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An operational model for a well-stirred membrane bioreactor: reactor performance analysis

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Abstract

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AN OPERATIONAL MODEL FOR A WELL-STIRRED MEMBRANE BIOREACTOR: REACTOR PERFORMANCE ANALYSIS

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ABSTRACT

We investigate the behavior of a reaction described by Michaelis-Menten kinetics in an immobilised enzyme reactor (IER). The IER is treated as a well-stirred flow reactor, in which the bound and unbounded enzyme species are immobilized and therefore constrained to remain within the reaction vessel. The product species leaves the bioreactor either in the reactor outflow or by permeating through the semi-permeable reactor wall. We explore how the concentration of recovered product and the reactor productivity vary with process parameters, particularly those associated with the separation of the product from the substrate through the semi-permeable reactor wall.

We show that at low residence times membrane extraction through the reactor walls increases the total product concentration recovered whereas at high residence times membrane extraction decreases the total product concentration. We also show that the reactor productivity is maximised at high residence times. For reactor productivity the key control variable is the ratio of the reactor volume to the jacket volume (V^*). If this ratio is greater than one, then membrane extraction increases the productivity. If this ratio is less than one, then membrane extraction decreases the productivity.

INTRODUCTION

Immobilised enzyme technology (IET) is attractive to process industries in which either the enzyme (biocatalysts or microorganisms) involved are expensive or a large throughput of substrate is required. When using IET the enzymes are constrained to remain within the reactor. This may be achieved by enclosing the reactor in a membrane which is permeable to the substrate and product and impermeable to the enzymes. Alternatively, the enzyme may be attached to solid constraints, such as the reactor walls, using techniques such as adsorption, chemical bonding, polymer lattice entrapment *etc.*

We investigate a model for an enzyme-catalysed reaction, obeying Michaelis-Menten kinetics, occurring in an immobilised enzyme reactor (IER). The Michaelis-Menten mechanism is widely used in modeling studies because it has been shown to provide a

useful interpretation of kinetic data from many enzyme-catalysed reactors (Campbell, 1988). We consider both a reactor-only system, in which both the substrate and product flows out of the reactor, and a separator-reactor system, in which the product species is also removed from the reactor using a permselective membrane. The permselective membrane is located on the jacket that surrounds the reactor. Thus, the separator-reactor system integrates membrane separation with biological transformation.

We consider the reaction scheme:

$$[E]+S \underset{k_{-1}}{\overset{k_1}{\rightleftarrows}} [E-S] \xrightarrow{k_2} [E]+P \tag{1}$$

The process represented by equation (1) is the conversion of a substrate species (S) to a product species (P) via the Michaelis-Menten mechanism. The first step of this conversion is the reversible formation of a bounded enzyme species, an enzyme-substrate complex, ($[E-S]$). The final step is the irreversible decay of the bounded enzyme species ($[E-S]$) to give an activated site ($[E]$) and the product. We interpret this process as representing the synthesis of a desired chemical species.

Previous investigations [Lee & Lim, 1999; Xiu et al, 2002; Namjoshi et al, 2003] have indicated that steady-state analysis may be used in planning optimum bioreactor operation. We use this technique to investigate how the performance of the separator-reactor system depends upon process parameters that are associated with the separation of the product from the substrate through the semi-permeable reactor wall, and compare its performance against that of a reactor-only system. The chosen indicators for reactor performance are the concentration of recovered product and the reactor productivity.

MODEL EQUATIONS

We model the IER as a perfectly mixed bioreactor. The substrate (S) flows through the IER whilst the reaction product (P), and unused substrate, are discharged from it. It is assumed that the reactor wall is only permeable to the product, so that the product is also recovered by flowing an inert solvent through the jacket side of the bioreactor. It is assumed that the flow of sweep liquid through the jacket is high enough to prevent a concentration gradient arising along the length of the jacket membrane. Fouling of this membrane is neglected. A key feature of IET is the retention of an immobilised enzyme within a zone of a flow reactor. It is therefore assumed that the unbounded and bounded enzymes are constrained to remain within the reactor. Thus there are no flow terms in equations (2) – (3). We further assume that there are no enzyme complexes initially present in reactor, equations (8). The dimensional and dimensionless forms of the governing equations for the model are stated in the next two sections respectively.

Dimensional Model

Enzyme associated species:

$$V\frac{d}{dt}[E]=V(k_{-1}[E-S]-k_1[E]S+k_2[E-S]) \tag{2}$$

$$V\frac{d}{dt}[E-S]=V(k_1[E]S-k_{-1}[E-S]-k_2[E-S]) \tag{3}$$

Non-enzyme associated species:

$$V\frac{dS}{dt}=q(S_0-S)+V(k_{-1}[E-S]-k_1[E]S) \tag{4}$$

$$V\frac{dP}{dt}=q(P_0-P)+Vk_2[E-S]-AU(P-P_j) \tag{5}$$

$$V_j\frac{dP_j}{dt}=q(P_{j,0}-P_j)+AU(P-P_j) \tag{6}$$

Initial conditions:

$$[E](t=0)=E_0\ ,\ [E-S](t=0)=S(t=0)=P(t=0)=P_j(t=0)=0 \tag{7}$$

In equations (2) – (7): A is the surface area of the semi-permeable reactor jacket (m^2); $[E]$ is the concentration of immobilised enzyme $(\mathrm{mol\,m^{-3}})$; $[E]_0$ is the concentration of immobilised enzyme at time $t=0$ $(\mathrm{mol\,m^{-3}})$; $[E-S]$ is the concentration of immobilised species $E-S$ $(\mathrm{mol\,m^{-3}})$; P is the concentration of the product species $(\mathrm{mol\,m^{-3}})$; P_j is the concentration of the product species in the jacket $(\mathrm{mol\,m^{-3}})$; $P_{j,0}$ is the concentration of the product species flowing into the jacket $(\mathrm{mol\,m^{-3}})$; P_0 is the concentration of the product species flowing into the IER $(\mathrm{mol\,m^{-3}})$; S is the concentration of substrate $(\mathrm{mol\,m^{-3}})$; S_0 is the concentration of substrate in the feed $(\mathrm{mol\,m^{-3}})$; U is the membrane mass transfer coefficient $(\mathrm{m\,s^{-1}})$; V is the volume of the reactor (m^3); V_j is the volume of the jacket (m^3); k_1 is the forward reaction-rate constant $(\mathrm{m^3mol^{-1}s^{-1}})$; k_{-1} is the backwards reaction-rate constant $(\mathrm{s^{-1}})$; k_2 is the reaction-rate constant $(\mathrm{s^{-1}})$; q is the flow rate $(\mathrm{m^3s^{-1}})$; and, t is time (s).

Adding equations (2) and (3), integrating, and applying the initial conditions we obtain $[E]+[E-S]=E_0$, for all time t. This relationship enables the elimination of one of the enzyme species, $[E]$ or $[E-S]$, from the model. In what follows, we choose to eliminate the unbounded enzyme species $[E]$.

Dimensionless Equations

We scale the concentration of enzyme and non-enzyme species relative to the initial concentration of free enzyme in the reactor (E_0). Time is scaled using the reaction-rate constant k_{-1}. A feature of our scaling is that there is a one-to-one correspondence between scaled and non-scaled variables. Therefore in what follows we write, for

example, mass transfer number rather than dimensionless mass transfer number. The relationship $[E]+[E-S]=E_0$ becomes $\theta_S(t^*)+\theta(t^*)=1$.

Enzyme associated species:

$$\frac{d\theta_S}{dt}=k_1^*(1-\theta_S)S^*-(1+k_2^*)\theta_S \tag{8}$$

Non-enzyme associated species:

$$\frac{dS^*}{dt^*}=\frac{1}{\tau^*}(S_0^*-S^*)+\theta_S-k_1^*(1-\theta_S)S^* \tag{9}$$

$$\frac{dP^*}{dt^*}=\frac{1}{\tau^*}(P_0^*-P^*)+k_2^*\theta_S-U^*(P^*-P_j^*) \tag{10}$$

$$\frac{dP_j^*}{dt^*}=\frac{1}{\tau_j^*}(P_{j,0}^*-P_j^*)+U^*V^*(P^*-P_j^*) \tag{11}$$

Initial conditions:

$$\theta_S(t^*=0)=S^*(t^*=0)=P^*(t^*=0)=P_j^*(t^*=0)=0 \tag{12}$$

For a given reactor design, that is for specified values for the parameters U^* and V^*, the dimensionless residence times, τ^* and τ_j^*, are the main experimentally controllable parameters. The scaled parameters are: the product concentration ($P^*=P/[E]_0$); the product species concentration in the jacket ($P_j^*=P_j/[E]_0$); the concentration of product species flowing into the jacket ($P_{j,0}^*=P_{j,0}/[E]_0$); the concentration of the product species in the feed ($P_0^*=P_0/[E]_0$); the substrate concentration ($S^*=S/[E]_0$); the substrate concentration in the feed ($S_0^*=S_0/[E]_0$); the mass transfer coefficient ($U^*=A\,U/(V\,k_{-1})$); the reactor volume ratio ($V^*=V/V_j$); the forward reaction-rate constant ($k_1^*=k_1[E]_0/k_{-1}$); the reaction-rate constant ($k_2^*=k_2/k_{-1}$); time ($t^*=k_{-1}\,t$); the proportion of the enzyme present in the form of unbounded enzyme ($\theta=[E]/[E]_0$); the proportion of the enzyme present in the form of substrate-enzyme complex ($\theta_S=[E-S]/[E]_0$); the residence time ($\tau^*=V\,k_{-1}/q$); and, the residence time in the jacket ($\tau_j^*=V_j\,k_{-1}/q$).

RESULTS

In our analysis we assume that the concentrations of product entering the reactor and the jacket are zero, i.e. $P_0=P_0^*=P_{j,0}=P_{j,0}^*=0$. In the next section we show that the system has a unique, physically meaningful, steady-state solution. For a physically meaningful solution we require that the fractional coverage of the enzyme is between zero and one ($0\le\theta_S^*\le 1$) and that all other concentrations are non-zero ($S^*,P^*,P_j^*>0$). Later we show that this solution is always locally stable.

Steady-state analysis

The steady-state solutions of equations (8) – (11) are given by

$$(\theta_S, S^*, P^*, P_j^*) = \left(\theta_S, S_0^* - k_2^* \tau^* \theta_S, \frac{(1+U^*V^*\tau_j^*)k_2^*\tau^*\theta_S}{1+(\tau^*+V^*\tau_j^*)U^*}, \frac{U^*V^*\tau_j^*P^*}{1+U^*V^*\tau_j^*} \right) \tag{13}$$

where the steady-state value for the proportion of enzyme present as a substrate-enzyme complex (θ_S) solves the quadratic equation

$$G(\theta_S) = a(\theta_S)^2 + b\theta_S + c, \tag{14}$$

where $a = k_1^* k_2^* \tau^*$, $b = -\left[k_1^* k_2^* \tau^* + k_1^* S_0^* + (1+k_2^*)\right]$ and $c = k_1^* S_0^*$. The system (9) – (11) therefore has two steady-state solutions. We show that only one of these is physically meaningful. Calculation shows that $G(0) = k_1^* S_0^* > 0$ and $G(1) = -(1+k_2^*) < 0$. Consequently equation (14) has two positive solutions: one satisfying $0 < \theta_S < 1$ and one satisfying $\theta_S > 1$. Only the former is physically meaningful. As the coefficient a in equation (14) is strictly positive the physically meaningful solution to equation (14) is

$$\theta_S = \frac{-b - \sqrt{b^2 - 4ac}}{2a} \tag{15}$$

Combining the steady-state expression for the substrate concentration in equation (13) with the expression given in (15) we have that

$$S^* = S_0^* + \frac{b + \sqrt{b^2 - 4ac}}{2k_1^*} \tag{16}$$

It can be shown that the steady-state substrate concentration is strictly positive ($S^* > 0$), and that the eigenvalues are negative. Thus the unique, physically-meaningful, steady-state solution is locally stable.

DISCUSSION

Here we investigate how the product concentration leaving the reactor system and the reactor productivity vary as a function of the residence time and process parameters associated with the design of the separator unit.

Total product concentration leaving the reactor

The total steady-state product concentration leaving the reactor is given by

$$P_{\text{tot}}^* = P_j^* + P^* = \frac{\left(1+2U^*V^*\tau_j^*\right)k_2^*\tau^*}{1+\left(\tau^*+V^*\tau_j^*\right)U^*} \cdot \theta_S \tag{17}$$

where the values for θ_S is given by equation (15).

In the limit of small residence times ($\tau^* \ll 1$) it follows from equation (17) that we have

$$P_{\text{tot}}^* = P_0^* \frac{\left(1+2U^*V^*\tau_j^*\right)k_1^*k_2^*S_0^*}{\left(1+U^*V^*\tau_j^*\right)\left(1+k_2^*+k_1^*S_0^*\right)} \cdot \tau^* + O\left(\tau^{*2}\right)$$

From this expression we see that, at small residence times, the total production concentration is an increasing function of the product $U^* V^* \tau_j^*$ and that, at small residence times, membrane extraction increases the total product concentration recovered up to a maximum of twice the concentration recovered in the absence of membrane extraction ($U^* = 0$). In the limit of high residence times ($\tau^* >> 1$) and *no* membrane extraction ($U^* = 0$) we have from equation (17) that

$$P_{\text{tot}}^* = S_0^* - \frac{\left(1+k_2^*\right)S_0^*}{k_1^* k_2^*} \cdot \frac{1}{\tau^*} + O(\frac{1}{\tau^2})$$

In the limit of high residence times ($\tau^* >> 1$) and membrane extraction ($U^* > 0$) we have

$$P_{\text{tot}}^* = \left(2V^*\tau_j^* + \frac{1}{U^*}\right) S_0^* \cdot \frac{1}{\tau^*} + O(\frac{1}{\tau^2})$$

Thus at sufficiently high residence times the concentration of product recovered in a separator-reactor system is *lower* than that recovered in a reactor-only system. Thus there is a critical value of the residence time below which the separator-reactor system is the superior configuration and above which the reactor-only system is the superior configuration. It can be shown that the switch over in reactor behaviour occurs at a critical value of the residence time τ_{cr}^* given by $\tau_{\text{cr}}^* = V^* \tau_j^*$.

Figure 1 illustrates the results described above. For the chosen parameter values the critical value of the residence time is $\tau_{\text{cr}}^* = 5$. In figure 1(a) we see that over the range $0 < \tau_{\text{cr}}^* < 5$ the performance of the separator-reactor configuration is superior to that of the reactor-only configuration. Figure 1(b) shows that at high residence times the performance of the separator-reactor system asymptotes to zero whereas that of the reactor-only system asymptotes to the value S_0^*, which in this case is one.

The proportion of the total concentration of recovered product that is recovered through the jacket is given by

$$C_c = \frac{P_j^*}{P_j^* + P^*} = \frac{U^* V^* \tau_j^*}{1 + 2U^* V^* \tau_j^*}$$

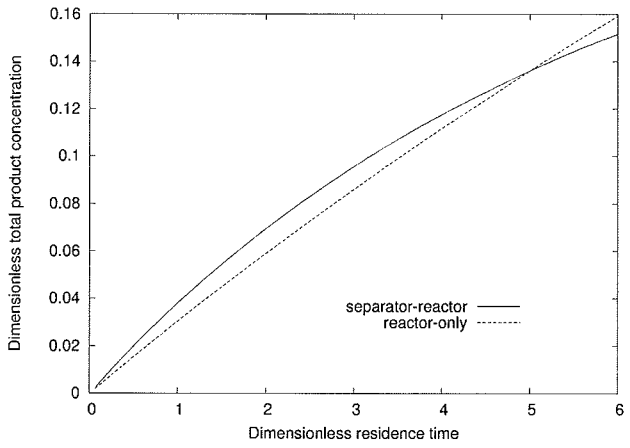
It follows that the maximum value of C_c is 0.5 (50% recovery through the jacket) and occurs in the limit $U^* V^* \tau_j^* \to \infty$.

Total reactor productivity

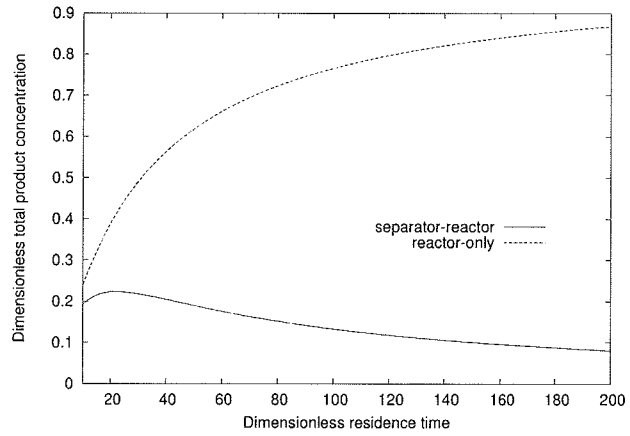
The dimensionless reactor productivity (Pr^*) under steady-state operation is given by

$$\text{Pr}^* = \frac{P^*}{\tau^*} + \frac{P_j^*}{\tau_j^*} = \frac{\left[1 + \left(\tau_j^* + \tau^*\right)U^* V^*\right] k_2^*}{1 + \left(\tau^* + V^* \tau_j^*\right) U^*} \cdot \theta_S \qquad (18)$$

where the value for θ_S is given by equation (15). Differentiating equation (18) we obtain



(a)



(b)

Fig 1: Total product concentration leaving the reactor as a function of dimensionless residence time. In the separator-reactor system $U^* = 0.1$ whereas in the reactor-only system $U^* = 0$. Other parameter values: $S_0^* = 1, V^* = 10,\ k_1^* = 0.1,\ k_2^* = 0.5,\ \tau_j^* = 0.5$.

$$\frac{\mathrm{dPr}^*}{\mathrm{d}V^*} = \frac{k_2^*\left[1+\left(\tau^*+\tau_j^*\right)U^*\right]U^*\tau^*}{\left[1+\left(\tau^*+V^*\tau_j^*\right)V^*\right]^2}\cdot\theta_S > 0$$

Thus the reactor productivity is a strictly increasing function of the reactor volume ratio. Now note that $\mathrm{Pr}^*\left(V^*=1\right) = \mathrm{Pr}^*\left(U^*=0\right)$. As the reactor productivity is a strictly *increasing* function of the reactor volume ratio it follows that the separator-reactor configuration is superior (inferior) to the reactor-only system when the reactor volume ratio is greater (less) than one.

In the limit of small residence times ($\tau^* \ll 1$) it follows from equation (18) that we have

$$\mathrm{Pr}^* = \frac{k_1^*k_2^*S_0^*}{1+k_2^*+k_1S_0^*}\left\{1+\left[\frac{\left(V^*-1\right)U^*}{1+U^*V^*\tau_j^*} - \frac{k_1^*k_2^*\left(1+k_2^*\right)}{\left(1+k_2^*+k_1S_0^*\right)^2}\right]\cdot\tau^*\right\} + O\left(\tau^{*2}\right) \qquad (19)$$

Equation (19) shows that in the limit of small residence time ($\tau^* \to 0$) the performances of the separator-reactor and reactor-only configurations are identical. Equation (19) can be written in the form

$$\Pr^*\left(U^* > 0\right) = \Pr^*\left(U^* = 0\right) + \frac{k_1^* k_2^* S_0^*}{1 + k_2^* + k_1^* S_0^*} \cdot \frac{\left(V^* - 1\right)U^*}{1 + U^* V^* \tau_j^*} \cdot \tau^* + O\left(\tau^{*2}\right)$$

where $\Pr^*\left(U^* = 0\right)$ is the formula obtained from equation (18) by setting the mass transfer number U^* to zero. This equation shows that the critical parameter in determining the performance of the separator-reactor system is the value of the reactor volume ratio. The separator-reactor configuration is superior (inferior) to the reactor-only system depending upon if the reactor volume ratio is greater (less) than one.

Assuming that the volume ratio is greater than one, than the maximum increase in reactor productivity as a function of the dimensionless membrane mass transfer coefficient occurs in the limit when $U^* \to \infty$. We have

$$\lim_{U \to \infty} \left[\Pr^*\left(U^* > 0\right) - \Pr^*\left(U^* = 0\right) \right] = \frac{k_1^* k_2^* S_0^*}{1 + k_2^* + k_1^* S_0^*} \cdot \frac{V^* - 1}{V^*} \cdot \frac{\tau^*}{\tau_j^*}$$

In the limit of high residence times ($\tau^* \gg 1$) and *no* membrane extraction ($U^* = 0$) we have

$$\Pr^*\left(U^* = 0\right) \approx \frac{S_0^*}{\tau^*} - \frac{\left(1 + k_2^*\right) S_0^*}{k_1^* k_2^*} \cdot \frac{1}{\tau^{*2}} + O\left(\frac{1}{\tau^{*3}}\right) \tag{20}$$

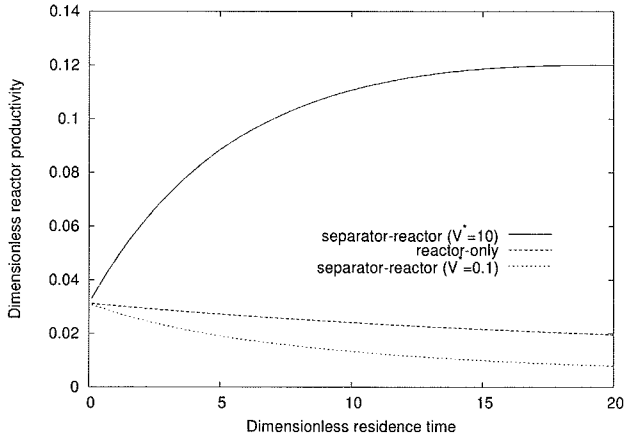
In the limit of high residence times ($\tau^* \gg 1$) and membrane extraction ($U^* > 0$) we have

$$\Pr^*\left(U^* > 0\right) \approx \frac{S_0^* V^*}{\tau^*} - \left[\frac{V^* - 1}{U^*} + \left(V^* - 1\right) V^* \tau_j^* + \frac{\left(1 + k_2^*\right) V^*}{k_1^* k_2^*} \right] \cdot \frac{S_0^*}{\tau^{*2}} + O\left(\frac{1}{\tau^{*3}}\right) \tag{21}$$

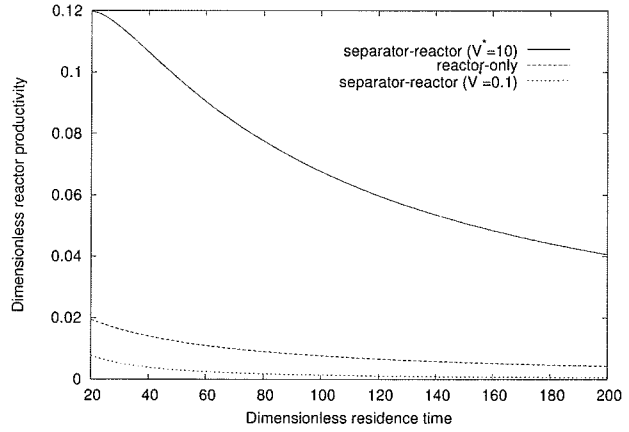
To leading order the performance of a separator-reactor system does not depend upon the value of the dimensionless membrane mass transfer number (U^*) but it does depend upon the value of the reactor volume ratio (V^*). Comparing equation (20) with equation (21) we again see that at leading order a separator-reactor is more efficient than a reactor-only system if the reactor volume ratio is greater than one ($V^* > 1$) and less efficient if the reactor volume ratio is less than one ($V^* < 1$). We also see that at sufficiently high residence times the increase in productivity for a separator-reactor system with a reactor volume ratio greater than one is simply the reactor volume ratio. Equation (21) also shows that the productivity of the separator-reactor system is an increasing (decreasing) function of the dimensionless membrane mass transfer coefficient if the reactor volume ratio is greater (less) than one.

Figure 2 illustrates the results described above. We see that the performance of the separator-reactor configuration with $V^* = 10$ is superior to that of the reactor-only system which in turn is superior to that of the separator-reactor configuration with $V^* = 0.1$. Figure 2(b) shows that at high residence times the performance of all three reactors asymptotes to zero. Observe that for the reactor-only system and for the

separator-reactor system with $V^* = 0.1$ that the productivity is a strictly decreasing function of residence time. Thus the productivity is maximised at zero residence time, i.e. infinite flow rates. Conversely, the productivity of the separator-reactor system with $V^* = 10$ is maximised at a finite residence time.



(a)



(b)

Fig 2: Reactor productivity as a function of dimensionless residence time. Parameter values: $S_0^* = 1$, $V^* = 10$, $k_1^* = 0.1$, $k_2^* = 0.5$, $\tau_j^* = 0.5$.

It is also of interest to know the proportion of the total reactor productivity that is recovered through the jacket. This is given by

$$C_{\text{pr}} = \frac{P_j^* / \tau_j^*}{P_j^* / \tau_j^* + P^* / \tau^*} = \frac{U^* V^* \tau}{1 + U^* V^* \left(\tau^* + \tau_j^*\right)} \tag{22}$$

It follows that the maximum value of C_{pr} is 1.0 (100% recovery through the jacket) and occurs in the limit when $\tau^* \to \infty$. A second limit of interest is the case $U^* V^* \to \infty$ when we have $C_{\text{pr}} = \dfrac{\tau^*}{\tau^* + \tau_j^*}$. It follows from differentiation of equation (22) that C_{pr} is an increasing function of the residence time (τ^*) and the product $U^* V^*$ and a decreasing function of the jacket residence time (τ_j^*).

CONCLUSIONS

We have investigated the performance of an immobilised enzyme reactor in a separator-reactor configuration, in which product is removed from the reactor using a semi-permeable membrane, and a reactor-only system without a membrane extraction unit. The kinetic scheme used was the Michaelis-Menten mechanism. Both configurations have a unique physically-meaningful steady-state solution that is locally stable.

In the limit of small residence times the concentration of product recovered is an increasing function of the mass transfer number: a separator-reactor system outperforms the reactor-only system and the maximum possible increase in performance in a separator-reactor system is twice that obtained in an equivalent reactor-only system. However, in the limit of high residence times the reactor-only system outperforms the separator-reactor system. The critical value of the residence time, below which the separator-reactor system gives the superior performance, is given by equation (20). Interestingly, the critical value is independent of the reaction kinetics and the mass transfer rate through the membrane.

The productivity of a separator-reactor system is determined by the value of the reactor volume ratio. If this is greater than one then the reactor-separator configuration outperforms a reactor-only system. If this is less than one, then the reactor-only system is superior to the separator-reactor configuration. If the reactor volume ratio is greater than one, than the productivity is an increasing function of the mass transfer number.

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