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ORIGINAL PAPER

Dynamic behaviour of electrochemical reactors for a step change in inlet concentration under potentiostatic control according to the dispersion model

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Abstract The dynamic modelling of electrochemical reactors under potentiostatic control is performed according to the dispersion model. The response of the system in concentration and current is discussed for a step change in the inlet concentration in terms of the Peclet number. The stirred tank and plug flow behaviours are analyzed as limiting cases of the dispersion model.

Keywords Dynamic behaviour · Electrochemical engineering · Stability analysis

Nomenclature

a_e	Specific surface area (m^{-1})
A	Magnitude of step change in the inlet concentration ($mol\ m^{-3}$)
B_n	Constant given by Eq. 35 ($mol\ m^{-3}$)
C	Concentration ($mol\ m^{-3}$)
C_0	Inlet concentration ($mol\ m^{-3}$)
C_f	Outlet concentration ($mol\ m^{-3}$)
C_s	Steady-state concentration ($mol\ m^{-3}$)
C_s^{bp}	Steady-state concentration before perturbation ($mol\ m^{-3}$)
C_t	Transient concentration ($mol\ m^{-3}$)
Da	Damköhler number
D_L	Dispersion coefficient ($m^2\ s^{-1}$)
F	Faraday constant ($A\ s\ mol^{-1}$)
H	Heaviside shifting function
I	Total current (A)
I_s	Steady-state current (A)
I_t	Transient current (A)
j	Current density ($A\ m^{-2}$)
k	Kinetic constant ($m\ s^{-1}$)
k_f	Rate constant ($m\ s^{-1}$)
L	Electrode length (m)
m	Constant given by Eq. 24
n	Index number in the series
p	Constant
Pe	Peclet number = $vL/\varepsilon D_L$
t	Time (s)
T	Dimensionless time defined by Eq. 7
u	Function defined by Eq. 22 ($mol\ m^{-3}$)
v	Superficial liquid flow velocity ($m\ s^{-1}$)
W	Electrode width (m)
x	Axial coordinate (m)
y	Normalized axial coordinate given by Eq. 4
Y	Perturbation variable for outlet concentration ($mol\ m^{-3}$)

Greek characters

α	Constant given by Eq. 16
β	Constant given by Eq. 12
ε	Porosity
λ_n	Roots of Eq. 33
ν_e	Charge number of the electrode reaction
τ	Residence time (s)
Υ	Function defined by Eq. 31
Ψ	Function defined by Eq. 30

1 Introduction

The dispersion model is recognized as an appropriate way to design electrochemical reactors [1, 2] from which the

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behaviour of the idealised models, stirred tank and plug flow, can be obtained as limiting cases.

However, very little work has been done on the mathematical modelling of the dynamic behaviour of electrochemical reactors taking into account the dispersion model. Thus, Yeo and Fahidy [3] reported an analytical model with axial dispersion for steady-state and transient response in electrolysers assuming a constant mean current density along the electrodes. In [4] a more rigorous treatment of electrolysers with axial dispersion is treated using a numerical procedure. Nguyen et al. [5] presented a comprehensive mathematical treatment to determine the time behaviour in parallel plate electrochemical reactors. It was concluded that the effects of axial diffusion and axial migration should be taken into account when the aspect ratio is higher than 0.5.

In a previous paper [6] the dynamic behaviour of electrochemical reactors for a step change in the inlet concentration was reported, taking into consideration a cascade of continuous stirred tank electrochemical reactors. The aim of this work is to study the temporal variation in the outlet concentration and current of an electrochemical reactor under potentiostatic control for a step change in the inlet concentration according to the dispersion model.

2 Mathematical modelling

2.1 Concentration as a function of time

The temporal behaviour of an electrochemical reactor according to the dispersion model is given by

$$\tau\frac{\partial C(t,y)}{\partial t}=\frac{1}{\mathrm{Pe}}\frac{\partial^2 C(t,y)}{\partial y^2}-\frac{\partial C(t,y)}{\partial y}-\frac{j(t,y)a_{\mathrm{e}}L}{\nu\nu_{\mathrm{e}}F} \tag{1}$$

where

$$\tau=\frac{\varepsilon L}{v} \tag{2}$$

$$\mathrm{Pe}=\frac{vL}{\varepsilon D_{\mathrm{L}}} \tag{3}$$

and

$$y=\frac{x}{L} \tag{4}$$

Under potentiostatic operation the kinetics of a first order reaction at high electrode potential is given by [7]

$$j(t,y)=\nu_{\mathrm{e}}FkC(t,y) \tag{5}$$

where

$$k=\frac{k_{\mathrm{f}}}{1+\mathrm{Da}} \tag{6}$$

High values of the Damköhler number indicate an approach to mass transport control.

Defining a dimensionless time as

$$T=\frac{t}{\tau} \tag{7}$$

and introducing Eqs. 5–7 into Eq. 1 and rearranging results in

$$\frac{\partial C(T,y)}{\partial T}=\frac{1}{\mathrm{Pe}}\frac{\partial^2 C(T,y)}{\partial y^2}-\frac{\partial C(T,y)}{\partial y}-\beta C(T,y) \tag{8}$$

with the following initial and boundary conditions

$$T=0 \quad C(0,y)=C_{\mathrm{s}}^{\mathrm{bp}}(y) \tag{9}$$

$$y=0 \quad C_0+A=C(T,0)-\frac{1}{\mathrm{Pe}}\left.\frac{\partial C(T,y)}{\partial y}\right|_{y=0} \tag{10}$$

$$y=1 \quad \left.\frac{\partial C(T,y)}{\partial y}\right|_{y=1}=0 \tag{11}$$

being

$$\beta=\frac{ka_{\mathrm{e}}\tau}{\varepsilon} \tag{12}$$

Thus, β is a dimensionless number which lumps the electrochemical kinetics, k, electrode properties, a_{e} and ε, with the reactor residence time.

The solution of Eq. 8 will be assumed as the sum of a transient and a steady-state portion, thus

$$C(T,y)=C_{\mathrm{t}}(T,y)+C_{\mathrm{s}}(y) \tag{13}$$

Introducing Eq. 13 into 8 yields for the steady-state

$$\frac{1}{\mathrm{Pe}}\frac{\mathrm{d}^2 C_{\mathrm{s}}(y)}{\mathrm{d}y^2}-\frac{\mathrm{d}C_{\mathrm{s}}(y)}{\mathrm{d}y}-\beta C_{\mathrm{s}}(y)=0 \tag{14}$$

The solution of Eq. 14 with the boundary conditions 10 and 11 is [1]

$$\frac{C_{\mathrm{s}}(y)}{C_0+A}=\frac{2(\alpha+1)\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}(1-y)}+2(\alpha-1)\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}(1-y)}}{(\alpha+1)^2\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}}-(\alpha-1)^2\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}}}\mathrm{e}^{\frac{\mathrm{Pe}}{2}y} \tag{15}$$

where

$$\alpha=\sqrt{1+\frac{4\beta}{\mathrm{Pe}}} \tag{16}$$

The outlet concentration is given by [2]

$$\frac{C_{\mathrm{f}}}{C_0+A}=\frac{4\alpha \mathrm{e}^{\frac{\mathrm{Pe}}{2}}}{(\alpha+1)^2\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}}-(\alpha-1)^2\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}}} \tag{17}$$

which was obtained by evaluating Eq. 15 at $y = 1$.

The transient part is given by

$$\frac{\partial C_{\mathrm{t}}(T,y)}{\partial T}=\frac{1}{\mathrm{Pe}}\frac{\partial^2 C_{\mathrm{t}}(T,y)}{\partial y^2}-\frac{\partial C_{\mathrm{t}}(T,y)}{\partial y}-\beta C_{\mathrm{t}}(T,y) \tag{18}$$

with the following initial and boundary conditions

$$T=0 \quad C_{\mathrm{t}}(0,y)=C_{\mathrm{s}}^{\mathrm{bp}}(y)-C_{\mathrm{s}}(y) \tag{19}$$

$$y=0 \quad C_{\mathrm{t}}(T,0)-\frac{1}{\mathrm{Pe}}\frac{\partial C_{\mathrm{t}}(T,y)}{\partial y}\bigg|_{y=0}=0 \tag{20}$$

$$y=1 \quad \frac{\partial C_{\mathrm{t}}(T,y)}{\partial y}\bigg|_{y=1}=0 \tag{21}$$

By applying the exponential transformation

$$u(T,y)=C_{\mathrm{t}}(T,y)\mathrm{e}^{-\frac{\mathrm{Pe}}{m}T}\mathrm{e}^{-\frac{\mathrm{Pe}}{p}y} \tag{22}$$

Equation 18 is transformed into

$$\begin{aligned}\frac{\partial u(T,y)}{\partial T}=&\frac{1}{\mathrm{Pe}}\frac{\partial^2 u(T,y)}{\partial y^2}+\left(\frac{2}{p}-1\right)\frac{\partial u(T,y)}{\partial y}\\&+\left(\frac{\mathrm{Pe}}{p^2}-\frac{\mathrm{Pe}}{p}-\beta-\frac{\mathrm{Pe}}{m}\right)u(T,y)\end{aligned} \tag{23}$$

taking $p = 2$ and

$$m=-\frac{4\ \mathrm{Pe}}{\mathrm{Pe}+4\beta} \tag{24}$$

Equation 23 is simplified to

$$\frac{\partial u(T,y)}{\partial T}=\frac{1}{\mathrm{Pe}}\frac{\partial^2 u(T,y)}{\partial y^2} \tag{25}$$

with the following initial and boundary conditions

$$T=0 \quad u(0,y)=-A\frac{2(\alpha+1)\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}(1-y)}+2(\alpha-1)\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}(1-y)}}{(\alpha+1)^2\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}}-(\alpha-1)^2\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}}} \tag{26}$$

$$y=0 \quad u(T,0)-\frac{2}{\mathrm{Pe}}\frac{\partial u(T,y)}{\partial y}\bigg|_{y=0}=0 \tag{27}$$

$$y=1 \quad u(T,1)+\frac{2}{\mathrm{Pe}}\frac{\partial u(T,y)}{\partial y}\bigg|_{y=1}=0 \tag{28}$$

Equation 25 with the boundary conditions Eqs. 27 and 28 is also found in a heat conduction problem [8], longitudinal mixing in beds [9] and in electrochemical reactors with a constant value of current density along the electrode [3]. According to the method of separation of variables it is proposed

$$u(T,y)=\Psi(T)\Upsilon(y) \tag{29}$$

The differential equations for Ψ and Υ are

$$\frac{\mathrm{d}\Psi}{\mathrm{d}T}+\frac{\lambda^2}{\mathrm{Pe}}\Psi=0 \tag{30}$$

and

$$\frac{\mathrm{d}^2\Upsilon}{\mathrm{d}y^2}+\lambda^2\Upsilon=0 \tag{31}$$

where λ^2 is the separation constant. Considering the boundary conditions, Eqs. 27 and 28, there are an infinite number of solutions of Eq. 31. Thus, the solution of Eq. 25 is given by

$$u(T,y)=\sum_{\mathrm{n}=1}^{\infty}B_{\mathrm{n}}\left[\cos(\lambda_{\mathrm{n}}y)+\frac{\mathrm{Pe}}{2\lambda_{\mathrm{n}}}\sin(\lambda_{\mathrm{n}}y)\right]\mathrm{e}^{-\frac{\lambda_{\mathrm{n}}^2T}{\mathrm{Pe}}} \tag{32}$$

where λ_{n} are the positive roots in ascending order of the equation

$$\mathrm{tg}(\lambda_{\mathrm{n}})=\frac{\lambda_{\mathrm{n}}\mathrm{Pe}}{\lambda_{\mathrm{n}}^2-\frac{\mathrm{Pe}^2}{4}} \tag{33}$$

According to Carslaw and Jaeger [8] B_{n} is given by

$$B_{\mathrm{n}}=\frac{2\lambda_{\mathrm{n}}^2}{\lambda_{\mathrm{n}}^2+\frac{\mathrm{Pe}^2}{4}+\mathrm{Pe}}\int_0^1 u(0,y)\left[\cos(\lambda_{\mathrm{n}}y)+\frac{\mathrm{Pe}}{2\lambda_{\mathrm{n}}}\sin(\lambda_{\mathrm{n}}y)\right]\mathrm{d}y \tag{34}$$

Introducing Eq. 26 into Eq. 34, integrating and rearranging yields

$$B_{\mathrm{n}}=-\frac{2\lambda_{\mathrm{n}}^2}{\lambda_{\mathrm{n}}^2+\frac{\mathrm{Pe}^2}{4}+\mathrm{Pe}}\frac{A\ \mathrm{Pe}}{\lambda_{\mathrm{n}}^2+\left(\frac{\alpha\mathrm{Pe}}{2}\right)^2} \tag{35}$$

Combining Eqs. 22, 24, 32 and 35 results in

$$\begin{aligned}C_{\mathrm{t}}(T,y)=&-2A\mathrm{Pe}\ \mathrm{e}^{-\left(\frac{\mathrm{Pe}}{4}+\beta\right)T}\mathrm{e}^{\frac{\mathrm{Pe}}{2}y}\\&\times\sum_{\mathrm{n}=1}^{\infty}\frac{\lambda_{\mathrm{n}}}{\left(\lambda_{\mathrm{n}}^2+\frac{\mathrm{Pe}^2}{4}+\mathrm{Pe}\right)\left[\lambda_{\mathrm{n}}^2+\left(\frac{\alpha\mathrm{Pe}}{2}\right)^2\right]}\\&\times\left[\lambda_{\mathrm{n}}\cos(\lambda_{\mathrm{n}}y)+\frac{\mathrm{Pe}}{2}\sin(\lambda_{\mathrm{n}}y)\right]\mathrm{e}^{-\frac{\lambda_{\mathrm{n}}^2T}{\mathrm{Pe}}}\end{aligned} \tag{36}$$

Taking into account Eqs. 13, 17 and 36 evaluated at $y = 1$, the outlet concentration after a step perturbation of magnitude A in the inlet concentration is given by

$$
\begin{aligned}
C_{\mathrm{f}}(T) = & -2A\mathrm{Pe}\ \mathrm{e}^{-\left(\frac{\mathrm{Pe}}{4}+\beta\right)T}\mathrm{e}^{\frac{\mathrm{Pe}}{2}} \\
& \times \sum_{\mathrm{n}=1}^{\infty} \frac{\lambda_{\mathrm{n}}}{\left(\lambda_{\mathrm{n}}^{2}+\frac{\mathrm{Pe}^{2}}{4}+\mathrm{Pe}\right)\left[\lambda_{\mathrm{n}}^{2}+\left(\frac{\alpha\mathrm{Pe}}{2}\right)^{2}\right]} \\
& \times \left[\lambda_{\mathrm{n}}\cos(\lambda_{\mathrm{n}})+\frac{\mathrm{Pe}}{2}\sin(\lambda_{\mathrm{n}})\right]\ \mathrm{e}^{-\frac{\lambda_{\mathrm{n}}^{2}T}{\mathrm{Pe}}} \\
& + (C_0 + A)\frac{4\alpha\mathrm{e}^{\frac{\mathrm{Pe}}{2}}}{(\alpha+1)^{2}\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}}-(\alpha-1)^{2}\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}}}
\end{aligned}
\tag{37}
$$

and

$$
\begin{aligned}
\frac{Y(T)}{A} = & -2\mathrm{Pe}\ \mathrm{e}^{-\left(\frac{\mathrm{Pe}}{4}+\beta\right)T}\mathrm{e}^{\frac{\mathrm{Pe}}{2}} \\
& \times \sum_{\mathrm{n}=1}^{\infty} \frac{\lambda_{\mathrm{n}}}{\left(\lambda_{\mathrm{n}}^{2}+\frac{\mathrm{Pe}^{2}}{4}+\mathrm{Pe}\right)\left[\lambda_{\mathrm{n}}^{2}+\left(\frac{\alpha\mathrm{Pe}}{2}\right)^{2}\right]} \\
& \times \left[\lambda_{\mathrm{n}}\cos(\lambda_{\mathrm{n}})+\frac{\mathrm{Pe}}{2}\sin(\lambda_{\mathrm{n}})\right]\ \mathrm{e}^{-\frac{\lambda_{\mathrm{n}}^{2}T}{\mathrm{Pe}}} \\
& + \frac{4\alpha\ \mathrm{e}^{\frac{\mathrm{Pe}}{2}}}{(\alpha+1)^{2}\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}}-(\alpha-1)^{2}\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}}}
\end{aligned}
\tag{38}
$$

where $Y(T)$ is the difference between the outlet concentration at time t and the outlet concentration before the perturbation.

2.2 Current as a function of time

The current drained by the reactor is given by

$$
I(t) = WL\int_{0}^{1} j(t,y)\mathrm{d}y \tag{39}
$$

Combining Eqs. 5, 15 and 39 and solving them, the steady-state current after perturbation is

$$
I_{\mathrm{s}} = \frac{4v_{e}FkWL(C_0+A)}{\mathrm{Pe}(\alpha^{2}-1)}\left[1-\frac{4\alpha\ \mathrm{e}^{\frac{\mathrm{Pe}}{2}}}{(\alpha+1)^{2}\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}}-(\alpha-1)^{2}\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}}}\right] \tag{40}
$$

Likewise, the transient portion of the current is obtained by combining Eqs. 5, 36 and 39 and solving,

$$
\begin{aligned}
I_{\mathrm{t}}(T) = & -2Av_{\mathrm{e}}FkWL\mathrm{Pe}\ \mathrm{e}^{-\left(\frac{\mathrm{Pe}}{4}+\beta\right)T}\mathrm{e}^{\frac{\mathrm{Pe}}{2}} \\
& \times \sum_{\mathrm{n}=1}^{\infty} \frac{\lambda_{\mathrm{n}}\sin(\lambda_{\mathrm{n}})}{\left(\lambda_{\mathrm{n}}^{2}+\frac{\mathrm{Pe}^{2}}{4}+\mathrm{Pe}\right)\left[\lambda_{\mathrm{n}}^{2}+\left(\frac{\alpha\mathrm{Pe}}{2}\right)^{2}\right]}\ \mathrm{e}^{-\frac{\lambda_{\mathrm{n}}^{2}T}{\mathrm{Pe}}}
\end{aligned}
\tag{41}
$$

Combining Eqs. 40 and 41 the variation in the current related to the initial current is given by

$$
\frac{I(T)-I(0)}{I(0)} = \frac{A}{C_0}\left\{
\begin{aligned}
& 1-\frac{\mathrm{Pe}^{2}(\alpha^{2}-1)\mathrm{e}^{-\left(\frac{\mathrm{Pe}}{4}+\beta\right)T}\mathrm{e}^{\frac{\mathrm{Pe}}{2}}}{2\left[1-\frac{4\alpha\mathrm{e}^{\frac{\mathrm{Pe}}{2}}}{(\alpha+1)^{2}\mathrm{e}^{\frac{\alpha\mathrm{Pe}}{2}}-(\alpha-1)^{2}\mathrm{e}^{-\frac{\alpha\mathrm{Pe}}{2}}}\right]} \\
& \times \sum_{\mathrm{n}=1}^{\infty} \frac{\lambda_{\mathrm{n}}\sin(\lambda_{\mathrm{n}})\mathrm{e}^{-\frac{\lambda_{\mathrm{n}}^{2}T}{\mathrm{Pe}}}}{\left(\lambda_{\mathrm{n}}^{2}+\frac{\mathrm{Pe}^{2}}{4}+\mathrm{Pe}\right)\left[\lambda_{\mathrm{n}}^{2}+\left(\frac{\alpha\mathrm{Pe}}{2}\right)^{2}\right]}
\end{aligned}
\right\}
\tag{42}
$$

3 Limiting behaviour of electrochemical reactors

The dynamics of a continuous stirred tank electrochemical reactor under potentiostatic control for a step perturbation in the inlet concentration is [6]

$$
Y(T) = \frac{A}{1+\beta}\left[1-\mathrm{e}^{-(1+\beta)T}\right] \tag{43}
$$

and for the current

$$
\frac{I(T)-I(0)}{I(0)} = \frac{A}{C_0}\left[1-\mathrm{e}^{-(1+\beta)T}\right] \tag{44}
$$

For a plug flow reactor the temporal variation of concentration and current is given by Eqs. 45 and 46 respectively [6]

$$
Y(T,1) = A\ \mathrm{H}(t-\tau)\ \mathrm{e}^{-\beta} \tag{45}
$$

with

$$
\mathrm{H}(t-\tau) = 0 \quad t<\tau \tag{45a}
$$

$$
\mathrm{H}(t-\tau) = 1 \quad t\geq\tau \tag{45b}
$$

and

$$
\frac{I(T)-I(0)}{I(0)} = \frac{A}{C_0}\frac{1-\exp(-\beta T)}{1-\exp(-\beta)} \quad \text{for } 0\leq T\leq 1 \tag{46}
$$

4 Results and discussion

Figure 1 reports the steady-state concentration as a function of position in the reactor according to Eq. 15. For a Peclet number of 0.01 the dispersion model overlaps the stirred tank behaviour and for Peclet numbers higher than 100 the plug flow model is reached.

Figures 2 and 3 show the response of electrochemical reactors according to the dispersion model (Eq. 38) for two β values. Thus, the difference between the outlet concentration at time t and the outlet concentration before the perturbation related to the magnitude of the step change in

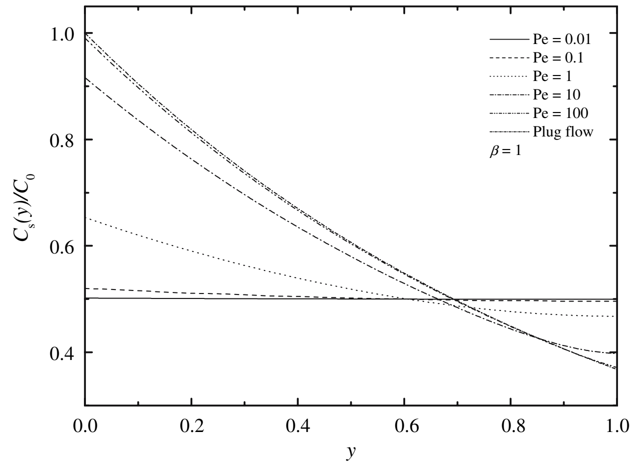

Fig. 1 Steady-state concentration as a function of position. $\beta = 1$

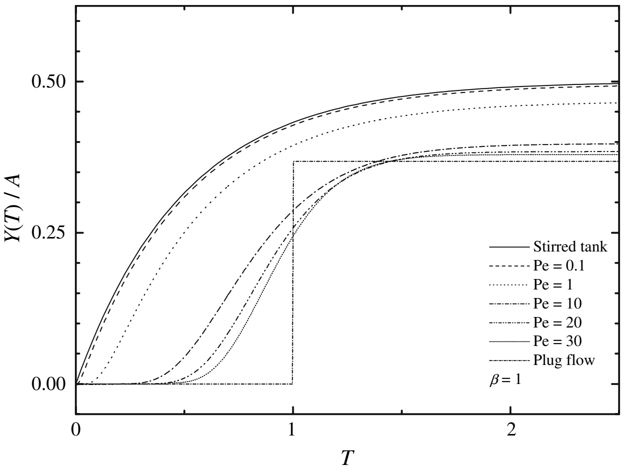

Fig. 2 Transient response in outlet concentration. $\beta = 1$

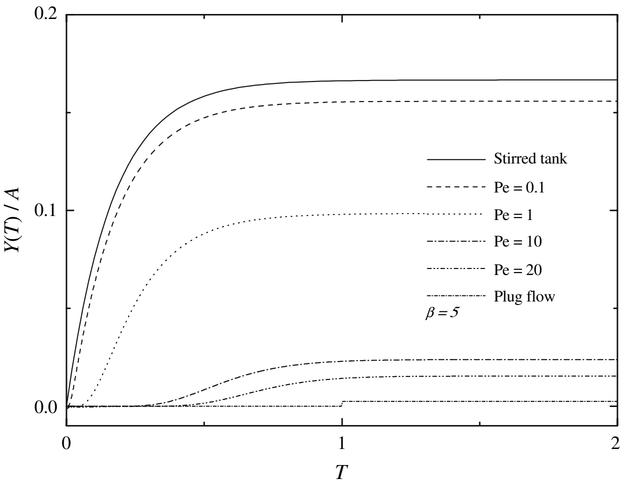

Fig. 3 Transient response in outlet concentration. $\beta = 5$

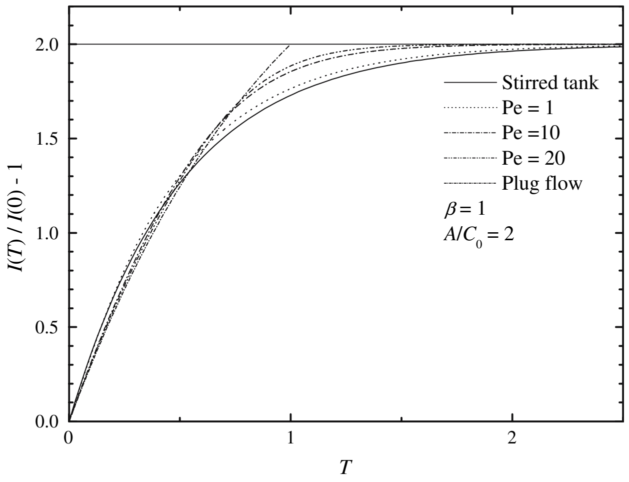

Fig. 4 Transient response in current. $\beta = 1$

the inlet concentration is plotted as a function of the normalized time, t/τ. The behaviour according to the continuous stirred tank model and the plug flow model under the same conditions (Eqs. 43 and 45) are also given. When the Peclet number approaches 0.1 the dispersion model agrees well with the prediction of the stirred tank model and when the Peclet number increases the behaviour of the plug flow model is approached. A similar performance was reported in [3] for the case of a constant mean current density along the electrode. Likewise, at high times the electrochemical system approaches a constant, which decreases when the Peclet number is increased and achieves a limiting behaviour in accordance with the plug flow model.

Figure 4 shows the response in current of electrochemical reactors under potentiostatic control for a step change in the inlet concentration. At t → ∞ the variation in the current related to the initial current approaches a constant, whose value is the same for different Peclet numbers. This limiting behaviour was also reported in previous work [6].

Thus, the reported theoretical treatment can be recognized as a helpful tool to analyse the temporal behaviour in concentration and current of electrochemical reactors under potentiostatic control.

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