ISSN : 0974 - 7435 Volume 9 Issue 9

BioTechnology
An Indian Journal

FULL PAPER

BTAIJ, 9(9), 2014 [353-362]

Computational investigation of microfluidic reactor with static mixers for analytical application

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ABSTRACT

A computational study of microchannel reactors with different static mixers has been carried out. The influence of static mixers (rectangular and elliptical static mixers) on the fluid velocity and conversion in a microchannel was studied for the case of enzyme catalyzed oxidation of glucose. Simulations were performed over a Reynolds number range of 0.1 – 100. The simulation results have been discussed along with the underlying mechanisms. Our results show that the shape of static mixer and its orientation with respect to the axis of microchannel influences substrate-enzyme contact and the substrate conversion. The study provides an insight on the application of microchannels with static mixers for bioanalytical application.

KEYWORDS

Micro mixers;
CFD;
Diffusion;
Conversion;
SIMPLE;
Microchannel.

INTRODUCTION

Micro-reaction engineering is a field that has seen rapid growth in the past decade. The idea, initially derived from the miniaturized silicon chips of the electronics industry has expanded its horizon, providing a variety of solutions in the chemical, biochemical and the biotechnological industries. One of the major factors that encourage research and development in this area is the advantage of process intensification offered by the micro-devices. This is a direct result of the differences that arise as a result of reduction in the geometry scale leading to increased gradients (thereby increased heat and mass transfer) as well as increased surface to volume ratio. Navier-Stokes equation, originally developed for flow through channels of diameter few mm & above, is valid for flow in micro-channels as well, provided the appropriate boundary conditions are used[1].

Though the micro world has proved promising, certain observed phenomena call for the modifications of design to suit the purpose. With a shift to operation in the low Reynolds number region owing to the decreased dimensions and flow rate,[2] viscous forces are dominant and consequently the flow is predominantly laminar. One parameter that is affected in laminar flow is the mass transfer of particles to the reaction site. Brody et al[3]. observed that diffusion remained low in micro flows as the time required for diffusion was not available at the specified fluid flow rate. Kamholz et al[4] reported the effect of channel width and the diffusion coefficient on channels dealing with surface reactions. Research focus on the study of transverse diffusion has been limited till date. Walter et al[5]. investigated the effect of channel dimensions and temperature on the mass transfer in micro channels. The general assumption used in mass transfer problems is to proceed considering only the primary concentration gradient in the radial direction. This results in a one-dimensional Einstein approximation of the diffusion equation. However the diffusion of solute varies at different axial positions, leading to introduction of modifications in the original profile for a considerable distance from the inlet[6].

To alleviate this problem of slow diffusion, mixers, both active and passive have been in use. Shu and Kang provided a comprehensive review on mixing in micro fluidic devices[7]. Both active and passive mixers can be used to address homogenization as well as transverse diffusion in the micro scale, while static mixers have the advantage of easier integration into the system[8]. Moreover, static mixers do not require external power to bring about mixing and rely on the driving force for flow. Hydrodynamic focusing using a continuous-flow mixer to reduce the diffusion times by manipulating the flow from different streams has also been reported[9]. The diffusion length between molecules to be mixed is reduced by flow focusing to achieve mixing at shorter lengths[10].

The effect of arrangement and the number of obstacles as mixers were tested in a Y-channel to investigate the best layout of the obstacles with circular cross section[11]. Around the same time Johnson et al[12] focused on the lateral transport of material in electro-osmotic flow in micro-channels. Both experimental and simulation works were carried out by different groups to check the effect of variations in the geometry on mixing in comparison to the conventional channels[13,14].

Yet another method to increase mixing is to force the fluid to take alternative path by the use of overlapping barriers increased mixing[15]. The use of herring bone shaped structures in the flow channels too have been reported to increase mixing[16]. Herring bone structures of higher depth were found to improve mixing more effectively than shallow herring bone structures[17]. Sotowa et al[18]. studied the behavior of baffles in micro channels. Stroock et al[19]. studied the use of ridges, inclined at an angle to the flow for effective mixing. Following these seminal studies, several static mixers for low Reynolds number flow have been modeled to work with an acceptable pressure drop[20].

The present study focuses on the effect of new types of static mixers with potential to enhance the transverse flow for the purpose of facilitating enzyme aided surface reactions. These reactions are widely used for bioanalytical applications, which when carried out in micro-fluidic platform can lead to smaller analyte volume and faster reaction.

MODEL

The flow length scale considered in the study makes the use of the equations of classical fluid mechanics valid in the region and partial differential equations, derived from continuum mechanics assumption are solved for momentum and mass transfer.

ASSUMPTIONS

The following assumptions have been made:

- The flow is steady, laminar (due to low velocity and lower channel hydraulic diameter) and incompressible
- Density and viscosity are constant throughout the domain. The presence of small quantities of analyte does not significantly affect these properties.
- The flow is isothermal, as the heat of the reaction is negligible due to low concentration of the analyte and low levels of enzyme per unit area in the channels.
- The reactions are heterogeneous and enzyme-based. The reactions occur only on the two side walls.

GOVERNING EQUATIONS

In light of the above assumptions, the governing equations are as follows:

The mass conservation or the continuity equation at constant density is given by,

$$\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y}=0 \qquad (1)$$

The momentum transport (Navier-stokes) equation (2) and the species balance equation (3) are as follows: [21]

$$\rho V.\nabla V = -\nabla P + \mu \nabla^2 V + \rho g \quad (2)$$

$$V.\nabla C_A = D_{AB}.\nabla^2.C_A + S \quad (3)$$

In Eqns. (1-3), V is the velocity vector and P is the pressure. The velocity components in x and y directions are u, v respectively. The bulk density and viscosity of the fluid are ρ and μ respectively and S is the source term in the mass transport equation.

In the species transport equation (Eqn (3)), C_A refers to the concentration of a particular species (gmole/m^3) and D_{AB} is the diffusivity of the analyte in the liquid (m^2/s).

The heterogeneous reaction taking place at the surface is represented by the Michaelis-Menton equation as follows:

$$-r_A = \frac{V_m.[S]}{k_m + [S]} \quad (4)$$

where [S] is the analyte concentration in gmole/m^3, K_m is the Michaelis constant and V_m is the maximum reaction velocity (U/mg). The reaction rate is appropriately included in the transport equation for species as the source term for the heterogeneous reaction occurring at the wall.

BOUNDARY CONDITIONS

The following boundary conditions are used to solve equations (1)-(4)

- At the inlet, constant velocity and concentration of the species (analyte)
  $u = C_1$, $C_A = C_2$, C_1 and C_2 are constants. C_2 is fixed in all the cases at $6.8e^{-4}$ (mass fraction of analyte glucose in blood), while C_1 varies based on the Reynolds number under consideration.
- Outlet is specified as pressure outlet
  $P = P_A$, where P_A is atmospheric pressure.
- A no slip boundary condition for velocity near the wall, $u - u_w = 0$
- Mass flux near the wall is equivalent to reaction rate (Michaelis-Menton) at that region

$$-D_{AB}\frac{\partial [S]}{\partial n} = \frac{V_m.[S]}{k_m + [S]}, \text{ if reaction takes place}$$

$$-D_{AB}\frac{\partial [S]}{\partial n} = 0, \text{ if no reaction occurs}$$

Geometry of static mixers

All the channels tested in the study were of dimensions 200 × 400 × 18000 μm (W × h × L). This results in hydraulic diameter of 266 μm and aspect ratio of 0.5.

The first type of static mixer tested was a rectangular block, 100 μm long and 10 μm in width and placed perpendicular to the flow direction to increase the flow towards the wall. They were placed alternately along the length of the channel with an offset of 90 μm between two blocks. The depth of the static mixers and the channels were maintained the same. This arrangement was made to facilitate the coating of the side walls of the channel with the respective enzyme.

The elliptical mixer had an aspect ratio of 1.66 (major axis = 100 μm, minor axis = 60 μm). The angle of orientation of the ellipse with respect to x-axis was changed, with distance between the centre of the ellipse and the side wall kept constant. The distance was maintained at 70 μm. Offset between the static mixers was 100 μm, measured from the centre of the mixer. Simulation experiments were performed for angles of 0, 30, 60, 90, 120 and 150 with the mixer placed at 0 being streamlined to the flow.

A schematic showing the different static mixers is shown in Figure 1.

SOLUTION ALGORITHM

The simulations were carried out using the solver FLUENT of the commercial software ANSYS 13. The pressure-velocity coupling based on SIMPLE algorithm,[22] and first order upwind scheme were used for the discretization. FLUENT uses a finite volume based code for solving the transport equations, the domain being broken down into a number of grids. The flow of the fluid is pressure-based, with the outlet pressure fixed.

Parameters used in the study

The micro-channels with static mixer were compared with the channel without static mixer to ascertain their efficiency in terms of mixing and transverse mass transfer. To explicate this effect, the glucose assay reaction has been chosen. Accordingly, conversion of glucose in micro-channels with different geometries of static mixers has been determined. Glucose oxidase

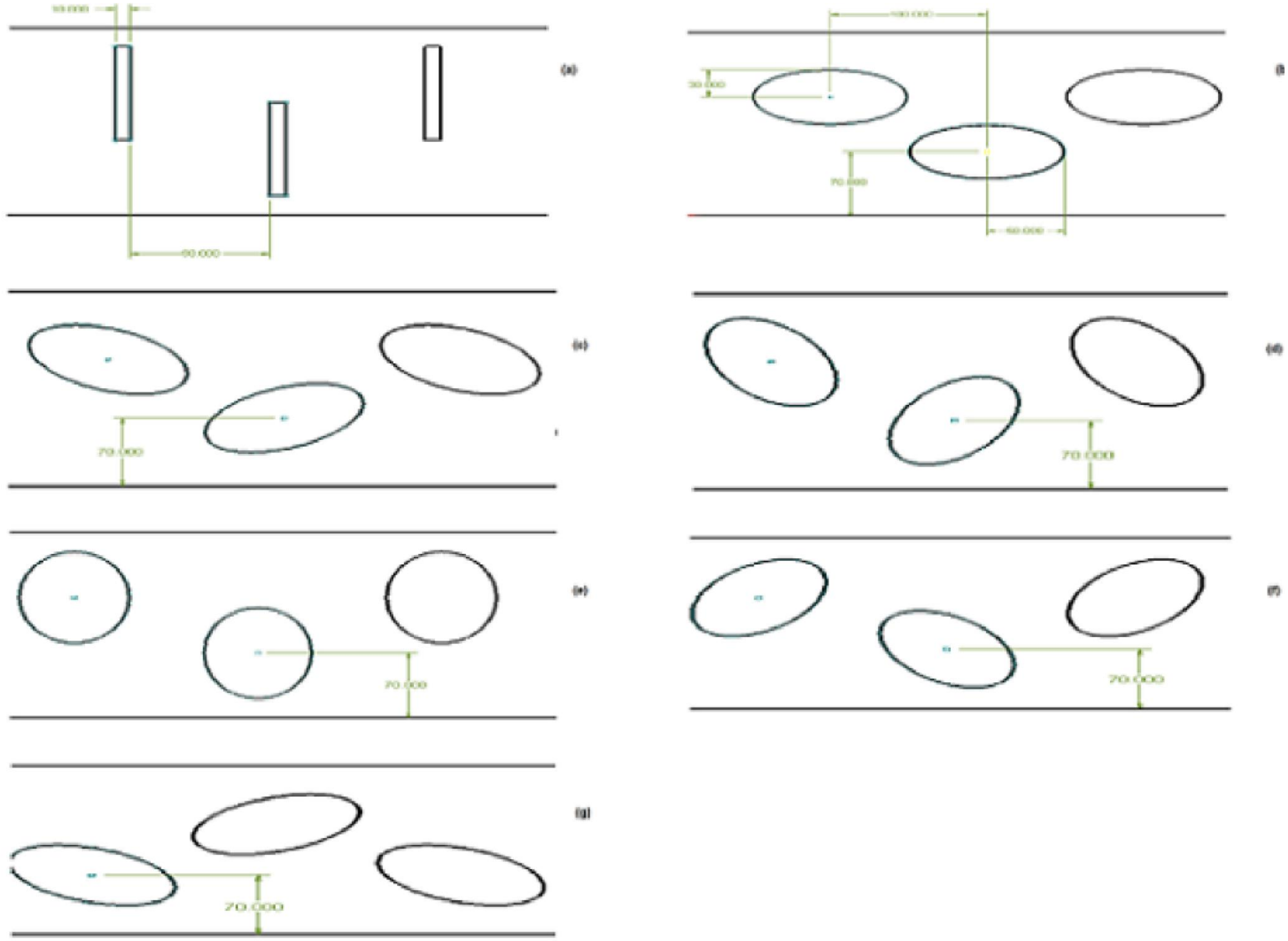

Figure 1 : Schematic of different static mixers shown over a section of the channel, a) rectangular static mixer, b) elliptical static mixer streamlined to the flow, c) elliptical static mixer at 30°, d) elliptical static mixer at 60°, e) elliptical static mixer at 90°, f) elliptical static mixer at 120°, g) elliptical static mixer at 150°.

enzyme is specific to the glucose assay test and the reaction catalyzed by the enzyme is as follows:

$$\text{Glucose} + H_2O + O_2 \xrightarrow{\text{Glucose oxidase}} \text{Gluconic acid} + H_2O_2$$

The K_m and V_m values are taken corresponding to glucose oxidase extracted from *A.niger* strain. The viscosity used corresponds to the viscosity of whole blood in physiological conditions. Use of whole blood viscosity facilitates testing the performance of such channels with static mixers in analytic devices where serum samples are often tested. The glucose concentration was fixed at 72 mg/dL which corresponds to a mass fraction of 0.0006792.

Grid sensitivity test

To ascertain the variation in the simulated results brought about by the grid sizing, three different grid sizes were tested for all the models. The element sizes corresponding to low, medium and high ranges were tested and the level of conversion at each range was compared. Structured grid was used for the micro-channel without static mixer, while unstructured meshing was adopted for those with static mixers.

RESULTS AND DISCUSSION

Grid sensitivity test

Grid sensitivity test was performed to ascertain number of grids required beyond which the simulation results are independent of the number of grids. This is required to ensure that the simulations results are not influenced by choice of number of grid elements or the grid size. The comparison between the simulation results for conversion (Figures 2 and 3) using different mesh sizes confirm that the difference between the predicted conversions using medium and higher number of mesh elements was negligible. Hence all the studies were carried out using the central range of element sizing.

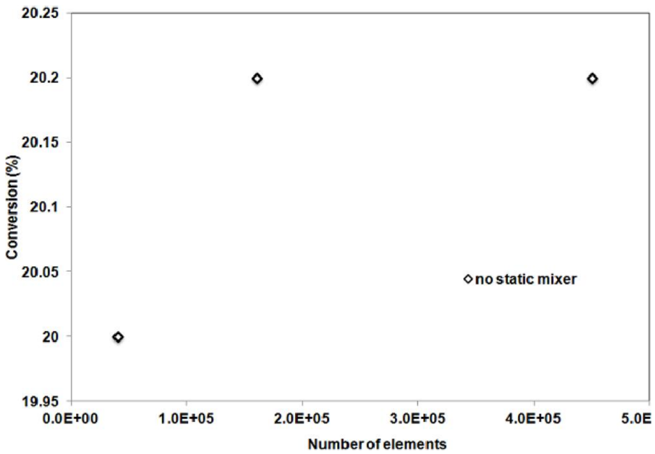

Figure 2 : Grid sensitivity test (Re = 0.1, channel without static mixer)

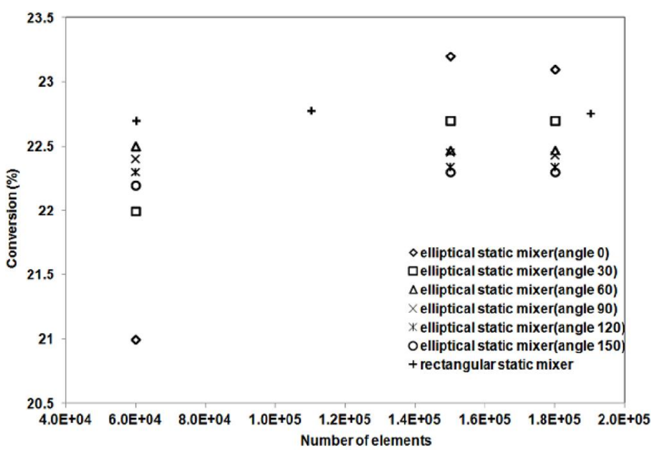

Figure 3 : Grid sensitivity test (Re = 0.1, channel with different static mixers)

Reynolds number of 0.1 has been taken as the model case for comparison of the different static mixers, as it lies in the range normally applicable to analytical devices. This reduces the computational time significantly to facilitate comparison of different geometries of static mixers. Under the conditions of simulation shown in TABLE 1, the conversion in microchannel reactor without any static mixer is 20 %.

Conversion in a micro-fluidic reactor can be increased by increasing the residence time either through the use of longer micro-channels or through use of lower fluid velocity. The use of a micro-channel with static mixers can lead to reduction in the analysis time circumventing the need for a longer reactor for the desired conversion.

TABLE 1 : Range of Reynolds number and values of other parameters used for simulations

Parameters	Values
Reynolds number range	0.01-100
Density of blood	1060 Kg/m^3
Viscosity of blood	0.0035 N/ms
Diffusivity of glucose	1E-9 m^2/s
K_m [23]	2.56 mM
V_m [23]	43.5 U/mg
Inlet glucose concentration	72 mg/dL

Residence time and analyte volume

A conventional micro-fluidic reactor with 18 mm long channel resulted in substrate conversion of 20 % at a Reynolds number of 0.1. Under these conditions, the residence time was found to be 14.4 s. Simulations carried out for reaction in micro-channels with rectangular static mixers of width 10 μm indicated that 20 % conversion was achieved within a residence of 11.78 s. The reduction in residence time for a fixed conversion (20 %) may be attributed to increased contact between the substrate and enzyme. The presence of rectangular static mixer has contributed to increasing the flux of substrate towards the wall on which enzyme was coated.

Figure 4 shows the effect of the mixer in increasing the lateral flux, where positive flux denotes flow towards one side wall and negative flux denotes the other side wall. The effect is shown for a length of 100 μm of the channel.

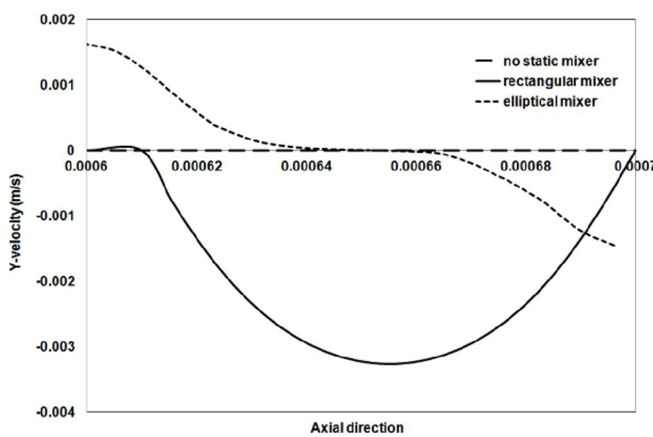

Figure 4 : Effect of mixers on lateral velocity. Simulation conditions Re = 0.1.

Due to reduction in the residence time, conversion of 20 % could be achieved in 15.5 mm long channel with rectangular static mixers, as compared to 18 mm long channel required in the absence of any static mixer.

The orientation of the rectangular static mixer with sharp edges in the direction perpendicular to the flow increases the total drag. There is a low pressure region created immediately behind the mixer which is more pronounced at higher velocities. The velocity vectors in Figure 5 for micro-channel with rectangular static mixer shows the presence of zones of lower velocity in lateral direction (y-component velocity) closer to the wall, behind the rectangular static mixers. This could be attributed to the sharp edges of static mixer. The presence of sharp edges leads to higher frictional losses and hence higher momentum loss for the fluid stream. This may be circumvented through the use of static mixers with rounded edges and surfaces. Hence, elliptical shaped static mixers have been chosen to study their influence of mixing and conversion in micro-channel reactors.

TABLE 2 shows the residence time required in micro-channel reactor equipped with elliptical static mixer, placed at different angles with respect to x-direction. It is evident that the residence required for 20 % conversion in micro-channel reactors with elliptical static mixers is in the range of 9.25-9.6s. Hence a reduction of 35.83-33.3 % in residence time can be achieved in micro-channels with elliptical mixers, when compared to micro-channels without any static mixer.

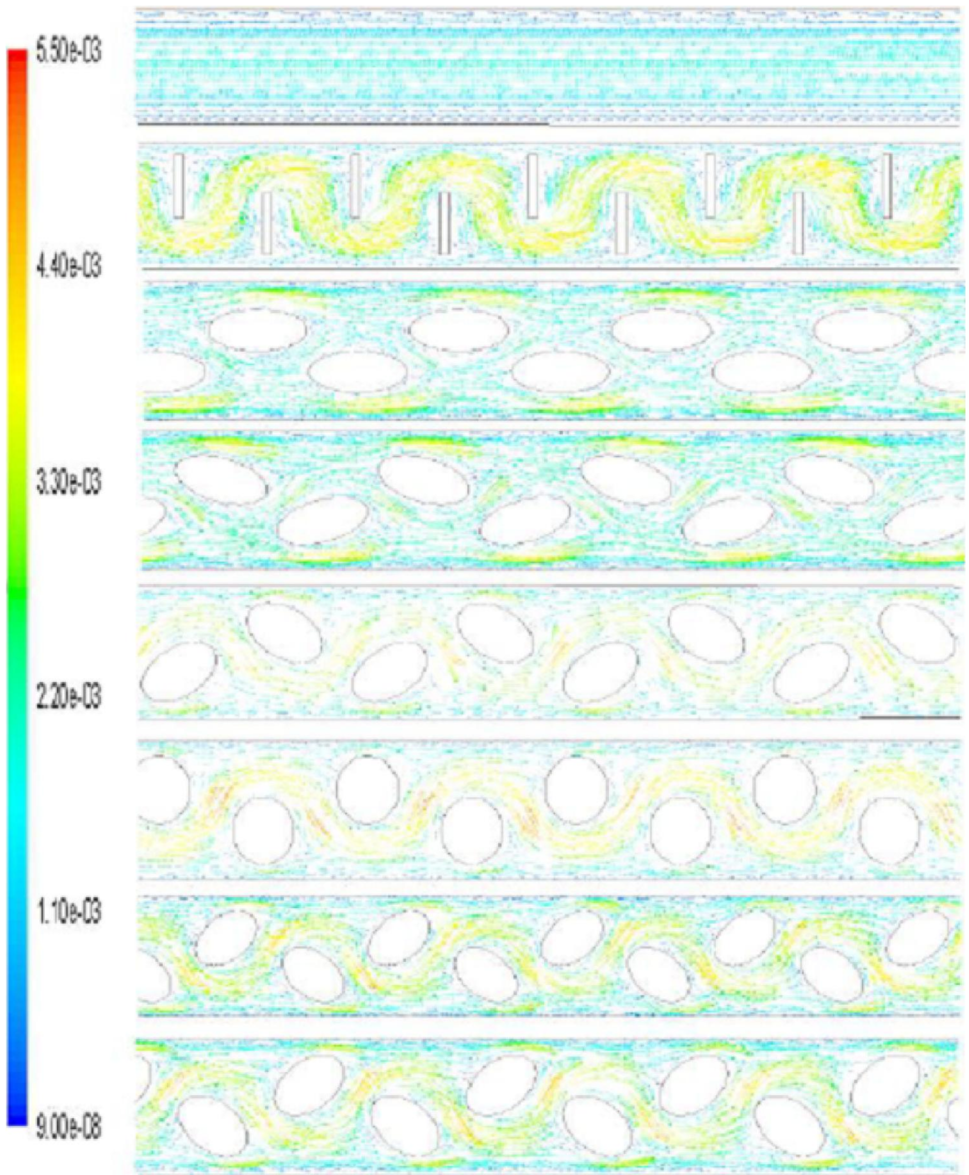

Figure 5 : Velocity vectors for Re = 0.1

TABLE 2 : Residence time and relative analyte volume to obtain a conversion of 20% in micro-channel reactors with static mixers. The simulation conditions are Re=0.1; Target conversion = 20 %. Relative analyte volume represents the analyte volume in a channel with static mixer with the channel without static mixer as reference.

Static mixer	Time (s)	Relative analyte volume (%)	Pressure drop (Pa)
No static mixer	14.4	100	19.7
Rectangular static mixer (15.5mm)	11.78	82	459
Elliptical static mixer (15.5mm)			
Angle 0	9.59	66.6	541
Angle 30	9.6	66.7	645
Angle 60	9.28	64.45	813
Angle 90	9.59	66.6	845
Angle 150	9.25	64.26	809
Angle 180	9.25	64.26	822

The reduction in analyte volume is also observed due to the presence of the static mixers. With 20 % conversion as the basis conversion, 18 % decrease in the required volume of the analyte was observed with the use of channel with the rectangular static mixers. A further decrease of 15-17.8% in the volume was observed with the use of the channel with elliptical static mixers. This makes the channel advantageous from three different aspects: Decrease in analysis time, requirement of a reduced amount of solution and reduction in the length of the channel.

Effect of Reynolds number (Re= 0.1 to 100)

Channel with no static mixers

A channel without static mixers gives the maximum possible conversion at low Reynolds number and when the velocity of the fluid is increased the conversion is reduced. The reduction in conversion was 99.8% when the Reynolds number was increased from 0.1 to 100. This effect is shown in Figure 6. Diffusion effects are predominant in comparison with convection at very low Reynolds number and hence the molecules are able to reach the surface of the channel for the reaction to take place. But at higher Reynolds number, the rapid flow of fluid limits the lateral diffusion.

Channel with rectangular static mixers

The conversion steadily drops in the channel with

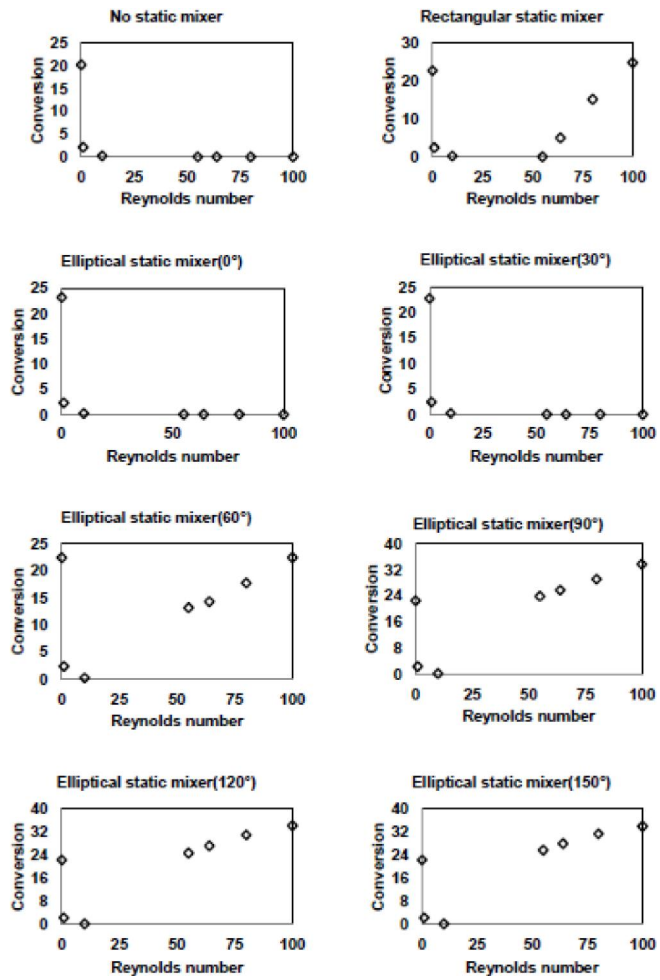

Figure 6 : Effect of change in Reynolds number on conversion, in the presence and absence of mixers.

rectangular static mixers till Reynolds number of 55 and increases after a Reynolds number of 64 as shown in Figure 6. A transition occurs in the mid region. At low Reynolds number the fluid remains attached to the mixers as it flows. With the presence of rectangular static mixers, the velocity is increased in the regions immediately adjacent to and in between mixers but the lateral diffusion is still limited with the flow near the wall largely undisturbed. The conversion is increased to 22.7 % as compared to 20 % in the channel without static mixer. This is complemented by the other advantages of decreased analyte volume and residence time.

At higher Reynolds number, the fluid as it moves past the mixer does not remain attached to it. This leads to the fluid being directed towards the wall and laterally moving fluid of higher velocity occurs in the region close to the wall. This leads to a conversion of 24.8 % in comparison to a conversion of 0.029 % at a Reynolds

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number of 100.

Channel with elliptical static mixer streamlined to the flow

In the microchannel with elliptical static mixer streamlined to the flow, the conversion steadily decreased with increasing Reynolds number. This effect was observed in the channel without static mixers also and is shown in Figure 6. At the lower Reynolds number 23 % conversion was attained. Also, the residence time was reduced in comparison to the channel without any mixer. The similarity in the conversion-Reynolds number relationship can be attributed to the fact that streamlining the body keeps the fluid attached to the surface of the mixer even at high Reynolds number. The conversion decreased by 99 % when a Reynolds number as high as 100 was used.

Channel with elliptical static mixer inclined at 30°

In the channel with elliptical static mixer inclined at 30° an inverse relationship was observed between conversion and Reynolds number with higher Reynolds number giving low conversions. However the conversion obtained at Reynolds number of 1 and 10 was higher by 4 % in comparison to the other positions of the elliptical static mixer. One of the important reasons for the observation of similarity in the conversion-Reynolds number relationship between the streamlined position and the static mixer at 30°, is the reduction of fluid flow in the central region.

Channel with elliptical static mixer inclined at 60°

In a micro-channel with elliptical static mixer inclined at 60°, the conversion reduces steadily till the tested Reynolds number of 10 and increases after the Reynolds number of 55. At Reynolds number of 55, the increase in conversion compared to the channel without static mixer is 99.7 % as the conversion seen is 13.176 % against a low conversion of 0.0477 % in a channel without static mixer. At higher Reynolds number, the conversion steadily increased yielding 22.5 % conversion at a Reynolds number of 100. It is observed that at low Reynolds number the elliptical static mixer fixed at 60° does not really affect the flow near the wall and the higher velocity regions are confined to the spaces between the static mixers. At higher Reynolds number however, there is an effect on the flow near the wall directing more fluid towards the wall. This relationship is seen in figure 6.

Channel with elliptical static mixer inclined at 90°

In a micro-channel with elliptical static mixer positioned at 90°, the conversion-Reynolds number relationship observed in figure 6 is such that there is a steady linear increase in conversion from Reynolds number of 55. The conversion observed at Reynolds number of 55 was 29 %, which is about 99.87 % increase when compared to the channel without static mixer. However higher pressure drop in comparison to all the orientations of the elliptical static mixer is observed.

Channel with elliptical static mixer inclined at 120°

The conversion-Reynolds number relationship in a microchannel with elliptical static mixer stationed at 120° shows a steady linear increase in the conversion from Reynolds number of 55. The behavior was observed with the elliptical static mixer oriented at 60° as can be seen in figure 6. A conversion as high as 31 % was observed at Reynolds number of 100, which may be attributed to pronounced effect on the flow behavior caused by the static mixer.

Channel with elliptical static mixer inclined at 150°

For all the Reynolds number above 55, the maximum conversion was observed in an elliptical static mixer stationed at angle of 150. The increase in conversion with Reynolds number shows an almost linear trend as observed in figure 6. Up to Reynolds number of 10, there is a decrease in conversion with increase in Reynolds number.

Figure 6 represents the effect of change in Reynolds number on the conversion in a microchannel reactor, in the presence and absence of static mixers.

Conversions at lower reynolds number (Re<0.1)

Simulations were performed at lower Reynolds number to ascertain the effectiveness of static mixers. The simulation results on the influence of Reynolds number on conversion in micro-channel without static mixer, micro-channel with rectangular mixer and micro-channel with streamlined elliptical static mixer are shown in Figure 7. The simulations were confined to these geometries due to computational expense involved at lower Reynolds numbers. The conversion in all the three channels simulated increased with reduction in Reynolds number. This is attributed to the lower fluid velocity and higher residence time. The rectangular static mixer provided an increase of 10.3 % and 6.24 % in conversion, with respect to the microchannel without static mixer at Re = 0.036 and 0.018 respectively while the channel with elliptical static mixer showed an increase in conversion of 10 % and 5.5 %. The analysis time reduction of 23.5% and 5.8% was observed in the channel with elliptical mixer and rectangular mixer respectively.

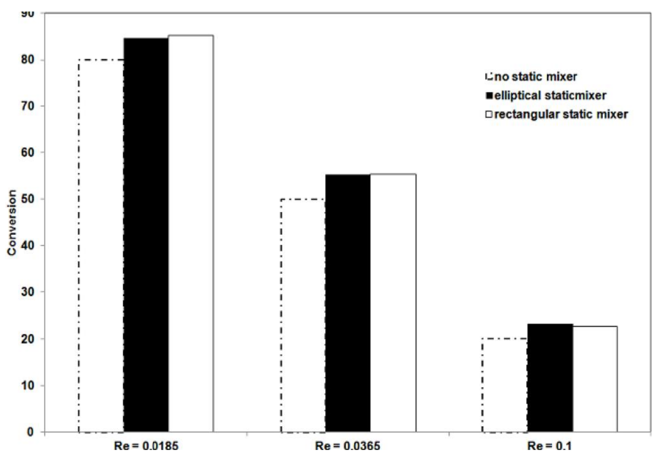

Figure 7 : Effect of mixers on conversion at lower Reynolds number.

CONCLUSIONS

From the present study on computational investigations of enzyme catalyzed reactions in microchannel reactors with static mixers, it can be concluded that the microchannel reactor with streamlined elliptical mixer resulted in higher conversion at lower residence times leading to lower requirement of analyte volume. A very high conversion was obtained at a Reynolds number of 0.1, which falls in the region of Reynolds number of interest for bioanalytical application. At Reynolds number < 55, the elliptical static mixer yielded the highest conversion when oriented at 30°. Above the Reynolds number of 55, the elliptical mixer inclined at 150° yielded the highest conversion. The angle of orientation at which the highest conversion was achieved is a function of Reynolds number due to variation of pressure drag with Reynolds number. Streamlining reduces pressure drop and may contribute to improving the structural integrity.

ACKNOWLEDGEMENTS

This work is supported by Grant No: ERIP/ER/0903807/M/01/1227 of Defence Research and Development Organisation, Govt. of India. The authors thank SASTRA University for the infrastructural support extended during the work.

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