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Modeling and experimental validation of hydrodynamics in an ultrasonic batch reactor

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ABSTRACT

Simulation of hydrodynamics in ultrasonic batch reactor containing immobilized enzymes as catalyst is done. A transducer with variable power and constant frequency (24 kHz) is taken as source of ultrasound (US). Simulation comprises two steps. In first step, acoustic pressure field is simulated and in second step effect of this field on particle trajectories is simulated. Simulation results are compared with experimentally determined particle trajectories using PIV Lab (particle image velocimetry). Effect of varying ultrasonic power, positioning and number of ultrasonic sources on particle trajectories is studied. It is observed that catalyst particles tend to orientate according to pattern of acoustic pressure field. An increase in ultrasonic power increases particle velocity and also brings more particles into motion. Simulation results are found to be in agreement with experimentally determined data.

1. Introduction

In heterogeneously catalyzed reactions high-intensity ultrasound produces agitation, which can be utilized to disperse particles [1]. At liquid/solid interfaces, acoustic waves cause turbulence known as “acoustic streaming” [2,3]. This reduces the diffusion boundary layer, increases the convective mass transfer, and considerably accelerates diffusion [4]. These effects influence external resistance and should affect mass transfer in the same way as mechanical stirring [5].

Use of enzymes for catalyzing chemical reactions is becoming popular. The key advantages associated with enzymes include their high activity and specificity (thus minimizing or eliminating side product formation) along with capability to operate at milder operating conditions (thus decreasing the energy consumption of process) [6]. For industrial applications enzymes are bound on some porous carrier material termed as immobilized enzymes (this facilitates separation of enzyme from product). Study of enzyme catalyzed reactions is a major topic of research at Institute of Process and Plant Engineering TU Hamburg [7–12]. Research involves both experimental as well as modeling aspects. Objective of the research is to develop strategies for efficient utilization of enzymes for industrial scale applications. This requires investigation of the methods to intensify the activity of enzymes. One such method is application of ultrasound. A review of literature shows that application of ultrasound to enzyme catalyzed reactions can bring an improvement of up to 200% [13–17]. In order to investigate this phenomenon in detailed and systematic manner a research project is started at institute. This project will involve experimental study leading to the development of a mathematical model for an enzyme catalyzed batch ultrasonic reactor. Current work presents the first step towards the development of such a model.

One of the requirements for effective utilization of US is that the applied US should be uniformly dispersed in the medium (acoustic pressure field). Meeting this requirement helps in improving the efficiency of chemical reaction by achieving the highest possible process intensification. This requires a thorough understanding that how different influencing parameters effect this acoustic pressure field (which ultimately governs the reactor performance). Some of the influencing parameters include ultrasonic power, frequency, position of ultrasonic source, type of reacting medium, reactor geometry, material of construction of reactor, temperature etc. Experimentally determining the influence of these parameters on acoustic pressure field for a given reaction requires a lot of time and investment. Moreover doing so, experimentally for every new reaction and reactor geometry is certainly impractical. A practical approach is to develop a mathematical model that can predict the performance of an ultrasonic reactor as a function of afore mentioned parameters. This model can be validated for a small scale lab reactor. Once validated, it can be adapted for predicting the performance of new reactors and geometries [18].

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Simulation of ultrasonic reactors using COMSOL has been presented in a number of publications e.g. [19–24]. In all of these publications, simulations are limited to the distribution of sound pressure (acoustic pressure field) in reactor. To the best of our knowledge, it is first ever study to simulate the effect of ultrasound on immobilized enzyme particles. In the present study a simplified method for simulation of an enzyme catalyzed (immobilized) ultrasonic batch reactor is presented. Effect of ultrasonic power, transducer position and use of multiple Sonotrodes is studied. To validate the simulation results "particle image velocimetry (PIV)" has been used. Once validated, model can be used to predict the distribution of particle trajectories as a function of afore mentioned parameters. From the simulation it is possible to predict the trajectories of catalyst particles in reaction medium resulting from sonication. The hydrodynamic information thus obtained can be used to estimate rate of mass transfer. In this way performance for newly proposed ultrasonic reactors can be analyzed quickly and conveniently.

Patidar and Kalva [25,35] have modeled the phenomenon of cavitation in sonicated enzyme catalyzed reaction systems. They have shown that cavitation bubbles can influence reaction through physical (turbulence, shock wave) and chemical (radical formation) effects. They stated that cavitation has an adverse effect on enzyme action (due to shock waves) and therefore should be avoided. For a given power input of ultrasound, detrimental effects of cavitating ultrasound can be avoided by using a higher hydrostatic pressure [25,36]. Our experiments in lab have also shown that cavitating US damages Novozymes-435 particles (used as example for present study). Novozyme-435 was sonicated (80 W) in lab at 40 °C in equimolar mixture of oleic acid and n-hexanol for two hours. As can be seen in Fig. 1 enzyme particles got damaged due to cavitation and therefore it was not possible to reuse them. However under same conditions but in absence of cavitation the enzyme particles remained undamaged. Due to these facts cavitation phenomenon is undesirable and therefore not considered in simulation.

2. Experimental setup

Example batch reactor used in this study consists of a transducer (ultrasonic source) and a glass reactor (Fig. 2). Transducer used is UP400S from Hielscher US Technology and is capable of operating at a constant frequency of 24 kHz (400 W). Ultrasonic generator and transducer are integrated into one assembly. For transfer of US into reaction medium ultrasonic horns are attached to the transducer. The transducer assembly can be mounted on a stand to sonicate the reaction medium. To hold the reaction contents a glass reactor from NORMAG Labor- und Prozesstechnik GmbH is employed. The glass reactor has a heating/cooling jacket around it to maintain the required temperature. Ultrasonic horn can be inserted into reaction medium at desired positions (vertically and horizontally).

Immobilized enzyme used in this study was Novozym-435. In Novozym-435 lipase enzyme is immobilized on Lewatit VP OC 1600, a divinylbenzene-cross-linked poly (methyl methacrylate) resin produced by Lanxess Germany. The average particle size is around 0.6 mm with pore size around 4.5 nm [26].

3. Modeling

In current study application of ultrasound to a suspension of immobilized enzyme particles in reaction medium is to be modeled. For this two-step modeling approach is adopted as

1. *Acoustic modeling:* Simulates the dispersion of ultrasound in reaction medium.
2. *Modeling of particle trajectories:* Simulates how particles are affected by the acoustophoretic force (resulting from acoustic pressure).

In the following paragraphs follows a detailed explanation about each model.

3.1. Acoustic modeling

The governing equation for propagation of sound inside a medium is the Helmholtz wave equation given by [18,20]

$$\nabla\left(\frac{1}{\rho}\nabla \mathrm{p}\right) - \frac{\omega^2}{\rho c^2}\cdot \mathrm{p} = 0 \qquad (1)$$

where ρ = density of medium [kg/m^3], p = acoustic pressure [Pa], c = speed of sound in medium [m/s], $\omega = 2\pi f$ (angular frequency)[rad/s], f = frequency [Hz].

The above mentioned equation is in time-harmonic formulation. Sound wave travels in harmonic manner, so the time dependence can be taken out of the equation. Using this equation, sound field is described and solved by the pressure p. The pressure represents the acoustic variations (or excess pressure) to the ambient stationary pressure. Acoustic pressure gives the acoustophoretic force (F_{aco}) for particle movement in Section 3.2.

3.1.1. Boundary conditions for acoustics modeling [27]

- *Sound hard boundary (reactor walls):* This type of boundary assumes that incident sound wave is perfectly reflected back and is in phase with the incident one ($R = 1$). Wall of reactor

Fig. 1. Effect of non-cavitating and cavitating US on immobilized enzymes.