1, c_2 and, in particular, oscillations may be present near X^* also asymptotically. However varying order of the parameters γ_1, γ_2 one can observe that:

i) if $\gamma_1 > \gamma_2 > 0$ then $F(x) = c_0(x) + \dfrac{\gamma_2}{\gamma_1 - \gamma_2} c_{10}(x) \ge c_0(x) > 0$.

Then **H1** implies that assumption **H4** is satisfied and $r \to 0$ uniformly for $t \to \infty$, that is only the isotope 2 is present asymptotically.

ii) if $\gamma_1 = \gamma_2 = \gamma > 0$ then $F(x) = \gamma c_{10}(x)$.

Then in assumption **H5** we have that $M_\infty = \int_{-L}^{L} F > 0$ depends only on the isotope 1 and the asymptotic expansion of $m(x,t)$ is

$$te^{-\gamma t}\frac{x+L}{2L}M_\infty.$$

However for any initial data satisfying **H1** we have $r \leq \dfrac{1}{\gamma t}$ for $t > 1$ and $x \in \Omega$, so that $r \to 0$ uniformly for $t \to \infty$, that is there exists a uniform asymptotic distribution of r in Ω, independently of the possible vanishing of F in a subset of Ω.

Let us remark that if assumption **H5** does not hold, that is if $F \equiv 0$ in Ω, the isotope 1 is initially absent in the explicit solution and the first asymptotic term of m is

$$e^{-\gamma t}\frac{x+L}{2L}\int_{-L}^{L} c_0(\xi)\, d\xi, \;\; c_0 \equiv c_{20}.$$

This example shows that depending on the form of the matrix Λ there can be three different asymptotic behaviours:

case I for any initial data satisfying **H1**, $F(x)$ is always strictly positive, and hence hypothesis **H4** holds. Then $\mathbf{r} = (r_1, ..., r_{n-1})$ has an asymptotic distribution in the whole Ω (see [8] and (12));

case II assuming hypothesis **H5**, there exists an asymptotic distribution of $\mathbf{r}$ in the whole Ω;

case III assuming hypothesis **H5**, there does not exists in general an asymptotic distribution of $\mathbf{r}$ in the whole Ω.

These three possible behaviours are present in the general case of n species with different evolutive laws. We will present some of them, interesting from a physical point of view.

case I

example Ia) The matrix Λ is a multiple of the identical matrix, defined by:

$$\dot{c}_i = -\gamma c_i, \quad i = 1, ..., n, \quad \gamma \geq 0. \tag{38}$$

This example describes both sets of stable isotopes, i.e. with $\gamma = 0$, e.g. of the couple (Cl^{37}, Cl^{35}), and of radiative isotopes that decade out of the element with the same coefficients of decay ($\gamma > 0$), e.g. the couple (U^{235}, U^{238}).

In this case we have that $F(x) = c_0(x) > 0$ because of hypothesis **H1**.

Let us remark that in this case the asymptotic distribution of $\mathbf{r}$ strongly depends on the initial conditions, since it is given explicitely by:

$$\lim_{t\to\infty} r_i(x,t) = \frac{c_{i0}(X_\infty^{-1}(x))}{c_0(X_\infty^{-1}(x))}, \quad i = 1, ..., n. \tag{39}$$

example Ib) The matrix Λ is defined by

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1, \\ \dot{c}_i = \gamma_{i-1} c_{i-1} - \gamma_i c_i, \quad i = 2, ..., n-1, \\ \dot{c}_n = \gamma_{n-1} c_{n-1}, \end{cases} \tag{40}$$

with $\gamma_i > 0, \ i = 1, ..., n-1$.

This case describes the evolution of a chain of n isotopes such that the i^{th} one decades into the $(i+1)^{th}$ one, for $i = 1, ..., n-1$, while the n^{th} one is stable.

It is shown in [8] that also in this example $F(x) = c_0(x)$, however in this case

$$\lim_{t\to\infty} r_i(x,t) = 0 \quad i = 1, ..., n-1, \tag{41}$$

uniformly in Ω, then the unique isotope asymptotically present is the n^{th} one, that is the unique stable isotope.

example Ic) The matrix Λ is defined by

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1, \\ \dot{c}_i = \gamma_{i-1} c_{i-1} - \gamma_i c_i, \quad i = 2, ..., n. \end{cases} \tag{42}$$

with $\gamma_i > 0, \ i = 1, ..., n$ and $\gamma_n = \min \gamma_i, \mu(-\gamma_n) = 1$.

This is a generalization of the couple (U^{238}, U^{234}): we have a chain of n isotopes of which the i^{th} one decades into the $(i+1)^{th}$ one, for $i = 1, ..., n-1$, and the n^{th} one decays out of the element. In [8, Example 2, Section 4] we have shown that

$$\mathbf{F} = F(x)\mathbf{v}^n, \ \mathbf{v}^n = (0, ..., 0, 1), \qquad F(x) \geq c_0(x).$$

Then, again, for any datum satisfying **H1**, $F(x)$ is strictly positive and

$$\lim_{t\to\infty} r_i(x,t) = 0 \quad i = 1, ..., n-1, \tag{43}$$

uniformly in Ω, and the unique isotope asymptotically present is the n^{th} one.

Let us remark that the estimate on $F(x)$ can be derived directly, without a detailed analysis of the eigenvalues-eigenvectors of Λ.

In fact in this case the ODE system $\dot{\tilde{\mathbf{C}}} = \tilde{\mathbf{\Lambda}}\tilde{\mathbf{C}}$ is given by

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1, \\ \dot{c}_i = \gamma_{i-1}c_{i-1} - \gamma_i c_i, \quad i = 2, ..., n \\ \dot{c} = -\gamma_n c + \gamma_n \sum_{i=1}^{n-1} c_i(t). \end{cases} \tag{44}$$

Then the $c_i(t)$, $i = 1, ..., n-1$, can be obtained from the first $n-1$ equations and depend only on $c_{i0}(t)$, $i = 1, ..., n-1$, and the total concentration consequently is given by

$$ce^{\gamma_n t} = c_0 + \gamma_n \int_0^t e^{\gamma_n \tau} \sum_{i=1}^{n-1} c_i(\tau)\, d\tau. \tag{45}$$

The hypotheses $\gamma_n = \min \gamma_i$, $\mu(-\gamma_n) = 1$ ensure that the integral in (45) is bounded for $t \to \infty$, since c_i, $i = 1, ..., n-1$, behave at most like $e^{-\gamma_i t}Q(t)$, with $Q(t)$ polynomial in t of degree less or equal to $n-1$ (equal if the γ_i, $i = 1, ..., n-1$, are all identical).

Since $c_i \geq 0$, we have $\lim_{t\to\infty} ce^{\gamma_n t} = F(x) \geq c_0$,

in particular $F(x) = c_0$ if $c_{i0} = 0$, $i = 1, ..., n-1$, that is if initially the unique isotope present is the n^{th} one.

Let us remark that if $\gamma_n = \min \gamma_i$, but $\mu(-\gamma_n) > 1$ then in general F is not positive everywhere. Indeed even in the case $n = 2$ we have seen that $F = \gamma_1 c_{10}$, and in general, for $n > 2$ we have, from (45) and since $\mu(-\gamma_n) = h(-\gamma_n) > 1$:

$$F = \lim_{t\to\infty} t^{-(h(-\gamma_n)-1)} e^{\gamma_n t} c = \lim_{t\to\infty} \gamma_n t^{-(h(-\gamma_n)-1)} \int_0^t e^{\gamma_n \tau} \sum_{i=1}^{n-1} c_i(\tau)\, d\tau.$$

If $c_{i0} = 0$, $i = 1, ..., n-1$ and $c_{n0} > 0$, then the initial data satisfy **H1** but $F = 0$.

case II

This case occurs when **H1** does not imply that $F(x)$ is positive in $\overline{\Omega}$, but **r** has a unique asymptotic limit for all data satisfying **H1**, as solution of an ODE. In this class we can find the example with Λ given by (42) with $\gamma_i = \gamma > 0,\ i = 1, ..., n$. Under hypothesis **H1**, in this case we have that, $\forall x \in \Omega$ and for $t > 1$:

$$\begin{aligned} &F_i(x) = \frac{\gamma^{n-1}}{(n-1)!} c_{10}(x)\delta_i^n, \quad && i = 1, ..., n, \\ &0 \le r_i \le \frac{i}{\gamma t}, && i = 1, ..., n-1, \end{aligned} \tag{46}$$

where δ_i^n is the Kronecker symbol.
Then for any initial datum satisfying hypothesis **H1**, we have that

$$\lim_{t\to\infty} \mathbf{r} = \mathbf{0},$$

uniformly in Ω, that is asimptotically the unique isotope present is the n^{th} one, however M_∞ depends only on the 1^{st} isotope (see Theorem 3.1).

To prove (46) we remark that Λ is multiple of a Jordan normal form and the solution can be explicitly written as follows:

$$\begin{cases} e^{\gamma t} c_i = \sum_{j=1}^{i} c_{j0} \dfrac{(\gamma t)^{i-j}}{(i-j)!}, & i = 1, ..., n-1, \\ e^{\gamma t} c = \sum_{i=1}^{n-1} c_{i0} \sum_{j=1}^{n-i} \dfrac{(\gamma t)^j}{(j)!} + c_0. \end{cases} \tag{47}$$

Then, recalling that $h(-\gamma_n) = n$ and $\lim_{t\to\infty} t^{-(h(-\gamma_n)-1)} e^{\gamma t} \tilde{\mathbf{C}} = \mathbf{F}$, (46) follows.

Let us remark that for any initial data such that $c_{10} > 0$ we have for $t \to \infty$:

$$r_i \simeq \frac{(n-1)!}{(i-1)!} (\gamma t)^{-(n-i)}, \qquad i = 1, ..., n-1,$$

that is the estimate (46) is almost sharp.

case III

This case occurs when **H1** does not imply that $F(x)$ is positive, and **r**, as solution of an ODE, does not have a unique asymptotic limit, for all the data satisfying **H1**.

example IIIa) The matrix Λ is diagonal, with eigenvalues not all equal. This is the case of a set of isotopes which decade out of the element with coefficients of decay not all equal.

Assuming the isotopes to be ordered with $\gamma_1 \leq \gamma_2 \leq ... \leq \gamma_n$, $\gamma_1 < \gamma_n$, from the explicit solution one can directly observe that, denoting $\mu(-\gamma_1) = j < n$:

$$F(x) = \sum_{i=1}^{j} c_{i0}(x),$$

and if $F(x_0) > 0$ then $\mathbf{r}(X(t;x_0),t)$ tends asymptotically to a limit which can depend on the initial data, if $j > 1$, but it is such that $\sum_{i=1}^{j} r_i$ tends to 1 and r_i tends to 0 for $i > j$, as $t \to \infty$.

On the other hand, we have that if $c_{10}(x_0) = ... = c_{(n-1)0}(x_0) = 0$, $c_{n0}(x_0) > 0$ then $F(x_0) = 0$ and $\mathbf{r}(X(t;x_0),t) \equiv 0$, that is a different limit from the previous one.

Then in general there does not exist a limit for $\mathbf{r}$ in the whole Ω.

example IIIb) Let us consider the example (42) assuming now that the γ_i are not all equal and that $-\gamma_n$ is not the maximum eigenvalue. Then if we choose the initial data $c_{i0} = 0$, $i = 1, ..., n-1$, $c_{n0} > 0$, satisfying hypothesis **H1**, we have the solution:

$$\tilde{\mathbf{C}} = c_0 e^{-\gamma_n t}\mathbf{v}^n, \;\; \mathbf{v}^n = (0, ..., 0, 1).$$

Denoted by $\lambda_1 = -\min_{i=1,...,n} \gamma_i$ the maximum eigenvalue, say $-\gamma_k$, $k \neq n$, then, for this initial condition we have:

$$\mathbf{F} = \lim_{t\to\infty} t^{-(h(\lambda_1)-1)} e^{-\lambda_1 t}\tilde{\mathbf{C}} = \lim_{t\to\infty} t^{-(h(\lambda_1)-1)} e^{-(\gamma_n-\gamma_k)t} c_0 \mathbf{v}^n = \mathbf{0}.$$

Moreover, on any characteristic $X(t;x_0)$ with x_0 such that $c_{i0} = 0$, $i = 1, ..., n-1$, $c_{n0} > 0$, we have $\mathbf{r} = \mathbf{0}$.

On the other hand we can show, see [8, Example 2], that in this case $\mathbf{F}(x) = \beta(x)\mathbf{v}^k$, where $\mathbf{v}^k$ is given by:

$$\begin{array}{lll} v^{k,i} = 0, & i = 1, ...k-1, & \text{if } k > 1, \\ v^{k,i} = \prod_{j=i}^{n-1} \dfrac{\gamma_{j+1} - \gamma_k}{\gamma_j}, & i = k, ..., n-1, & \\ v^{k,n} = 1 + \sum_{i=1}^{n-1} \prod_{j=i}^{n-1} \dfrac{\gamma_{j+1} - \gamma_k}{\gamma_j}. & & \end{array} \tag{48}$$

Then if $F(x_0)$ is positive on the characteristic starting in x_0 we have

$$\lim_{t\to\infty} \mathbf{r} = \mathbf{r}_E \neq \mathbf{0}, \quad \mathbf{r}_{E,i} = \frac{v^{k,i}}{v^{k,n}}, \quad i = 1, ...n-1,$$

that is in general a limit for $\mathbf{r}$ does not exist in the whole Ω.

In particular if $k = 1$, all the components of $\mathbf{r_E}$ are positive and $\sum_{i=1}^{n-1} \mathbf{r}_{E,i} < 1$, that is, from a physical point of view, we have the so called secular equilibrium of all the n isotopes.

Acknowledgments. The authors wish to thank Claudio Pescatore for his helpful suggestions and comments.

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Received March 19, 2012
Revised June 25, 2012