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Simulation and analysis of the performance of tubular enzymatic membrane reactors under different configurations, kinetics and mass transport conditions

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ABSTRACT

A model was developed to simulate tubular enzymatic membrane reactors under three different configurations: dead-end, tangential flow with a porous enzymatic membrane and a non-permeable enzymatic wall. The simulations were applied to analyze the influence of reactor configuration, kinetics and mass transport conditions over the reactor performance in order to identify the main aspects to be taken into consideration for attaining optimal designs. The non-permeable enzymatic wall configuration under the evaluated conditions seems to be more valuable than the dead-end case in terms of substrate conversion and the tangential configuration looked more favorable to promote the best conversion in the permeate but not in the retentate. It was demonstrated that for a similar value, the Damköhler number can result in very dissimilar performances. The simulated results demonstrated that the most significant variable of the global performance of the enzymatic membrane reactors is the reaction kinetics: fast reactions attained very considerable conversion values under very different conditions.

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1. Introduction

Enzymes are gaining relevance as biocatalysts in industrial processes, as their capacities have been adapted during the last 50 years for successful production of some commodities, fine chemicals and pharmaceuticals in an environmentally friendly way (high reaction rates at room temperature and pressure without auxiliary chemicals). Enzymes are specifically useful to facilitate the synthesis and modification of organic molecules by chemo-, regio-, and stereoselective bond-forming and -breaking reactions [1,2]. But enzymes have to compete economically with inexpensive traditional chemical processes which are nowadays industrially established. Therefore, the recovery and reuse of enzymes, which are generally expensive, becomes really essential in most cases in order to assure the economic viability of the processes [3,4]. Indeed, immobilization of enzymes is generally required in industry.

In addition, immobilization can also improve enzyme performance by increasing their activity, stability and selectivity and allowing optimal process reaction conditions, such as acidity, alkalinity, organic solvents or elevated temperatures [5,6]. Many immobilization methods and protocols have been put into practice and are compiled in bibliography [7], but in a broad framework, all different immobilization strategies can be categorized in one of the following groups: adsorption, covalent linking, entrapment–encapsulation, cross-linking or affinity [8]. Membranes are considered as very attractive supports for immobilizing enzymes since they can be placed in multifunctional membrane reactors, which integrate biocatalytic reaction, product separation and enzyme recovery [9–11]. Enzymatic membrane reactors (EMRs) can overcome some of the negative aspects that are related to immobilization and show some advantages over other alternatives, but some disadvantages are still apparent as summarized in Table 1 [10].

EMRs contribute to the implementation of continuous operation mode and avoid the intrinsic discontinuous nature of batch processes and their problems. Nevertheless, although several continuous enzymatic processes have been fully developed, improvement of the entire processes is still required to maximize the benefits derived from their use [9–12]. Reaction engineering must provide solutions to improve enzymatic processes at the commercial level [13]. Decisions made in reactor design have a very significant impact on the enzymatic process performance, but there are no simple and direct procedures available to determine the optimal conditions [14]. These reaction conditions also determine the downstream processing and, under the consideration of the high importance of the costs due to downstreaming, have a deep impact in the economic viability of the whole process [15].

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Table 1
Advantages and drawbacks of EMRs. Adapted from [10].

Advantages	Drawbacks
Effective retention and reuse of catalyst	Enzyme activity decay
Continuous mode operation	Heterogeneous reaction conditions
Reduction in substrate/product inhibition	Polarization layers
Final product free of enzyme	Membrane fouling
Enhanced enzyme stability when grafted	Possible less enzyme activity when grafted
Decrease of diffusional limitations	
Process intensification (single-step reaction and separation)	

Modeling and simulation are among the most useful tools to be applied to reaction and process engineering. They complement purely empirical procedures, accelerate and simplify process development and help to have a better process understanding [16], so these tools should be applied to the complete set of chemical and biochemical processes [15,17], including some reluctant sectors such as the pharmaceutical or biotechnological ones [18,19]. As modeling allows experimental effort reduction by avoiding time, money and resources consuming experiments, it has to be recommended for the investigation and analysis of the influence of the different factors that have to be taken into account in the design of reactors based on enzymatic activity [20].

The objective of this work is the employment of computer aided process engineering tools such as modeling and simulation to advance in the knowledge of the performance of enzymatic membrane reactors under different design and operation conditions by building a three dimensional (3D) model of the system. The obtained results for the different analyzed configurations provided a very valuable support to identify the most relevant aspects that have to be taken into account for attaining optimal conditions for each application. Steady state one dimensional models (1D) of membrane bioreactors utilizing free enzymes or confined in a volume have been developed by different groups [21–23]. Among these models, one of the most interesting model is the model of Salzman et al. [23], who make a fine analysis of the effect of diffusional limitations that may prevail in this type of reactor with the aid of two dimensionless groups, an effectiveness factor (related with reaction rates) and a second dimensionless number related with the mass transport through the membrane. In the literature, have been reported more recent 1D or 2D models analyzing the influence of the main operation variables on the performance of hollow-fibers membrane reactors with entrapped enzymes inside the porosity of the membranes [24–28]. All of them are based on interesting analyses of the balance between the reaction rates and mass transport with explicit equations and through various dimensionless numbers (Thiele, Peclet, Bodenstein).

In a first published part of this work [29], we have reported a preliminary 3D model which was developed for a dead-end configuration and the enzymatic hydrolysis of butyl acetate by a lipase as model reaction. The enzymes were considered to be grafted on the internal surface of the membranes. The model was based on the coupling of mass balances, hydrodynamics of the tubular membranes and enzymatic kinetics. The results in this preliminary work were reported in terms of reaction productivity which is not an interesting magnitude for chemical engineering and design purposes. In addition any analysis with respect to other configurations or the controlling phenomena (enzymatic kinetics or mass transfers) has not been carried out. In this second part and for the same hydrolysis reaction, we complete the previous reported model in order to consider other EMR configurations which are more usual at industrial scale like the tangential configuration. Moreover, a detailed theoretical analysis was carried out in order to study the main process variables controlling the global performance and conversion of EMRs. For this purpose we based our simulations on the different enzymatic kinetics reported in the literature. This analysis was carried out considering initially the Damköhler number, and then carrying out a careful analysis of the influence of the different parameters on the conversion of enzymatic membrane reactors in tangential configuration.

2. Process modeling and case study resolution

2.1. Process modeling and presentation of the case study

In a preliminary 3D model for the simulation of enzymatic membrane reactors working in dead-end mode (feed stream leaves as permeate stream) [29], we were able to represent, in a successful way, the hydrodynamics inside the membrane reactors and the resulting flux profiles, as well as the reactive regions through membranes length and the corresponding concentration profiles. In this previous work, [29] the model was validated with experimental results of the hydrolysis of butyl acetate with a lipase immobilized on the internal surface of tubular ceramic membranes. The model presented here is inspired in the previous one but it has been modified and completed in order to simulate the behavior of other different configurations of the EMR, like an enzymatic non-permeable wall reactor (feed stream leaves only as retentate stream) and in tangential mode (feed stream is split between permeate and retentate streams). The configurations considered are shown in Fig. 1.

Although the configuration under tangential mode could be considered as the most relevant for industrial purposes, the other configurations can also have promising applications. For instance, the dead-end configuration can be employed for situations with quite clean streams (no filtration is needed) where the permeation through the membrane involves a significant improvement through the "flow through membrane reactor concept" [9]. The opposite case can be applied to the non-permeable wall: the permeation of the fluid should be avoided because of, for instance, very high solids content which will drive to membrane fouling, in such case the membrane acts only as a catalyst support, this case can be approached to the honeycomb supports.

The 3D developed models were built with COMSOL Multiphysics™. The modeling work was focused on mono-channel membranes because these membranes do not require long calculation times. In addition to the classical model considerations which integrate the flux through the inner compartments, like it has been reported in the previous work named above; we studied here the relative influence of the permeation rate (transmembrane flux through the different membrane layers in radial direction) and the reaction kinetics. These both parameters are essential in order to determine theoretically the cases when the use of EMRs is interesting in terms of reaction conversion.

As illustrating case study, the butyl acetate hydrolysis reaction to acetic acid and butanol catalyzed by immobilized *Candida antarctica* lipase B (CALB) was selected:

$$C_6H_{12}O_2 + H_2O \rightarrow C_2H_4O_2 + C_4H_{10}O \quad (1)$$

This reaction was selected because of its very well-known Michaelis–Menten kinetics, which has facilitated the appearance of previous studies about its implementation in EMRs.

The mono-channel tubular membranes considered have 13 cm of effective length, and external and internal diameters of 1 and 0.7 cm respectively. They present an asymmetric structure with

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