ON A MODEL FOR THE CODIFFUSION OF ISOTOPES

E. Comparini^{*}, A. Mancini^{*}, C. Pescatore⁺, M. Ughi^o

* Dipartimento di Matematica "U. Dini" Università degli Studi di Firenze, Italy

⁺ OECD/Nuclear Energy Agency Issy-les-Moulineaux, France

^o Dipartimento di Matematica e Geoscienze, Università di Trieste, Italy

FBP 2012

different isotopes of the same chemical element A should not be considered as distinct chemical species and should contribute jointly to the chemical potential of A.

The total flux J_i of a particular isotope A_i of an element A has two components:

- one representing the effects of interaction of A_i with the solvent molecules B (classical Fick's law):

A - B interaction \rightarrow flux $= -\tilde{D}_i \nabla c_i$,

- the other representing the interaction with the kin isotopes, depending on the total concentrations of the A-molecules:

$$A - A$$
 interaction \rightarrow flux $= -D_i \frac{c_i}{c} \nabla c$,

where c_i is the concentration of A_i , c is the total concentration of A, D_i is the diffusivity of A in the solvent B, D_i is a measure of the mobility of the A_i molecules due to the A - A interactions within the solvent B.

diffusion of n species of isotopes of the same element

the flux of the i component J_i is given by

$$J_i = -\left(\tilde{D}_i \nabla c_i + D_i \frac{c_i}{c} \nabla c\right), \qquad i = 1, \dots, n, \qquad c = \sum_{i=1}^n c_i.$$

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

$$\frac{d\underline{C}}{dt} = \Lambda \underline{C}, \qquad \underline{C} \in \mathbb{R}^n,$$

with Λ a suitable $n \times n$ constant matrix.

general case of positive diffusion coefficients with Dirichlet boundary conditions

$$\begin{cases} \frac{\partial c_i}{\partial t} = -\operatorname{div} J_i + \sum_{j=1}^n \Lambda_{ij} c_j, & \text{in } \Omega \times (0, T), \\ c_i \big|_{\partial \Omega} = f_i, & \text{in } \partial \Omega \times (0, T), \\ c_i(x, 0) = c_{i0}(x) & \text{in } \overline{\Omega}, \end{cases}$$

$$J_i = -\left(\tilde{D}_i \nabla c_i + D_i \frac{c_i}{c} \nabla c\right), \quad i = 1, ..., n$$

 Ω bounded domain of \mathbb{R}^n with regular boundary $\partial \Omega$.

existence and uniqueness of classical solutions are proved in the physically relevant assumption that

$$K \ge c_i \ge 0, \quad i = 1, ..., n, \qquad c \ge k > 0,$$

k, K constant.

The system can be written as a quasilinear parabolic system in separated divergence form

$$\begin{cases} \frac{\partial \tilde{\mathbf{C}}}{\partial t} = \sum_{j,k=1}^{n} \frac{\partial}{\partial x_{j}} \left(\mathcal{A}_{jk}(\tilde{\mathbf{C}}) \frac{\partial \tilde{\mathbf{C}}}{\partial x_{k}} \right) + \tilde{\Lambda} \tilde{\mathbf{C}}, & \text{in } \Omega \times (0,T) \\ \tilde{\mathbf{C}} = (c_{1}, \dots, c_{n-1}, c), \quad \mathcal{A}_{jk}(\tilde{\mathbf{C}}) = A(\tilde{\mathbf{C}}) \delta_{jk}, \ j,k = 1, \dots, n \end{cases}$$
$$\begin{pmatrix} \tilde{D}_{1} & 0 & \dots & 0 & D_{1} \frac{c_{1}}{c} \\ 0 & \tilde{D}_{2} & \dots & 0 & D_{2} \frac{c_{2}}{c} \\ \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & \tilde{D}_{n-1} & D_{n-1} \frac{c_{n-1}}{c} \\ \tilde{D}_{1} - \tilde{D}_{n} & \tilde{D}_{2} - \tilde{D}_{n} & \dots & \tilde{D}_{n-1} - \tilde{D}_{n} & \tilde{D}_{n} + D_{n} + \sum_{i=1}^{n-1} (D_{i} - D_{n}) \frac{c_{i}}{c} \end{cases} \end{cases}$$

Space operator NOT STRONGLY ELLIPTIC but satisfies a normal ellipticity condition (see [1], [2]),

$$n-1$$
 eigenvalues $\simeq \tilde{D}_i$, 1 eigenvalue $\simeq D_i + \tilde{D}_i$

$\tilde{D}_i << D_i \longrightarrow$ appearance of a "hyperbolic" behaviour for the c_i

setting $\tilde{D}_i = 0$ and $D_1 = \max D_j, D_n = \min D_j, \quad j = 1, ..., n, \quad c_n = c - \sum_{j=1}^{n-1} c_j$

$$\begin{cases} c_{it} = \left(D_i \frac{c_i}{c} c_x\right)_x + \sum_{j=1}^{n-1} \beta_{ij} c_j + \beta_{in} c, & i, j = 1, \dots n-1 \\ c_t = (a(c, c_1, \dots c_{n-1}) c_x)_x + \sum_{j=1}^{n-1} \beta_{nj} c_j + \beta_{nn} c, \\ a = D_n + \sum_{j=1}^{n-1} (D_j - D_n) \frac{c_i}{c} \end{cases}$$

 β_{ij} constants depending on Λ_{ij} .

 $0 \le c_i \le c, \longrightarrow c$ satisfies a uniformly parabolic quasilinear equation in divergence form $(0 < D_n \le a \le D_1, \forall c_i, c \ge 0)$.

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.

 c_i will have finite speed of propagation and will in general be non smooth for t > 0 (in qualitative accordance with experimental results).



smpIP - N 1.0D-1 1.0D-1 1.0D0 1.0D0 0.0D0 0.0D0 1 0.1 0.4 +0.0 0.0 0 0.2 0.80 0.0 0.0 1.0 1.0D-3 - t=0.000



smpIP - N 1.0D-1 1.0D-1 1.0D0 1.0D0 0.0D0 0.0D0 1 0.1 0.4 +0.0 0.0 0 0.2 0.80 0.0 0.0 1.0 1.0D-3 - t=0.0500



smpIH - N 1.0D-6 1.0D-6 1.0D0 1.0D0 0.0D0 0.0D0 1 0.1 0.4 +0.0 0.0 0 0.2 0.80 0.0 0.0 1.0 1.0D-3 - t=0.0500

If $\Lambda_{ij} \equiv 0$ with $D_i = 1 \ \forall i$, after scaling on t, the system reduces to

$$\begin{cases} c_{it} = \left(\frac{c_i}{c}c_x\right)_x, & i = 1, \dots n - 1\\ c_t = c_{xx}. \end{cases}$$

diffusion of the total concentration c governed by the classical heat equation,

system for the c_i uncoupled; each equation is a linear first order equation for c_i .

similar situation also for $\Lambda_{ij} \neq 0$ in the following examples:

Example 1. All the species decay with almost the same coefficient λ (i.e. $\Lambda_{ij} = -\lambda \delta_{ij}$) \longrightarrow same system, with c_i, c replaced by $c_i e^{\lambda t}, c e^{\lambda t}$.

Example 2. A triplet c_1, c_2, c_3 with decay law respectively $-\lambda_1 c_1, \ \lambda_1 c_1 - \lambda_2 c_2, \ \lambda_2 c_2$.

$$\begin{cases} c_{1t} = \left(\frac{c_1}{c}c_x\right)_x - \lambda_1 c_1, \\ c_{2t} = \left(\frac{c_2}{c}c_x\right)_x + \lambda_1 c_1 - \lambda_2 c_2, \\ c_t = c_{xx}. \end{cases}$$

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 related to the "activity ratio".

problem without decay:

$$r_{it} = r_{ix} \frac{c_x}{c}, \qquad i = 1, \dots n - 1,$$

 \longrightarrow r_i are constant on the characteristics.

problem with decay ($D_i \equiv 1, a = 1$)

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

 P_i polinomial of 2 degree.

 \longrightarrow r_i evolve on each characteristic independently of c.



smplH - N 1.0D-6 1.0D-6 1.0D0 1.0D0 0.0D0 0.0D0 1 0.1 0.4 +0.0 0.0 0 0.2 0.80 0.0 0.0 1.0 1.0D-3 - t=1.0000



decadH - N 1.0D-6 1.0D-6 1.0D0 1.0D0 1.0D-3 1.0D0 1 0.1 0.4 +0.0 0.0 0 0.2 0.80 0.0 0.0 1.0 1.0D-3 - t=1.0000

Example 3: couple
$$(U^{238},\,U^{234})$$

decay law respectively

$$\dot{c}_1 = -\lambda_1 c_1, \quad \dot{c}_2 = \lambda_1 c_1 - \lambda_2 c_2, \qquad \lambda_1 \ll \lambda_2.$$

then

$$\begin{cases} c_t = c_{xx} + \lambda_2 c(r_1 - 1), & \text{in } \Omega \times (0, T), \\ r_{1t} = r_{1x} \frac{c_x}{c} + \lambda_2 r_1 (r_E - r_1), & r_E = \frac{\lambda_2 - \lambda_1}{\lambda_2}. \end{cases}$$

along the characteristics:

$$r_{1}(0) = 0 \rightarrow r_{1}(t) \equiv 0, \quad t > 0$$

$$0 < r_{1}(0) \le 1 \rightarrow r_{1}(t) = \frac{r_{E}r_{1}(0) \exp(\lambda_{2} - \lambda_{1})t}{r_{E} - r_{1}(0) + r_{1}(0) \exp(\lambda_{2} - \lambda_{1})t},$$

$$r_{1}(t) \rightarrow r_{E} \text{ as } t \rightarrow \infty,$$

in accordance with the physical fact that for the couple (U^{238}, U^{234}) has a "secular equilibrium" positive and attractive (i.e. normally the two isotopes are found in a precise positive ratio).

characteristics

- define the characteristic for any $(x_0,t_0),\,t_0>0$ fixed, $x_0\in\Omega$,
- think r_i given on $t=t_0$ ($t=t_0$ is not a characteristic itself)
- extend it in a neighborhood of $t=t_0$

 $X(t; x_0, t_0)$ denotes the characteristic starting in x_0, t_0 , whose equation is

$$\begin{cases} \left. \frac{dX(t;x_0,t_0)}{dt} = -\frac{c_x}{c} \right|_{x=X(t;x_0,t_0)}, \quad (x_0,t_0) \in \Omega \times (0,T), \\ X(t_0;x_0,t_0) = x_0, \end{cases}$$

main fact:

the masses between two characteristics behave as the solution of the ODE.

$$\mathbf{m}_{1,2}(t) = \int_{X_1(t)}^{X_2(t)} \tilde{\mathbf{C}}(\xi, t) \, d\xi, \quad \tilde{\mathbf{C}} = (c_1, ..., c_{n-1}, c)$$

is solution of ODE:

$$\begin{cases} \dot{\mathbf{Y}} = \tilde{\Lambda} \mathbf{Y}, \\ \mathbf{Y}(0) = \int_{X_1(0)}^{X_2(0)} \tilde{\mathbf{C}}_0(\xi) \, d\xi. \end{cases}$$

 $\Omega = (-L,L)$ bounded then there will be:

•

. a set $\Omega_1(t_0) = (-L, l_1)$ such that the characteristics starting from (x_0, t_0) , $x_0 \in \Omega_1$, $t_0 > 0$, will reach the lateral boundary x = -L,

. a set $\Omega_2(t_0)=(l_1,l_2)$ such that they go to the initial set $\Omega imes\{t=0\}$,

. a set $\Omega_3(t_0) = (l_2, L)$ such that they go to x = L.

Assume that two characteristics, denoted by $X_1(t), X_2(t)$ with $X_1(t_0) < X_2(t_0)$ starting from $t_0 > 0$ reach t = 0, then:

$$X_1(0) < X_2(0), \quad c_0(x) \neq 0, \quad x \in (X_1(0), X_2(0))$$

In other words if $c_0(x) \equiv 0, x \in I$, I interval of Ω , then there cannot be two distinct characteristics ending in I. This fact is due to the "infinite speed of propagation" of the total concentration i.e. to the fact that $c(x,t) > 0, \forall t > 0, x \in \Omega$.

"holes" of the initial data for \boldsymbol{c}

• Let us assume that

$$c_0(x) \equiv 0, \quad x \in I_0 = (a, b) \subset \Omega,$$

$$c_0(x) > 0, \quad x \in I = (a - \delta, b + \delta) - \overline{I}_0, \quad \delta > 0 \text{ such that } I \subset \Omega,$$

Then there exists a curve $\boldsymbol{x}=\boldsymbol{s}(t)$ separating two regions

 $C_{-} = \{\text{characteristics from } (a - \delta, a)\},\$ $C_{+} = \{\text{characteristics from } (b, b + \delta)\}.$

• Let us assume that $\Omega = (-L, L)$,

$$c_0(x) > 0, \quad x \in [-L, a], \qquad c_0(x) \equiv 0, \quad x \in (a, L],$$

 $a \in \Omega$, then $\Omega_2(t) \to (-L, a)$ as $t \to 0$.

• Assume $\Omega = (-L, L)$ and $c_0 \equiv 0$ in Ω , then Ω_2 is at most one curve, moreover:

- if the boundary data are such that both Ω_1 and Ω_3 are not empty, that is the case e.g. of incoming flux at both boundaries, then Ω_2 is precisely a line;

- if either Ω_1 or Ω_3 is empty (e.g. incoming flux from only one boundary) then Ω_2 is empty.

EXAMPLE, stable isotopes

$$c_0(x) = \begin{cases} c_L, & x < a, \\ 0, & a < x < b, \\ c_R, & b < x, \end{cases} \quad 0 < c_L \le c_R,$$

either Cauchy Problem or Homogeneous Neumann Problem with -L < a < b < L

$$s(t) \rightarrow \bar{x} = \frac{a+b}{2}$$
 as $t \rightarrow 0^+$.

For Cauchy Problem
$$ar{x} - lpha t \leq s(t) \leq ar{x}, \,\, lpha = rac{2}{b-a} \ln rac{c_R}{c_L}$$

similar estimates for Homogeneous Neumann Problem.

Exact solutions confirm that if instead $c_L = 0 < c_R$ then all the characteristics starts in $\{x > b\}$.

Other results on initial behaviour of the characteristics for x = 0, which confirm the a priori results

- $c_0 \operatorname{smooth} \longrightarrow X(t;0) \simeq -\frac{c'_0(0)}{c_0(0)}t$
- jump in c'_0 e.g.

$$c_0(x) = c_0(0) + \begin{cases} \gamma_-, & x < 0 \\ \gamma_+, & x > 0 \end{cases}$$

- $\begin{array}{l} \mathbf{1.} \ c_0(0) > 0 \longrightarrow \ X(t;0) \simeq -\frac{\gamma_+ + \gamma_-}{2c_0(0)} t, \\ \\ \mathbf{2.} \ c_0(0) = 0, \gamma_- < 0 \longrightarrow \ X(t;0) \simeq -2\alpha\sqrt{t}, \alpha \text{ depending explicitely on } \gamma_+, \gamma_-, \\ \\ \mathbf{3.} \ c_0(0) = 0, \gamma_- = 0 \longrightarrow \text{ all characteristics start from } x > 0. \end{array}$
- jump in c_0 e.g.

$$c_0(x) = \begin{cases} c_L, & x < 0 \\ c_R, & x > 0, \\ c_L < c_R \end{cases}$$

1. $c_L > 0 \longrightarrow X(t; 0) \simeq -2\alpha\sqrt{t}$, α depending explicitly on c_L, c_R , 2. $c_L = 0 \longrightarrow$ all characteristics start from x > 0.

"holes" of the initial data for c_i (stable isotopes)

- $r_i = \frac{c_i}{c}$ constant along the characteristics $\longrightarrow c_i(x, t)$ inside the domain can be recovered from the initial and boundary data,

- the oscillations of initial and boundary data persist in the interior of the domain (consistent with the experimental results).

- possible existence of interior regions depleted of c_i

the "holes" of c_i remain in time only if they do not coincide with the "holes" of c_0 .

-if $c_i(x,0) \equiv 0$ in $I = (a,b) \subset \Omega$ and $c_0(x) \not\equiv 0$ in I, then, for any time t there exists an interval \tilde{I} where $c_i(x,t) \equiv 0$ for $x \in \tilde{I}$.

-On the contrary, suppose that $\Omega_1 = \Omega_3 = \emptyset$ and that supp $c_i(x, 0) \equiv \text{supp } c_0(x)$, then $c_i(x, t) > 0$ a.e. for any t > 0.



nZsCS-4 - N 1.0D-4 1.0D-4 1.0D0 1.0D0 0.0D0 0.0D0 2 1.0D-3 0.5 -0.2 0.0 0 1.0D-4 0.50 0.0 0.0 1.0 1.0D-4 - t=0.000



nZsCS-4 - c1+c2



nZsCS-4 - c1



nZsCS-4 - c2



nZsCS-4 - c1/(c1+c2)

case with decay ($\Lambda \neq 0$)

c depending on c_i , but the characteristics for each c_i are defined as before; the behaviour of the solution does not change, provided the assumptions:

- (H1) \exists ! solution of the equation $\underline{\dot{u}} = \Lambda \underline{u}, \ u \in \mathbb{R}^n$ for any initial datum \underline{u}_0 .
- (H2) if $u_{i0} \ge 0$, then $u_i(t) \ge 0$, i = 1, ...n. (positive property)

 $0 \le r_{i0} \le 1 \longrightarrow 0 \le r_i(X(t), t) \le 1$

therefore c(x,t) satisfies the following equation

$$c_t = c_{xx} + c(\beta_{nn} + \sum_{i=1}^{n-1} \beta_{ni} r_i) = c_{xx} + b(x, t)c,$$
(1)

with

$$|b(x,t)| < B, \qquad \overline{\Omega} \times [0,T].$$
 (2)

From the classical theory we get then explicit bounds for *c*.

As for the behaviour of the "holes" of c_i , we have similar results as in the case without decay iff the elements of Λ satisfy the following assumption

• (H3) if $u_{i0} = 0$ and $u_{j0} \ge 0$, $u_{j0}(x) \ne 0$ $j \ne i$, then $u_i(t) = 0$ for any t.

example: cauchy problem for the couple $(U^{238},\,U^{234})$

suppose c_1 and c_2 initially separated, i.e.

$$c_0(x) > 0,$$
 $c_{10}(x) = c_0(x)H(x),$

where H(x) is the Heaviside function,

then $\left(c(x,t),s(t)\right)$ can be regarded as solution of the free boundary problem

$$\begin{cases} c_t = c_{xx} + \lambda_2 c(\tilde{r}(t)H(x - s(t)) - 1), \\ \dot{s}(t) = -\frac{c_x}{c}, \\ s(0) = 0, \quad c(x, 0) = c_0(x). \end{cases}$$

where

$$\tilde{r}(t) = \frac{r_E \exp(\lambda_2 - \lambda_1)t}{r_E - 1 + \exp(\lambda_2 - \lambda_1)t}, \quad r_E = \frac{\lambda_2 - \lambda_1}{\lambda_2}.$$

Once the couple $\left(c,s\right)$ is given

$$c_1(x,t) = \tilde{r}(t)c(x,t)H(x-s(t)),$$

 $c_2(x,t) = c(x,t) - c_1(x,t),$

with c_1 discontinuous across x = s(t) and zero for x < s(t).

remark: if we drop the assumption of c_0 positive, e.g. we take $c_0(x) \equiv 0, x < 0$ so that $c_{10}(x) = c_0(x), x \in R$ then we will have instead

$$c_1(x,t) = \tilde{r}c(x,t), \quad x \in \mathbb{R}, \quad t \ge 0,$$
$$c_2(x,t) = (1 - \tilde{r}(t))c(x,t),$$

and c(x,t) is the known solution of

$$c_t = c_{xx} - \lambda_2 c(1 - \tilde{r}(t)), \quad \text{in } \mathbb{R} \times (0, t),$$

therefore c can be calculated explicitly and c_1 will be strictly positive everywhere.

regularity and weak solutions

- irregular initial data give irregular solutions, (i.e. no parabolic effect)

- also if the data are C^∞ the solution can have discontinuities.

Example 4 Cauchy problem without decay

$$c_0 \in C^{\infty}, \ c_0(x) \equiv 0, \ x \in I = [-L, L], \ c_0(x) > 0, \ x \in \mathcal{R}/I,$$

 c_0 symmetric w.r.to x = 0 (hence x = 0 is a characteristic) and for a given i

$$c_{i0}(x) = \begin{cases} \gamma_{1i}c_0(x), & x \le -L \\ 0, & x \in I, \\ \gamma_{2i}c_0(x), & x \ge L, \end{cases}$$

with $\gamma_{1i}\not\equiv\gamma_{2i}$ constants in [0,1].

For t > 0 we have the explicit solution

$$c_{i}(x,t) = \begin{cases} \gamma_{1i}c(x,t), & x < 0, \\ \gamma_{2i}c(x,t), & x > 0, \end{cases}$$

so that c_i has a jump $\forall t>0$ in x=0 increasing in time

$$\gamma[c_i^+ - c_i^-] = (\gamma_{2i} - \gamma_{1i})c(0, t) \neq 0.$$

a similar behaviour can be showed if c_0 is not symmetric and in the case with $\Lambda \neq 0$.



ZcCS-4 - D 1.0D-4 1.0D-4 1.0D0 1.0D0 0.0D0 0.0D0 0 1.0D-4 0.5 +0.2 0.0 1 1.0D-4 0.50 -0.2 0.0 1.0 1.0D-3 - t=0.000



ZcCS-4 - D 1.0D-4 1.0D-4 1.0D0 1.0D0 0.0D0 0.0D0 0 1.0D-4 0.5 +0.2 0.0 1 1.0D-4 0.50 -0.2 0.0 1.0 1.0D-3 - t=0.0500



ZcCS-4 - D 1.0D-4 1.0D-4 1.0D0 1.0D0 0.0D0 0.0D0 0 1.0D-4 0.5 +0.2 0.0 1 1.0D-4 0.50 -0.2 0.0 1.0 1.0D-3 - t=1.0000

hyperbolic model as limit of the parabolic one

if the total concentration is strictly positive, 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 $\tilde{D} \rightarrow 0$.

The numerical simulations all confirm the convergence also if c_0 is allowed to become zero.

In the limit, boundary layers will appear (see again Ref. BLN).

For smooth data one can prove existence and uniqueness of classical solution.

asymptotic behaviour, $t \to \infty$, Homogeneous Neumann Problem

It depends strongly on the decay law $\dot{\tilde{C}} = \tilde{\Lambda} \tilde{C}, ~~ \tilde{C} = (c_1,...,c_{n-1},c).$

Natural assumptions:

- $\tilde{\Lambda}$ has real nonpositive eigenvalues $\sigma_s < \ldots < \sigma_1 \leq 0$
- if $\sigma_1 = 0$ then it is semisimple.

Denote:

• $h(\sigma_1) =$ dimension of the generalized autospace $E(\sigma_1)$

•
$$\hat{B}\mathbf{Y} = \frac{1}{(h(\sigma_1) - 1)!} (\tilde{\Lambda} - \sigma_1 Id)^{h(\sigma_1) - 1} \mathbf{Y}_{0,1}, \ \mathbf{Y}_{0,1} \in E(\sigma_1), \ \mathbf{Y} \in \mathbb{R}^n$$

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

The total mass $m(x,t) = \int_{-L}^x c(\xi,t)\,d\xi$ behaves as

$$m(x,t) \simeq t^{h(\sigma_1)-1} e^{\sigma_1 t} \frac{x+L}{2L} M_{\infty}, \ M_{\infty} = \int_{-L}^{L} F(\xi) d\xi$$

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

• characteristics:

$$X(t, x_0) \to X_{\infty}(x_0) = \frac{2L}{M_{\infty}} \int_{-L}^{x_0} F(\xi) \, d\xi - L \text{ as } t \to \infty$$

• ratioes
$$r_i = \frac{c_i}{c}$$
:
 $F(x) > 0 \implies$
 $r_i(x, t) \to r_{i\infty}(x) = \frac{F_i(X_{\infty}^{-1}(x))}{F(X_{\infty}^{-1}(x))}, \ i = 1, ..., n - 1, \text{ as } t \to \infty$
 $F(x) \ge 0 \implies$

in general one cannot have an asymptotic distribution in the whole $\overline{\Omega}$.

EXAMPLES

1. stable isotopes

$$F = c_0 > 0, \ M_{\infty} = \int_{-L}^{L} F(\xi) \, d\xi, \ m \simeq \frac{x+L}{2L} M_{\infty}, \quad r_i \simeq \frac{c_{i0}(X_{\infty}^{-1}(x))}{c_0(X_{\infty}^{-1}(x))}, \ i = 1, ..., n-1,$$

"asymptotic localization property"

2. chain of the type U^{238}, U^{234} :

$$\dot{c}_1 = -\lambda_1 c_1, \ \dot{c}_2 = \lambda_1 c_1 - \lambda_2 c_2, \ 0 < \lambda_1 < \lambda_2$$

$$F = (1 - \frac{\lambda_1}{\lambda_2})c_{10}, \ M_{\infty} = \int_{-L}^{L} F(\xi) \, d\xi, \ m \simeq e^{-\lambda_1 t} \frac{x + L}{2L} M_{\infty},$$

if $c_{10} > 0 \implies r \simeq r_E = 1 - \frac{\lambda_1}{\lambda_2}$, secular equilibrium
if $c_{10} \ge 0 \implies$ no limit near the points identified by the graph X_{∞}^{-1}

Still example 2 but

$$\begin{aligned} -0 < \lambda_2 < \lambda_1 &\Longrightarrow \\ F = c_0 + \frac{\lambda_2}{\lambda_1 - \lambda_2} c_{10} \ge c_0 > 0, \ M_\infty = \int_{-L}^{L} F(\xi) \, d\xi, \\ m &\simeq e^{-\lambda_2 t} \frac{x + L}{2L} M_\infty, \ r = \frac{c_1}{c} \to 0, \text{ only isotope } 2 \text{ is present at } t \to \infty \\ -0 < \lambda_1 = \lambda_2 = \lambda &\Longrightarrow \\ F = \lambda c_{10}, \ M_\infty = \int_{-L}^{L} F(\xi) \, d\xi \quad m \simeq t e^{-\lambda t} \frac{x + L}{2L} M_\infty \\ \text{but } \forall c_{10} \ge 0 \implies r = \frac{c_1}{c} \to 0. \end{aligned}$$

References

- [1] H. Amann, 'Dynamic theory of quasilinear parabolic systems. III Global existence', Math.Z, 202, (1989), pp. 219–250.
- [2] H. Amann 'Dynamic theory of quasilinear parabolic systems. II Reaction-Diffusion systems, Diff. and Int. Eq., 3, n.1 (1990), pp. 13–75.
- [3] D. Ambrosi, L. Preziosi, On the closure of mass balance models for tumor growth, *Math.Models Methods Appl. Sci.* 12 (2002) 737–754.
- [4] M. Bertsch, M.E. Gurtin, D. Hilhorst, 'On the interacting populations that disperse to avoid crowding: the case of equal dispersal velocities'. Nonlinear Anal.Th.Meth.Appl. vol II, **4**, (1987), pp. 493–499.
- [5] H.F. Bremer, E.I. Cussler, 'Diffusion in the Ternary System d-Tartaric Acid c-Tartaric Acid Water at 25°C'. AIChE Journal, 16, 9 (1980) pp. 832–838.
- [6] C. Bardos, A.Y. Leroux, J.C. Nedelec, 'First order quasilinear equations with boundary conditions'.
 Comm. In PDE, 4, 9 (1979) pp.1017 –1034.
- [7] E. Comparini, R. Dal Passo, C. Pescatore, M. Ughi *On a model for the propagation of isotopic disequilibrium by diffusion* Math.Models Methods Appl. Sci. **19**, 8 (2009) pp 1277–1294.
- [8] E. Comparini, A. Mancini, C. Pescatore, M. Ughi, *Numerical results for the Codiffuson of Isotopes*, Communications to SIMAI Congress, vol. 3, ISSN: 1827-9015 (2009).

- [9] E. Comparini, C. Pescatore, M. Ughi *On a quasilinear parabolic system modelling the diffusion of radioactive isotopes*, RIMUT XXXIX (2007) 127–140.
- [10] E. Comparini, M. Ughi Large time behaviour of the solution of a parabolic-hyperbolic system modelling the codiffusion of isotopes, to appear on Adv. in Math. Sc. and Appl. (2011).
- [11] E. Comparini, M. Ughi On the asymptotic behaviour of the characteristics in the codiffusion of radioactive isotopes with general initial data, to appear on RIMUT (2012).
- [12] A. Friedmann, Partial differential equations of parabolic type, Englewood Cliffs: Prentice-Hall, (1964).
- [13] G.E. Hernandez, 'Existence of solutions in a population dynamics problem', Quarterly of Appl. Math., vol. XLIII 4, (1986) pp. 509–521.
- [14] G.E. Hernandez, 'Localization of age-dependent anti-crowding populations', Quarterly of Appl. Math., vol. LIII 1, (1995) pp. 35–52.
- [15] T. Gimmi, H.N. Waber, A. Gautschi, A. Riibel, 'Stable water isotopes in pore water of Jurassic argillaceous rocks as tracers for solute transport over large spatial and temporal scales', WATER RESOURCES RESEARCH, vol. 43, (2007).
- [16] KASAM Nuclear Waste state of the art reports 2004', Swedish Government Official Reports SOU 2004-67, (2005).
- [17] R.C. MacCamy, 'A population model with nonlinear diffusion', J.Diff.Eq. 39, (1981), pp. 52–72.

- [18] C. Pescatore, Discordance in understanding of isotope solute diffusion and elements for resolution, Proceedings OECD/NEA "Radionuclide retention in geological media", Oskarsham, Sweden, (2002) pp. 247–255.
- [19] A. Terracina, A free boundary problem for scalar conservation laws. SIAM J. of Math.Anal. 30 5, (1999) pp. 985–1009.
- [20] R. Wang, P. Keast, P. H. Muir Algorithm 874:BACOLR spatial and temporal error control software for PDEs based on high-order adaptive collocation ACM Transactions on Mathematical Software (TOMS) 34, 3 (2008).