A study of MHD-based Chaotic Advection to Enhance Mixing in Microfluidics using Transient Three Dimensional CFD simulations

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Abstract

Building on our previous work on transient, two dimensional simulations of the Navier-Stokes equations to investigate mixing enhancement by introducing the Lorentz force in MHD as a control parameter to create turbulent-like chaotic advection, this paper describes transient, three-dimensional simulations. Our approach differs from many previous analytical investigations by other workers based on potential flow and linearized stokes flow. A shallow disk-ring cylindrical microfluidic cell with gold electrodes deposited on the floor serves as a representative lab-on-a-chip. By applying a voltage across specific disk electrodes and a ring counter electrode, a current is established in the weak conductive solution. The current interacts with an externally applied magnetic field generating a Lorentz force that causes fluid motion. Velocity vectors, electric potential distributions and ionic current lines are presented. By switching on and off a pair of disk electrodes with a certain period T , a “blinking vortex” that induces chaotic advection is produced. Various particle trajectory-based analyses using extensive post-processing of the simulation results show that the period T plays an important role in generating chaotic advection. Large periods provide efficient stirring which improves mixing performance. Taking a step further, we show that by having two pairs of disk electrodes that were subjected to a different on/off switching scheme, more complex chaotic motion can be generated, and the mixing region can be extended to almost the entire fluid domain. This study establishes CFD simulation of MHD at the microscale as a robust tool to develop efficient strategies for mixing by chaotic advection. The techniques developed in the present work are also applicable in MHD-based flow control in microfluidics for other applications such as pumping and steering fluid to target locations.

Key words: MHD, microfluidics, mixing, chaotic advection, stirring.

1. Introduction

In recent years, there is a growing interest in lab-on-a-chip (LOAC) devices which hold promise in revolutionizing the diagnosis of illnesses, personalizing medical treatment, detection of chemicals in the environment, and synthesis of materials. In many of these applications, the core purpose is to accomplish pumping, mixing and flow control functions. Among them, efficient and rapid mixing is an especially important task since it has an effect on chemical reaction rates when multiple species are present for purposes such as medical diagnostics and chemical detection by chemical reactions, which are often mass transfer limited. However, due to the small size of these devices which are usually of centimeter or even millimeter order, the flows are always laminar and turbulent mixing techniques would not be applicable.

Fortunately, there is a variety of approaches that can be implemented to enhance mixing in lab-on-a-chip. Generally, these strategies can be categorized as either passive or active methods [1]. Passive micro-mixers are designed to use specific channel geometry configurations to increase the interface between the different constituents [2-4], while active ones are designed to control the flow by introducing non-intrusive driving forces or by actuating mechanical components to introduce flow patterns that would result in more efficient mixing. However, equally due to the small size of these applications, it becomes a real challenge to rely on moving mechanical components because of manufacturing complexity, high manufacturing cost and the increased likelihood of mechanical failures. Therefore, to introduce driving forces by other means to move fluid along desired trajectories to enhance mixing would be desirable. Electrostatic force and electromagnetic force are two main types of non-intrusive driving forces

that have been widely studied in recent years. Compared to electromagnetic force, electrostatic force usually requires a higher voltage to produce the same order of flow rate. Furthermore, significant Joule heating, bubble generation and electrode erosion are also the major drawbacks of the electrostatic technique. Electromagnetic force, namely the Lorentz force, provides a simple and flexible means to manipulate fluid flow in small devices, and the main requirement is that the fluid should be slightly conductive which can be easily met by most biological and chemical solutions [5].

Using electromagnetic force to manipulate fluid flow is by no means new. Magnetohydrodynamics (MHD) has been used in the past for pumping and flow control of highly conductive liquid metals and plasma [6]. In recent years, it has also been used to induce flow in weakly conductive electrolyte solution in redox (reduced species-oxidized species) MHD based systems. Electrochemical MHD based on redox electrode reactions has advantages such as negligible Joule heating and the absence of bubble generation. Experiments and CFD simulations show that the flow velocities of $\mu\text{m/s}$ or even mm/s are feasible by applying electric potential of $\sim 1\text{V}$ and magnetic flux density $B \sim 0.5\text{T}$ [7-11]. Undoubtedly, using electromagnetic force becomes an effective tool to manipulate flow in LOAC devices.

Before we discuss how to improve mixing by using MHD in microfluidics, it is important to clarify the mechanisms underlying the terms “stirring” and “mixing.” Generally speaking, the two phrases “stirring” and “mixing” imply very different physical processes. As Eckert observed in 1948 [12], advection alone increases the mean value of any initial gradient, and this effect of advection is appropriately called stirring. On the other hand, the effect of conduction or diffusion is to decrease the mean value of the gradient, and this is called mixing. Viscosity tends to slow down stirring which leads to increased mixing [13]. In other words, if we want to mix two initially separated constituents by stirring, the early stage of the process should be dominated by advection to stretch and fold the fluid elements to increase the interface area between two constituents to increase the concentration gradient and then allow mixing to take place by diffusion which will reduce the concentration gradient [14]. Researchers find it very insightful to use these two phrases to distinguish the “larger-scale” and “molecular-scale” processes that underlie mixing. Now the question is how to efficiently stir the fluid before molecular diffusion can smoothen the gradients. The first answer immediately comes to mind is to make the flow turbulent. Indeed, for large scale mixing, turbulent flow that provides the chaotic motion is useful and efficient. However due to the small size of the micro LOAC devices we study here, turbulent flow would not develop; instead a turbulent-like laminar flow which can produce similar chaotic motion would be of interest. This behavior of laminar flow is implied by the term “chaotic advection.”

The term “chaotic advection” was first introduced some thirty years ago by Aref as an outgrowth of the work on interacting point vortices in incompressible inviscid fluid [15]. A point vortex agitator inside a circular domain along with its image on the wall provided an unsteady potential flow. With the agitator being fixed at a certain position, the system is integrable and regular, and the system does not stir very efficiently. If, on the other hand, the agitator is moved in such a way (blinks between left and right repeatedly) that the potential flow is unsteady, chaotic motion can be induced and the system can stir very efficiently. This manner of agitation, now called “blinking vortex,” is a very simple way to produce chaotic motion, and has been the inspiration for many subsequent chaotic advection studies. Following Aref’s model, another model, the “journal bearing flow” has been widely investigated by experiments and numerical simulations by Aref [16], Swanson and Ottino [17] and Chaiken et al [18]. The devices are made up of an outer and inner cylinder that can rotate successively with a time period, and the process is repeated for several periods. With a specific range of time periods and rotation speeds, chaotic motion can be generated. Yet another type called “cavity flows” has been studied by Chien et al [19]. The “cavity flows” model relates to a two dimensional rectangular device with moving upper and lower walls which are switched on and off to start or stop successively with a time period, and the process is repeated for several periods. The common feature of the above three models is that they fall under potential flow [15] and Stokes flow [16-19], and their two dimensional flow fields can be exactly obtained by analytical tools. Once the velocity field is obtained, particle trajectories-based

analyses can be conducted to investigate chaotic motion. The on/off switching scheme is a simple way to produce an unsteady flow. The investigators of the above studies recognized that the two dimensional kinematics of advection by an incompressible flow is equivalent to the Hamiltonian dynamics of a one degree of freedom system which has been well understood as chaotic since the mid-1960s. These observations have helped build the theoretical bridge between chaotic advection in fluid mechanics and chaos in classical mechanics.

It is not surprising that chaotic advection driven by electromagnetic force in microfluidics has been studied in the past decade since Lorentz force provides the possibility to manipulate fluid flow in a controlled manner instead of moving mechanical agitators or walls. Yi et al [20] perhaps were the first group to the best of our knowledge to study chaotic advection by using electromagnetic force. They investigated a microscale cylindrical cell with its axial dimension much smaller than the diameter. Switching the positions of the point electrodes placed on the bottom surface could produce the “blinking vortex” in their MHD stirrer. The governing equations under the Stokes flow and quasi-steady assumptions were solved and compared to their experiments. They also studied several rectangular ducts with electrodes deposited on the bottom or side walls to trigger chaotic motion by using switching schemes [21,22]. Another interesting work has been done by Rossi et al [23]. They conducted experiments in a small flat rectangular cell with a magnet moving underneath. By doing so, three typical flow sequences were created, and lamination, stretching and mixing performance were investigated. Dufour et al [24] performed experiments in a shallow cavity and compared their results to those using linearized equations under the Stokes flow assumption. Gopalakrishnan and Thess [25] studied glass melt homogenization by stirring and mixing of flow in a pipe mixer subjected to electromagnetic forces by using computational methods. Yuan and Isaac [26] studied mixing in microfluidics by chaotic advection by applying a sinusoidal potential difference across the electrodes by performing unsteady, two-dimensional CFD simulations. They found that off-axis placement of the working electrode cylinders and using various switching schemes made the flow more chaotic and enhanced mixing.

Dukhin [27] and Squires and Bazant [28] have studied an electro-osmotic effect caused by induced charge on dipolar surfaces. This effect arises from the inhomogeneous surface charge induced on the electrode in order to maintain an equipotential surface in the presence of an applied field. Squires and Bazant [28] give an expression for the slip velocity at the surface of a conducting cylinder as $U_0 \sim \varepsilon E_0^2 a / \mu$, where ε is the dielectric constant of the electrolyte, E_0 is the applied electric field gradient, a is the cylinder radius, and μ is the dynamic viscosity. The authors then give qualitative descriptions of how such effect could be present on planar metallic surfaces, but do not outline a method to quantify the effect. It is beyond the scope of the present work to incorporate this effect as is evident from models for spheres and cylinders studied by Squires and Bazant [28]. However, this effect might be important, judging from the vast amount of literature on the topic. From the E_0^2 -dependence we see that this lesser known electro-osmotic effect is non-linear, and strongly dependent on the magnitude of electric field gradient E_0 . Also note that, in the context of mixing, this effect would aid mixing, since it introduces a tangential velocity on the metal surface at its floating potential, unlike the vortical flow around the active electrodes caused by the Lorentz force which leads to the formation of islands of less mixed state.

In this work, three dimensional numerical experiments are conducted in a microfluidic cell to investigate mixing performance by chaotic advection driven by electromagnetic force. An uncapped shallow cylindrical cell with gold electrodes deposited on the bottom surface is used (Fig. 1). By judiciously choosing the number of active electrodes and the switching scheme, a “blinking vortex” and other types of chaotic motion are produced. Once the flow field is obtained by numerical solution of the Navier-Stokes equations, particle trajectories-based analyses are conducted to investigate the degree of chaotic advection and mixing performance, including Poincaré maps and particle concentration maps. Comparisons to selected Stokes flow simulations are also included.

2. Mathematical model and governing equations

This section briefly describes the mathematical model for the Magnetohydrodynamics (MHD) system. The electromagnetic force, namely the Lorentz force, is defined as followed

$$\mathbf{F}_L = \mathbf{J} \times \mathbf{B} \quad (1)$$

Where $\mathbf{B}$ is the magnetic flux density and $\mathbf{J}$ is the current density vector being carried in the flow by the ionic species when an external potential is applied. The current flux vector $\mathbf{J}$ has two contributions as given by equation (2).

$$\mathbf{J} = \sigma(-\nabla\phi + \mathbf{V} \times \mathbf{B}) \quad (2)$$

Here σ is the electrical conductivity of the electrolyte, ϕ is the electric potential, and $\mathbf{V}$ is the velocity vector. The first term on the right hand side of equation (2) is the migration term while the second term is the so-called induced electric field term that can be neglected in most studies involving weak electrolytes. Therefore the Lorentz force now can be written in the simpler form as

$$\mathbf{F}_L = -\sigma \nabla\phi \times \mathbf{B} \quad (3)$$

We neglect the thin double layer region, and therefore the electric potential ϕ satisfies the Laplace equation in the conducting solution outside the double layer.

$$\nabla^2 \phi = 0 \quad (4)$$

The flow is assumed to be incompressible, and therefore the velocity field is divergence-free.

$$\nabla \cdot \mathbf{V} = 0 \quad (5)$$

Just like the gravitational force, the Lorentz force is a body force which is included in the momentum equation

$$\rho \frac{\partial \mathbf{V}}{\partial t} + (\rho \mathbf{V} \nabla) \cdot \mathbf{V} = -\nabla p + \mu \nabla^2 \mathbf{V} + \mathbf{F}_L \quad (6)$$

where ρ is the fluid density, p is the pressure, and μ is the dynamic viscosity. Since the current density generated in this system is quite low, Joule heating effect can be neglected. Therefore it is appropriate not to solve the energy equation. Also because of negligible Joule heating, buoyancy due to temperature gradients can be neglected. Therefore the gravity term is not included in equation (6). However, density gradients and natural convection can exist due to the tendency to attain electroneutrality in a conducting medium. Isaac et al [7] have proposed a non-dimensional parameter called the TN number to help determine if natural convection will be important in mixed convection problems. Including a model for this effect requires careful consideration, and it is beyond the scope of this work, but is a topic of ongoing effort.

Since the analyses in the following sections are based mostly on particle trajectories, accurate positions of the particles which are initially injected into the computation domain are required, and need to be updated at each time step. The motion of a passive particle can be tracked by numerically integrating the advection equation shown below

$$\frac{d\mathbf{X}(t)}{dt} = \mathbf{V}(\mathbf{X}, t) \quad (7)$$

where $\mathbf{X}(t)$ is the particle position vector at time t which has the initial condition $\mathbf{X}(0) = \mathbf{x}_0$. The velocity vector $\mathbf{V}$ is first obtained by solving equations (4), (5) and (6) with appropriate boundary conditions, and equation (7) is then integrated to obtain the position vector.

3. Simulation model Description

This section describes the simulation model and numerical method we used in this work. A three dimensional uncapped cylindrical microfluidic cell with a ring serving as the counter electrode and four disks serving as the working electrodes is used. The electrodes all made of gold are deposited on the bottom surface. This cylindrical cell has a radius $r_l = 3\text{mm}$, the outer and inner radii of the ring counter electrode, respectively, are $r_2 = 2.4\text{mm}$ and $r_3 = 2\text{mm}$, respectively, and four disks with a radius $r_d = 0.16\text{mm}$ are located at 3, 6, 9 and 12 O'clock positions with their centers located on a circle concentric with the circular bottom face having a radius $r_4 = 1\text{mm}$. The cell height $H = 0.5\text{mm}$. The model geometry is shown in Fig. 1. By selectively activating the disks (working electrodes) labeled A, B, C and D, regular or chaotic flows can be achieved. For example, by switching on and off disks A and C and keeping disks B and D are at their floating potential, the “blinking vortex” can be reproduced. Because electrodes B and D are at their floating potential, no current will be flowing through them.

In all simulations, the cell is filled with NaCl solution. The simulation data are given in Table 1. Note that the potential ϕ for the electrode boundary condition in the simulations is the electric potential at the outer edge of the double layer. The electrical double layer is a very thin layer (~ 1 to $\sim 100\text{nm}$ thick) across which the electric potential drops dramatically. By using Gouy-Chapman Theory and Poisson-Boltzman equation, we calculate the potential at the outer edge of the double layer to specify the electrode boundary condition to solve equation (4). The ionic concentrations around a central ion is assumed to be related to the potential by the Boltzmann distribution

$$c_i = c_i^* \exp\left(-\frac{z_i F \phi}{RT}\right) \quad (8)$$

where c_i^* is the average concentration of species i in the electrolyte solution, and ϕ is the electrostatic potential established around the central ion. The product $z_i F \phi$ is the electric interaction energy per mole; other contributions to the interaction energy are ignored. Poisson's equation relates the potential variation to the charge density. The potential distribution has contributions from other ions which is described by Poisson's equation

$$\nabla^2 \phi = -\frac{\rho_e}{\epsilon} = -\frac{F}{\epsilon} \sum_i z_i c_i = -\frac{F}{\epsilon} \sum_i z_i c_i^* \exp\left(-\frac{z_i F \phi}{RT}\right) \quad (9)$$

For the double layer we use the one-dimensional form of Poisson's equation.

$$\frac{d^2 \phi}{dx^2} = -\frac{F}{\epsilon} \sum_i z_i c_i^* \exp\left(-\frac{z_i F \phi}{RT}\right) \quad (10)$$

where x is the distance from the electrode. The boundary conditions are

$$\phi = \phi_0 \quad \text{at} \quad x = 0 \quad (11)$$

and

$$\frac{d\phi}{dx} \rightarrow 0 \quad \text{as} \quad x \rightarrow \infty \quad (12)$$

The solution to equation (10) with the boundary conditions, equations (11) and (12), is

$$\frac{\tanh(K\phi)}{\tanh(K\phi_0)} = \exp(-\kappa x) \quad (13)$$

where $K = Fz / (4RT)$ and $\kappa = (2F^2 / \varepsilon RT)^{1/2} z \sqrt{c^*}$, z is the magnitude of the charge number, ε is the permittivity of the medium, and T is the temperature. For dilute aqueous solutions, the ratio of the permittivity to the permittivity of free space at 25°C, $\varepsilon/\varepsilon_0 = 78.49$. Substituting for the constants, we get the following expression for κ (cm⁻¹).

$$\kappa = 0.329 \times 10^8 z \sqrt{c^*} \quad (14)$$

where c^* is in M (moles/liter). Using this model, we estimated $\phi = 0.0379V$ at the outer edge of the double layer with $c^* = 0.1M$, $x = 1 \text{ nM}$ (10^{-7} cm), and externally applied potential $\phi_0 = 1V$, indicating a ~96% potential drop in the double layer. Because it is only an estimate, we rounded off ϕ to 0.04V. Our approach appears to be reasonable based on satisfactory agreement with reported chronoamperometry experiments that facilitated comparison of velocity magnitudes in a disk-ring microfluidic cell [29].

The magnetic field is in + z direction with an intensity $B = 0.36T$. Previous experimental studies have shown that a uniform magnetic field in the cell region of the size similar to ours is possible by using a rectangular ~25 mm x ~25 mm x ~12.5 mm, Nd-Fe-B permanent magnet [10]. Their measurements showed a maximum variation of ~5% in B_z over the cell height of 650 μm . Note that our cell height, $H = 500 \mu\text{m}$. Due to the larger lateral dimension of the magnet compared to the 3 mm radius of our circular cell floor, variation in the B_x and B_y components across the cell diameter would be much smaller.

As we mentioned, the “blinking vortex” is a simple model that provides chaotic advection in Stokes flow proposed by Aref [16]. Inspired by the “blinking vortex” model problem, we have designed the disk and ring configuration. By activating the ring and disk A during the first half of the period, a clockwise rotating flow is generated, which can be treated as an agitator in Aref’s model [16]. During the second half of the period, deactivating disk A while activating disk C produce another clockwise rotating flow on the right, which can be considered as another agitator. This process is repeated over several periods to achieve chaotic flow. Both disks B and D are at their floating potential in these simulations. Mathematically, the potential boundary conditions for equation (4) are

$$\begin{cases} \phi_A = 0.04V & \phi_C = 0 & kT < t < kT + \frac{T}{2} \\ \phi_A = 0 & \phi_C = 0.04V & kT + \frac{T}{2} < t < (k+1)T \end{cases} \quad (15)$$

where $k=0,1,2,\dots$ is an integer and the resulting flow field is periodic in time with period T . The time period T is an important parameter which determines whether the flow is regular or chaotic. Zero current flux ($\nabla\phi = 0$) boundary condition is specified on all surfaces except the active electrodes. This is also the boundary condition for the electrodes at their floating potentials. For the hydrodynamic boundary condition, the no-slip boundary condition is used at the walls and the top surface is treated as a free surface with no motion perpendicular to it.

Equations (4)-(6) with the above boundary conditions and appropriate initial conditions are simultaneously solved by the commercial software package ANSYS FLUENT [30]. The specific aspects of MHD simulations such as the presence of Lorentz force in the momentum equation and time dependent boundary conditions can be implemented by using UDFs (user defined functions) which can be added to the standard FLUENT solver. The

Laplace equation (4) for the electric potential is solved as a UDS (user defined scalar). Since time is an important variable in mixing and flow control problems, a time-accurate, transient simulation is required. An unsteady, second-order implicit method is used for time discretization.

4. Results and Discussion

4.1 Overall Features of Flow and Electric Fields

Scheme1: In this scheme, disks A and C are activated. Fig. 2 below shows velocity vectors (a), velocity profile along the x-axis (b), electrical potential contours and ionic current flux vectors (c) in the $z = 0.4$ mm plane, and the potential contours, current flux lines, velocity vectors in the $y = 0$ plane (d), and velocity profile in the $x = 0$ plane (e), for case $T = 4$ s at the end of the first half period. From Figs. 2c and 2d, in top view and side view, respectively, the electric potential has the largest value at disk A and decays toward disk C and the ring. The ionic current flux vectors are shown in Figs. 2c and 2d, and the ionic current flux lines which are perpendicular to the equipotential lines are shown in Fig. 2d. From Fig. 2c, we see that that current flux vectors are pointing outward from the disk, and with the magnetic field vector $\mathbf{B}$ pointing in the $+z$ direction, a clockwise Lorentz force is generated by virtue of the cross-product rule (equation 3), and causes a clockwise rotating flow as shown in Fig. 2a. As we expect, we see from Fig. 2d that the current flows from the activated disk A to the ring and disk C. The total current has a value $I = 30.1\mu\text{A}$. The maximum and minimum values of the z -component of the velocity are $\sim \pm 1\text{e-}7$ m/s, much lower compared to the x - and y -components that can be seen in Fig. 2e and Fig. 2b. This order of magnitude difference in the z -component velocity magnitude reflects corresponding smaller z -dimension of the cell compared to its radius. This behavior is as would be expected in a shallow liquid layer unless perturbed by forces that would create large-amplitude waves. A feature that arises from the z -component of velocity is a weak vortex that forms in the x - z plane that extends from Electrode A to Electrode C (graphical results not included in the figures).

Fig. 3 shows the potential contours and current lines when electrodes A and C are active and all other electrodes are at their floating potential. As can be seen from the potential contours, the induced charge electro-osmotic effect discussed in the *Introduction* (Section 1) would be smaller at the locations of Electrodes B and D in Fig. 3, compared to placing them closer to Electrodes A and B where the field gradients will be larger.

Scheme 2: In order to create more chaotic flow which will cause more efficient mixing, we introduce another on and off switching scheme that employs all four working electrodes. The corresponding electric potential boundary conditions are

$$\begin{cases} \phi_A = 0.04\text{V} & \phi_C = -0.04\text{V} & \nabla\phi_B = \nabla\phi_D = 0 & kT < t < kT + \frac{T}{2} \\ \nabla\phi_A = \nabla\phi_C = 0 & \phi_B = 0.04\text{V} & \phi_D = -0.04\text{V} & kT + \frac{T}{2} < t < (k+1)T \end{cases} \quad (16)$$

By doing so, during the first half period, disks A and C generate a pair of counter-rotating flows, and then by activating disks B and D during the second half period, another pair of counter-rotating flows is generated. For simulations of this nature, the choices are infinite. Here we do not address the optimization of the schemes, which is beyond the scope of the present work. However, our experience shows that various switching schemes lead to interesting flow patterns, indicating the strong dependence of mixing performance on the number of electrodes and the switching schemes. One important contribution of this work is in the use of larger number of electrodes and more complex switching schemes than used by previous investigators. Fig. 4 presents the flow properties for Scheme 2 at the end of the first half period. Fig. 4c shows the equipotential contours and the current flux vector map at the end of the first half period, while the corresponding current flux lines are shown in Fig. 4d in cross-sectional view.

The root mean square velocity and the maximum velocity in the computation domain for schemes 1 and 2 are $U_{rms} \sim 0.1 \text{ mm/s}$ and $U_{max} \sim 0.5 \text{ mm/s}$, which gives the Reynolds number based on U_{rms} , $Re_H = U_{rms}H/\nu \sim 1$. For this order of magnitude of Re_H , the flow is clearly laminar which guarantees that the chaotic characteristics arising in the cases considered in this study are not contributing to the development of turbulent flow. The Hartmann number appropriate for the present work can be defined as $Ha = \sqrt{\sigma \phi B H / \mu U_{rms}}$, the square root of the ratio M of the Lorentz force to viscous force. In our case, $Ha \sim 1$ which indicates that both the forces exert more or less equal influence on the flow.

As we mentioned, all the particle trajectory based analyses in the following sections are computed by post-processing of the flow field obtained by performing CFD simulations first. In order to integrate the advection equation (7), the Discrete Phase Model (DPM) available in FLUENT [30] is used. In DPM, massless particles are injected into the computational domain at specific initial positions selected by the user. After the main solver yields the flow field, equation (7) is integrated by using a 5th order Runge-Kutta method to obtain the successive particle positions. The next section presents the analyses on mixing performance including the Poincaré map and concentration of numerical particles.

The Navier-Stokes simulation results are compared to the Stokes flow results under potential step for 3 values of the Reynolds number Re_H in Figs. 5-7. In these potential step simulations, a potential difference is applied between Electrodes A and C. Electrodes B and D are inactive and they are treated the same as the glass substrate for the boundary conditions. Experimentally, however, a separate cell without electrodes B and D would need to be used to conform to this boundary condition. The velocity vectors and the streamlines at $t = 8 \text{ s}$ show clear differences between the two as the Reynolds number increases. The loss of symmetry about the $y = 0$ plane is clear in the Navier-Stokes results compared to the Stokes flow results. These results show similarity to those from flow over a cylinder. When Stokes flow assumptions are valid, the streamlines show symmetry between the upstream. The explanation for this behavior is that Stokes flow is dominated by the diffusion of vorticity, whereas at higher Reynolds numbers, vorticity is swept downstream by convection resulting in loss of symmetry, also evident in Fig. 7 for $Re_H = 14.98$. However, since these results are given only to demonstrate the differences between the two models, a comparison of mixing quality would require simulations using Schemes 1 and 2 for larger values of t , which is beyond the scope of this paper.

4.2 Poincaré map

A Poincaré map shows the long term behavior of trajectories of several different particles starting from their initial positions. If the initial points are selected appropriately they will indicate the behavior of all particles originating from everywhere in the domain. After the initial points are selected, the new particle positions at the end of each time period will be recorded after each time period and then all particle positions at each time period will be aggregated in one map. Generally, there are two types of behavior a Poincaré map can indicate, ‘regular’ and ‘chaotic’. A ‘regular’ behavior implies that the particle trajectories are confined to regular curves. While a ‘chaotic’ behavior is indicated by the particle positions that are scattered throughout the domain. It is worth mentioning that a Poincaré map shows regions where the initially placed particles go through or are forbidden from entering regardless of how many time periods are considered. If the system is chaotic, the forbidden regions will get smaller and finally disappear as the number of periods increases.

To create Poincaré maps, five initial points are selected as follows: $P_1(x=0.2, y=0, z=0.4) \text{ mm}$, $P_2(x=0.5, y=0, z=0.4) \text{ mm}$, $P_3(x=0.8, y=0, z=0.4) \text{ mm}$, $P_4(x=1.2, y=0, z=0.4) \text{ mm}$ and $P_5(x=1.5, y=0, z=0.4) \text{ mm}$ (Fig. 8a). Figs. 8b – 8f show the Poincaré maps with different time period $T=1 \text{ s}$, 2 s , 4 s , 8 s and 10 s . The corresponding elapsed periods are shown in Table 2. From Fig. 8b, we see regular behavior for case $T=1 \text{ s}$ since all five tracks are confined in five distinct regular curves. Note that if $T=0$, the system yields the steady two fixed-agitator system with its pathlines and streamlines coinciding. The streamlines of the two fixed-agitator system are similar to those in Fig. 8b. Therefore,

as the time period T approaches zero, the system will look more and more like the two fixed-agitator system, hence the similarities between the Poincaré map in Fig. 8b and the streamlines of the two fixed-agitator system. For case $T=2s$, the system becomes a little more chaotic since the innermost particles start to scatter to the outer region, hence the innermost regular curves begin to become more irregular while the outer curve still remains regular. This behavior has good agreement with the results from Aref's "blinking vortex" experiments [15]. For case $T=4s$, the system becomes more chaotic with the particles appearing mostly everywhere in the part of the domain inside the ring. However, two distinct islands mostly devoid of particles can be found. The appearance of the particle-free islands indicates that the particles placed at the locations to create the maps do not enter the islands regardless of how many periods are considered. This is likely to be due to the disk electrode being of finite size whereas Aref [15] used point vortices. Furthermore, one can see that the particles distinguished by their colors are restricted in some specific tracks though some hint of dispersion can be recognized (black circles). However the existences of this kind of clustering is not found neither in Aref's [15] results nor in Bau's results [20], which is not surprising since Aref [15] and Bau [20] relied on the results of their approximate analytical models which might have missed some features that appear in the present CFD simulations based on the full Navier-Stokes equations. It is reasonable to expect that when the full Navier-Stokes equations are considered, the system would be more prone to chaos since non-linear systems have a greater tendency toward chaos. For cases $T=8s$ and $10s$, the system can be considered globally chaotic, and the particles are dispersed almost everywhere in the region inside the ring, and no distinct clusters are found.

4.3 Concentration of numerical particles

In order to quantify mixing performance, we designate the two halves of the domain randomly seeded with particles of two different colors. Totally 500,000 black and grey numerical particles which represent two non-diffusive dyes are seeded into the $z=0.4$ mm plane. The black and grey particles, equal in number, are initially distributed evenly in the left and right halves (Fig. 9a) of the domain, and then the particles are tracked at each time step through integrating the advection equation (7). Figs. 9b – 9f present the particle distributions for different time periods T at $t=200s$. A good agreement between the particle distribution maps and Poincaré maps can be found. For case $T=1s$ (Fig. 9b), we see a regular structure which is very similar to that in the corresponding Poincaré map. Obviously, for this case, the mixing is not efficient since the black and grey particles are segregated into bands. For case $T=2s$ (Fig. 9c), the situation becomes better since some particles in the interior begin to scatter and mix in the region enclosing the two disks, which is also reflected in the corresponding Poincaré map (Fig. 8c). For case $T=4s$, mixing performance is higher, and the mixed region is larger. It is interesting that the islands seen in the Poincaré maps also appear in the particle distribution maps. For cases $T=8s$ and $10s$, the mixed region continues to grow. Gopalakrishnan and Thess [25] have presented similar maps for a range 0 -1000 of M , the square of the Hartmann number defined in section 4.1. As their results show, mixing performance improves at higher values of M . For our system $M \sim 1$, and the flow is driven only by the Lorentz force without any inflow or outflow boundaries. As a result, only a qualitative comparison between the two is possible, which shows that the concentration maps of the passive scalars are similar in both.

In order to quantify the mixing performance, a parameter called mixing quality has been defined [22, 26]:

$$\alpha(t) = 1 - \frac{\delta^2(t)}{\delta^2(0)} \quad (17)$$

Where $\delta^2(t)$ is the standard deviation of the dimensionless concentration C at time t which can be expressed as:

$$\delta^2(t) = \iint_s [C(x, y, t) - \bar{C}]^2 dx dy, \quad (18)$$

where $C(x,y,t)$ is the local concentration that varies with time and $\bar{C}$ is the constant average concentration. The concentration is computed from the particle number density displayed in Fig. 9.

Fig. 10 shows the time-evolution of mixing quality $\alpha(t)$ for different values of T . The set of curves shows that increasing the time period T improves $\alpha(t)$. The reason why the mixing quality is not close to unity even in the best case ($T = 10$ s) for which it is slightly less than 0.4, is that the integration domain in equation (18) is the entire rectangular cut surface in the vertical plane passing through the center. However, mixing is mostly confined to the region inside the ring counter electrode even for the best case, and little mixing occurs near the side walls even for the longest time, $t=200$ s (Fig. 10). A number fraction f_{black} of the black particles in a given cell area in the $z = 0.4$ mm plane can be defined as

$$f_{black} = n_{black} / (n_{black} + n_{grey}) \quad (19)$$

where n with the subscripts represent the number of black and grey particles in a given cell area. When mixing is complete in a given cell area $n_{black} = n_{grey}$, and $f_{black} = f_{grey} = 0.5$. Table 3 lists the mixed area (mm^2), defined as the sum of the cell areas in the $z = 0.4$ mm plane for which $f_{black} = 0.5 \pm 0.02$, and its percentage of the total area for different cases.

4.4 Features Revealed by CFD Simulations of the Navier-Stokes Equations

It'd be helpful to compare our results to Dufour et al [24] and highlight similarities and differences. They also used a shallow cylindrical cavity, but of a larger radius (17.5 mm), and the electrodes were non-intrusively placed on the cavity floor. Based on the cavity and the electrode dimensions, their device can be classified as mm-scale. Their Reynolds number is of the same order (~ 1) as ours. Their time periods and mixing times are an order higher compared to ours. As to the model itself, they used the linearized Stokes flow model, whereas we used the full Navier-Stokes model. Their magnetic flux density is 0.05T whereas ours is 0.36T. There is also another major difference between the works of Dufour et al [24] and ours in the electric potential boundary conditions. They used an electrode potential of 0.4 V in their simulations, whereas we used 0.04V after accounting for the steep double layer drop. Considering that the conductivities are similar (~ 1 S/m), the total current should scale as electrode area under the same potential boundary conditions. The kinematic viscosity of their electrolyte solution is ~ 3.6 times that of ours. The maximum velocity in their work is larger by a factor of 10. They took into account the variation of the z -component of the magnetic flux density B_z with radius. Since the radius of the cavity in our case is only 3 mm, we used a constant value for the magnetic flux density component B_z . Isaac et al [7] have shown that including all the three components of the magnetic field when it is non-uniform and not pointing along the symmetry axis significantly affects the flow field.

Our results show that efficient mixing is possible in microscale devices with time periods of potential switching and elapsed time, a surrogate for a lab-on-a-chip response time, an order of magnitude less than reported by Dufour et al [24]. The main reason for the shortening of the mixing time is the choice of the number of disk electrodes, their placement and the switching schemes. As suggested by previous investigators, unsteady, three-dimensional flow fields lead to better mixing compared to unsteady two-dimensional flow fields, even though the latter is more amenable to visual interpretation and comparison to Hamiltonian systems. As in other optimization problems involving geometric parameters, optimizing system parameters for mixing in microfluidics is challenging due to the large number of parameters such as actuator eccentricity, number of actuators, and switching schemes. Nevertheless the present MHD-based mixing schemes are promising due to its simplicity and the ability to provide actuation without relying on mechanical stirring.

5. Conclusions

In this study, numerical simulations are conducted to investigate the mixing performance enhancement by chaotic advection by using MHD in a disk-ring microfluidic device. The externally applied potential and the magnetic flux density, 1V and 0.36T, respectively, are smaller compared to those in other works, and the maximum velocity is also correspondingly smaller ($\sim 0.5\text{mm/s}$ vs. 10mm/s), it is still possible for the described MHD microfluidic cell to achieve good mixing by creating chaotic advection. By switching on and off two disks (working electrodes), flow pattern similar to that in the “blinking vortex” model has been created. For small periods, the flow patterns are regular and the mixing performance is poor. By increasing the period, more chaotic motion can be generated, thus improving the stretching and deformation of material lines and enhancing mixing performance. By having two pairs of electrodes switched according to the prescribed scheme, more complex chaotic advection has been generated that led to enhanced mixing. The time period T in this scheme is an important parameter that dictates whether the flow will be regular or chaotic. For small T , the motion is regular which is reflected by its regular curves shown in the corresponding Poincaré map. The particle concentration maps of the small T cases also reveal the poor mixing performance even after many switching cycles. By increasing T , the motion becomes more chaotic, and the particle markers in the corresponding Poincaré maps begin to scatter from inside to outside and spread to the whole domain. However, two islands form for some specific T values. Results show that little mixing occurs in these islands though the general mixing performance is improved. Continuing to increase T results in an even more uniformly distributed Poincaré map, and greater stretching of the material lines. As a result, mixing performance improves.

It is shown that greater chaotic motion along with the corresponding increase in mixing performance can be achieved by implementing more elaborate potential switching schemes and electrode placement. Instead of just cycling between the two disk electrode “agitators” with a given period, two counter-rotating flows are generated and the sequence is repeated by alternatively activating two disk electrode pairs. The four-disk scheme we have presented has significant advantage due to the large mixing regions achieved within a short time.

The choices for the number and locations of the electrodes, magnitude and orientation of the magnetic flux density, type of electrolyte and the switching scheme, provide a large set of options to choose from. Narrowing down the selections from the vast array of options would become an optimal problem for the specific LOAC application which is not be addressed in this work. The results presented here suggest that even more complex flow structures leading to better mixing performances can be created by straight forward extensions of the technique used in this study. However, other aspects such as energy consumption, feasibility of fabrications and cost should be also taken into account when designing MHD micro stirrers. Though excellent works on MHD mixing by chaotic advection have been reported previously by other researchers, our present work brings additional insights facilitated by the power of CFD simulations, high resolution graphics and powerful post-processing of the simulation results. The present work lays out a promising approach to gain insight into the complex optimal problem of designing the LOAC device to optimize mixing and flow control.

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Fig. 1. Cylindrical cell in top view and side view. One ring electrode and four disk electrodes are deposited on the bottom surface. The dimensions are: $r_1 = 3$ mm, $r_2 = 2.4$ mm, $r_3 = 2$ mm, $r_4 = 1$ mm, $r_d = 0.16$ mm, $H = 0.5$ mm. The Nd-Fe-B permanent magnet is placed underneath the cell as shown. The direction of the magnetic field B is also shown.

Fig. 2 (a): velocity vectors. (b): y-component of velocity vs. x. (c): electrical potential contours and current flux vectors (black) in plane $z = 0.4\text{mm}$. (d): electrical potential contours (blueish), current flux vectors and current flux lines (red) in cross-sectional view in plane $y = 0$. x and z units are m. (e): x-component of velocity vs. y. $T = 4\text{s}$.

Fig. 3 Equipotential lines and current flux lines from potential step simulation in $z = 0.4\text{mm}$ plane. Current paths are in red.

Fig. 4 (a), (b): velocity vector maps at the end of the first and second half periods, respectively. (c): electric potential contours (V) and current flux vector map in $z=0.4\text{mm}$ plane at the end of the first half period. (d): electrical potential (V), current flux vector map and current flux lines in cross-sectional view in $y=0$ plane. $T=2\text{s}$.

Fig.5 Velocity vector maps and streamlines from Navier-Stokes ((a) and (c)) and Stokes flow ((b) and (d)) in $z=0.4\text{mm}$ plane. Potential step solution. The boundary conditions are: $\phi_A = 0.1\text{ V}$, $\phi_C = 0\text{ V}$. All the other surfaces are electrically insulated ($\bar{\nabla}\phi \cdot \hat{n} = 0$). $t = 8\text{ s}$. $\text{Re}_H = 0.1$.

Fig.6 Velocity vector maps and streamlines from Navier-Stokes ((a) and (c)) and Stokes flow ((b) and (d)) in $z=0.4\text{mm}$ plane. Potential step solution: The boundary conditions are: $\phi_A = 1\text{ V}$, $\phi_C = 0\text{ V}$. All the other surfaces are electrically insulated ($\bar{\nabla}\phi \cdot \hat{n} = 0$). $t = 8\text{s}$. $\text{Re}_H = 1.07$.

Fig.7 Velocity vector maps and streamlines from Navier-Stokes ((a) and (c)) and Stokes flow ((b) and (d)) in $z=0.4\text{mm}$ plane. Potential step solution: The boundary conditions are: $\phi_A = 20\text{ V}$, $\phi_C = 0\text{ V}$. All the other surfaces are electrically insulated ($\bar{\nabla}\phi \cdot \hat{n} = 0$). $t = 8\text{s}$. $\text{Re}_H = 14.98$.

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Fig. 8 Poincaré maps for different time periods T in plane $z=0.4$ mm. (a): Initial conditions, (b): $T=1$ s, (c): $T=2$ s, (d): $T=4$ s, (e): $T=8$ s and (f): $T=10$ s.

Fig. 9 Numerical particle distributions for different time period T at $t=200s$ at plane $z=0.4$ mm. The black and grey particles represent two non-diffusive dyes. (a): Initial distribution, (b): $T=1s$, (c): $T=2s$, (d): $T=4s$, (e): $T=8s$ and (f): $T=10s$.

Fig. 10 Evolution of mixing quality with time t for different time periods T at plane $z=0.4$ mm

Table 1. Simulation Data

Density (kg/m ³)	1000
Viscosity (kg/(m.s))	0.001
NaCl solution concentration	0.1M
Electrode potential difference (V)	0.04
Solution conductivity (S/m)	1.29
Magnetic field intensity (T)	0.36

Table 2. Number of elapsed periods for each case in Fig. 8(b-f).

T(s)	Number of periods (n)
1	75
2	100
4	300
8	400
10	400

Table 3. Mixed area in plane $z=0.4$ mm at $t=200$ s for T ranging from 1s to 10s.

Time period T (s)	Mixed area (mm ²)	% of total area
1	0.523075E-05	18.5
2	0.124047E-04	43.9
4	0.130527E-04	46.2
8	0.135999E-04	48.2
10	0.140859E-04	49.8