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## Application of a novel type impinging streams reactor in solid–liquid enzyme reactions and modeling of residence time distribution using GDB model



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#### ABSTRACT

Solid–liquid enzyme reactions constitute important processes in biochemical industries. The isomerization of D-glucose to D-fructose, using the immobilized glucose isomerase (Sweetzyme T), as a typical example of solid–liquid catalyzed reactions has been carried out in one stage and multi-stage novel type of impinging streams reactors. Response surface methodology was applied to determine the effects of certain pertinent parameters of the process namely axial velocity (A), feed concentration (B), nozzles' flow rates (C) and enzyme loading (D) on the performance of the apparatus. The results obtained from the conversion of glucose in this reactor were much higher than those expected in conventional reactors, while residence time was decreased dramatically. Residence time distribution (RTD) in a one-stage impinging streams reactor was investigated using colored solution as the tracer. The results showed that the flow pattern in the reactor was close to that in a continuous stirred tank reactor (CSTR). Based on the analysis of flow region in the reactor, gamma distribution model with bypass (GDB) was applied to study the RTD of the reactor. The results indicated that RTD in the impinging streams reactor could be described by the latter model.

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### 1. Introduction

Solid–liquid catalytic reactions are widely used in chemical industries. An example is the isomerization of glucose to fructose using an immobilized glucose isomerase enzyme. This reaction is one of the most widely used processes in food industry in producing dietetic "light" foods and drinks, because it improves the sweetening, color, and hygroscopic characteristics as well as reducing viscosity of the products. In addition, fructose is about 75% sweeter than sucrose. Also it is absorbed more slowly than glucose, and metabolized without intervention of insulin. Also, crystallization of glucose in concentrated solutions can make prior handling and processing difficult. For all these reasons, this process has a lot of room for improvement utilizing both cells and free or immobilized enzymes [1–5].

From the perspective of chemical kinetics, isomerization of glucose to fructose is a reversible reaction in the order of 5 kJ/mol [6] and, consequently, the equilibrium product contains roughly a 1:1

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ratio of glucose to fructose and thus less temperature sensitive; so that at  $55 \,^{\circ}$ C the fructose content at equilibrium is 50%, and at higher temperatures of 60, 70, 80, and 90 °C, is 50.7%, 52.5%, 53.9%, and 55.6%, respectively. However by increasing the temperature, stability and the enzyme half-life and consequently, productivity of the process decline. Consequently, most industrial plants run at 58–60 °C, a temperature with low risk for microbial contamination [7].

In 1974, the first immobilized form of glucose isomerase was introduced. Since that time several reactor designs including continuous stirred tank reactors (CSTR), fixed-bed reactors, simulated moving beds, and fluidized-bed reactors have been applied in various modes of operation for this conversion process. Isomerization of glucose is normally carried out in tubular reactors packed with immobilized enzyme with a space velocity of  $0.2-2 h^{-1}$  [8–14].

Beck et al. investigated the effect of diffusion limitations within the immobilized enzyme particles on the fructose concentration leaving the reactor. It is observed that in this reaction external mass transfer resistance is more dominant relative to that of internal diffusion [2,15].

The impinging jets (IJ) technique is a unique flow configuration. The first patent for such a technique was published by Bower. The IJ method was applied by Elperin for gas-solid suspensions and

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further developed by Tamir in various chemical engineering processes [16,17]. The IJ technique has been successfully applied to various chemical processes such as desorption, absorption, solid–liquid enzyme reactions, copper extraction and stripping, liquid–liquid reactions, precipitation, and crystallization with a calcium oxalate model system [18].

The RTD is one of the major informative flow characteristics pattern in a chemical reactor. It provides information on the duration of stay of various elements within the reactor and allows for thorough comparisons between systems having different shapes and configuration [19]. For reactor design and scale-up purposes, it is essential to have information on the RTD.

Similar to other non-ideal reactors, RTD of impinging streams reactor can be represented by following models: (1) compartment models consisting of a number of ideal reactors (e.g. CSTR, PFR) connecting in series or parallel; (2) mixing models combining perfect mixing, short-circuiting and plug flow regions in various arrangements. Guo et al. [20], Royaee and Sohrabi [21] used Markov chain model to describe the RTD of impinging streams apparatus. Kitron et al. [22], Rajaie and Sohrabi [23] simulated the RTD of an impinging streams contactor by Monte Carlo method.

The main objectives of the present study are as follows:

- Investigation of the possibility of utilization of a new type of multi stage impinging-jets reactor in solid–liquid catalytic reactions.
- (2) Modeling of the RTD of an impinging stream reactor, using gamma distribution model with bypass.

### 2. Materials and methods

#### 2.1. Chemicals

All chemicals used in the present investigation were of analytical grade; D-glucose in crystalline form was supplied by Merck Co. (Germany). The immobilized enzyme, Sweetzyme T, was provided as a gift by Novo Nordisk (Iran). The immobilized glucose isomerase (IGI) enzyme particles were of cylindrical shape, with  $0.0002-0.0004\,m$  in diameter,  $0.001-0.0015\,m$  length and having an activity of  $350\,IGIU/g.$ 

Sodium di-hydrogen phosphate  $(NaH_2PO_4)$ , di-sodium hydrogen phosphate  $(Na_2HPO_4)$ , and magnesium sulfate  $(MgSO_4)$  were all supplied by Merck Co. (Germany). Reactive Red 198 was supplied by Youhao Co. (China) and used in RTD determination.

#### 2.2. Analytical technique

The determination of glucose concentration was carried out by both glucose oxidase/peroxidase method, applying Glucose assay kit supplied by Ziest Chem Diagnostics Co. (Iran) and spectroscopic technique using a visible spectrophotometer (Dr 2800 Hach Co., USA) at a wavelength of 490 nm. Concentration of C.I. Reactive Red 198 was also measured by the latter UV–vis spectrophotometer at the wavelength of 500 nm.

#### 2.3. Reactor

#### 2.3.1. One-stage reactor

A single stage reactor was applied to determine the optimum operating conditions. A schematic diagram of the impinging reactor used in this study is shown in Fig. 1. The apparatus consisted of a cylindrical vessel made of Plexiglas and equipped with two pressure nozzles mounted on the reactor in front of each other and tangentially injecting two jets of glucose solution toward the reaction zone. The novelty of this reactor is in plug-up flow stream of glucose solution along the reactor height while being impinged by the tangential flows from the nozzles. The objective of this study is to investigate the influence of such an axial flow on the substrate conversion.

The reactor consisted of three separate parts. The middle part of the reactor, containing the immobilized enzyme, comprised the reaction compartments. Two pressure nozzles were mounted in front of each other within this part. The top and bottom sections were both filled with inert packing in order to promote the plug flow of the axial solution stream.



Fig. 1. The one-stage impinging streams reactor dimensions. (a) front view, (b) nozzle, and (c) top view.

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