# Transient Sorption by Symmetrical Multilaminate Slabs in Well-Stirred Semiinfinite and Finite Baths 

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## Synopsis

Equations describing the sorption kinetics of symmetrical slabs with 2- $N$ laminae each with constant diffusion and partition coefficients in well-stirred semiinfinite and finite baths are presented. Their application is illustrated using slabs with $N=3$.

## INTRODUCTION

Equations have been reported describing transient sorption by homogeneous slabs and by symmetric laminate slabs of two components in well-stirred semiinfinite and finite baths, sometimes imposing constant concentration gradients, with constraints of constant diffusion and partition coefficients in each lamina. ${ }^{1-10}$ This article provides general equations for the transient sorption by symmetrical slabs with $2 N$ laminae each with constant diffusion and partition coefficients in well-stirred semiinfinite and finite baths. Equations for a specific system are obtained directly by inserting the system parameters into the general equations. They are formulated in a manner suitable for computer evaluation as the specific quantities can be obtained by systematic evaluation of determinants and summations and are applicable in the range of moderate to large time. The procedure is illustrated by application to a symmetrical laminate slab with $N=3$.

## DIFFUSION EQUATIONS FOR SEMIINFINITE BATH SYSTEMS

The basic system is a symmetrical laminate slab of $2 N$ laminae in contact with a semiinfinite well-stirred bath, schematically presented and indexed in Figure 1. The system is also equivalent to an $N$-laminate slab with the exposed face of the $N$ th layer rendered impermeable.

The permeate concentration in the semiinfinite bath is a constant, $c^{0}$. The concentration in each lamina prior to exposure to $c^{0}$ is uniform, $C_{j}^{i}$ in lamina $j$, and is related to the initial concentration in the bath, $c^{i}$, by the partition coefficient $K_{j}=C_{j}^{i} / c^{i}$. Equilibrium is maintained at each phase interface described by $K_{1}=C_{1}^{0} / c^{0}$ at $x=x_{0}$ and by $K_{j-1, j}=C_{j-1} / C_{j}$ at $x=x_{j-1}$, for $j=2, \ldots, N$. For the symmetrical laminate, the interface at $x=x_{N}$ is treated as impermeable. Each lamina is also described by a constant diffusion coefficient $D_{j}$ and a thickness $X_{j}=x_{j}-x_{j-1}, j=1, \ldots, N$. The total thickness of the free slab is $L=2 \Sigma_{j=1}^{N} X_{j}$.

The differential equations and boundary conditions are


Fig. 1. Schematic representation of a symmetrical laminate membrane system of $2 N$ laminae in a semiinfinite bath.

$$
\begin{gather*}
\frac{\partial^{2} C_{j}}{\partial x^{2}}=\frac{1}{D_{j}} \frac{\partial C_{j}}{\partial t} \quad x_{j-1}<x<x_{j}, j=1, \ldots, N  \tag{1}\\
C_{1}\left(x_{0}, t\right)=C_{1}^{0} \quad t \geq 0 ; \quad\left(\frac{\partial C_{N}}{\partial x}\right)_{x=x_{N}}=0 \quad t \geq 0  \tag{2}\\
C_{j}(x, 0)=C_{j}^{i} \quad x_{j-1}<x<x_{j}, j=1, \ldots, N  \tag{3}\\
K_{j-1, j}=\frac{K_{j-1}}{K_{j}}=\frac{C_{j-1}}{C_{j}} \quad \mathbf{x}=x_{j-1}, j=2, \ldots, N
\end{gather*}
$$

and

$$
\begin{gather*}
K_{1}=C_{1}^{0} / c^{0} \quad x=x_{0}, t \geq 0  \tag{4}\\
D_{j-1} \frac{\partial C_{j-1}}{\partial x}=D_{j} \frac{\partial C_{j}}{\partial x} \quad x=x_{j-1}, t \geq 0, j=2, \ldots, N \tag{5}
\end{gather*}
$$

Application of the Laplace transform method ${ }^{4}$ using the inversion theorem provides solutions
$C_{j}(x, t)=C_{j}^{0}+\left(C_{1}^{0}-C_{1}^{i}\right) \sum_{n=1}^{\infty} \frac{2\left[A_{n}^{1,3 j-1} Y_{n, j, 2 j-1}(x)+A_{n}^{1,2 j} Y_{n, j, 2 j}(x)\right] e^{-D_{N} \alpha_{N n}^{2} t}}{\alpha_{N n}\left(\frac{\partial|A|}{\partial \alpha_{N}}\right)_{n}}$

$$
\begin{equation*}
j=1, \ldots, N \tag{6}
\end{equation*}
$$

where $C_{j}^{0}$ is the concentration in lamina $j$ in equilibrium with the bath concentration $c^{0}$ and $|A|$ is the determinant of the elements $A_{l k}$ of order $2 N$ generated by applying $j$ in sequence $1, \ldots, N$ as follows:

$$
\begin{gather*}
j=1, \quad l=2 j-1 ; \quad A_{11}=i \sin \alpha_{1} x_{0}, A_{12}=\cos \alpha_{1} x_{0} \\
j=2 \ldots N, \quad l=2 j-2 ; \quad A_{l, l-1}=-i \sin \alpha_{j-1} x_{j-1}, A_{l, l}=-\cos \alpha_{j-1} x_{j-1}, \\
A_{l, l+1}=i K_{j-1, j} \sin \alpha_{j} x_{j-1}, A_{l, l+2}=K_{j-1, j} \cos \alpha_{j} x_{j-1} \quad(7)  \tag{7}\\
j=2 \ldots N, \quad l=2 j-1 ; \quad A_{l, l-2}=\delta_{j-1, \mathrm{j}} \cos \alpha_{j-1} x_{j-1}, \\
A_{l, l-1}=i \delta_{j-1, j} \sin \alpha_{j-1} x_{j-1}, A_{l, l}=-\cos \alpha_{j} x_{j-1}, A_{l, l+1}=-i \sin \alpha_{j} x_{j-1} \\
j=N, \quad l=2 N ; \quad A_{2 N, 2 N-1}=\cos \alpha_{N} x_{N}, A_{2 N, 2 N}=i \sin \alpha_{N} x_{N}
\end{gather*}
$$

and all other $A_{l k}=0$, with $\delta_{j-1, j}=\left(D_{j-1} / D_{j}\right)^{1 / 2}$. The $A^{1,2 j-1}$ and $A^{1,2 j}$ are the cofactors of $A_{1,2 j-1}$ and $A_{1,2 j}$, respectively, and $Y_{j, 2 j-1}(x)=i \sin \alpha_{j} x, Y_{j, 2 j}(x)$ $=\cos \alpha_{j} x, j=1, \ldots, N$.

The $\alpha_{\mathrm{N}}$ are the nonzero positive roots of

$$
\begin{equation*}
|A|=0 \tag{8}
\end{equation*}
$$

indexed as $\alpha_{N n}$, where $\alpha_{j}=\alpha_{N} / \delta_{j N}$.
For the reduced change in the diffusant mass in the membrane, $F(t)$, one obtains

$$
\begin{align*}
& F(t)=\frac{M(t)-M^{0}}{M^{i}-M^{0}} \\
& =\frac{C_{1}^{i}-C_{1}^{0}}{M^{i}-M^{0}} \sum_{n=1}^{\infty} \frac{2 e^{-D_{N} \alpha_{N n}^{2} t}}{\alpha_{N n}\left(\frac{\partial|A|}{\partial \alpha_{N}}\right)_{n}} \cdot \sum_{j=1}^{N} \frac{1}{\alpha_{j n}}\left[i A_{n}^{i, 2 j-1}\left(\cos \alpha_{j n} x_{j}-\cos \alpha_{j n} x_{j-1}\right)\right. \\
& \left.-A_{n}^{1,2 j}\left(\sin \alpha_{j n} x_{j}-\sin \alpha_{j n} x_{j-1}\right)\right] \tag{9}
\end{align*}
$$

where $M^{0}=\sum_{j=1}^{N} C_{j}^{0} X_{j}, M^{i}=\sum_{j=1}^{N} C_{j}^{i} X_{j}$, and $M(t)=\sum_{j=1}^{N} \int_{x_{j-1}}^{x_{j}} C_{j}(x, t) d x$.
In some applications, lamina 1 is a film formed in the bath medium in which the laminate $j=2, \ldots, N$ is placed to initiate sorption, which in this case commences at $x=x_{1}$. The differential equations and boundary conditions are provided in eqs. (1)-(5), with eq. (3) replaced by

$$
\begin{gather*}
C_{1}(x, 0)=C_{1}^{0} \quad x_{0} \leq x<x_{1} \\
C_{j}(x, 0)=C_{j}^{i} \quad x_{j-1} \leq x \leq x_{j}, j=2, \ldots, N \tag{10}
\end{gather*}
$$

The solutions are

$$
\begin{align*}
& C_{j}(x, t)=C_{j}^{0}+\left(C_{1}^{0}-C_{1}^{i}\right) \sum_{n=1}^{\infty} \\
& \times \frac{2\left[A_{n}^{2,2 j-1} Y_{n, j, 2 j-1}(x)+A^{2,2 j} Y_{n, j, 2 j}(x)\right] e^{D_{N \alpha_{N n}}^{2} t}}{\alpha_{N n}\left(\frac{\partial|A|}{\partial \alpha_{N}}\right)_{n}} \quad j=1, \ldots, N \tag{11}
\end{align*}
$$

where $|A|$ is again defined and evaluated as in eq. (7). The reduced change of the diffusant mass in the membrane, $j=2, \ldots, N$, plus in the film, $j=1$, is

$$
\begin{align*}
& F(t)=\frac{C_{1}^{i}-C_{1}^{0}}{M^{i}-M^{0}} \sum_{n=1}^{\infty} \frac{2 e^{-D \alpha_{N n}^{2} t}}{\alpha_{N n}\left(\frac{\partial|A|}{\partial \alpha_{N}}\right)_{n}} \sum_{j=1}^{N} \frac{1}{\alpha_{j n}} \\
& \quad \times x\left[i A_{n}^{2,2 j-1}\left(\cos \alpha_{j n} x_{j}-\cos \alpha_{j n} x_{j-1}\right)-A_{n}^{2,2 j}\left(\sin \alpha_{j n} x_{j}-\sin \alpha_{j n} x_{j-1}\right)\right] \tag{12}
\end{align*}
$$

## DIFFUSION EQUATIONS FOR FINITE BATH SYSTEMS

The basic system is a symmetrical laminate slab of $2 N$ layers in contact with a well-stirred bath of volume $V$ which replaces the semiinfinite bath of the previous system. The initial diffusant concentration in the bath contacting the slab at $x=x_{0}$ is $c^{0}$. The remainder of the system is as described for the system in Figure 1.

The differential equations and boundary conditions are given by eqs. (1)-(5), with eq. (2) replaced by

$$
\begin{equation*}
\frac{\partial C_{1}}{\partial x}=\frac{X_{1}}{D_{1} H_{1}} \cdot \frac{\partial C_{1}}{\partial t}, \quad x=x_{0}, t \geq 0 ;\left(\frac{\partial C_{N}(x, t)}{\partial x}\right)_{x-x_{N}}=0 \quad t \geq 0 \tag{13}
\end{equation*}
$$

where $H_{j}=K_{j} V_{j} / V, j=1, \ldots, N$, is the ratio of the amount of diffusant in lamina $j$ to the amount in $V$ at equilibrium.

Application of the Laplace transform method ${ }^{4}$ provides the solutions

$$
\begin{align*}
C_{j}(x, t)=C_{j}^{i}+\frac{\left(C_{j}^{0}-C_{j}^{i}\right)}{1+\sum_{j=1}^{N} H_{j}}+2\left(C_{1}^{i}-C_{1}^{0}\right) & X_{1} \\
& \times \sum_{n=1}^{\infty} \frac{i\left[A_{n}^{1,2 j-1} Y_{n, j, 2 j-1}(x)+A_{n}^{1,2 j} Y_{n, j, 2 j}(x)\right]}{\delta_{1, N}\left(\frac{\partial|A|}{\partial \alpha_{N}}\right)_{n}} \\
& \quad \times x e^{-D_{N a_{N n}^{2} t}^{2}} \quad j=1, \ldots, N
\end{align*}
$$

where $|A|$ is the determinant generated according to eq. (7), except that $A_{11}$ and $A_{12}$ are replaced by

$$
\begin{gather*}
j=1, l=2 j-1 ; A_{11}=H_{1} \cos \alpha_{1} x_{0}+\alpha_{1} X_{1} \sin \alpha_{1} x_{0} \\
A_{12}=i\left[H_{1} \sin \alpha_{1} x_{0}-\alpha_{1} X_{1} \cos \alpha_{1} x_{0}\right] \tag{15}
\end{gather*}
$$

Evaluating eq. (14) at $x=x_{0}$, using $K_{1}=C_{1} / c$, and rearranging, one obtains for the reduced change in the diffusant concentration in the bath volume $V$

$$
\begin{equation*}
G(t)=\frac{c(t)-c^{f}}{c^{0}-c^{i}}=\sum_{n=1}^{\infty} Z_{n} e^{-D_{N \alpha_{N n}}^{2} t} \tag{16}
\end{equation*}
$$

where

$$
\begin{equation*}
Z_{n}=2 i X_{1} \frac{\left[A_{n}^{11} Y_{n, 11}\left(x_{0}\right)+A_{n}^{12} Y_{n, 12}\left(x_{0}\right)\right]}{\delta_{1, N}\left(\frac{\partial|A|}{\partial \alpha_{N}}\right)_{n}} \tag{17}
\end{equation*}
$$

Other useful reduced concentration changes are given by

$$
\begin{equation*}
S(t)=\frac{c(t)-c^{f}}{c^{f}-c^{i}}=\left(1+\sum_{j=1}^{N} H_{j}\right) G(t) \tag{18}
\end{equation*}
$$

and

$$
\begin{equation*}
T(t)=\frac{c(t)-c^{f}}{c^{0}-c^{f}}=\frac{\left(1+\sum_{j=1}^{N} H_{j}\right)}{\sum_{j=1}^{N} H_{j}} G(t) \tag{19}
\end{equation*}
$$

When lamina 1 is a film formed in the medium in which the slab $j=2, \ldots, N$ is placed to initiate sorption, the boundary conditions are expressed by eqs. (1), (4), (5), (10), and (13), and the solutions are

$$
\begin{align*}
C_{j}(x, t)= & C_{j}^{i}+\frac{C_{j}^{0}-C_{j}^{i}}{1+\sum_{j=1}^{N} H_{j}}+2\left(C_{1}^{i}-C_{1}^{0}\right) X_{1} \\
& \times \sum_{n=1}^{\infty} \frac{i\left[A_{n}^{2,2 j-1} Y_{n, j, 2 j-1}(x)+A_{n}^{2,2 j} Y_{n, j, 2 j}(x)\right]}{\delta_{1, N}\left(\frac{\partial|A|}{\partial \alpha_{N}}\right)_{n}} \times e^{-D_{N \alpha_{N n}}^{2} t} \\
& j=1, \ldots, N \tag{20}
\end{align*}
$$

Evaluating $C\left(x_{0}, t\right)$ and rearranging yields

$$
\begin{equation*}
G(t)=\sum_{n=1}^{\infty} W_{n} e^{-D_{N} \alpha_{N n}^{2} t} \tag{21}
\end{equation*}
$$

where

$$
\begin{equation*}
W_{n}=\frac{2 i X_{1}\left[A_{n}^{21} Y_{n, 11}\left(x_{0}\right)+A_{n}^{22} Y_{n, 12}\left(x_{0}\right)\right]}{\delta_{1, N}\left(\frac{\partial|A|}{\partial \alpha_{N}}\right)_{n}} \tag{22}
\end{equation*}
$$

## APPLICATION TO LAMINATES WITH $\boldsymbol{N}=\mathbf{3}$

The laminate with $N=3$ is used here to illustrate the reduction of general expressions to specific systems.

## Semiinfinite Bath

The determinant generated according to eq. (7) with $N=3$ is
$|A|=$
$\left|\begin{array}{llllll}i \sin \alpha_{1} x_{0} & \cos \alpha_{1} x_{0} & 0 & 0 & 0 & 0 \\ -i \sin & -\cos & i K_{12} \sin & K_{12} \cos & 0 & 0 \\ \alpha_{1} x_{1} & \alpha_{1} x_{1} & \alpha_{2} x_{1} & \alpha_{2} x_{1} & & 0 \\ \delta_{12} \cos & i \delta_{2} \sin & -\cos & -i \sin & 0 & 0 \\ \alpha_{1} x_{1} & \alpha_{1} x_{1} & \alpha_{2} x_{1} & \alpha_{2} x_{1} & i K_{23} \sin & K_{23} \cos \\ 0 & 0 & -i \sin & -\cos & \alpha_{2} x_{2} & \alpha_{3} x_{2} \\ 0 & 0 & \alpha_{2} x_{2} & \alpha_{3} x_{2} \\ 0 & 0 & \delta_{23} \cos & i \delta_{23} \sin & -\cos & -i \sin \\ 0 & 0 & 0 & \alpha_{2} x_{2} & \alpha_{2} x_{2} & \alpha_{3} x_{2} \\ \alpha_{3} x_{2} \\ 0 & 0 & & & \cos \alpha_{3} x_{3} & i \sin \alpha_{3} x_{3}\end{array}\right|$
which reduces to

$$
\begin{align*}
& |A|=\sin \alpha_{1} X_{1} \cos \alpha_{2} X_{2} \sin \alpha_{3} X_{3} \\
& \quad+\delta_{23} K_{23} \sin \alpha_{1} X_{1} \sin \alpha_{2} X_{2} \cos \alpha_{3} X_{3}+\delta_{12} K_{12} \cos \alpha_{1} X_{1} \sin \alpha_{2} X_{2} \sin \alpha_{3} X_{3} \\
& \quad-\delta_{13} K_{13} \cos \alpha_{1} X_{1} \cos \alpha_{2} X_{2} \cos \alpha_{3} X_{3} \tag{24}
\end{align*}
$$

The $\alpha_{j n}$ are determined from $|A|=0$, and $\left(\partial|A| / \partial \alpha_{3}\right)_{n}$ becomes

$$
\begin{align*}
&\left(\frac{\partial|A|}{\partial \alpha_{3}}\right)_{n}=\left(\frac{X_{1}}{\delta_{13}}+\frac{\delta_{12} K_{12} X_{2}}{\delta_{23}}+\delta_{13} K_{13} X_{3}\right) \cos \alpha_{1 n} X_{1} \cos \alpha_{2 n} X_{2} \sin \alpha_{3 n} X_{3} \\
&+\left(\frac{\delta_{23} K_{23} X_{1}}{\delta_{13}}+\frac{\delta_{13} K_{13} X_{2}}{\delta_{23}}+\delta_{2} K_{12} X_{3}\right) \cos \alpha_{1 n} X_{1} \sin \alpha_{2 n} X_{2} \cos \alpha_{3 n} X_{3} \\
&-\left(\frac{\delta_{12} K_{12} X_{1}}{\delta_{13}}+\frac{X_{2}}{\delta_{23}}+\delta_{23} K_{23} X_{3}\right) \sin \alpha_{1 n} X_{1} \sin \alpha_{2 n} X_{2} \sin \alpha_{3 n} X_{3} \\
&+\left(\frac{\delta_{12} K_{13} X_{1}}{\delta_{13}}+K_{23} X_{2}+X_{3}\right) \sin \alpha_{1 n} X_{1} \cos \alpha_{2 n} X_{2} \cos \alpha_{3 n} X_{3} \tag{25}
\end{align*}
$$

and the necessary cofactors are

$$
\begin{aligned}
& A_{n}^{11}=i\left[\cos \alpha_{1 n} x_{1} \cos \alpha_{2 n} X_{2} \sin \alpha_{3 n} X_{3}\right. \\
& \quad+\delta_{23} K_{23} \cos \alpha_{1 n} x_{1} \sin \alpha_{2 n} X_{2} \cos \alpha_{3 n} X_{3} \\
& \quad-\delta_{12} K_{12} \sin \alpha_{1 n} x_{1} \sin \alpha_{2 n} X_{2} \sin \alpha_{3 n} X_{3} \\
& \left.\quad+\delta_{13} K_{13} \sin \alpha_{1 n} x_{1} \cos \alpha_{2 n} X_{2} \cos \alpha_{3 n} X_{3}\right]
\end{aligned}
$$

$$
\begin{gather*}
A_{n}^{12}=\sin \alpha_{1 n} x_{1} \cos \alpha_{2 n} X_{2} \sin \alpha_{3 n} X_{3}+\delta_{23} K_{23} \sin \alpha_{1 n} x_{1} \sin \alpha_{2 n} X_{2} \cos \alpha_{3 n} X_{3} \\
\quad+\delta_{12} K_{12} \cos \alpha_{1 n} x_{1} \sin \alpha_{2 n} X_{2} \sin \alpha_{3 n} X_{3} \\
\quad-\delta_{13} K_{13} \cos \alpha_{1 n} x_{1} \cos \alpha_{2 n} X_{2} \cos \alpha_{3 n} X_{3} \\
A_{n}^{13}=i \delta_{12}\left[\cos \alpha_{2 n} x_{2} \sin \alpha_{3 n} X_{3}+\delta_{23} K_{23} \sin \alpha_{2 n} x_{2} \cos \alpha_{3 n} X_{3}\right] \\
A_{n}^{14}=\delta_{12}\left[\sin \alpha_{2 n} x_{2} \sin \alpha_{3 n} X_{3}-\delta_{23} K_{23} \cos \alpha_{2 n} x_{2} \cos \alpha_{3 n} X_{3}\right] \\
A_{n}^{15}=i \delta_{13} \sin \alpha_{3 n} x_{3}  \tag{26}\\
A_{n}^{16}=-\delta_{13} \cos \alpha_{3 n} x_{3}
\end{gather*}
$$

Substituting eqs. (25) and (26) into eq. (9) and using the relation

$$
\begin{equation*}
\frac{\mathrm{C}_{1}^{i}-\mathrm{C}_{1}^{0}}{M^{i}-M^{0}}=\frac{K_{13}}{\left(K_{13} \lambda_{13}+K_{23} \lambda_{23}+1\right) X_{3}} \tag{27}
\end{equation*}
$$

yields

$$
\begin{equation*}
F(t)=\frac{2 \delta_{13} K_{13}}{K_{13} \lambda_{13}+K_{23} \lambda_{23}+1} \sum_{n=1}^{\infty} U_{n} \mathrm{e}^{-D_{3} \alpha_{3 n}^{2} t} \tag{28}
\end{equation*}
$$

where $X_{1}=\lambda_{13} X_{3}$ and $X_{2}=\lambda_{23} X_{3}$ and the $U_{n}$ are given by $U_{n}=\left(\tan \alpha_{3 n} X_{3}+\delta_{23} K_{23} \tan \alpha_{2 n} X_{2}-\delta_{12} K_{12} \tan \alpha_{1 n} X_{1} \tan \alpha_{2 n} X_{2} \tan \alpha_{3 n} X_{3}\right.$ $\left.+\delta_{13} K_{13} \tan \alpha_{1 n} X_{1}\right) / \alpha_{3 n}^{2} X_{3}\left[\left(X_{1} / \delta_{13}+\delta_{12} K_{12} X_{2} / \delta_{23}+\delta_{13} K_{13} X_{3}\right) \tan \alpha_{3 n} X_{3}\right.$

$$
\begin{array}{r}
+\left(\frac{\delta_{23} K_{23} X_{1}}{\delta_{13}}+\frac{\delta_{13} K_{13} X_{2}}{\delta_{23}}+\delta_{12} K_{12} X_{3}\right) \tan \alpha_{2 n} X_{2} \\
-\left(\frac{\delta_{12} K_{12} X_{1}}{\delta_{13}}+\frac{X_{2}}{\delta_{23}}+\delta_{23} K_{23} X_{3}\right) \tan \alpha_{1 n} X_{1} \tan \alpha_{2 n} X_{2} \tan \alpha_{3 n} X_{3} \\
\left.\quad+\left(K_{13} X_{1}+K_{23} X_{2}+X_{3}\right) \tan \alpha_{1 n} X_{1}\right] \tag{29}
\end{array}
$$

It is convenient to use $\alpha_{3 n} X_{3}=R_{n}, \alpha_{2 n} X_{2}=\lambda_{23} R_{n} / \delta_{23}$ and $\alpha_{1 n} X_{1}=\lambda_{13} R_{n} / \delta_{13}$ to give

$$
\begin{equation*}
F(t)=\frac{2 \delta_{13} K_{13}}{\lambda_{13} K_{13}+\lambda_{23} K_{23}+1} \sum_{n=1}^{\infty} Z_{n} e^{-D_{3} R_{n}^{2} t / X_{3}^{2}} \tag{30}
\end{equation*}
$$

where

$$
\begin{align*}
& Z_{n}=\left[\tan R_{n}+\delta_{23} K_{23} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}}-\delta_{12} K_{12} \tan \frac{\lambda_{13} R_{n}}{\delta_{13}} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}} \tan R_{n}\right. \\
& \left.+\delta_{13} K_{13} \tan \frac{\lambda_{13} R_{n}}{\delta_{13}}\right] / R_{n}^{2}\left[\left(\frac{\lambda_{13}}{\delta_{13}}+\frac{\delta_{12} K_{12} \lambda_{23}}{\delta_{23}}+\delta_{23} K_{23}\right) \tan R_{n}\right. \\
& \quad+\left(\frac{\delta_{23} K_{23} \lambda_{13}}{\delta_{13}}+\frac{\delta_{13} K_{13} \lambda_{23}}{\delta_{23}}+\delta_{12} K_{12}\right) \tan \frac{\lambda_{23} R_{n}}{\delta_{23}}  \tag{31}\\
& \quad-\left(\frac{\delta_{12} K_{12} \lambda_{13}}{\delta_{13}}+\frac{\lambda_{23}}{\delta_{23}}+\delta_{23} K_{23}\right) \tan \frac{\lambda_{13} R_{n}}{\delta_{13}} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}} \tan R_{n} \\
& \\
& \\
& \left.+\left(K_{13} \lambda_{13}+K_{23} \lambda_{23}+1\right) \tan \frac{\lambda_{13} R_{n}}{\delta_{13}}\right]
\end{align*}
$$

The $R_{n}$ are the nonzero positive roots of

$$
\begin{align*}
{\left[\tan R_{n}+\delta_{23} K_{23} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}}\right] } & \tan \frac{\lambda_{13} R_{n}}{\delta_{13}} \\
& +\delta_{12} K_{12}\left[\tan R_{n} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}}-\delta_{23} K_{23}\right]=0 \tag{32}
\end{align*}
$$

## Finite Bath

The determinant $|A|$ for the system with $N=3$ generated according to eq. (7) is given by eq. (23), with terms $A_{11}$ and $A_{12}$ replaced by the expressions provided in eq. (15). The determinant reduces to

$$
\begin{align*}
& |A|=i\left\{\left[H_{1} \cos \alpha_{1 n} X_{1}-\alpha_{1 n} X_{1} \sin \alpha_{1 n} X_{1}\right]\right. \\
& \quad \times\left[\cos \alpha_{2 n} X_{2} \sin \alpha_{3 n} X_{3}+\delta_{23} K_{23} \sin \alpha_{2 n} X_{2} \cos \alpha_{3 n} X_{3}\right] \\
& \quad \quad+\delta_{12} K_{12}\left[H_{1} \sin \alpha_{1 n} X_{1}+\alpha_{1 n} X_{1} \cos \alpha_{1 n} X_{1}\right] \\
& \left.\quad \times\left[\sin \alpha_{2 n} X_{2} \sin \alpha_{3 n} X_{3}+\delta_{23} K_{23} \cos \alpha_{2 n} X_{2} \cos \alpha_{3 n} X_{3}\right]\right\} \tag{33}
\end{align*}
$$

The $\alpha_{j n}$ are determined from $|A|=0$. Finally, following eq. (16) using $R_{n}=$ $\alpha_{3 n} X_{3}$,

$$
\begin{equation*}
G(t)=\sum_{n=1}^{\infty} Z_{n} \mathrm{e}^{-D_{3} R_{n}^{2} t / X_{3}^{2}} \tag{34}
\end{equation*}
$$

where

$$
\begin{align*}
Z_{n}=2 \lambda_{13}\left(\tan \frac{\lambda_{13} R_{n}}{\delta_{13}} \tan R_{n}\right. & +\delta_{23} K_{23} \tan \frac{\lambda_{13} R_{n}}{\delta_{13}} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}} \\
& \left.+\delta_{12} K_{12} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}} \tan R_{n}-\delta_{13} K_{13}\right) / L_{n} \tag{35}
\end{align*}
$$

and

$$
\begin{aligned}
L_{n}=\delta_{13}\{ & -\left[\left(1+H_{1}\right) \frac{\lambda_{13}}{\delta_{13}}+H_{1} \delta_{12} K_{12} \frac{\lambda_{23}}{\delta_{23}}+H_{1} \delta_{13} K_{13}\right] \\
& \times \tan \frac{\lambda_{13} R_{n}}{\delta_{13}} \tan R_{n}-\left[\left(1+H_{1}\right) \delta_{23} K_{23} \frac{\lambda_{13}}{\delta_{13}}+H_{1} \delta_{13} K_{13} \frac{\lambda_{23}}{\delta_{23}}\right.
\end{aligned}
$$

$$
\begin{align*}
& \left.+H_{1} \delta_{12} K_{12}\right] \tan \frac{\lambda_{13} R_{n}}{\delta_{13}} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}} \\
& -\left[\left(1+H_{1}\right) \delta_{12} K_{12} \frac{\lambda_{13}}{\delta_{13}}+H_{1} \frac{\lambda_{23}}{\delta_{23}}+H_{1} \delta_{23} K_{23}\right] \tan \frac{\lambda_{23} R_{n}}{\delta_{23}} \tan R_{n} \\
& +\left[\left(1+H_{1}\right) K_{13} \lambda_{13}+H_{1} K_{23} \lambda_{23}+H_{1}\right]-\left[\frac{\lambda_{13} R_{n}}{\delta_{13}}\right]\left[\left(\frac{\lambda_{13}}{\delta_{13}}+\delta_{12} K_{12} \frac{\lambda_{23}}{\delta_{23}}\right.\right.  \tag{36}\\
& \left.+\delta_{13} K_{13}\right) \tan R_{n}+\left(\delta_{23} K_{23} \frac{\lambda_{13}}{\delta_{13}}+\delta_{13} K_{13} \frac{\lambda_{23}}{\delta_{23}}+\delta_{12} K_{12}\right) \tan \frac{\lambda_{23} R_{n}}{\delta_{23}} \\
& \quad+\left(K_{13} \lambda_{13}+K_{23} \lambda_{23}+1\right) \tan \frac{\lambda_{13} R_{n}}{\delta_{13}}-\left(\delta_{12} K_{12} \frac{\lambda_{13}}{\delta_{13}}+\frac{\lambda_{23}}{\delta_{23}}\right. \\
& \left.\left.\left.\quad+\delta_{23} K_{23}\right) \tan \frac{\lambda_{13} R_{n}}{\delta_{13}} \tan \frac{\lambda_{23} R_{n}}{\delta_{23}} \tan R_{n}\right]\right\}
\end{align*}
$$

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