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**PADLOC, A ONE-DIMENSIONAL
COMPUTER PROGRAM FOR
CALCULATING COOLANT AND PLATEOUT
FISSION-PRODUCT CONCENTRATIONS**

PART II

by
W. W. HUDRITSCH

Prepared Under
Contract DE-AT03-76ET35301
for the San Francisco Operations Office
Department of Energy

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ABSTRACT

The behavior of some of the prominent fission products along their convection pathways is dominated by the interaction of other species with them. This gave rise to the development of a plateout code capable of analyzing coupled species effects. The single species plateout computer program PADLOC is described in Part I of this report. The present Part II is concerned with the extension of PADLOC to MULTI*PADLOC, a multiple species version of PADLOC.

MULTI*PADLOC is designed to analyze the time and one-dimensional spatial dependence of the concentrations of interacting (fission product) species in the carrier gas and on the surrounding wall surfaces on an arbitrary network of flow channels. The problem solved is one of mass transport of several impurity species in a gas, including the effects of sources in the gas and on the surface, convection along the flow paths, decay interaction, sorption interaction on the wall surfaces, and chemical reaction interactions in the gas and on the surfaces. These phenomena are governed by a system of coupled, nonlinear partial differential equations. The solution is achieved by, a) linearizing the equations about an approximate solution and employing a Newton-Raphson iteration technique, b) employing a finite difference solution method with an implicit time integration, and c) employing a substructuring technique to logically organize the systems of equations for an arbitrary flow network.

1. INTRODUCTION

The need for a multiple species plateout computer program arises from the fact that the behavior of some of the prominent fission products is dominated by the interaction of other species with them. As an illustration three important species interactions, namely

- precursor effects,
- isotopic exchange on surfaces, and
- chemical reactions on surfaces,

are discussed below.

1.1 PRECURSOR EFFECTS

Consider, e.g., the formation of two typical condensible nuclides, Sr^{90} and Cs^{137} , from fission gases according to

and

where the cumulative fission yields (U^{235} fissions) are indicated in parentheses. It is seen that most of the yield is due to the decay of precursors. Cs^{137} and Sr^{90} are introduced into the primary coolant both, by release from the core and by decay of their precursors in the

primary coolant. Neglecting the precursor decay time* would result in an exponential decrease of coolant and surface activities along the path with appreciable concentrations only for a short distance. By taking account of the formation of the condensable nuclides along their path by decay of precursors, the picture looks quite different. Since the (gaseous) precursors are carried along with the coolant before there is appreciable decay,** the metals are now plating out over distances much larger than those predicted by neglecting precursor effects. This is indicated in the sketch below

- a) Based on increased coolant source at core exit to account for the decay of precursors.
- b) Based on coolant source from core release and precursor sources in the coolant along its way.

* Increasing the source rate of the metal at the core exit.

** Coolant transit times are short compared to the half-life of the gaseous precursors.

1.2 ISOTOPIC EXCHANGE ON SURFACES

Here the case of the important ^{131}I isotope that is produced with a cumulative yield of $\approx 3\%$ is considered. Iodine has six other fission isotopes with yields between $\approx 0.4\%$ and $\approx 6.4\%$ that range all the way from stable ^{127}I to $0.88\text{ h }^{134}\text{I}$. Since all of these isotopes exhibit the same sorption behavior, they all compete for sorption sites on the conduit surfaces. An analysis considering ^{131}I on its own thus grossly over predicts the amount of plateout of this isotope. On the other hand, a multiple species code would account for the site competition and predict the exchange of isotopes as a function of space and time.

1.3 CHEMICAL REACTIONS

Under LOFC conditions some of the conduit surfaces reach temperatures high enough to allow metal iodide formation. These iodides are less volatile than elemental iodine, thus binding the iodine to the surfaces. It is clear that neglecting this chemical reaction over-predicts the amount of iodine in the coolant.

1.4 MULTI*PADLOC COMPUTER PROGRAM

Above examples clearly show the need for a multiple species plateout code. For this reason MULTI*PADLOC was developed. This computer program is a straightforward generalization of the single species code PADLOC to a multiple species version. Apart from the fact that MULTI*PADLOC has a restart option and that the input and the printout of results are different from PADLOC, all other aspects of the code, including the solution method, have essentially not changed. It is assumed that the reader is familiar with the theory and applications of PADLOC, described in Part I of this report. Since detailed discussions of most features of the single species code are directly applicable to the multiple species version they will not be repeated here.

2. THE MATHEMATICAL MODEL

The governing equations for the coolant and surface concentrations, C_i and S_i of species i , containing now additional precursor and chemical reaction terms, can, for a single channel of length L , be written in the form

$$\begin{aligned} \frac{\partial C_i(x,t)}{\partial t} = & [\dot{q}_c(x,t)]_i + f_i[\vec{C}(x,t)] + \sum_{j=1}^i a_{ij}^* C_j(x,t) - \\ & - \frac{P(x) h_i(x,t)}{A(x)} \left\{ C_i(x,t) - B_i[\vec{S}(x,t)] \right\} - \frac{1}{A(x)} \frac{\partial [A(x)v(x,t)C_i(x,t)]}{\partial x} \end{aligned} \quad (1)$$

$$\begin{aligned} \frac{\partial S_i(x,t)}{\partial t} = & [\dot{q}_s(x,t)]_i + g_i[\vec{S}(x,t)] + \sum_{j=1}^i a_{ij}^* S_j(x,t) + \\ & + h_i(x,t) \left\{ C_i(x,t) - B_i[\vec{S}(x,t)] \right\} , \end{aligned} \quad (2)$$

$$\left. \begin{aligned} C_i(o,t) &= C_{io}(t) , \quad v > 0 \\ C_i(L,t) &= C_{iL}(t) , \quad v < 0 \end{aligned} \right\} \text{Boundary condition,} \quad (3)$$

$$\left. \begin{aligned} C_i(x,o) &= C_i^*(x) \\ S_i(x,o) &= S_i^*(x) \end{aligned} \right\} \text{Initial conditions,} \quad (4)$$

$$i=1,2,\dots,n .$$

Here,

$x(\text{cm})$, is the distance along the channel,

$t(\text{sec})$, is the time,

$v(\text{cm/sec})$, is the average carrier gas velocity,

$P(\text{cm})$, is the channel perimeter length,

$A(\text{cm}^2)$, is the channel cross section area,

$C_i \left(\frac{\mu\text{g}}{\text{cm}^3} \right)$, is the bulk coolant concentration,

$B_i \left(\frac{\mu\text{g}}{\text{cm}^3} \right)$, is the boundary layer concentration,

$S_i \left(\frac{\mu\text{g}}{\text{cm}^2} \right)$, is the (wall) surface concentration,

$h_i \left(\frac{\text{cm}}{\text{sec}} \right)$, is the mass transfer coefficient between the boundary layer and the surface,

$(\dot{q}_c)_i \left(\frac{\mu\text{g}}{\text{cm}^3 \text{sec}} \right)$, is the direct source rate in the carrier gas,

$(\dot{q}_s)_i \left(\frac{\mu\text{g}}{\text{cm}^2 \text{sec}} \right)$, is the direct source rate on the surface.

The terms involving the "depletion matrix" a^* are source and removal rates resulting from decay. Specifically, with

$\lambda_i (\text{cm}^{-1})$, being the decay constant, and

$0 \leq q_{ji} \leq 1$, being the decay fraction (branching ratio) of species j to species i ,

$$a_{ii}^* \equiv -\lambda_i \quad , \quad (5)$$

$$a_{ij}^* \equiv q_{ji} \lambda_j \quad , \quad j=1,2,\dots,i-1 \quad .$$

The rates involving the difference between the coolant and the boundary layer concentrations are source or removal rates, depending on the sign of the net flux $= h_i [C_i - B_i]$ from the coolant to the surface. $B_i(\vec{S})$ in this connection is shorthand for*

$$B_i = B_i[S_1(x,t), S_2(x,t), \dots, S_n(x,t)] \quad .$$

This function, known as sorption relation, is in general nonlinear. Specific forms will be given in Chapter 8.

The last term on the right hand side of (1),

$$- \frac{1}{A} \frac{\partial (AvC_i)}{\partial x} \quad ,$$

is the source rate resulting from convection in positive x direction.

Finally, the rates $f_i(\vec{C})$ and $g_i(\vec{S})$ represent the net source rates resulting from chemical reactions in the carrier gas and on the conduit surface. These functions are, in general, nonlinear and will be discussed in Appendix A2.

The method employed to solve Eqs. (1) and (2) for a general network of interconnected flow channels or branches** is outlined in the following chapter.

* The same applies for $f_i(\vec{C})$ and $g_i(\vec{S})$.

** A branch is defined as an assembly of $n_c \geq 1$ identical parallel flow channels connected at both ends.

3. SOLUTION METHOD

If there were only one branch (with $n_c=1$), the $2n$ coupled equations (1) and (2), together with the n boundary conditions (3) and the $2n$ initial conditions (4) would have to be solved for all the $C_i, S_i, i=1,2,\dots,n$. In a general network, however, there are n_B coupled branches and the boundary conditions for a downstream branch depend on the branch end coolant concentrations of the connected upstream branches. For a network of n_B branches there are then $2nn_B$ coupled equations to solve. The way this is done is to reduce the equation system of each branch to a system involving only branch-end coolant concentrations (or mass flow rates). Each of the reduced systems is then complemented with a set of mixing equations for the upstream branch end. The $2n_B$ sets of global equations are then solved for branch-end coolant concentrations. These branch-end coolant concentrations are the boundary conditions for the different branches. Knowing the boundary conditions, the coolant concentrations within each of the branches can be solved for. Finally, the surface concentrations are computed from the known coolant concentrations.

The reduction of the equation system of a single channel involves the following steps:

- a. Linearization of the nonlinear terms so that solution can be achieved by iteration and updating of the concentration dependent linearization coefficients (Newton-Raphson method).
- b. Time discretization.
- c. Solving for $\vec{S} \approx \vec{S}(\vec{C})$ in the discretized equations (2).*

* $\vec{C}$ and $\vec{S}$ are column vectors of n components $i=1,2,\dots,n$.

- d. Eliminating $\vec{S}$ in favor of $\vec{C}$ in the discretized equations (1).
- e. Spatial discretization of equation system (1) which has as its only unknown the vector $\vec{C}$.
- f. Reduction of the (matrix) equation for $\vec{C}$ to a form involving only the end concentrations $\vec{C}(0,t)$ and $\vec{C}(L,t)$.

All the above steps will be discussed in the following chapter.

4. REDUCTION OF THE SINGLE CHANNEL EQUATIONS

4.1 LINEARIZATION OF THE NONLINEAR TERMS

Linearization about the last solution guess $\vec{C}_o, \vec{S}_o$ leads to

$$B_i(\vec{S}) \approx \phi_i^*(\vec{S}_o) + \sum_{j=1}^n \psi_{ij}^*(\vec{S}_o) S_j, \quad (6)$$

$$f_i(\vec{C}) \approx \xi_i^*(\vec{C}_o) + \sum_{j=1}^n \eta_{ij}^*(\vec{C}_o) C_j, \quad (7)$$

$$g_i(\vec{S}) \approx \sigma_i^*(\vec{S}_o) + \sum_{j=1}^n \omega_{ij}^*(\vec{S}_o) S_j, \quad (8)$$

where the dependence on $\vec{V}$ means that the function depends on all V_i , $i=1,2,\dots,n$. The actual concentration dependence of the linearization coefficients will be derived in Appendix A.

To facilitate a compact representation of Equations (1) and (2), the following parameters are introduced:

$$(\dot{Q}_c)_i \equiv (\dot{q}_c)_i + \frac{P}{A} h_i \phi_i^* + \xi_i^*, \quad (9)$$

$$\psi_{ij} \equiv \frac{P}{A} h_i \psi_{ij}^*, \quad (10)$$

$$a_{ij} \equiv a_{ij}^* + \eta_{ij}^* - \delta_{ij} \frac{p}{A} h_j, \quad (11)$$

$$(\dot{Q}_s)_i \equiv (\dot{q}_s)_i - h_i \phi_i^* + \sigma_i^*, \quad (12)$$

$$b_{ij} \equiv a_{ij}^* - h_i \psi_{ij}^* + \omega_{ij}^*. \quad (13)$$

Here the x and time dependencies are, for convenience, suppressed, and

$$\delta_{ij} = \begin{cases} 0, & j \neq i \\ 1, & j = i \end{cases}.$$

With these parameters, the linearized equations (1) and (2) take the form

$$\frac{\partial C_i}{\partial t} = (\dot{Q}_c)_i + \sum_{j=1}^n \psi_{ij} S_j + \sum_{j=1}^n a_{ij} C_j - \frac{1}{A} \frac{\partial (A v C_i)}{\partial x}, \quad (14)$$

$$\frac{\partial S_i}{\partial t} = (\dot{Q}_s)_i + h_i C_i + \sum_{j=1}^n b_{ij} S_j, \quad (15)$$

or

$$\frac{d\vec{C}}{dt} = \vec{\dot{Q}}_c + [\psi] \vec{S} + [a] \vec{C} - \frac{1}{A} \frac{\partial (A v \vec{C})}{\partial x} = \vec{F}_1(\vec{C}, \vec{S}), \quad (16)$$

$$\frac{\partial \vec{S}}{\partial t} = \vec{\dot{Q}}_s + [\sim h \sim] \vec{C} + [b] \vec{S} = \vec{F}_2(\vec{C}, \vec{S}). \quad (17)$$

$\vec{C}$ and $\vec{S}$ are $n \times 1$ vectors, the matrices $[a]$, $[b]$, $[\psi]$ and $[h]$ are $n \times n$.

The first three are, in general, full matrices, whereas $[\sim h \sim]$ is diagonal as indicated.

4.2 TIME DISCRETIZATION

Discretization of the time t in Eq. (16) and (17) leads to

$$\vec{C}^\ell = \vec{C}^{\ell-1} + [(1-\kappa) \vec{F}_1^{\ell-1}(\vec{C}^{\ell-1}, \vec{S}^{\ell-1}) + \kappa \vec{F}_1^\ell(\vec{C}^\ell, \vec{S}^\ell)] \Delta t_\ell, \quad (18)$$

$$\vec{S}^\ell = \vec{S}^{\ell-1} + [(1-\kappa) \vec{F}_2^{\ell-1}(\vec{C}^{\ell-1}, \vec{S}^{\ell-1}) + \kappa \vec{F}_2^\ell(\vec{C}^\ell, \vec{S}^\ell)] \Delta t_\ell, \quad (19)$$

$$\Delta t_\ell \equiv t_\ell - t_{\ell-1}, \quad (20)$$

where ℓ is the time point index ($\ell \geq 1$), $\vec{F}_1$ and $\vec{F}_2$ are the right hand sides of (16), (17), and

$$0 \leq \kappa \leq 1, \quad (21)$$

is the time integration parameter. For different values of κ the integration schemes are known as

$$\begin{aligned} \text{explicit} & \quad , \quad \kappa = 0, \\ \text{implicit} & \quad , \quad 0 < \kappa \leq 1, \quad (\text{Crank-Nicholson}, \kappa = 0.5) \\ & \quad \quad \quad (\text{fully implicit}, \kappa = 1.0) \end{aligned}$$

We now solve (19) for $\vec{S}^\ell$, yielding

$$\vec{S}^\ell = [D^\ell] \left\{ \vec{S}^{\ell-1} + \Delta t_\ell \left[(1-\kappa) \vec{F}_2^{\ell-1} + \kappa \left(\vec{Q}_s^\ell + [-h^\ell] \vec{C}^\ell \right) \right] \right\}, \quad (22)$$

$$[D^\ell] = \left([U] - \kappa \Delta t_\ell [b^\ell] \right)^{-1}, \quad (23)$$

$[U]$ is the unit matrix.

For known coolant concentrations $\vec{C}^\ell$, Eq. (22) will be used to compute the surface concentrations $\vec{S}^\ell$. For the time however, we want to obtain an equation for $\vec{C}^\ell$. Substitution with Eq. (22) in $\vec{F}_1^\ell$ of (18) yields this equation:

$$[G^\ell] \vec{C}^\ell + \kappa \Delta t_\ell \frac{1}{A} \frac{\partial (A v \vec{C}^\ell)}{\partial x} = \vec{H}^{\ell-1,\ell} . \quad (24)$$

Here,

$$[G^\ell] = [U] - \kappa \Delta t_\ell \left(\kappa \Delta t_\ell [\psi^\ell] [D^\ell] [h^\ell] + [a^\ell] \right) , \quad (25)$$

$$\vec{H}^{\ell-1,\ell} = \vec{C}^{\ell-1} + \Delta t_\ell \left[(1-\kappa) \vec{F}_1^{\ell-1} + \kappa (\vec{Q}_c^\ell + \vec{F}_3^{\ell-1,\ell}) \right] , \quad (26)$$

$$\vec{F}_3^{\ell-1,\ell} = [\psi^\ell] [D^\ell] \left\{ \vec{S}^{\ell-1} + \Delta t_\ell \left[(1-\kappa) \vec{F}_2^{\ell-1} + \kappa \vec{Q}_s^\ell \right] \right\} , \quad (27)$$

and the superscript $\ell-1,\ell$ indicates a dependence on the past and present time points.

Because of the convection term in Eq. (24) the next step is spatial discretization. This will be done in the next paragraph.

4.3 SPATIAL DISCRETIZATION

We assume $n_x \geq 3$ discrete points x_k , $k=1,2,\dots,n_x$ along the channel length L . Then

$$x_1 = 0 ,$$

$$x_{n_x} = L , \text{ and} \quad (28)$$

$$\Delta x_k = x_{k+1} - x_k , \quad k=1,2,\dots,n_x-1$$

To avoid singularities of the system matrix, we use backward differences for $v>0$ and forward differences if $v<0$. This leads to

$$\frac{1}{A} \frac{\partial (A v \vec{C})}{\partial x} = \begin{cases} \gamma_{k,k-1} \vec{C}_{k-1} + \gamma_{k,k} \vec{C}_k, & 2 \leq k \leq n_x, v>0 \\ \gamma_{kk} \vec{C}_k + \gamma_{k,k+1} \vec{C}_{k+1}, & 1 \leq k \leq n_x-1, v<0 \end{cases} \quad (29)$$

and the following coefficients γ :

	$\gamma_{k,k-1}$	γ_{kk}	$\gamma_{k,k+1}$
backward d. $v>0$	$-\frac{A_{k-1} v_{k-1}}{A_k \Delta x_{k-1}}$	$\frac{v_k}{\Delta x_{k-1}}$	
forward d. $v<0$		$-\frac{v_k}{\Delta x_k}$	$\frac{A_{k+1} v_{k+1}}{A_k \Delta x_k}$

(30)

With this information, we proceed to put Eq. (24) into its final form before reduction.

4.3.1 The Case $v>0$

Substitution with Eq. (29) in Eq. (24) for the nodal points $k=2,3,\dots,n_x$ leads to

$$[\varepsilon^\ell]_{k,k-1} \vec{C}_{k-1}^\ell + [\varepsilon^\ell]_{kk} \vec{C}_k^\ell = \vec{H}_k^{\ell-1,\ell}, \quad 2 \leq k \leq n_x, \quad (31a)$$

where the matrices

$$[\varepsilon^\ell]_{k,k-1} = \kappa \Delta t_\ell \gamma_{k,k-1}^\ell [U], \quad (32a)$$

$$[\varepsilon^\ell]_{kk} = [G^\ell]_k + \kappa \Delta t_\ell \gamma_{kk}^\ell [U], \quad (33a)$$

are of size $n \times n$ and $\vec{C}$ and $\vec{H}$ of size $n \times 1$.* Eq. (31a) written out takes the form

$$\begin{matrix} k = & 1 & 2 & 3 & \cdot & n_x-1 & n_x \\ & \left[\begin{array}{c|c|c|c|c|c} \hline [\epsilon^\ell]_{21} & [\epsilon^\ell]_{22} & & & & \\ \hline & [\epsilon^\ell]_{32} & [\epsilon^\ell]_{33} & & & \\ & & & \cdot & & \\ & & & & \cdot & \\ & & & & [\epsilon^\ell]_{n_x-2}^{n_x-1} & [\epsilon^\ell]_{n_x-1}^{n_x-1} \\ \hline & & & & [\epsilon^\ell]_{n_x}^{n_x-1} & [\epsilon^\ell]_{n_x}^{n_x-1} \\ \hline \end{array} \right] & \left\{ \begin{array}{c} \vec{C}_1^\ell \\ \vec{C}_2^\ell \\ \vec{C}_3^\ell \\ \cdot \\ \vec{C}_{n_x-1}^\ell \\ \vec{C}_{n_x}^\ell \end{array} \right\} = \left\{ \begin{array}{c} \vec{H}_2^{\ell-1, \ell} \\ \vec{H}_3^{\ell-1, \ell} \\ \cdot \\ \vec{H}_{n_x-1}^{\ell-1, \ell} \\ \vec{H}_{n_x}^{\ell-1, \ell} \end{array} \right\} \quad (34a)
 \end{matrix}$$

and represents n_x-1 matrix equations in the n_x unknown coolant concentration vectors $\vec{C}_k^\ell$, $k=1,2,\dots,n_x$. We now turn to reducing (34a) to an equation containing only the channel end concentrations $\vec{C}_1^\ell$ and $\vec{C}_{n_x}^\ell$.

First (34a) is rewritten with the partitioned matrices and vectors as indicated by the dashed lines and where the definitions of the new quantities are obvious.

$$\left[\begin{array}{c|c|c} \hline [\Sigma^\ell]_{21} & [\Sigma^\ell]_{22} & \\ \hline & [\Sigma^\ell]_{32} & [\Sigma^\ell]_{33} \\ \hline \end{array} \right] \left\{ \begin{array}{c} \vec{C}_1^\ell \\ \hline \vec{C}_{\text{inside}}^\ell \\ \hline \vec{C}_{n_x}^\ell \end{array} \right\} = \left\{ \begin{array}{c} \vec{H}_{\text{inside}}^{\ell-1, \ell} \\ \hline \vec{H}_{n_x}^{\ell-1, \ell} \end{array} \right\} \quad (35a)$$

From the first of these equations we obtain

$$\vec{C}_{\text{inside}}^\ell = [\Sigma^\ell]_{22}^{-1} \left(\vec{H}_{\text{inside}}^{\ell-1, \ell} - [\Sigma^\ell]_{21} \vec{C}_1^\ell \right), \quad (36a)$$

* n is the number of fission product species.

and substitution for $\vec{C}_{\text{inside}}^\ell$ in the second equation yields

$$\begin{pmatrix} -[\Sigma^\ell]_{32} & [\Sigma^\ell]_{22}^{-1} & [\Sigma^\ell]_{21} \end{pmatrix} \vec{C}_1^\ell + [\Sigma^\ell]_{33} \vec{C}_{n_x}^\ell = \vec{H}_{n_x}^{\ell-1,\ell} - [\Sigma^\ell]_{32} [\Sigma^\ell]_{22}^{-1} \vec{H}_{\text{inside}}^{\ell-1,\ell} \quad (37a)$$

Equation (37a) is the reduced equation for a single channel containing only channel end concentration vectors.

We now introduce the notion of a branch. It often happens that a number of identical parallel channels, connected at both ends, need to be analyzed. Such an assembly with $n_c \geq 1$ channels is called a branch. Branch end points are global points. A connection of two branches at a global point of index IG_2 is shown in the sketch.

To find the boundary condition for each upstream branch end, it is of advantage to use branch end fission product flow rates. These are the flow rates at the end nodes 1 and n_x :

$$\begin{aligned}\vec{M}_1^{*\ell} \left(\frac{\mu g}{\text{sec}} \right) &= n_c A_1 v_1 \vec{C}_1^\ell, \\ \vec{M}_{n_x}^{*\ell} \left(\frac{\mu g}{\text{sec}} \right) &= n_c A_{n_x} v_{n_x} \vec{C}_{n_x}^\ell.\end{aligned}\tag{38}$$

Eq. (37a) is now transformed by multiplication with $n_c A_1 v_1^\ell A_{n_x} v_{n_x}^\ell$, resulting in

$$[\Sigma^{*\ell}]_{11} \vec{M}_1^{*\ell} + [\Sigma^{*\ell}]_{n_x n_x} \vec{M}_{n_x}^{*\ell} = \vec{H}^{*,\ell-1,\ell},\tag{39a}$$

where

$$[\Sigma^{*\ell}]_{11} = - A_{n_x} v_{n_x}^\ell \left([\Sigma^\ell]_{32} [\Sigma^\ell]_{22}^{-1} [\Sigma^\ell]_{21} \right),\tag{40a}$$

$$[\Sigma^{*\ell}]_{n_x n_x} = A_1 v_1^\ell [\Sigma^\ell]_{33},\tag{41a}$$

$$\vec{H}^{*,\ell-1,\ell} = n_c A_1 v_1^\ell A_{n_x} v_{n_x}^\ell \left(\vec{H}_{n_x}^{\ell-1,\ell} - [\Sigma^\ell]_{32} [\Sigma^\ell]_{22}^{-1} \vec{H}_{\text{inside}}^{\ell-1,\ell} \right).\tag{42a}$$

Equation (39a) is the reduced branch equation. Equations of this type, together with mixing equations, will be used to establish a global equation system for the branch-end impurity mass flow rates $\vec{M}^{*}$.

4.3.2 The Case $v < 0$

With the application of Eq. (29) for $v < 0$ (using for convenience the same symbols* as before) we obtain in a similar fashion:

$$[\epsilon^\ell]_{kk} \vec{C}_k^\ell + [\epsilon^\ell]_{k,k+1} \vec{C}_{k+1}^\ell = \vec{H}_k^{\ell-1,\ell} \quad (31b)$$

$$[\epsilon^\ell]_{kk} = [G^\ell]_k + \kappa \Delta t_\ell \gamma_{kk}^\ell [U] \quad (32b)$$

$$[\epsilon^\ell]_{k,k+1} = \kappa \Delta t_\ell \gamma_{k,k+1}^\ell [U] \quad (33b)$$

$$\begin{array}{c}
 k= \\
 \begin{array}{cccccc}
 1 & 2 & 3 & & n_x-1 & n_x \\
 1 & [\epsilon^\ell]_{11} & [\epsilon^\ell]_{12} & & & \\
 2 & & [\epsilon^\ell]_{22} & [\epsilon^\ell]_{23} & & \\
 \cdot & & & \cdot & \cdot & \\
 n_x-2 & & & [\epsilon^\ell]_{n_x-2, n_x-2} & [\epsilon^\ell]_{n_x-2, n_x-1} & \\
 u_x-1 & & & [\epsilon^\ell]_{n_x-1, n_x-1} & [\epsilon^\ell]_{n_x-1, n_x} & [\epsilon^\ell]_{n_x, n_x}
 \end{array}
 \end{array}
 \left[\begin{array}{c}
 \vec{C}_1^\ell \\
 \vec{C}_2^\ell \\
 \vec{C}_3^\ell \\
 \cdot \\
 \vec{C}_{n_x-1}^\ell \\
 \vec{C}_{n_x}^\ell
 \end{array} \right] = \left[\begin{array}{c}
 \vec{H}_1^{\ell-1,\ell} \\
 \vec{H}_2^{\ell-1,\ell} \\
 \vec{H}_3^{\ell-1,\ell} \\
 \cdot \\
 \vec{H}_{n_x-1}^{\ell-1,\ell} \\
 \vec{H}_{n_x}^{\ell-1,\ell}
 \end{array} \right] \quad (34b)$$

* Correctly, different symbols for $[\epsilon]$, $[\Sigma]$, $[\Sigma^*]$ and $\vec{H}^*$ should be used since the coefficients γ are not the same for $v > 0$ and $v < 0$.

$$\begin{bmatrix} [\Sigma^\ell]_{11} & [\Sigma^\ell]_{12} & \\ & [\Sigma^\ell]_{22} & [\Sigma^\ell]_{23} \end{bmatrix} \begin{Bmatrix} \vec{C}_1^\ell \\ \vec{C}_{\text{inside}}^\ell \\ \vec{C}_{n_x}^\ell \end{Bmatrix} = \begin{Bmatrix} \vec{H}_1^{\ell-1,\ell} \\ \vec{H}_{\text{inside}}^{\ell-1,\ell} \end{Bmatrix} \quad (35b)$$

$$\vec{C}_{\text{inside}}^\ell = [\Sigma]_{22}^{-1} \left(\vec{H}_{\text{inside}}^{\ell-1,\ell} - [\Sigma^\ell]_{23} \vec{C}_{n_x}^\ell \right) \quad (36b)$$

$$[\Sigma^\ell]_{11} \vec{C}_1^\ell - [\Sigma^\ell]_{12} [\Sigma^\ell]_{22}^{-1} [\Sigma^\ell]_{23} \vec{C}_{n_x}^\ell = \vec{H}_1^{\ell-1,\ell} - [\Sigma^\ell]_{12} [\Sigma^\ell]_{22}^{-1} \vec{H}_{\text{inside}}^{\ell-1,\ell} \quad (37b)$$

$$[\Sigma^{*\ell}]_{11} \vec{M}_1^{*\ell} + [\Sigma^{*\ell}]_{n_x n_x} \vec{M}_{n_x}^{*\ell} = \vec{H}^{*\ell-1,\ell} \quad (39b)$$

$$[\Sigma^{*\ell}]_{11} = A_n v_n^\ell [\Sigma^\ell]_{11} \quad (40a)$$

$$[\Sigma^{*\ell}]_{n_x n_x} = -A_1 v_1^\ell [\Sigma^\ell]_{12} [\Sigma^\ell]_{22}^{-1} [\Sigma^\ell]_{23} \quad (41a)$$

$$\vec{H}^{*\ell-1,\ell} = n_c A_1 v_1^\ell A_n v_n^\ell \left(\vec{H}_1^{\ell-1,\ell} - [\Sigma^\ell]_{12} [\Sigma^\ell]_{22}^{-1} \vec{H}_{\text{inside}}^{\ell-1,\ell} \right) \quad (42a)$$

Each reduced branch equation (39) contains two unknown branch end flow rates $\vec{M}_1^*$ and $\vec{M}_{n_x}^*$. For each branch a second equation involving these unknowns is therefore required. This second equation is either a given global boundary condition which has to be supplied for each free upstream branch end, or it is a mixing equation involving impurity end flow rates of connected branches. These mixing equations are discussed in the following chapter.

5. MIXING EQUATIONS

Consider a global point to which several branches are connected. $i=1,2,\dots,I$ branches have flows coming into the global point and $j=1,2,\dots,J$ have

flows coming out of the global point. $(\vec{M}_{n_x})_i$ is the fission product flow rate at the last nodal point n_x of the i -th incoming branch. $\vec{X}_j$ is the fission product flow rate at nodal point 1 of the j -th outgoing branch. $\vec{m}_j^*$ (g/sec) is the branch flow rate of the carrier gas. If we assume perfect mixing in the global point so that the concentrations $(\vec{C}_1)_j$ are the same for all outgoing branches, then

$$\vec{X}_j = \frac{\vec{m}_j^*}{\sum_{j=1}^J \vec{m}_j^*} \sum_{j=1}^J \vec{X}_j$$

and since for continuity

$$\sum_{j=1}^J \vec{\dot{X}}_j = \sum_{i=1}^I (\dot{M}_{n_x}^*)_i ,$$

$$\vec{\dot{X}}_j = \frac{\dot{m}_j^*}{\sum_{j=1}^J \dot{m}_j^*} \sum_{i=1}^I (\dot{M}_{n_x}^*)_i . \quad (42a)$$

Eq. (42a) is the mixing equation. In the case of a free upstream global point, where there would only be outgoing branches, the right hand sum is replaced by the boundary mass flow rate $\dot{Z}_{IG}$ ($\mu\text{g}/\text{sec}$) for the particular global point IG,

$$\vec{\dot{X}}_j = \frac{\dot{m}_j^*}{\sum_{j=1}^J \dot{m}_j^*} \dot{Z}_{IG} . \quad (42b)$$

Equations (42a) or (42b), with one exception, are used to complement the reduced branch equations for each branch in the network. No mixing equation is really needed if there is only one incoming and one outgoing branch. In this case,

$$\vec{\dot{X}}_1 = (\dot{M}_{n_x}^*)_1 \quad (42c)$$

and $\vec{\dot{X}}$ in the global system can be replaced by $\vec{\dot{M}}_{n_x}$ of the corresponding reduced equation.

6. GLOBAL EQUATIONS

An example of the system of global equations for a particular network was given in Part I of this report. To generalize the results given there on page 47* to multiple species one only has to replace

$$X_j \rightarrow \dot{X}_j ,$$

$$Z_j \rightarrow \dot{Z}_j ,$$

$$(H^*)_i \rightarrow \vec{H}_i^* ,$$

$$(B_{11}^*)_i \rightarrow ([\Sigma^*]_{11})_i ,$$

$$(B_{nn}^*)_i \rightarrow ([\Sigma^*]_{n_x n_x})_i ,$$

where i is the branch index and j is the index for the unknown branch end point flow rate.

* Errata: $-\alpha_{32}$ should be in the third row and second column instead in the third row and third column.

7. SOLUTION STEPS

According to the last two sections MULTI*PADLOC performs the following calculational steps for each time step.

1. Repeat steps 1.1 through 1.6 for each branch of the network.
 - 1.1 Compute the linearization coefficients of B_i , f_i and g_i (see (6), (7), (8)) according to the formulae given in Appendix A.
 - 1.2 Establish the matrices $[a]$, $[b]$, $[\psi]$ and the source rates $\vec{Q}_c$, $\vec{Q}_s$ according to (9) through (13).
 - 1.3 Compute $[D]$, $[G]$ and $\vec{H}$ using (23), (25), and (26).
 - 1.4 Compute the system matrix $[\epsilon]$ using (30), (32), (33).
 - 1.5 Reduce the branch system to the form (39) by computing $[\Sigma^*]_{11}$, $[\Sigma^*]_{n_x n_x}$ and $\vec{H}^*$ according to (40), (41), (42).
 - 1.6 Complement the reduced branch equation (39) by the mixing equation (42) for the upstream end of the branch, thereby establishing the global equation system for the branch-end fission product mass flow rates.
2. Solve the global system for the branch-end fission product mass flow rates.

3. Repeat steps 3.1 through 3.4 for each branch.
 - 3.1 Transform branch-end impurity mass flow rates to channel-end carrier gas concentrations.
 - 3.2 Check convergence of the end-concentrations by comparison with those of last iteration cycle.
 - 3.3 Use (36) to compute carrier gas concentrations at the inner nodal points.
 - 3.4 Use (22) to compute the surface concentrations.
4. If all channel-end concentrations are converged go to step 5. Otherwise go to step 1 and start another iteration cycle.
5. If desired, print results for this time step. Repeat the entire process for the next time step.

8. USER'S MANUAL

8.1 INTRODUCTION

Some improvements to simplify the input to PADLOC have been made since its introduction. These improvements were also adopted for MULTI*PADLOC. Some of them are:

1. A branch can consist of $n_c \geq 1$ parallel identical channels connected at both ends.
2. Time dependent parameters are only read for certain time points. Floating point values are linearly interpolated between input times. Integer values (flags) are kept constant until a new value is read.
3. Time step sizes are read in advance.
4. The sorption pressure can now be the sum of two terms that are nonlinear in the surface concentrations.
5. Initial concentrations, source rates, boundary conditions and sorption parameters are input in terms of μg instead of atoms.

The output has also been improved considerably. Calculated quantities such as carrier gas densities, viscosities, diffusion constants, loop transfer times, etc. have been added for editing. MULTI*PADLOC also has a restart option that is useful for problems that require long running times.

8.2 EXPLANATION OF INPUT DATA

This section explains certain details of the input to MULTI*PADLOC that may not be obvious from the card image descriptions which follow. In particular, time dependent input and sorption parameters are discussed.

8.2.1 Time Dependent Input

8.2.1.1 Time Steps

For the above case there are

$NNDT=3$ time step sizes to cover the calculation time $0 \leq t \leq t_{\text{end}}$
 $NDT(1)=2$ time steps of size $DT(1)$
 $NDT(2)=2$ time steps of size $DT(2)$
 $NDT(3)=4$ time steps of size $DT(3)$

8.2.1.2 All Other Time Dependent Input With the Exception of Boundary Conditions

The time dependence of variable $V_I(t)$ is described by

- the number of input times $MTIN(I) \geq 1$,
- the input times $(TIN(J,I), J=1, MTIN(I))$, and
- the values of the variable $(V(J,I), J=1, MTIN(I))$.

If the variable is of floating point type, the code linearly interpolates for times between input times. For t greater than the maximum input time, it assumes the value of the variable specified for the maximum input time.

Consider, as an example the two floating point variables $V_1(t)$ and $V_3(t)$. For $V_1(t)$ we have four input times, or

$$\begin{aligned}
 \text{MTIN}(1) &= 4 \\
 \text{TIN}(1,1) &= t_1 (= 0)^* & V(1,1) &= f_1 \\
 \text{TIN}(2,1) &= t_2 & V(2,1) &= f_2 \\
 \text{TIN}(3,1) &= t_3 & V(3,1) &= f_3 \\
 \text{TIN}(4,1) &= t_4 & V(4,1) &= f_4, \text{ and}
 \end{aligned}$$

$$V_1(t_1 \leq t \leq t_2) = f_1 + (f_2 - f_1) * (t - t_1) / (t_2 - t_1),$$

$$V_1(t_2 \leq t \leq t_3) = f_2 + (f_3 - f_2) * (t - t_2) / (t_3 - t_2),$$

$$V_1(t_3 \leq t \leq t_4) = f_3 + (f_4 - f_3) * (t - t_3) / (t_4 - t_3),$$

$$V_1(t > t_4) = f_4$$

* $\text{TIN}(1,1)$ is actually not read since it is assumed to be zero.

For variable $V_3(t)$ there is only one input value:

$$MTIN(3) = 1$$

$$TIN(1,3) = t_1 (=0)$$

$$V(1,3) = g_1, \text{ and}$$

$$V_3(t \geq 0) = g_1$$

Integer variables, such as option flags always assume the last input value.
If, e.g., $INT_2(t)$ should behave as in the sketch below,

There are three input values needed.

$$MTIN(2) = 3$$

$$TIN(1,2) = t_1 (=0) \quad INT(1,2) = k_2$$

$$TIN(2,2) = t_2 \quad INT(2,2) = k_1$$

$$TIN(3,2) = t_3 \quad INT(3,2) = k_3, \text{ and}$$

$$\text{INT}_2(t_1 \leq t < t_2) = k_2 \quad ,$$

$$\text{INT}_2(t_2 \leq t < t_3) = k_1 \quad ,$$

$$\text{INT}_2(t \geq t_3) = k_3 \quad .$$

If $\text{INT}_2(t) = \text{constant}$, only one value need be supplied:

$$\text{MTIN}(2) = 1$$

$$\text{TIN}(1,2) = t_1 (=0)$$

$$\text{INT}(1,2) = \text{const}, \quad \text{and} \quad \text{INT}_2(t \geq t_1) = \text{const}.$$

8.2.1.3 Boundary Conditions

The number of required boundary conditions for the network* can, depending on flow conditions, change with time. During each interval in which the number of boundary conditions is a constant, the fission product mass flow rates coming into the global boundary points may also change with time. An example is shown in the sketch on the following page. The number of boundary conditions (number of global boundary points) are the hatched lines. The full lines are the fission product mass flow rates (f_1) coming into the indicated global points IG. Seven input parameters describe the situation.

* Here the original network without broken junctions is meant. If junctions break, additional boundary conditions may have to be supplied. This is explained in the card image input instructions.

- a. $MTIN(7)$ = the total number of time points required to specify the total number of boundary conditions in each interval M .
- b. $MTIN(8)$ = the maximum number of time points required to describe the mass flow rates in any interval M .
- c. $(NGBC(M), M=1, MTIN(7))$ = the total number of boundary conditions in each time interval M .
- d. $(MTINB(M), M=1, MTIN(7))$ = the total number of time points required to describe the mass flow rates in each interval M . $MTIN(8) = \text{MAX}(MTINB(M))$.

- e. $((\text{TINB}(\text{L},\text{M}), \text{L}=1, \text{MTINB}(\text{M})) = \text{the input times (sec) for the mass flow rates in each interval M.}$
 $, \text{M}=1, \text{MTIN}(7))$
- f. $((\text{IGX}(\text{N},\text{M}), \text{N}=1, \text{NGBC}(\text{M})) = \text{the indices of the global points receiving the flow rates.}$
 $, \text{M}=1, \text{MTIN}(7))$
- g. $((\text{XBAFR}(\text{L},\text{M}, \text{IGX}(\text{N},\text{M})) = \text{the fission product mass flow rates (the species index was dropped for convenience).}$
 $, \text{L}=1, \text{MTINB}(\text{M}))$
 $, \text{N}=1, \text{NGBC}(\text{M}))$
 $, \text{M}=1, \text{MTIN}(7))$

The values of these parameters for the example on the previous page are given in the table below.

$$\text{MTIN}(7) = 3$$

$$\text{MTIN}(8) = \text{MAX}(\text{MTINB}(\text{M})) = 5$$

M	NGBC (M)	N	IGX (N,M)	MTINB (M)	L	TINB (L,M)	XBAFR (L,M, IGX (N,M))
1	3	1	20	3	1 2 3	t_1 t_2 t_3	$\begin{matrix} \cdot \\ f_6 \\ f_6 \\ f_6 \\ f_6 \end{matrix}$
		2	1		1 2 3	t_1 t_2 t_3	$\begin{matrix} \cdot \\ f_4 \\ f_5 \\ f_5 \\ f_5 \end{matrix}$
		3	4		1 2 3	t_1 t_2 t_3	$\begin{matrix} \cdot \\ f_1 \\ f_2 \\ f_3 \\ f_3 \end{matrix}$
2	2	1	11	5	1 2 3 4 5	t_3 t_4 t_5 t_6 t_7	$\begin{matrix} \cdot \\ f_{11} \\ f_{11} \\ f_{11} \\ f_{12} \\ f_{13} \end{matrix}$
		2	9		1 2 3 4 5	t_3 t_4 t_5 t_6 t_7	$\begin{matrix} \cdot \\ f_7 \\ f_8 \\ f_7 \\ f_9 \\ f_{10} \end{matrix}$
3	1	1	3	2	1 2	t_7 t_8	$\begin{matrix} \cdot \\ f_{14} \\ f_{15} \end{matrix}$

8.2.2 Sorption Options

MULTI*PADLOC allows seven different sorption options for each chemical element IE in branch IB. The first five options are for regular sorption chosen by the flag NSORPT(IE,IB). The last two are for "adsorption only" and "desorption only" from a given time point on. For the discussion of these options the following nomenclature is assumed:

$T_c(^{\circ}\text{K})$... carrier gas temperature,

$T_s(^{\circ}\text{K})$... surface temperature,

$M_{i\ell}(\text{g/mol})$... molecular mass (i = species index, ℓ = element index),

$S_{i\ell}(\mu\text{g/cm}^2)$... surface concentration,

$S_{\ell}^*(\mu\text{g/cm}^2)$... surface saturation concentration,

$p_{i\ell}(\text{atm})$... sorption pressure above surface,

$B_{i\ell}(\mu\text{g/cm}^3)$... boundary layer concentration above surface.

$R = 82.06 (\text{cm}^3\text{atm})/(\text{mole}^{\circ}\text{K})$... gas constant.

a. NSORPT=1, Linear Sorption with Saturation

$$B_{i\ell}(S_{i\ell}) = k_{i\ell} p_{i\ell}(S_{i\ell}) \quad (43)$$

$$k_{i\ell} = \frac{10^6 M_{i\ell}}{RT_c} \quad , \quad (44)$$

$$p_{i\ell} = \begin{cases} \beta_{\ell} \frac{S_{i\ell}}{S_{\ell}^*} & , \quad S_{i\ell} < S_{\ell}^* \\ \beta_{\ell} & , \quad S_{i\ell} \geq S_{\ell}^* \end{cases} \quad , \quad (45)$$

$$\beta_{\ell} = e^{a_{\ell} + \frac{b_{\ell}}{T_s}} . \quad (46)$$

b. NSORPT=2, Linear or Quasilinear Sorption

$$B_{i\ell}(\vec{S}_{\ell}) = k_{i\ell} p_{i\ell}(\vec{S}_{\ell}) , \quad (47)$$

$$p_{i\ell}(\vec{S}_{\ell}) = \beta_{\ell} \left(\sum_{j=1}^{J_{\ell}} S_{j\ell} \right)^{\gamma_{\ell}-1} S_{i\ell} , \quad (48)$$

$$\gamma_{\ell} = c_{\ell} + \frac{d_{\ell}}{T_s} = \begin{cases} 1 , & \text{linear sorption} \\ \approx 1 , & \text{quasilinear sorption} \end{cases} . \quad (49)$$

The sum $j=1,2,\dots,J_{\ell}$ is over all species that belong to the chemical element ℓ .

c. NSORPT=3, Freundlich Sorption

Same as for NSORPT=2, except that $\gamma_{\ell} > 1$

d. NSORPT=4, Combination of Options NSORPT=2 and NSORPT=3

$$B_{i\ell}(\vec{S}_{\ell}) = k_{i\ell} \left\{ \left[p_{i\ell}(\vec{S}_{\ell}) \right]_2 + \left[p_{i\ell}(\vec{S}_{\ell}) \right]_3 \right\} \quad (50)$$

This option avoids discontinuities of $\partial p_{i\ell} / \partial S_{i\ell}$ as indicated in the sketch on the following page.

The full line represents the experimentally obtained sorption pressure.
Since

$$(p_{i\ell})_3 \ll (p_{i\ell})_2, \quad S < S_{\text{transition}}$$

$$(p_{i\ell})_3 \gg (p_{i\ell})_2, \quad S > S_{\text{transition}},$$

Eq. (50) is a good description of the actual sorption pressure.

e. NSORPT=5

Same as option 4 with the exception that $(p_{i\ell})_2$ is linear in $S_{i\ell}$ and that the transition concentration $S_{i\ell}^T$ is explicitly given.

$$(p_{i\ell})_2 = (\beta_\ell)_2 S_{i\ell}, \quad (51)$$

$$(p_{i\ell})_3 = (\beta_\ell)_3 \left(\sum_{j=1}^{J_\ell} S_{j\ell} \right)^{(\gamma_\ell)_3 - 1} S_{i\ell}. \quad (52)$$

From

$$\left(p_{i\ell} (S^T) \right)_2 = \left(p_{i\ell} (S^T) \right)_3 \quad (53)$$

follows

$$(\beta_\ell)_2 = (\beta_\ell)_3 \left(\sum_{j=1}^{J_\ell} S_{j\ell}^T \right)^{(\gamma_\ell)_3^{-1}} = (\beta_\ell)_3 \left(J_\ell S_\ell^T \right)^{(\gamma_\ell)_3^{-1}}, \quad (54)$$

since all isotopes have the same transition concentration S_ℓ^T . With $(\beta_\ell)_2$ so expressed,

$$p_{i\ell} (S_\ell) = (\beta_\ell)_3 \left[\left(J_\ell S_\ell^T \right)^{(\gamma_\ell)_3^{-1}} + \left(\sum_{j=1}^{J_\ell} S_{j\ell} \right)^{(\gamma_\ell)_3^{-1}} \right] S_{i\ell}. \quad (55)$$

$(\beta_\ell)_3$ and $(\gamma_\ell)_3$ are input in terms of $(a_\ell)_3$, $(b_\ell)_3$, $(c_\ell)_3$, $(d_\ell)_3$ (Eq. (46), (49)). The temperature dependence of the transition concentration,

$$S_\ell^T = e^{\sum_{k=0}^4 \alpha_{\ell k} T_s^k}, \quad (56)$$

is input in terms of the parameters $\alpha_{\ell k}$.

f. Adsorption Only

From a given time point (input) on it is assumed that the surface is a perfect sink, i.e., the boundary layer concentration B_i of Eq. (1) and (2) is set to zero.

g. Desorption Only

From a given time point (input) on, the residence time of fission products on the conduit surface is zero. Any flux coming from the coolant to the surface is instantaneously returned to the coolant. Consequently, the net flux between coolant and surface,

$$h_i [C_i - B_i] = 0 \quad ,$$

which the code accounts for by setting $h_i = 0$. If the surface has a non-zero source rate and/or a non-zero concentration at time t_d at which desorption starts, the fission product masses are transferred to the coolant via an equivalent coolant source rate. Consider first the coolant source due to a surface concentration $S_i(x, t_d) > 0$. The species mass contained in a surface element of length dx is

$$dM_i = S_i(x, t_d) P(x) dx = A(x) dx \int_{t_d}^{t_d + \Delta t} [\dot{q}_c^*(t)]_i dt \quad .$$

Assuming a linear change of $(\dot{q}_c^*)_i$ between $t_d \leq t \leq t_d + \Delta t$ as indicated,

it follows that

$$\dot{\xi}_i = 2 \frac{P(x)}{A(x)} \frac{S_i(x, t_d)}{\Delta t} , \quad (57)$$

which is the coolant source rate applied at $t_d + \Delta t$. From this time on the surface concentration is set to zero. The equivalent coolant source for a surface source follows from

$$[\dot{q}_s(x, t)]_i \, dt \, P(x) \, dx = [\dot{q}_c^{**}(x, t)]_i \, dt \, A(x) \, dx ,$$

or

$$[\dot{q}_c^{**}(x, t)]_i = \frac{P(x)}{A(x)} [\dot{q}_s(x, t)]_i . \quad (58)$$

The total equivalent coolant source for $t \geq t_d + \Delta t$ is thus

$$[\dot{q}_c(x, t)]_{i, \text{equ}} = \frac{P(x)}{A(x)} \left\{ [\dot{q}_s(x, t)]_i + 2 \frac{S_i(x, t)}{\Delta t} \right\} \quad (59)$$

$$S_i(x, t) \begin{cases} > 0 & , \quad t = t_d \\ = 0 & , \quad t > t_d \end{cases} , \quad (60)$$

and Δt is the time step between t_d and the next time point.

8.3 EXECUTION OF MULTI*PADLOC

MULTI*PADLOC may be executed using the following run stream:

VASG,AX	FILE*WRITEREAD//WRITEKEY.	} required if estimated running time $\geq$ 10 min. and/ or if IRSTRT of card 1 is 1.
VUSE	13.,FILE*WRITEREAD.	
VASG,A	MULTI*PADLOC.	
VPRT,T	MULTI*PADLOC. (optional, but should always be used)	
VXQT	MULTI*PADLOC.PADLOC	

Input Data

VPMD,REL (optional)

If the estimated run time is $\geq$ 10 min and the job can not be completed in that time, the results of the last completed time step are printed out and written onto unit 13. A message quoting the time point and real time is also printed. For restart, the only change required in the input data is that IRSTRT=1 (instead of IRSTRT=0 for a regular run).

8.4 CARD IMAGES

Word	1	2	3	4	5	6
Column	1-6	7-12	13-18	19-24	25-30	31-36
Format	I6	I6	I6	I6	I6	I6
Card 1	Number of species. NN>1	Total number of chemical elements to which species belong 1<NE<NN	Number of branches. For a closed loop NB>2, otherwise NB>1	Number of global points 2<NG<NB+1	Max. number of branches connected at any global point 1<NBGP<NB	Max. number of nodal points in any branch. NDMX>3.
Symbol	NN	NE	NB	NG	NBGP	NDMX

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Word	7	8	9	10	11	12
Column	37-42	43-48	49-45	55-60	61-66	67-72
Format	I6	I6	I6	I6	I6	I6
Card 1 (Cont)	Total number of time step sizes. NNDT>1	Number of branches with chemical reactions in coolant. 0<NBCR(1)<NB	Same for the surface 0<NBCR(2)<NB	NDIFF=0	Total number of time points at which junction breaks at global points occur. NDISCT>0	IRSTRT=0 for regular run. IRSTRT=1 if restart values are to be read.
Symbol	NNDT	NBCR(1)	NBCR(2)	NDIFF	NDISCT	IRSTRT

Word	1	2	3	4	5	6
Column	1-6	7-12	13-18	19-24	25-30	31-36
Format	I6	I6	I6	I6	I6	I6
Card 2	Number of time points for input of branch carrier gas mass flow rates. MTIN1>1	Same for flow flags MTIN2>1	Carrier gas pressure MTIN3>1	Gas and surface temperatures MTIN4>1	Gas source rates MTIN5>1	Surface source rates MTIN6>1
Symbol	MTIN1	MTIN2	MTIN3	MTIN4	MTIN5	MTIN6

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Word	7	8				
Column	37-42	43-48				
Format	I6	I6				
Card 2 (Cont)	Same for the number of boundary conditions. The number of boundary conditions may change if flow reversals occur. MTIN7>1	Max. number of time points for input of species mass flow rates at boundary global points. Max, refers to any number of boundary conditions. MTIN8>1				
Symbol	MTIN7	MTIN8				

Word	1	2	etc.			12
Column	1-6					67-72
Format	A6	A6	etc.			A6
Card 3	Alphanumeric Job Description					
Symbol	TITLE1	TITLE2	etc.	etc.	etc.	TITLE12

Use as many cards as required. The last card 3 is a blank card.

04

Word	1	2	3			
Column	1-12	13-24	25-36			
Format	I12	E12.0	E12.0			
Card 4	Number of time steps between printout of results. NEDIT>1	Time integration parameter. Use $\kappa \leq 0.5$ for transients and $\kappa = 1$ if only steady-state results are of interest. 0<KAPPA<1	Absolute value of relative error for convergence of gas concentrations. ECONVGE>0			
Symbol	NEDIT	KAPPA	ECNVGE			

Word	1	2		2*NNDT-1	2*NNDT	
Column	1-12	13-24				
Format	I12	E12.0		I12	E12.0	
Card 5	Number of time steps of size DT(1)	Time step size, sec DT>0		Number of time steps of size DT (NNDT)		
Symbol	NDT(1)>1	DT(1)>0	etc.	NDT(NNDT)>1	DT(NNDT)>0	

Use as many cards
as required.

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Read card 6 in a loop over species I=1,NN

Word	1	2	3	4		
Column	1-6	6-12	13-24	25-36		
Format	A6	I6	E12.0	E12.0		
Card 6	Alphanumeric species name. Start in column 1	Index of chemical element to which species belongs 1<IEL(I)<NN	Decay con- stant (sec ⁻¹) XLAM(I)>0	Molecular mass (g/mol) WMNU(I)>0		
Symbol	XNAME(I)	IEL(I)	XLAM(I)	WMNU(I)		

Skip card 7 if all decay constants are zero or if $NN=1$.
 Otherwise read in a loop $I=1, NN-1$.

Word	1	2		$NN-I$		
Column	1-12	13-24	etc.	etc.		
Format	E12.0	E12.0		E12.0		
Card 7	Decay fraction of species I to I+1	I to I+2	etc.	I to NN		
Symbol	$DCYFR(I, I+1) \geq 0$	$DCYFR(I, I+2) \geq 0$	etc.	$DCYFR(I, NN) \geq 0$		

Use as many cards
 as required for
 each index I.

42

Skip cards 8 through 11 if $NBCR(1) = NBCR(2) = 0$. Otherwise read card 8 in a loop $IR=1, 2$.
 $IR=1 \rightarrow$ carrier gas region, $IR=2 \rightarrow$ surface region.

Word	1	2		$2 \cdot NBCR(I) - 1$	$2 \cdot NBCR(IR)$	
Column	1-3	4-6				
Format	I3	I3				
Card 8	Branch for which there is chemical reaction in region IR.	$IBCR(IR, IB) = 1$		etc.	etc.	
Symbol	IB	$IBCR(IR, IB)$	etc.	IB	$IBCR(IR, IB)$	

Skip card 8 if
 $NBCR(IR) = 0$.

Read the set of cards 9, 10 and 11 in a loop

I=1,NN within the region loop IR=1,2. Skip these cards if NBCR(IR)=0

Word	1	2				
Column	1-12	13-24				
Format	E12.0	E12.0				
Card 9	$k_{i1}^* \begin{matrix} \geq \\ < \end{matrix} 0$ of (A14), page 86.	$k_{i2}^* \begin{matrix} \geq \\ < \end{matrix}$ of (A14), page 86.				
Symbol	RTECO(I,1,IR)	RTECO(I,2,IR)				

Skip if NBCR(IR)=0

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Word	1	2				
Column	1-12	13-24				
Format	E12.0					
Card 10	Activation energy $\left(\frac{\text{cm}^3 \text{atm}}{\text{mol}}\right)$ $Q_{i1} \begin{matrix} \geq \\ < \end{matrix} 0$ of (A14), page 86.	Same for index ℓ_2 . If $\ell_2=1$ skip word 2.	L1=1, L2=2 if both RTECO(I,1,IR)≠0 RTECO(I,2,IR)≠0 If RTECO(I,1,IR)=0 RTECO(I,2,IR)≠0 → L1=2, L2=2 If RTECO(I,1,IR)≠0 RTECO(I,2,IR)=0 → L1=1, L2=1			
Symbol	RTECQ(I,L1,IR)	RTECQ(I,L2,IR)				

Skip if NBCR(IR)=0
or if RTECO(I,1,IR)
= RTECO(I,2,IR)
= 0

Word	1	2	etc.	NN		
Column	1-12	13-24		etc.	etc.	etc.
Format	E12.0	E12.0	etc.	E12.0	E12.0	E12.0
Card 11	Exponent d_{i1l_1} of (A13), page 86.	d_{i2l_1}	Definition of L1 and L2 same as for card 10.	d_{i,n,l_1}	d_{i1l_2} If L2=1	d_{i2l_2} skip
Symbol	EXPCR(I,1, L1,IR)	EXPCR(I,2, L1,IR)	etc.	EXPCR(I,NN, L1,IR)	EXPCR(I,1, L2,IR)	EXPCR(I,2, L2,IR)

Read card 11 only
if card 10 was
read.

4

Word		2*NN				
Column		etc.				
Format		E12.0				
Card 11 (Cont)		d_{inl_2}				
Symbol	etc.	EXPCR(I,NN, L2,IR)				

Word	1	2		NE		
Column	1-3	4-6				
Format	I3	I3	I3	I3		
Card 12	Total number of branches with adsorption only or desorption only for element 1 $0 \leq \text{NBADD}(\text{IE}) \leq \text{NB}$	for element 2		for element NE		
Symbol	NBADD(1)	NBADD(2)		NBADD(NE)

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Skip Card 13 if $\text{MAX}(\text{NBADD}(\text{IE}))=0$. Otherwise read in a loop $\text{IE}=1, \text{NE}$,
 IE is the chemical element index.

Word	1	2	3	$3 * \text{NBADD}(\text{IE}) - 2$	$3 * \text{NBADD}(\text{IE}) - 1$	$3 * \text{NBADD}(\text{IE})$
Column	1-6	6-12	13-24	etc.	etc.	etc.
Format	I6	I6	I12	I6	I6	I12
Card 13	Index of 1st branch with adsorption only or desorption only.	IAD=1, adsorption only. IADD=-1, desorption only.	Time point from which on this should happen. $\text{NTSAD} \geq 1$.	Same information for the $\text{NBADD}(\text{IE})$ branch.		
Symbol	IB	IAD(IE, IB)	NTSAD(IE, IB)	IB	IAD(IE, IB)	NTSAD(IE, IB)

Skip card 13 if
 $\text{NBADD}(\text{IE})=0$

Skip Card 14 if NDISCT=0.

Word	1	2			2*NDISCT	
Column	1-12	13-24		etc.	etc.	
Format	I12	I12		I12	I12	
Card 14	1st time point number at which branch junctions (global points) break.	Total number of global points that break at 1st time point.		Same for the last (NDISCT) time point number.		
Symbol	ITDISC(1) $\geq$ 2	NDISC(1)	etc.	ITDISC(NDISCT) $\geq$ 2	NDISC(NDISCT)	

Branch end global point indices. If there is a closed loop of NLOOP branches within the network, its branches should be numbered consecutively as IB=1,2,3,...NLOOP. The global points should be labeled IBEGIN(IB)=IB, IEND(IB)=IB+1 for IB=1,2,...NLOOP-1. IBEGIN(NLOOP)=NLOOP, IEND(NLOOP)=1. Otherwise the designations are arbitrary.

Word	1	2	2*Nb-1	2*Nb		
Column	1-3	4-6	etc.	etc.		
Format	I3	I3	I3	I3		
Card 15	Index of global point at x=0 of branch IB=1.	Index of global number of x=L of branch IB=1.	Same for branch Nb.			
Symbol	IBEGIN(1)	IEND(1)	IBEGIN(Nb)	IEND(Nb)		

<u>Comments</u>
Max(IBEGIN)=NG
Max(IEND)=NG

Read Card 16 in a loop L=1,6

Input times (sec) for carrier gas mass flow rates (L=1)

Flow Flags (L=2)

Gas Pressure (L=3)

Gas and Surface Temp (L=4)

Gas Source Rate (L=5)

Surface Source Rate (L=6)

Word	1	2		MTINL		
Column	1-12	13-24				
Format	E12.0	E12.0		E12.0		
Card 16						
Symbol	TIN(2,L)>0	TIN(3,L)>0	etc.	TIN(MTINL,L)>0		

Skip card 16 if
MTINL=1

17

Cards 17 through 32 are read in a loop over branches IB=1,NB

Word	1	2	3	12		
Column	1-6	7-12	etc.	67-72		
Format	A6	A6	A6	A6		
Card 17	Alphanumeric branch identification. 72 characters maximum					
Symbol	BRNCHN(1,IB)	BRNCHN(2,IB)		BRNCHN(12,IB)		

Word	1	2	3	4	5	6
Column	1-12	13-24	25-36	37-39	40-42	43-45
Format	E12.0	E12.0	I12	I3	I3	I3
Card 18	Branch length (cm)	1. ENTL=hydrodynamic entry length (cm) if $KGFY \leq 3$ or $KGNFP=0$. (See 27 for $KGFY$ and $KGNFP$). 2. ENTL=BLENGT otherwise.	Number of identical parallel channels in branch IB $NCHAN(IB) \geq 1$.	Number of nodes (including end nodes) for branch. $NNODES(IB) \geq 3$.	Flag for initial gas concentration. INIT=0, C=0 INIT=1, C>0 uniform along branch, INIT=2, $C=a+bx > 0$ along branch, INIT=3 for arbitrary varying C along branch.	Same for initial surface concentration S.
Symbol	BLENGT(IB)	ENTL(IB)	NCHAN(IB)	NODES(IB)	INIT(1,IB)	INIT(2,IB)

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Word	7	8	9			
Column	46-48	49-51	52-54			
Format	I3	I3	I3			
Card 18 (Cont)	=0, if all Δx within branch are of same length. =1, otherwise.	=1 for arbitrary cross section (including circular) =0 for circular cross section whose hydraulic diameter changes linearly along branch.	0 = for uniform conduit geometry along X. =1, otherwise Leave blank if $KCIR(IB)=0$			
Symbol	IX	KCIR(IB)	NHD(IB)

Sorption option indicators for the different chemical elements.

Word	1	2		NE		
Column	1-3	4-6	etc.	etc.		
Format	I3	I3		I3		
Card 19						
Symbol	NSORPT(1,IB)	NSORPT(2,IB)	etc.	NSORPT(NE,IB)		

$1 \leq \text{NSORPT}(\text{IE}, \text{IB}) \leq 5$
see also 8.2.2.

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Cards 20 and 21 are read in a loop over chemical elements (IE=1,NE) within the branch loop IB=1,NB.
Sorption parameters for $[S] = \mu\text{g}/\text{cm}^2$ and $[p] = \text{atm}$.

Word	1	etc.		4 or 8	5 or 9	
Column						
Format						
Card 20	Read the first 4 words for J=J1,J2. J1=J2=1 for NSORPT 1,2,3 J1=1,J2=2 for NSORPT=4 J1=J2=2 for NSORPT=5					
	a_{IE} of (46)	b_{IE} of (46)	c_{IE} of (49)	d_{IE} of (49)	S_{IE}^* of (45)	
Symbol	ACP(J,IE,IB)	BCP(J,IE,IB)	CCP(J,IE,IB)	DCP(J,IE,IB)	SSAT(IE,IB)	

Use as many cards
as required.
See also 8.2.2.

Skip card 21 if NSORPT≠5.

Word	1	2	3	4		
Column	1-12	13-24	25-36	37-48		
Format	E12.0	E12.0	E12.0	E12.0		
Card 21	$\alpha_{IE,1}$ of (56)	$\alpha_{IE,2}$	$\alpha_{IE,3}$	$\alpha_{IE,4}$		
Symbol	TRC(1,IE,IB)	TRC(2,IE,IB)	TRC(3,IE,IB)	TRC(4,IE,IB)		

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[illegible]

Skip Card 22 if IX=0 (of Card 18)
Spatial discretization of branch IB.

Word	1	2	etc.	N1		
Column	1-12	13-24	etc.			
Format	E12.0	E12.0	E12.0	E12.0		
Card 22	= Δx (cm) at the upstream end of the branch.	etc.		= Δx (cm) at the downstream end of branch.		
Symbol	DELX(1,IB)	DELX(2,IB)		DELX(N1,IB)		

Comments

N1 = number of nodes minus 1.

Use as many 6-word cards are required per branch.

51 Skip Card 23 if KCIR(IB)=1 (of Card 18)
Circular pipe with linear change of diameter DHD.

Word	1	2				
Column	1-12	13-24				
Format	E12.0	E12.0				
Card 23	Inner pipe diameter (cm) at upstream end of branch.	Inner pipe diameter (cm) at downstream end of branch. Leave blank for uniform DHD along branch.				
Symbol	DHD(1,IB)	DHD(2,IB)				

Skip Card 24 if KCIR(IB)=0 (of Card 18)
Arbitrary conduit (including circular)

Word	1	2	etc.	2*N-1	2*N	
Column	1-12	13-24	etc.	etc.	etc.	
Format	E12.0	E12.0	E12.0	E12.0	E12.0	
Card 24	Hydraulic diameter (cm) at upstream end of branch. (See Part I, Appendix A for definition).	Cross section area (cm ²) at upstream end of branch.		Hydraulic diameter (cm) at downstream end of branch.	Cross section area (cm ²) at downstream end of branch.	
Symbol	DHD(1,IB)	CROSSA(1,IB)	etc.	DHD(N,IB)	CROSSA(N,IB)	

Comments

$$N = \begin{cases} 1 & \text{if} \\ & \text{NHD(IB)=0} \\ & \text{NODES(IB)} \\ & \text{if} \\ & \text{NHD(IB)=1.} \end{cases}$$

Use as many cards as required per branch.

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Skip Card 25a if INIT(1,IB)=0 (of Card 18). Otherwise read in a loop over species I=1,NN
Initial carrier gas concentration (μg/cm³)

Word	1	2		N		
Column	1-12	13-24		etc.		
Format	E12.0	E12.0		E12.0		
Card 25a	Initial concentration at x=0 of branch IB.			Initial concentration at x=L of branch IB.		
Symbol	C(I,1,IB)	C(I,2,IB)	etc.	C(I,N,IB)		

$$N = \begin{cases} 1, \text{INT}(1, \text{IB})=1 \\ 2, \text{INIT}(1, \text{IB})=2 \\ \text{NODES, INIT} \\ (1, \text{IB})=3 \end{cases}$$

Skip Card 25b if INIT(2,IB)=0 (of Card 18). Otherwise read in a loop over species I=1,NN
Initial surface concentration ($\mu\text{g}/\text{cm}^2$) of species I.

Word	1	2		N		
Column						
Format						
Card 25b	Initial concentration at $x=0$ of branch IB.			Initial concentration at $x=L$ of branch IB.		
Symbol	$S(I,1,IB)$	$S(I,2,IB)$	etc.	$S(I,N,IB)$		

$$N = \begin{cases} 1, \text{INIT}(2,IB)=1 \\ 2, \text{INIT}(2,IB)=2 \\ \text{NODES}, \\ \text{INIT}(2,IB)=3 \end{cases}$$

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Branch Mass Flow Rates (g/sec) of carrier gas for Input Times (TIN(J,1),J=1,MTIN1)

Word	1	2	etc.	MTIN1		
Column	1-12	13-24	etc.	etc.		
Format	E12.0	E12.0	E12.0	E12.0		
Card 26	Mass flow rate for $t=0$	$t>0$	$t>0$	$t>0$		
Symbol	$XBRFR(1,IB)$	$XBRFR(2,IB)$	etc.	$XBRFR(MTIN1,IB)$		

Comments

Use as many cards as required per branch.

Coolant flow condition flags for input times (TIN(J,2),J=1,MTIN2)

Word					2*MTIN2	2*MTIN2+1
Column	1-3	4-6		etc.	etc.	etc.
Format	I3	I3		I3	I3	I3
Card 27	=0, natural convection. 1<KGFY<4, forced convection.	=0, forced convection. 1<KGNV<3, natural convection.		etc.	etc.	=1,KNNY>2 0, in all other cases.
Symbol	KGFY(1,IB)	KGNV(1,IB)	etc.	KGFY(MTIN2,IB)	KGNV(MTIN2,IB)	KGNFP(IB)

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Coolant Pressures (atm) for Input Times (TIN(J,3),J=1,MTIN3)

Word	1	2	etc.	2*MTIN3		
Column	1-12	13-24	etc.	etc.		
Format	E12.0	E12.0	E12.0	E12.0		
Card 28	Pressure at x=0.	Pressure at x=L (Branch length) (leave blank if pressure is uniform along branch).	Pressure for x=0.	Pressure for x=L. (leave blank if pressure is uniform along branch).		
Symbol	XCPRE(1,1,IB)	XCPRE(1,2,IB)	XCPRE(MTIN3,1,IB)	XCPRE(MTIN3,2,IB)		

Comments

Use as many cards as required per branch.

Coolant Temperature (°C) for Input Times (TIN(J,4),J=1,MTIN4)

Word	1	2	etc.	2*MTIN4		
Column	1-12	13-24	etc.	etc.		
Format	E12.0	E12.0	E12.0	E12.0		
Card 29	Coolant temperature at x=0.	Coolant temperature at x=L (leave blank if coolant temperature is uniform along branch).	etc.	etc.		
Symbol	XCTEM(1,1,IB)	XCTEM(1,2,IB)	XCTEM(MTIN4, 1,IB)	XCTEM(MTIN4, 2,IB)		

Comments

Use as many cards as required per branch.

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Wall Temperature (°C) for Input Times (TIN(J,4),J=1,MTIN4)

Word	1	2	etc.	2*MTIN4		
Column	1-12	13-24	etc.	etc.		
Format	E12.0	E12.0	E12.0	E12.0		
Card 30	Wall temperature at x=0 (leave blank if wall temp is to be same as coolant temp).	Wall temperature at x=L (leave blank if word 1 is blank or if wall temp is to be uniform along branch)	etc.	etc.		
Symbol	XWTEM(1,1,IB)	XWTEM(1,2,IB)	XWTEM(MTIN4, 1,IB)	XWTEM(MTIN4, 2,IB)		

Comments

Use as many cards as required per branch.

Source rate flags.

Word	1	2				
Column	1-3	4-6				
Format	I3	I3				
Card 31	JQC=0, QC=0 JQC=1, QC>0 and uniform along branch. JQC=2, QC=a+bx ₀ along branch. JQC=3 for arbitrary varying QC(x).	Same for surface source rates.				
Symbol	JQC(IB)	JQS(IB)				

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[illegible]

Skip Card 32a if JQC(IB)=0, otherwise read in a loop I=1,NN within the loop J=1,MTIN5 within the branch loop.
 Gas source rates ($\mu\text{g}/(\text{cm}^3\text{sec})$) for input times (TIN(J,5),J=1,MTIN5)

Word	1	2		N		
Column	1-12	13-24		etc.		
Format	E12.0	E12.0		E12.0		
Card 32a	Source rate at x=0.			Source rate at x=L.		
Symbol	XQC(J,I,1,IB)	XQC(J,I,2,IB)	etc.	XQC(J,I,N,IB)		

Use as many cards
as required.

$$N = \begin{cases} 1, JQC(IB)=1 \\ 2, JQC(IB)=2 \\ \text{NODES}, JQC(IB) \\ =3 \end{cases}$$

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Skip Card 32b if JQS(IB)=0, otherwise read within the loop I=1,NN within the loop J=1,MTIN6 within the branch loop.
 Surface source rates ($\mu\text{g}/(\text{cm}^2\text{sec})$) for input times (TIN(J,6),J=1,MTIN6)

Word	1	2	etc.	N		
Column	1-12	13-24		etc.		
Format	E12.0	E12.0		E12.0		
Card 32b	Source rate at x=0.			Source rate at x=L.		
Symbol	XQS(J,I,1,IB)	XQS(J,I,2,IB)	etc.	XQS(J,I,N,IB)		

$$N = \begin{cases} 1, JQS(IB)=1 \\ 2, JQS(IB)=2 \\ \text{NODES}, JQS(IB) \\ =3 \end{cases}$$

End of Branch Loop

Read Cards 33 through 37 in a loop over input time intervals MT=1,MTIN7 for the number of boundary conditions in time interval MT.

Word	1					
Column	1-3					
Format	I3					
Card 33	Total number of global points for which incoming species mass flow rates are to be read. 0<NBGCX<NG					
Symbol	NGBCX(MT)					

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Skip Cards 34 through 37 if NGBCX(MT)=0

Word	1					
Column	1-3					
Format	I3					
Card 34	Number of time points required to describe the set of NGBCX(MT) global point species mass flow rates in time interval MT MTINB(MT) ≥ 1					
Symbol	MTINB(MT)					

Skip card 35 if MT=1 and MTINB(MT)=1. Otherwise read (TINB(J,MT),J=M1,M2).

$$M1 = \begin{cases} 2, MT=1 \\ 1, MT>1 \end{cases}$$

$$M2 = MTINB(MT)$$

Times (sec) for which species mass flow rates for the NGBCX(MT) global points in time interval MT are to be supplied.

Word	1	2	etc.	M2		
Column	1-12	13-24	etc.	etc.		
Format	E12.0	E12.0	E12.0	E12.0		
Card 35						
Symbol	TINB(M1,MT)	TINB(M1+1,MT)	etc.	TINB(M2,MT)		

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Read Card 36 in a loop over global points N=1,NGBCX(MT) within the loop MT=1,MTIN7

Word	1					
Column	1-3					
Format	I3					
Card 36	Global point number for which boundary condition is to be supplied $1 \leq IGX(N,MT) \leq NG$					
Symbol	IGX(N,MT)					

Read Card 37 in a loop over species I=1,NN within the global point loop N=1,NGBCX(MT)
 XBAFR = species mass flow rate ($\mu\text{g}/\text{sec}$) coming into global point IG from outside the network.

Word	1	2	etc.			
Column	1-12	13-24	etc.			
Format	E12.0	E12.0	E12.0			
Card 37						
Symbol	XBAFR(1,MT,I, IG)	XBAFR(2,MT,I, IG)	etc.	XBAFR(M2,MT,I, IG)		

IG=IGX(N,MT)
 M2=MTINB(MT)

or

Skip Cards 38 and 39 if NDISCT=0 (Card 1). Otherwise read the set of cards 38, 39 in a loop over global points L=1,NDISC(J) within the loop J=1,NDISCT.

Word	1					
Column	1-12					
Format	I12					
Card 38	New global point number for which boundary condition has to be supplied.					
Symbol	IG					

Species mass flow rates ($\mu\text{g}/\text{sec}$) coming into global point IG from outside the network.

Word	1	2	etc.	NN		
Column	1-12	13-24	etc.	etc.		
Format	E12.0	E12.0	E12.0	E12.0		
Card 39						
Symbol	BAFRTE(1,IG)	BAFRTE(2,IG)	etc.	BAFRTE(NN,IG)		

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[illegible]

9. SAMPLE PROBLEMS

9.1 INTRODUCTION

In this section a few sample problems will be presented. The intention is to verify MULTI*PADLOC by comparison with analytical solutions. For this purpose a few exact solutions based on simplifying assumptions will be derived. These solutions will also give some insight to the general plateout problem. Two cases will be considered. The first case will deal with pure time dependence by assuming all parameters in a closed loop to be uniform. The second one will deal with pure spatial dependence, i.e., steady-state solutions. The comparison of MULTI*PADLOC results with the exact solutions will then be presented in Section 9.3.

9.2 ANALYTIC SOLUTIONS

First we assume linear sorption and linear chemical reactions (Eq. (16) and (17)). Furthermore we assume all parameters to be uniform in time and space. If we then consider only time or spatial dependence of the unknowns $\vec{C}$ and $\vec{S}$, we are led to a general equation of the form

$$\frac{d\vec{Z}(y)}{dy} + [V] \vec{Z}(y) = \vec{W}(y) \quad , \quad (61)$$

$$\vec{W}(y) = \vec{O}_{\Omega} + \sum_{\ell=1}^n \ell_{\Omega}^{\ell} e^{-\omega_{\ell} y} \quad , \quad (62)$$

$$\ell_{\Omega_i}^{\ell} = 0 \quad , \quad \ell > i \quad , \quad i=1,2,\dots,n \quad , \quad (63)$$

(n = the total number of species)

which will be solved before doing specific sample problems.

y in the above equation is either time or a spatial position. $\vec{Z}$ is the unknown (n×1) column vector. $\vec{W}$, $\vec{\Omega}^0$ and $\vec{\Omega}^l$ are (n×1) column vectors, and [V] is an (n×n) lower triangular matrix.

For the homogeneous equation

$$\frac{d\vec{Z}^*(y)}{dt} + [V] \vec{Z}^*(y) = 0 \quad , \quad (64)$$

the trial solution

$$\vec{Z}^*(y) = \vec{E} e^{-\mu t} \quad , \quad (65)$$

leads to the eigenvalue problem

$$([V] - \mu_r [U]) \vec{E}^r = 0 \quad , \quad r=1,2,\dots,n \quad . \quad (66)$$

([U] is an n×n unit matrix)

Since the system matrix of (66) is lower triangular, the eigenvalues are

$$\mu_r = V_{rr} \quad , \quad r=1,2,\dots,n \quad . \quad (67)$$

Consequently the solution of (64) is

$$\vec{Z}^*(y) = \sum_{r=1}^n \vec{E}^r e^{-V_{rr} y} \quad , \quad (68)$$

where the eigenvectors $\vec{E}^r$ are to be determined from (66).

The system matrix of (66) is of rank $n-1$ and one component of $\vec{r}_E$ can be arbitrarily chosen. If we choose the r -th component r_{E_r} for this purpose, we can solve (66) for the rest of the $n-1$ components. Because of the lower triangular nature of the matrix, these components can be obtained recursively, according to

$$r_{E_i} = \begin{cases} 0 & , 1 \leq i < r \\ r_{E_r} = \text{arbitrary} & , i = r \\ \frac{1}{v_{rr} - v_{ii}} \sum_{j=r}^{i-1} v_{ij} r_{E_j} & , r < i \leq n \end{cases} , r=1,2,\dots,n \quad (69)$$

Repetitive substitution for r_{E_j} in the third of these equations leads to

$$r_{E_i} = r_{f_i} r_{E_r} , \quad (70)$$

where

$$r_{f_i} = \begin{cases} 0 & , i < r \\ 1 & , i = r \\ r_{f_i}(V) & , i > r \end{cases} \quad (71)$$

For $n=3$, the eigenvectors are, for example,

$$\vec{1}_E = \begin{pmatrix} 1_{E_1} \\ 1_{E_2} = \frac{1}{v_{11} - v_{22}} v_{21} 1_{E_1} \\ 1_{E_3} = \frac{1}{v_{11} - v_{33}} (v_{31} 1_{E_1} + v_{32} 1_{E_2}) \end{pmatrix} = \begin{pmatrix} 1 \\ 1_{f_2}(V) \\ 1_{f_3}(V) \end{pmatrix} 1_{E_1}$$

$${}^2\vec{E} = \begin{pmatrix} {}^2E_1 = 0 \\ {}^2E_2 \\ {}^2E_3 = \frac{1}{v_{22}-v_{33}} v_{32} {}^2E_2 \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \\ {}^2f_3(v) \end{pmatrix} {}^2E_2 ,$$

$${}^3\vec{E} = \begin{pmatrix} {}^3E_1 = 0 \\ {}^3E_2 = 0 \\ {}^3E_3 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} {}^3E_3 , \quad (72)$$

and

$${}^1f_2(v) = \frac{v_{21}}{v_{11}-v_{22}} \quad (73)$$

$${}^1f_3(v) = \frac{1}{v_{11}-v_{33}} \left(v_{31} + v_{32} \frac{v_{21}}{v_{11}-v_{22}} \right) , \quad {}^2f_3(v) = \frac{v_{32}}{v_{22}-v_{33}} .$$

With (67) and (69), the solution (68) containing the integration constants r_{E_r} is given.

We now turn to the solution of the inhomogeneous equation (61). If we assume all eigenvalues v_{rr} of (68) to be different from all $\omega_{\ell\ell}$ of (62)*; a particular solution of (61) must be of the form

$$\vec{z}^{**}(y) = \vec{o}_A + \sum_{\ell=1}^n \vec{\ell}_A e^{-\omega_{\ell\ell} y} . \quad (74)$$

* This will be the case in the applications.

Substitution for $\vec{Z}(y)$ of (61) with (74) yields the desired equations for ${}^0\vec{A}$ and ${}^\ell\vec{A}$,

$$[V] {}^0\vec{A} = {}^0\vec{\Omega} \quad (75)$$

$$([V] - \omega_{\ell\ell}[U]) {}^\ell\vec{A} = {}^\ell\vec{\Omega} \quad , \quad \ell=1,2,\dots,n \quad (76)$$

The solutions for the components are

$${}^0A_i = \frac{1}{V_{ii}} \left({}^0\Omega_i - \sum_{j=1}^{i-1} V_{ij} {}^0A_j \right) = \sum_{j=1}^i g_{ij} {}^0\Omega_j \quad , \quad (77)$$

$${}^\ell A_i = \begin{cases} \frac{1}{V_{ii} - \omega_{\ell\ell}} \left({}^\ell\Omega_i - \sum_{j=1}^{i-1} V_{ij} {}^\ell A_j \right) = \sum_{j=\ell}^i g_{ij}^* {}^\ell\Omega_j \quad , \quad i \geq \ell \\ 0 \quad , \quad i < \ell \text{ (because of (63))} \end{cases} \quad (78)$$

where

$$g_{ij} = ([V]^{-1})_{ij} \quad , \quad (79)$$

$$g_{ij}^* = \left\{ ([V] - \omega_{\ell\ell}[U])^{-1} \right\}_{ij} \quad . \quad (80)$$

With these coefficients the general solution of (61), (62), (63) is

$$\vec{Z}(y) = \vec{Z}^*(y) + \vec{Z}^{**}(y) = {}^0\vec{A} + \sum_{r=1}^n \left(\vec{r}_E e^{-V_{rr}y} + \vec{r}_A e^{-\omega_{rr}y} \right) \quad . \quad (81)$$

The integration constants follow from the boundary value $\vec{Z}(y_0)$ to be

$$r_{E_r} = e^{V_{rr} y_0} \left\{ Z_r(y_0) - {}^0A_r - \sum_{\ell=1}^r {}^\ell A_r e^{-\omega_{\ell\ell} y_0} - \sum_{\ell=1}^{r-1} {}^\ell f_r(V) {}^\ell E_\ell e^{-V_{\ell\ell} y_0} \right\} \quad (82)$$

For $n=3$, Eq. (81) would read

$$\begin{aligned} \begin{Bmatrix} Z_1(y) \\ Z_2(y) \\ Z_3(y) \end{Bmatrix} &= \begin{Bmatrix} {}^0A_1 \\ {}^0A_2 \\ {}^0A_3 \end{Bmatrix} + \begin{Bmatrix} 1 \\ {}^1f_2(V) \\ {}^1f_3(V) \end{Bmatrix} {}^1E_1 e^{-V_{11} y} + \begin{Bmatrix} 0 \\ 1 \\ {}^2f_3(V) \end{Bmatrix} {}^2E_2 e^{-V_{22} y} + \begin{Bmatrix} 0 \\ 0 \\ 1 \end{Bmatrix} {}^3E_3 e^{-V_{33} y} + \\ &+ \begin{Bmatrix} {}^1A_1 \\ {}^1A_2 \\ {}^1A_3 \end{Bmatrix} e^{-\omega_{11} y} + \begin{Bmatrix} 0 \\ {}^2A_2 \\ {}^2A_3 \end{Bmatrix} e^{-\omega_{22} y} + \begin{Bmatrix} 0 \\ 0 \\ {}^3A_3 \end{Bmatrix} e^{-\omega_{33} y} . \end{aligned} \quad (83)$$

If we now substitute for 1E_1 in $Z_1(y)$, we obtain finally

$$\begin{aligned} Z_1(y) &= {}^0A_1 + \sum_{\ell=1}^{1-1>1} {}^\ell f_1(V) {}^\ell E_\ell e^{-V_{\ell\ell} y} + e^{-V_{11}(y-y_0)} \left\{ Z_1(y_0) - {}^0A_1 - \sum_{\ell=1}^1 {}^\ell A_1 e^{-\omega_{\ell\ell} y_0} - \sum_{\ell=1}^{1-1>1} {}^\ell f_1(V) {}^\ell E_\ell e^{-V_{\ell\ell} y_0} \right\} + \\ &+ \sum_{\ell=1}^1 {}^\ell A_1 e^{-\omega_{\ell\ell} y} , \end{aligned} \quad (84)$$

which for $y=y_0$ obviously reduces to the boundary values $Z_i(y_0)$. Eq. (84) is the solution of (61), (62), (63).

9.2.1 Time Dependence Only

For this case we set the convection term in (1) to zero. If we neglect chemical reactions and assume all parameters to be uniform in time and space, (1) and (2) reduce to

$$\frac{dC_i(t)}{dt} = (\dot{q}_c)_i - \frac{P}{A} h_i [C_i(t) - B_i(t)] + \sum_{j=1}^i a_{ij}^* C_j(t) , \quad (85)$$

$$\frac{dS_i(t)}{dt} = (\dot{q}_s)_i + h_i [C_i(t) - B_i(t)] + \sum_{j=1}^i a_{ij}^* S_j(t) . \quad (86)$$

To solve this coupled systems we will first solve for

$$T_i = C_i + \frac{P}{A} S_i , \quad (87)$$

and then separately for C_i and S_i . For this purpose (86) is multiplied by P/A and added to (85). The result is an equation for the $(n \times 1)$ column vector $\vec{T}$,

$$\frac{d\vec{T}(t)}{dt} - [a^*] \vec{T}(t) = \vec{\dot{q}} , \quad (88)$$

where $[a^*]$ is the $(n \times n)$ lower triangular depletion matrix and the elements of $\vec{\dot{q}}$ are

$$\dot{q}_i = (\dot{q}_c)_i + \frac{P}{A} (\dot{q}_s)_i . \quad (89)$$

The solution of (88), satisfying the initial condition

$$\vec{T}(t=0) = \vec{T}(0) , \quad (90)$$

can be obtained from the solution for our general system (61), (62), (63).
For $n=3$ it is

$$\vec{T}(t) = \begin{Bmatrix} T_1(t) \\ T_2(t) \\ T_3(t) \end{Bmatrix} = \begin{Bmatrix} {}^0A_1 \\ {}^0A_2 \\ {}^0A_3 \end{Bmatrix} + \begin{Bmatrix} 1 \\ {}^1f_2(-a^*) \\ {}^1f_3(-a^*) \end{Bmatrix} {}^1E_1 e^{-\lambda_1 t} + \begin{Bmatrix} 0 \\ 1 \\ {}^2f_3(-a^*) \end{Bmatrix} {}^2E_2 e^{-\lambda_2 t} + \begin{Bmatrix} 0 \\ 0 \\ 1 \end{Bmatrix} {}^3E_3 e^{-\lambda_3 t} \quad (91)$$

where $a_{ii}^* = -\lambda_i$ has been used. In (91)

$${}^0\vec{A} = \begin{Bmatrix} -\frac{1}{a_{11}^*} \dot{q}_1 \\ -\frac{1}{a_{22}^*} (\dot{q}_2 + a_{21}^* {}^0A_1) \\ -\frac{1}{a_{33}^*} (\dot{q}_3 + a_{31}^* {}^0A_1 + a_{32}^* {}^0A_2) \end{Bmatrix} , \quad (92)$$

$${}^1f_2(-a^*) = \frac{a_{21}^*}{a_{11}^* - a_{22}^*} \quad (93)$$

$${}^1f_3(-a^*) = \frac{1}{a_{11}^* - a_{33}^*} \left(a_{31}^* + \frac{a_{32}^* a_{21}^*}{a_{11}^* - a_{22}^*} \right) , \quad {}^2f_3(-a^*) = \frac{a_{32}^*}{a_{22}^* - a_{33}^*} ,$$

$${}^1E_1 = T_1(o) - {}^oA_1, \quad r=1 \quad (94)$$

$${}^rE_r = T_r(o) - {}^oA_r - \sum_{\ell=1}^{r-1} {}^\ell f_r(-a^*) {}^\ell E_\ell, \quad r>1$$

Given $T_i(t)$ we now turn to obtain $C_i(t)$. Assuming the linear sorption relation

$$B_i(S_i) = \psi_{ii}^* S_i, \quad (95)$$

and substituting for S_i via (87), Eq. (85) becomes

$$\frac{dC_i(t)}{dt} + \sum_{j=1}^i v_{ij} C_j(t) = \varepsilon_i(t), \quad i=1,2,\dots,n, \quad (96)$$

where

$$v_{ij} = \delta_{ij} h_j \left(\frac{P}{A} + \psi_{jj}^* \right) - a_{ij}^*, \quad (97)$$

$$\varepsilon_i(t) = (\dot{q}_c)_i + h_i \psi_{ii}^* T_i(t).$$

With the form (91) for T_i , (96) can also be written as

$$\frac{d\vec{C}}{dt} + [v] \vec{C} = \vec{\varepsilon}(t), \quad (98)$$

$$\vec{\varepsilon}(t) = {}^o\vec{\Omega} + \sum_{\ell=1}^n {}^\ell\vec{\Omega} e^{-\lambda_\ell t}, \quad (99)$$

$$o_{\Omega_i} = (\dot{q}_c)_i + h_i \psi_{ii}^* o_{A_i} , \quad (100)$$

$${}^{\ell}\Omega_i = h_i \psi_{ii}^* {}^{\ell}f_i {}^{\ell}E_{\ell} e^{-\lambda_{\ell} t} , \quad (101)$$

with ${}^{\ell}f_i$ defined in (71) and (73).

The solution of (98) satisfying the initial condition $\vec{C}(t=0) = \vec{C}(0)$ can immediately be written down. It is

$$\vec{C}(t) = o_D + \sum_{r=1}^n \left(r_F e^{-v_{rr} t} + r_D e^{-\lambda_r t} \right) , \quad (102)$$

$$o_{D_i} = \frac{1}{v_{ii}} \left(o_{\Omega_i} - \sum_{j=1}^{i-1} v_{ij} o_{D_j} \right) , \quad (103)$$

$$r_{D_i} = \begin{cases} \frac{1}{v_{ii} - \lambda_r} \left(r_{\Omega_i} - \sum_{j=1}^{i-1} v_{ij} r_{D_j} \right) , & i \geq r \\ 0 & , \quad i < r \end{cases} \quad (104)$$

$$r_{F_i} = r_{f_i} r_{F_r} , \quad (105)$$

$$r_{f_i} = \begin{cases} 0 & , \quad i < r \\ 1 & , \quad i = r \\ r_{f_i}(v) & , \quad i > r \end{cases} \quad (106)$$

$$r_{F_r} = c_r(o) - o_{D_r} - \sum_{\ell=1}^r \ell_{D_r} - \sum_{\ell=1}^{r-1} \ell_{f_r}(\nu) \ell_{F_\ell}, \quad (107)$$

and $r_{f_i}(\nu)$ as defined by (73). For $n=3$ species, combination of (102), (105), (106) and (107) yields

$$\begin{aligned} \begin{Bmatrix} c_1(t) \\ c_2(t) \\ c_3(t) \end{Bmatrix} &= \begin{Bmatrix} o_{D_1} \\ o_{D_2} \\ o_{D_3} \end{Bmatrix} + \begin{Bmatrix} c_1(o) - \sum_{\ell=0}^1 \ell_{D_1} \\ \ell_{f_2}(\nu) \ell_{F_1} \\ \ell_{f_3}(\nu) \ell_{F_1} \end{Bmatrix} e^{-\nu_{11}t} + \\ &\quad \begin{Bmatrix} 0 \\ c_2(o) - \sum_{\ell=0}^2 \ell_{D_2} - \ell_{f_2}(\nu) \ell_{F_1} \\ \ell_{f_3}(\nu) \ell_{F_2} \end{Bmatrix} e^{-\nu_{22}t} + \\ &\quad \begin{Bmatrix} 0 \\ 0 \\ c_3(o) - \sum_{\ell=0}^3 \ell_{D_3} - \sum_{\ell=1}^2 \ell_{f_3}(\nu) \ell_{F_\ell} \end{Bmatrix} e^{-\nu_{33}t} + \end{aligned}$$

(Continued)

$$+ \begin{Bmatrix} 1_{D_1} \\ 1_{D_2} \\ 1_{D_3} \end{Bmatrix} e^{-\lambda_1 t} + \begin{Bmatrix} 0 \\ 2_{D_2} \\ 2_{D_3} \end{Bmatrix} e^{-\lambda_2 t} + \begin{Bmatrix} 0 \\ 0 \\ 3_{D_3} \end{Bmatrix} e^{-\lambda_3 t} \quad (108)$$

where

$$v_{ii} = \lambda_i + \left(\frac{P}{A} + \psi_{ii}^* \right) h_i \quad (109)$$

Given $T_i(t)$ and $C_i(t)$, the surface concentration $S_i(t)$ can then be obtained from (87).

9.2.2 Spatial Dependence Only

Under the assumption of negligible chemical reactions and uniform parameters in space and time, Eqs. (1) and (2) reduce to

$$(\dot{q}_c)_i + \sum_{j=1}^i a_{ij}^* C_j(x) - \frac{P}{A} h_i [C_i(x) - B_i(x)] - v \frac{dC_i(x)}{dx} = 0 \quad (110)$$

$$(\dot{q}_s)_i + \sum_{j=1}^i a_{ij}^* S_j(x) + h_i [C_i(x) - B_i(x)] = 0 \quad (111)$$

Here two cases are easily tractable. These are:

- a. Nonlinear sorption with no decay,
- b. Linear sorption with decay.

We will first discuss case a.

9.2.2.1 Nonlinear Sorption for Stable Species

Equations (110) and (111) reduce to

$$(\dot{q}_c)_{i\ell} - \frac{P}{A} h_{i\ell} [C_{i\ell}(x) - B_{i\ell}(\vec{S}_\ell)] - v \frac{dC_{i\ell}(x)}{dx} = 0 \quad , \quad (112)$$

$$(\dot{q}_s)_{i\ell} + h_{i\ell} [C_{i\ell}(x) - B_{i\ell}(\vec{S}_\ell)] = 0 \quad . \quad (113)$$

Note that the chemical element index ℓ has been introduced. The reason for this is that only species j belonging to the same element ℓ are assumed to compete for sorption sites. Combining (112) and (113) we obtain

$$\frac{dC_{i\ell}(x)}{dx} = \frac{1}{v} \left[(\dot{q}_c)_{i\ell} + \frac{P}{A} (\dot{q}_s)_{i\ell} \right] , \quad (114)$$

with the solution

$$C_{i\ell}(x) = C_{i\ell}(0) + \frac{(\dot{q}_c)_{i\ell} + \frac{P}{A} (\dot{q}_s)_{i\ell}}{v} x \quad , \quad (115)$$

$$B_{i\ell}(x) = \frac{(\dot{q}_s)_{i\ell}}{h_{i\ell}} + C_{i\ell}(x) \quad . \quad (116)$$

To solve for the surface concentration $S_{i\ell}$ we assume the familiar Freundlich sorption relation

$$B_{i\ell}(\vec{S}_\ell) = k_{i\ell} \beta_\ell \left(\sum_{j=1}^J S_{j\ell}(x) \right)^{\gamma_\ell - 1} S_{i\ell}(x) \quad , \quad (117)$$

$$k_{i\ell} = \frac{10^6}{RT_c} M_{i\ell} \quad , \quad (118)$$

$$\beta_{\ell} = e^{a_{\ell} + \frac{b_{\ell}}{T_s}} \quad (119)$$

$$\gamma_{\ell} = c_{\ell} + \frac{d_{\ell}}{T_s} > 1, \quad (120)$$

where T_c and T_s are the coolant and surface temperatures ($^{\circ}\text{K}$) and $M_{i\ell}$ (g/mol) is the molecular mass. $S_{i\ell}$ of (117) appears also in the sum over all js. Division of (117) by $k_{i\ell} \beta_{\ell}$ and summation over all isotopes yields

$$\sum_{i=1}^{J_{\ell}} \frac{B_{i\ell}}{k_{i\ell} \beta_{\ell}} = \left(\sum_{j=1}^{J_{\ell}} S_{j\ell} \right)^{\gamma_{\ell}-1} \sum_{i=1}^{J_{\ell}} S_{i\ell} = \left(\sum_{j=1}^{J_{\ell}} S_{j\ell} \right)^{\gamma_{\ell}}, \quad (121)$$

or

$$\left(\sum_{j=1}^{J_{\ell}} S_{j\ell} \right)^{\gamma_{\ell}-1} = \left(\sum_{i=1}^{J_{\ell}} \frac{B_{i\ell}}{k_{i\ell} \beta_{\ell}} \right)^{\frac{\gamma_{\ell}-1}{\gamma_{\ell}}} \quad (122)$$

Substitution with (122) in (117) gives then finally

$$S_{i\ell}(x) = \left(\sum_{j=1}^{J_{\ell}} \frac{B_{j\ell}}{k_{j\ell} \beta_{\ell}} \right)^{-\frac{\gamma_{\ell}-1}{\gamma_{\ell}}} \frac{B_{i\ell}}{k_{i\ell} \beta_{\ell}} \quad (123)$$

where $B_{i\ell}$ is given by (116) and $C_{i\ell}$ by (115). For the case of one isotope ($J_{\ell}=1$), (123) reduces to

$$S_{i\ell}(x) = \left(\frac{B_{i\ell}}{k_{i\ell} \beta_{\ell}} \right)^{\frac{1}{\gamma_{\ell}}}, \quad (124)$$

and for the linear case of $\gamma_\ell=1$, to

$$S_{i\ell}(x) = \frac{B_{i\ell}}{k_{i\ell} \beta_\ell} \quad (125)$$

To assess the influence of competition for sorption sites we compute the ratio $R_{i\ell}$ of the surface concentrations of isotope i for the case of one isotope (Eq. (124)) and J_ℓ isotopes (Eq. (123)),

$$R_{i\ell} = \frac{(S_{i\ell})_1}{(S_{i\ell})_{J_\ell}} = \left(\frac{k_{i\ell} \beta_\ell}{B_{i\ell}} \sum_{j=1}^{J_\ell} \frac{B_{j\ell}}{k_{j\ell} \beta_\ell} \right)^{\frac{\gamma_\ell-1}{\gamma_\ell}} \quad (126)$$

Now

$$\frac{k_{i\ell} \beta_\ell}{B_{i\ell}} \sum_{j=1}^{J_\ell} \frac{B_{j\ell}}{k_{j\ell} \beta_\ell} = \sum_{j=1}^{J_\ell} \frac{k_{i\ell}}{B_{i\ell}} \frac{B_{j\ell}}{k_{j\ell}} = 1 + \frac{k_{i\ell}}{B_{i\ell}} \sum_{j \neq i}^{J_\ell} \frac{B_{j\ell}}{k_{j\ell}} = 1 + q_{i\ell}^2 \quad (127)$$

Thus,

$$R_{i\ell} = \frac{(S_{i\ell})_1}{(S_{i\ell})_{J_\ell}} = (1 + q_{i\ell}^2)^{1 - \frac{1}{\gamma_\ell}} > 1 \quad (128)$$

for $\gamma_\ell > 1$, as expected. The surface concentration of the same isotope i is larger when it is the only isotope than when it has to compete with other isotopes.

9.2.2.2 Linear Sorption with Decay

Here we use the linearized equations (16) and (17)

$$\vec{Q}_c + [\psi] \vec{S} + [a] \vec{C} - v \frac{d\vec{C}}{dx} = 0 \quad , \quad (129)$$

$$\vec{Q}_s + [-h] \vec{C} + [b] \vec{S} = 0 \quad , \quad (130)$$

$\vec{S}$ follows from (130) to be

$$\vec{S} = - [b]^{-1} \left(\vec{\dot{Q}}_s + [h] \vec{C} \right). \quad (131)$$

Substitution in (129) yields the equation for $\vec{C}$

$$\frac{d\vec{C}}{dx} + [\kappa] \vec{C}(x) = \vec{R} \quad (132)$$

$$[\kappa] = \frac{1}{v} \left([\psi] [b]^{-1} [h] - [a] \right) \quad (133)$$

$$\vec{R} = \frac{1}{v} \left(\vec{\dot{Q}}_c - [\psi] [b]^{-1} \vec{\dot{Q}}_s \right) \quad (134)$$

Again, $[\kappa]$ is lower triangular and the solution of (132) satisfying the boundary condition $\vec{C}(x=0) = \vec{C}(0)$ is

$$\vec{C}(x) = \vec{A} + \sum_{r=1}^n \vec{r}_G e^{-\kappa_r x} \quad (135)$$

$$A_i = \frac{1}{\kappa_{ii}} \left(R_i - \sum_{j=1}^{i-1} \kappa_{ij} A_j \right) \quad (136)$$

$$r_{G_i} = r_{f_i} r_{G_r} \quad (137)$$

$$r_{f_i} = \begin{cases} 0 & , i < r \\ 1 & , i = r \\ r_{f_i}(\kappa) & , i > r \end{cases} \quad (138)$$

$$r_{G_r} = c_r(0) - A_r - \sum_{\ell=1}^{r-1} l_{f_r}(\kappa) l_{G_\ell} \quad (139)$$

and $r_{f_i}(\kappa)$ is defined in (73).

For $n=3$ species, (135) can be written as

$$\begin{pmatrix} c_1(x) \\ c_2(x) \\ c_3(x) \end{pmatrix} = \begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix} + \begin{pmatrix} c_1(0) - A_1 \\ l_{f_2}(\kappa) l_{G_1} \\ l_{f_3}(\kappa) l_{G_1} \end{pmatrix} e^{-\kappa_{11} x} +$$

$$\begin{pmatrix} 0 \\ c_2(0) - A_2 - l_{f_2}(\kappa) l_{G_1} \\ l_{f_3}(\kappa) l_{G_2} \end{pmatrix} e^{-\kappa_{22} x} + \begin{pmatrix} 0 \\ 0 \\ c_3(0) - A_3 - \sum_{\ell=1}^2 l_{f_3}(\kappa) l_{G_\ell} \end{pmatrix} e^{-\kappa_{33} x} \quad (140)$$

With $\vec{C}(x)$ given, $\vec{S}(x)$ can then be obtained from Eq. (131).

9.3 TEST CASES

A number of test problems were established and their solutions obtained using the formulae developed in paragraph 9.2. The same problems were also run with MULTI*PADLOC and perfect agreement with the analytical solutions was obtained in each case. The parameters used in these problems do not necessarily reflect real life situations. Rather, they were chosen in such a manner as to assure their clear influence on the results. A few large real life problems for HTGR primary loops* were also run and no inconsistencies in the results were found. These problems involved three coupled nuclides with several sorption options and up to 27 branches with up to 40 nodes per branch. The code has thus passed considerable testing and may be used with confidence.

The three test cases that were solved analytically and compared with the code results involved between three and six coupled nuclides each. The cases are

- Time dependence only, decay coupling, linear sorption,
- Spatial dependence only, no decay, nonlinear sorption coupling,
- Spatial dependence only, decay coupling, linear sorption.

The hand calculations and corresponding MULTI*PADLOC runs are preserved in Ref. 2. Here only the parameters used and the results are given.

9.3.1 Time Dependence, Decay Coupling, Linear Sorption

The following table for nomenclature corresponds to that of paragraph 9.2.1 and the units are those quoted in paragraph 2.

* As depicted on page 9 of Part I of this report.

i	$(\dot{q}_c)_i$	$(\dot{q}_s)_i$	h_i	$\psi_{ii}^* (\text{cm}^{-1})$	λ_i	a_{ij}^*		
						J=1	J=2	J=3
1	10^{-9}	10^{-10}	4.801886	5.788849	5	-5	0	0
2	2×10^{-9}	2×10^{-10}	4.281669	3.441000	10	2	-10	0
3	3×10^{-10}	3×10^{-10}	4.867107	1.729748	15	3	5	-15

In addition, $P/A = 2.5 \text{ cm}^{-1}$ and the initial concentrations were $S_i(x,0) = C_i(x,0) = 0$. From these parameters it follows that

$$\begin{Bmatrix} T_1(t) \\ T_2(t) \\ T_3(t) \end{Bmatrix} = 10^{-10} \left[\begin{Bmatrix} 2.5 \\ 3 \\ 4 \end{Bmatrix} - \begin{Bmatrix} 2.5 \\ 1 \\ 1.25 \end{Bmatrix} e^{-5t} - \begin{Bmatrix} 0 \\ 2 \\ 2 \end{Bmatrix} e^{-10t} - \begin{Bmatrix} 0 \\ 0 \\ 0.75 \end{Bmatrix} e^{-15t} \right]$$

$$\begin{Bmatrix} C_1(t) \\ C_2(t) \\ C_3(t) \end{Bmatrix} = \begin{Bmatrix} 1.774325 \\ 1.897314 \\ 2.205463 \end{Bmatrix} 10^{-10} + \begin{Bmatrix} -28.3500 \\ 9.209603 \\ 4.231904 \end{Bmatrix} 10^{-13} e^{-44.80211t} +$$

$$+ \begin{Bmatrix} 0 \\ -1.512249 \\ 2.471730 \end{Bmatrix} 10^{-11} e^{-38.64571t} - \begin{Bmatrix} 0 \\ 0 \\ 6.528396 \end{Bmatrix} 10^{-11} e^{-35.58664t}$$

$$- 10^{-10} \left[\begin{pmatrix} 1.745975 \\ 0.5969087 \\ 0.6128836 \end{pmatrix} e^{-5t} + \begin{pmatrix} 0 \\ 1.15839 \\ 0.8844337 \end{pmatrix} e^{-10t} + \begin{pmatrix} 0 \\ 0 \\ 0.306711 \end{pmatrix} e^{-15t} \right]$$

9.3.2 Spatial Dependence, No Decay, Nonlinear Sorption Coupling

In the following table the nomenclature is that used in paragraph 9.2.2.1. The units are those quoted in paragraph 2.

i	ℓ	$(\dot{q}_c)_{i\ell}$	$(\dot{q}_s)_{i\ell}$	$h_{i\ell}$	$M_{i\ell}$	β_ℓ	γ_ℓ	$C_i(o)$
1	2	10^{-9}	10^{-10}	4.801886	137	4.889424146×10^5	2.1	2.791320×10^{-14}
2	1	2×10^{-9}	2×10^{-10}	4.821699	90	4.42413392×10^5	2	2.791320×10^{-13}
3	3	3×10^{-9}	3×10^{-10}	4.867107	50	5.403649372×10^5	2.2	5.582640×10^{-14}
4	3	4×10^{-9}	4×10^{-10}	4.850207	60			8.373960×10^{-14}
5	1	5×10^{-9}	5×10^{-10}	4.789832	200			5.582640×10^{-14}
6	3	6×10^{-9}	6×10^{-10}	4.838042	70			8.373960×10^{-13}

In addition, $P/A = 2.5 \text{ cm}^{-1}$ and $v = 1.781808 \times 10^3 \text{ cm sec}^{-1}$. The results for $C_{i\ell}(x)$ and $S_{i\ell}(x)$ for three different x values are given on the following page.

i	ℓ	x (cm)	$C_{i\ell}$	$S_{i\ell}$
1	2	0 237.5 475	2.791320-14 1.666424-10 3.332569-10	4.386326-10 1.248161-09 1.689599-09
2	1	0 237.5 475	2.791320-13 3.335081-10 6.667370-10	1.880243-10 5.635049-10 7.744174-10
3	3	0 237.5 475	5.582640-14 4.998993-10 9.997427-10	1.445396-09 3.955511-09 5.283888-09
4	3	0 237.5 475	8.373960-14 6.665416-10 1.333000-09	1.611768-09 4.396748-09 5.872213-09
5	1	0 237.5 475	5.582640-14 8.331282-10 1.666201-09	2.116244-10 6.339754-10 8.712432-10
6	3	0 237.5 475	8.373960-13 1.000524-09 2.000211-09	2.089390-09 5.658107-09 7.553625-09

9.3.3 Spatial Dependence, Decay Coupling, Linear Sorption

In the following table the nomenclature is that used in paragraph 9.2.2.2. The units are then quoted in paragraph 2.

i	$(\dot{Q}_c)_i$	$(\dot{Q}_s)$	h_i	ψ_{ii} (cm/sec)	λ_i	$C_i(0)$	a_{ij}			b_{ij}		
							j=1	j=2	j=3	j=1	j=2	j=3
1	10^{-9}	10^{-10}	4.801886	6.949347+1	5	3×10^{-10}	-17.00471	0	0	-32.79739	0	0
2	2×10^{-10}	2×10^{-10}	4.281669	4.147867+1	10	2×10^{-10}	2	-22.05425	0	2	-26.59147	0
3	3×10^{-10}	3×10^{-10}	4.867107	2.104717+1	15	1×10^{-10}	3	5	-27.16777	3	5	-23.41887

In addition, $v = 1.781808 \times 10^3 \text{ cm sec}^{-1}$. The solution is

$$\begin{aligned} \begin{Bmatrix} c_1(x) \\ c_2(x) \\ c_3(x) \end{Bmatrix} &= 10^{-10} \left[\begin{Bmatrix} 1.774325 \\ 1.897314 \\ 2.205463 \end{Bmatrix} + \begin{Bmatrix} 1.225675 \\ 0.3909108 \\ 0.4068411 \end{Bmatrix} e^{-3.833258 \times 10^{-3} x} \right. \\ &\quad \left. - \begin{Bmatrix} 0 \\ 0.2882248 \\ 0.2028915 \end{Bmatrix} e^{-8.156395 \times 10^{-3} x} - \begin{Bmatrix} 0 \\ 0 \\ 1.409413 \end{Bmatrix} e^{-1.279238 \times 10^{-2} x} \right] \end{aligned}$$

APPENDIX A

So far we assumed the linearization coefficients of the sorption relation and the chemical reaction sources to be known. These coefficients depend, of course, on the particular functional form chosen for these phenomena. We will first discuss the sorption relations and their linearizations.

A.1 THE SORPTION RELATION

We consider the so-called "general" Freundlich sorption relation, for which*

$$B_i(T_c, T_s, \vec{S}) = k_i(T_c, T_s) G_i(T_s, \vec{S}) \quad , \quad (A1)$$

$$G_i(T_s, \vec{S}) = F_i(T_s, \vec{S}) S_i \quad , \quad (A2)$$

$$F_i(T_s, \vec{S}) = \left(\sum_{j=1}^n S_j \right)^{\gamma_i(T_s)-1} \quad (A3)$$

and the dependence on $\vec{S}$ means dependence on $S_1, S_2, \dots, S_1, \dots, S_n$.

Linearization of G_i about the last solution guess $\vec{S}_0$ gives

$$G_i(\vec{S}) \approx \alpha_i(\vec{S}_0) + \sum_{j=1}^n \beta_{ij}(\vec{S}_0) S_j \quad , \quad (A4)$$

* $T_c(^{\circ}\text{K})$ = coolant temperature, $T_s(^{\circ}\text{K})$ = surface temperature.

$$\beta_{ij}(\vec{S}_o) = \left(\frac{\partial G_i(\vec{S})}{\partial S_j} \right)_{\vec{S}_o}, \quad (A5)$$

$$\alpha_i(\vec{S}_o) = G_i(\vec{S}_o) - \sum_{j=1}^n \beta_{ij}(\vec{S}_o) S_{jo}. \quad (A6)$$

Taking the derivatives yields

$$\alpha_i^*(\vec{S}_o) = (\gamma_i^{-1}) \left(\sum_{j=1}^n S_{jo} \right)^{\gamma_i^{-2}}, \quad (A7)$$

$$\beta_{ij}(\vec{S}_o) = \alpha_i^*(\vec{S}_o) S_{io} + \delta_{ij} F_j(\vec{S}_o), \quad (A8)$$

$$\alpha_i(\vec{S}_o) = - \alpha_i^*(\vec{S}_o) S_{io} \sum_{j=1}^n S_{jo}. \quad (A9)$$

From (A3) it is seen that all $j=1,2,\dots,n$ species compete for the same sorption sites. For practical purposes, however, it is postulated that only species j belonging to the chemical element E to which species i belongs, compete. MULTI*PADLOC therefore considers only j values for which $j \in E$. With α_i and β_{ij} given, the linearized coefficients ϕ_i^* and ψ_{ij}^* of (6) are then,

$$\phi_i^*(\vec{S}_o) = k_i \alpha_i(\vec{S}_o), \quad (A10)$$

$$\psi_{ij}^*(\vec{S}_o) = k_i \beta_{ij}(\vec{S}_o). \quad (A11)$$

A.2 CHEMICAL REACTIONS

MULTI*PADLOC assumes that the function f_i (or g_i) of the governing equations (1), (2) can be expressed as

$$f_i(\vec{C}) = \sum_{\ell=1}^L k_{i\ell}(T) f_{i\ell}^*(\vec{C}) \quad , \quad 1 \leq L \leq 2 \quad , \quad (\text{A12})$$

$$f_{i\ell}^*(\vec{C}) = \prod_{k=1}^n C_k^{d_{ik\ell}} \quad (\text{A13})$$

$$k_{i\ell}(T) = k_{i\ell}^* e^{-\frac{Q_{i\ell}}{RT}} \quad , \quad (\text{A14})$$

where for brevity, the index differentiating the carrier gas and the surface regions, is suppressed. In practice the exponents usually follow the rule

$$d_{i\ell\ell} \begin{cases} = 0 \quad , \quad k_{i\ell}^* > 0 \\ > 0 \quad , \quad k_{i\ell}^* < 0 \end{cases}$$

and whenever

$$d_{ik1} > 0 \rightarrow d_{ik2} = 0$$

$$d_{ik1} = 0 \rightarrow d_{ik2} > 0$$

As an example, consider the reaction

for which the rate equations for the mass concentrations may be written as

$$\frac{dC_{H_2}}{dt} = \beta(T) C_{HI}^2 - \alpha(T) C_{H_2} C_{I_2} \quad , \quad (I=1)$$

$$\frac{dC_{I_2}}{dt} = \frac{M_{I_2}}{M_{H_2}} \beta(T) C_{HI}^2 - \frac{M_{I_2}}{M_{H_2}} \alpha(T) C_{H_2} C_{I_2} \quad , \quad (I=2)$$

$$\frac{dC_{HI}}{dt} = \frac{2M_{HI}}{M_{H_2}} \alpha(T) C_{H_2} C_{I_2} - \frac{2M_{HI}}{M_{H_2}} \beta(T) C_{HI}^2 \quad , \quad (I=3)$$

$$\alpha(T) = \alpha_o e^{-\frac{Q_\alpha}{RT}} \quad , \quad \beta(T) = \beta_o e^{-\frac{Q_\beta}{RT}} \quad ,$$

and where M is the molecular mass. For this system we would have $n=3$, $L=2$,

$$C_1 = C_{H_2} \quad , \quad k_{11}^* = \beta_o \quad , \quad k_{12}^* = -\alpha_o \quad , \quad Q_{11} = Q_\beta \quad , \quad Q_{12} = Q_\alpha$$

$$C_2 = C_{I_2} \quad , \quad k_{21}^* = \frac{M_{I_2}}{M_{H_2}} \beta_o \quad , \quad k_{22}^* = -\frac{M_{I_2}}{M_{H_2}} \alpha_o \quad , \quad Q_{21} = Q_\beta \quad , \quad Q_{22} = Q_\alpha$$

$$C_3 = C_{HI} \quad , \quad k_{31}^* = \frac{2M_{HI}}{M_{H_2}} \alpha_o \quad , \quad k_{32}^* = -\frac{2M_{HI}}{M_{H_2}} \beta_o \quad , \quad Q_{31} = Q_\alpha \quad , \quad Q_{32} = Q_\beta \quad ,$$

$$d_{112} = d_{122} = d_{212} = d_{222} = d_{321} = d_{34} = 1 \quad ,$$

$$d_{131} = d_{231} = d_{332} = 2 \quad .$$

and all exponents d of other subscripts zero.

Now we turn to linearizing (A13). This leads to

$$f_{i\ell}^*(\vec{C}) \approx u_{i\ell}(\vec{C}_o) + \sum_{j=1}^n v_{ij\ell}(\vec{C}_o) c_j, \quad (A15)$$

$$v_{ij\ell}(\vec{C}_o) = \left(\frac{\partial f_{i\ell}^*(\vec{C})}{\partial c_j} \right)_{\vec{C}_o} = \begin{cases} 0 & c_{jo} = 0 \\ f_{i\ell}^*(\vec{C}_o) \frac{d_{ij\ell}}{c_{jo}}, & c_{jo} \neq 0 \end{cases} \quad (A16)$$

$$u_{i\ell}(\vec{C}_o) = f_{i\ell}^*(\vec{C}_o) - \sum_{j=1}^n v_{ij\ell}(\vec{C}_o) c_{jo} = f_{i\ell}^*(\vec{C}_o) \left(1 - \sum_{j=1}^n d_{ij\ell} \right),$$

which is easily verified. It follows that the coefficients ξ_i^* , η_{ij}^* of (A7) are then

$$\xi_i^*(\vec{C}_o) = \sum_{\ell=1}^L k_{i\ell} f_{i\ell}^*(\vec{C}_o) \left(1 - \sum_{k=1}^n d_{ik\ell} \right) \quad (A17)$$

$$\eta_{ij}^* = \begin{cases} 0, & c_{jo} = 0 \\ \frac{1}{c_{jo}} \sum_{\ell=1}^L k_{i\ell} f_{i\ell}^*(\vec{C}_o) d_{ij\ell}, & c_{jo} \neq 0 \end{cases} \quad (A18)$$