LAWRENCE LIVERMORE NATIONAL LABORATORY

UCRL-TR-218892

Dynamic simulation tools for the analysis and optimization of novel collection, filtration and sample preparation systems

D. Clague, T. Weisgraber, J. Rockway, K. McBride

February 13, 2006

Disclaimer

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

FY05 Final Report

Dynamic simulation tools for the analysis and optimization of novel collection, filtration and sample preparation systems

LDRD Project Tracking Code: Tracking Code: 02-ERD-066

Principal Investigator
David S. Clague, EE/EETD
Co-investigators
Todd Weisgraber Engr/EETD
John Rockway (EE/DSED/Postdoctoral Fellow)
Kristal McBride (Graduate Student, GTU)

1.0 Abstract

The focus of research effort described here is to develop novel simulation tools to address design and optimization needs in the general class of problems that involve species and fluid (liquid and gas phases) transport through sieving media. This was primarily motivated by the heightened attention on Chem/Bio early detection systems, which among other needs, have a need for high efficiency filtration, collection and sample preparation systems. Hence, the said goal was to develop the computational analysis tools necessary to optimize these critical operations. This new capability is designed to characterize system efficiencies based on the details of the microstructure and environmental effects. To accomplish this, new lattice Boltzmann simulation capabilities where developed to include detailed microstructure descriptions, the relevant surface forces that mediate species capture and release, and temperature effects for both liquid and gas phase systems. While developing the capability, actual demonstration and model systems (and subsystems) of national and programmatic interest were targeted to demonstrate the capability. As a result, where possible, experimental verification of the computational capability was performed either directly using Digital Particle Image Velocimetry or published results.

2.0 Introduction/Background

With the increased priority on Chem/Bio-agent detection systems, LLNL programs have been aggressively responding. In advanced detection systems, the pressing need is for more effective sample collection, filtration and preparation. Because these sub-systems involve the transport of fluids (gases and liquids), particulates and some form of sieving medium, there are a number of scientific issues that need to be understood and addressed. Both the BASIS and APDS systems involve sample collection, filtration and preparation. Additionally, there are many filters, collection methods and separation methods that make use of sieving media (e.g., fibrous filters), but there are very few tools available to optimize these systems. In particular, optimize of the attachment and removal of samples from surfaces is not well understood. Therefore, it is of crucial importance that the National Laboratory has the necessary computational tools to properly support pressing programmatic and national needs. These problems have been explored using idealized representations, where the particle-particle and particle-medium hydrodynamic interactions are ignored. Proper accounting of these interactions is very difficult; however, it is these interactions, particularly in liquid systems that dictate particle transport behavior and sub-system efficiency. In this project, a suite of computational tools was developed to address optimization and design needs for these important systems by enabling the accurate characterization of fluid and particle transport in relevant microenvironments.

The objective of this effort was to develop computational tools that will enable rapid design analysis for novel filtration, collection and sample preparation systems. In all cases, gas and liquid phase samples containing particulate species, such as, spores, dust and pollen, are passed through a sieving medium, e.g., fibrous filter. The functionality and efficacy of these operations is highly dependent on the details of the microstructure and the surface properties of the sieving medium and the suspended species. Additionally, filters, collectors and sample preparation systems are deployed in actual environments. Therefore, this effort was focused on the development of a new computational capability that will provide the necessary tools to determine filter, collector and separator efficiencies as a function of the details of the microstructure, surface properties and environmental conditions.

Sample purification, collection and separation systems constitute a large class of problems involving fluid and particle transport through porous media. Commercially available software packages do not currently have the capabilities to handle such design problems. The primary competitors include CFD RC, and Coventor. Both CRD RC and Coventor use continuum methods and they treat suspended species as non-interacting point particles. Or if the particles are explicitly taken into account, these approaches typically use analytic approximations to account of the coupling forces of interaction. In contrast, our proposed capability will be an integrated approach that takes into account both the fluid and suspended species response in sieving media with surface interactions. Additionally, neither CFD-RC nor Coventor has developed capabilities to model porous or sieving media with mobile species.

This effort was focused to align with and to directly impact sub-systems that were topics of active research. This work directly aligned with computational needs in HSO, NAI, CBNP and MTP. This included projects that included the pillar chip. The pillar chip is also a sieving medium that is used to effect DNA collection and sample preparation. There are, however, optimization issues associated with medium geometry and surface properties for sample collection and separations. Furthermore, at the time, the artificial kidney work, which was an LDRD project in M-division, utilized functionalized porous silica to perform blood filtration. The computational tools developed are directly applicable and will be used to support the MTP effort as well. Therefore, the resulting capability will be used to impact on future important internal projects in this arena. This capability aligns with Engineering's core competency of developing computational design tools for real world problems that impact the laboratory mission.

3.0 Approach

Lattice Boltzmann capabilities developed by the PI and team on previous LDRD/ER and DARPA projects was leveraged and enhanced to include sieving media, relevant surface interactions, gas phase capabilities and environmental effects, that is, temperature effects. The lattice Boltzmann equation is a discrete form of the Boltzmann transport equation [1]

$$f_i(\underline{x}+e_i, t+1) = f_i(\underline{x}, t) - \frac{f_i(\underline{x}, t) - f_i^{eq}(\underline{x}, t)}{\tau} \quad . \tag{1}$$

Here f_i is the i^{th} component of the single particle distribution function, f^{eq}_i is the equilibrium distribution function, and τ is the relaxation parameter, which is directly related to the viscosity of the fluid. $\underline{x}$ denotes the position in the flow field, and t denotes time. e_i is the lattice velocity [2]. In the lattice Boltzmann formulation, the no-slip boundary condition is achieved using what is known as the bounce back condition. This boundary condition is particularly useful when studying complex, porous media. Shown in Figure 1 is a unit cell that is representative of the pillar arrangement in the pillar chip discussed in this proposal. Because the single particle distribution function, f, is a moment baring function, the lattice Boltzmann method easily accommodates the study of velocity, pressure and shear stress fields in complex media. In addition to being able to study fluid properties, the lattice Boltzmann method readily permits the inclusion of mobile species [3]. Sub-system efficiencies were initially characterized using spherical representations of target species. While the capability has been developed to model spherical particles, the particles are fully coupled, hydrodynamically, with each other and the surrounding microenvironment.

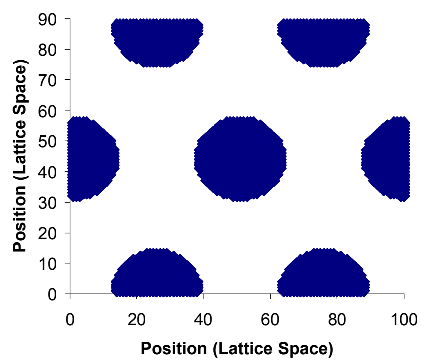


Figure 1. Unit cell for pillar arrangement in pillar chip.

The particle dynamics are accounted for through Newton's equations of motion, and the hydrodynamic force and torque, in the lattice Boltzmann representation, is accounted for using Ladd's half-link method [2]. The force and torque are related to the particle's relative velocity through the hydrodynamic resistance tensor, $\underline{\underline{R}}$. The resistance tensor can be further modified to account for near-field hydrodynamic forces, or lubrication forces:

$$\begin{bmatrix} \delta \underline{F} \\ \delta \underline{L} \end{bmatrix}_{Hydrodynamic} = \left(\underline{\underline{R}}_{Lub} - \underline{\underline{R}}_{2B}\right) \cdot \begin{bmatrix} \underline{U} \\ \underline{\Omega} \end{bmatrix}_{rel} \quad , \tag{2}$$

where $\left(\underline{\underline{R}}_{Lub} - \underline{\underline{R}}_{2B}\right)$ is the correction due to lubrication forces, $\underline{U}$ and $\underline{\Omega}$ are the particle translational and rotational velocities, and δF and δL are the force and torque corrections due to lubrication interactions respectively. The framework and approach for including the lubrication interaction provides a natural framework for including the relevant surface interactions,

$$\underline{F}_{Corr} = \underline{F}_0 + \delta \underline{F}_H + \delta \underline{F}_{vdw} + \delta \underline{F}_{Columbic} \tag{3}$$

Here, "0," "H," "*vdw*," and "Columbic" stand for lattice Boltzmann result, hydrodynamic correction for lubrication, van der Waals and Columbic forces respectively. $\underline{F}_{Corr}$ is the force acting on the target species corrected for all of the relevant interactions. Along with geometric details of the microstructure, the surface interactions dictate the efficiency and functionality of the sub-system, i.e., filter, collector and/or separator. Essentially, the efficiency is a measure of number densities, that is, a ratio of manipulated to total species present, or,

$$\eta = n / n_0 , \tag{4}$$

where n is the number density of, say, the captured species, and n_0 is the initial concentration of the species in the sample.

4.0 Summary of Research Activity

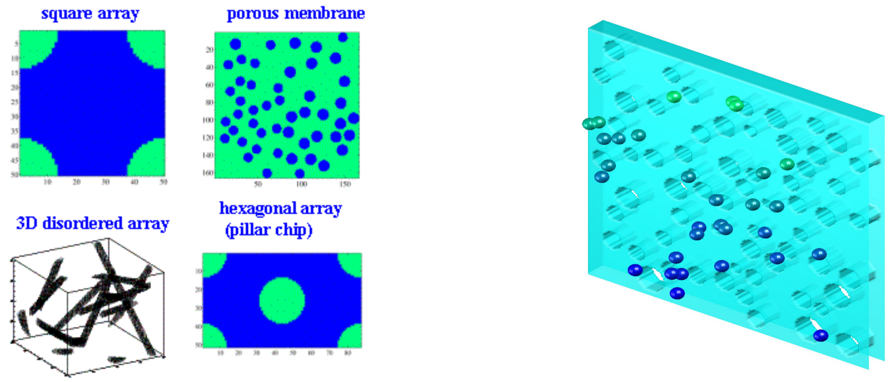
As discussed above, this effort was focused on development of simulation tools to optimize sample collection, filtration, sample preparation systems, i.e., all systems where the efficacy is driven by the details of the microstructure (and interactions with the microstructure). To this end, capabilities were developed to produce representations of media of interest, e.g., arrangement of cylinders and porous membranes, include mobile species with surface properties, include colloidal and Brownian forces for these media, and to include both viscous and non-viscous fluids, i.e., gas phase.

In the sections to follow, the new features of the capability and provide selected results are described. These new features include the ability to computationally generate representation of sieving media, the ability to include individual and suspensions of particles in the sieving media, the ability to characterize the convective transport coefficient, inclusion of temperature effects, and extension of these capabilities to characterize particulate transport in the gas phase.

5.0 Results/Technical Outcome

5.1 Media Module

To enable the study of fluid flow and particle interactions in complex microenvironments, a media module to generate ordered and random fibrous media in both two and three dimensions was developed. Additionally, the ability to generate three dimensional porous substrates to mimic membranes with a user specified pore size distribution. The sieving media representation is included as part of the computational domain, see Figure 2 below.

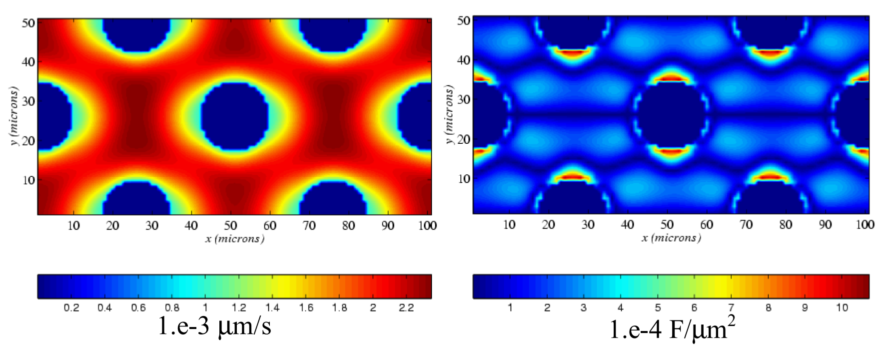


(a) (b)

Figure 2: **a**. Media Module, media generated as specified by the user. Three arrangements of cylinders including the pillar chip, and a porous membrane with specified porosity and pore size distribution. **b**. Three dimensional perspective of porous membrane with mobile species partitioning through the membrane.

As depicted above in Figure 2, fluid velocity and shear fields can be characterized, and more importantly particle velocities, trajectories, convective transport coefficients and collection efficiencies can be characterized.

Given the timing and importance of the pillar chip, much of the work performed in the first year and a half of the work presented here was focused on characterization of fluid and particulate transport through the chip. Shown below in Figure 3 are images of the fluid velocity and shear fields resulting from pressure driven flow through the pillar chip.



(a) (b)

Figure 3: (a) velocity, and (b) shear stress field for the pillar chip geometry in a 50 µm deep channel.

The images in Figure 3 are two dimensional slices from a three dimensional flow domain. The dark blue "circles" are the cylindrical pillars, and the flow is from left to right, orthogonal to the pillars. The Reynolds number based on the pillar radius of 4 µm is 0.026. The large shear at the top and bottom of the pillars would prevent particles from attaching to these surfaces. Capture is most likely to occur at the leading edge. To convert from lattice space to real space, only the viscosity, kinematic viscosity and lattice spacing in microns are necessary. Digital Particle Image Velocimetry (DPIV) experiments to measure the velocity profile through the pillar arrangement to validate the code were performed by a summer student, see Figure 4.

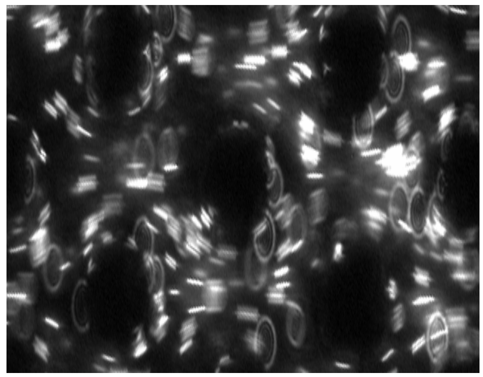


Figure 4: Example DPIV image of tracer particles advecting through a pillar chip.

Using DPIV imaging software, the velocity fields predicted by the new capability and those predicted by DPIV were in excellent agreement.

5.2 Temperature Effects (Energy Equation)

To accommodate particle interactions and environmental effects, two new physics modules were incorporated, that is, the colloidal surface interactions and thermal transport modules. The colloidal module incorporates electrostatic, van der Waals, hydration and hydrophobic/hydrophilic forces as described above in Eq(3). The thermal module incorporates a second, kinetic evolution equation into the lattice-Boltzmann algorithm to account for heat transport in the systems. The new kinetic equation for energy/temperature [4] was shown to be in exact agreement with theory for coupled heat transport in Couette flow.

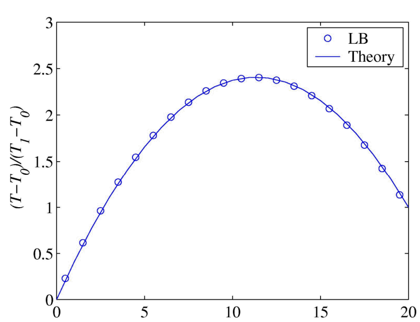
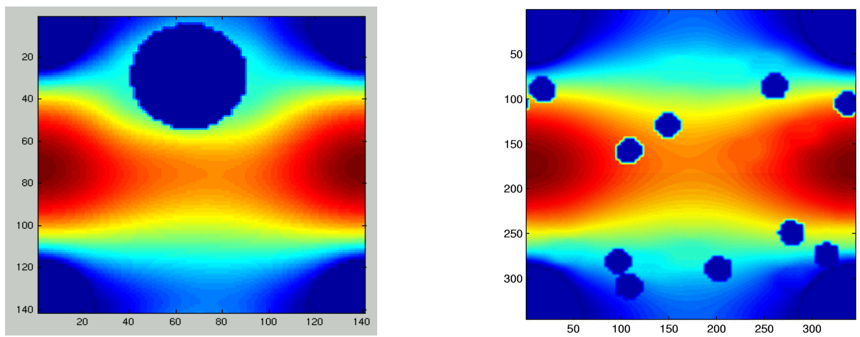


Figure 5: Dimensionless temperature distribution between two temperatures in Couette flow.

The dimensionless temperature is plot as a function of dimensionless channel width. The open circles are results from the new capability and the line is from exact theory.

5.3 Species Transport (Convective Transport Coefficient)

This capability will allow determination of the role of temperature in the capture and filtration of target species. In addition, species diffusive and convective transport through different pillar geometries for both single particles and suspensions were characterized (Figure 6).



(a) (b)

Figure 6: Convective transport simulations of a) single particle and b) suspension in a periodic array of cylinders. Particle locations are superposed on pseudo-color representation of velocity field.

In Figure 6, the pillars are at the corners of the periodic simulation cell. The mobile species is (are) shown as solid blue circle (s), and the magnitude of the fluid velocity is shown. The fluid flow is from left to right. As intuitively expected, the fluid velocity orthogonal to the applied flow direction and between pillars is nearly stagnant, and as expected, the maximum fluid velocity occurs between pillars coincident to the applied flow field.

Shown below in Figure 7 is a validation study using the new capability to characterize the convective transport coefficient for a single mobile species in a typical pillar arrangement. More specifically, the new capability is used to characterize the average particle velocity between pillars in a pillar arrangement for many realizations of particle starting position. The average particle velocity is then divided by the average fluid velocity to form the convective transport coefficient.

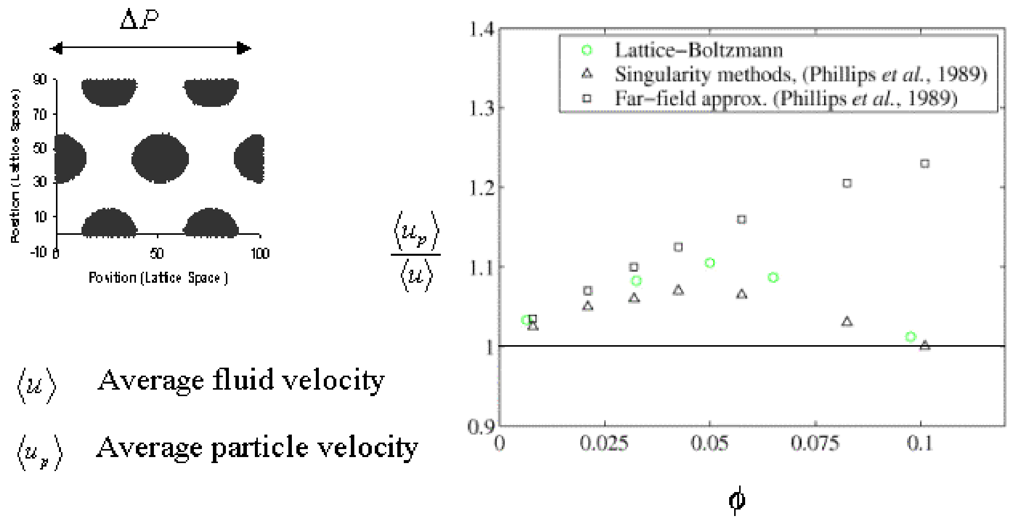


Figure 7: Comparison of convective transport coefficients predicted using the new Lattice Boltzmann capability and the singularity result of Phillips et al., 1989 [5].

As shown above our results exhibit very good agreement with the results of Phillips et al, 1989 [5]. The differences are due to the fact that Phillips et al. used spheres to construct their cylinders and in this LDRD/ER cylinders were explicitly used and the new capability did not have lubrication correction for tangential approach to a cylinder.

5.4 Particle Transport, Gas Phase

The particle dynamics capability was augmented to account for particle Reynolds number effects in aerosol transport phenomena in gas phase [6]. This advancement enabled the study of bulk phase gas and particle dynamics; however, in such flow conditions, the fluid experience increasing slip as the Knudsen number increases; therefore to ensure accurate prediction of fluid and particle transport properties, the boundary condition at sold surfaces must account for a Knudsen number dependent slip condition [7]. A major accomplishment was the development of a novel, Knudsen number dependent, boundary condition that enables accurate prediction of fluid slip velocities for a wide range of conditions, see Figure 8 below.

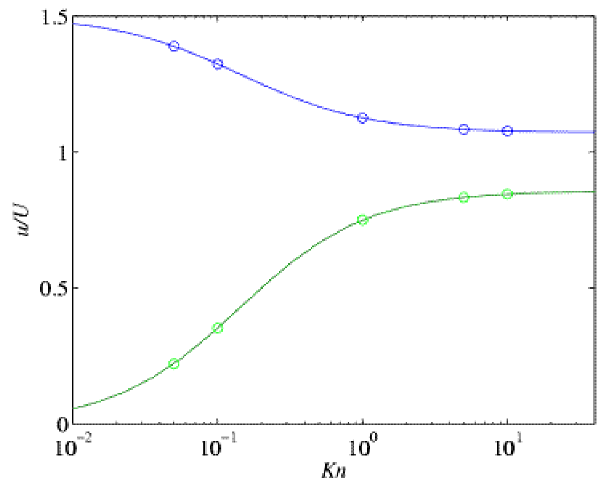


Figure 8: Dimensionless fluid, slip velocity in channel flow predicted using the new lattice Boltzmann boundary condition compared with predictions from theory as a function of Knudsen number, *Kn*.

As shown above our results exhibit nearly exact agreement with theory. Prior to this break-through, lattice Boltzmann boundary conditions were accurate to $Kn < 1$ (10^0). The above result constitutes a significant improvement over the current the state of the art.

Gas phase fluid and particle simulation capability were developed a to enable characterization of flow and aerosol transport in complex porous media. The new capability uses the media module and particle inertial [7] and colloidal effects. Furthermore, a fourth order aerosol particle tracking routine was developed and incorporated. (Figure 9).

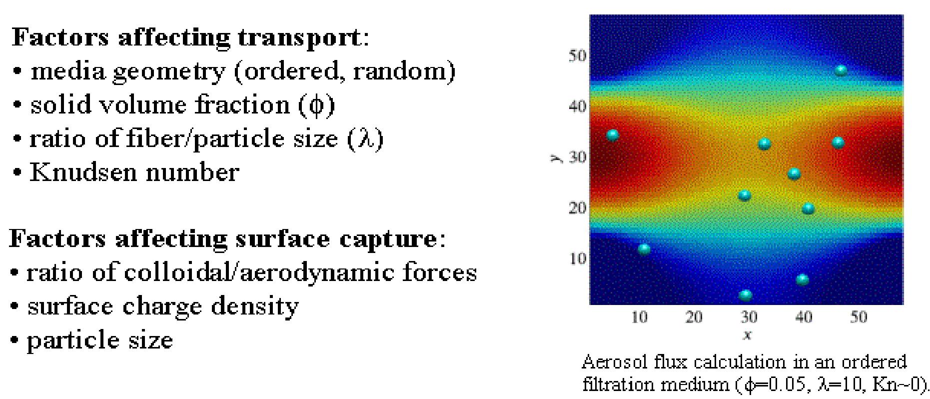


Figure 9: A two dimensional cross-section at a particular height of Aerosol transport through a regular array of cylinders. The colors represent the magnitude of the fluid velocity. The cylinder quarters are in each corner of the periodic simulation cell.

The particle dynamics/tracking capability was further advanced to handle complex geometries, and to demonstrate this new advancement, the capability was used to explore and optimize particle focusing (sample preparation/conditioning) for Mass Spectrometry, i.e., particle transport and focusing in rarified gas conditions. Shown below in Figure 10 are results from actual design simulations that were later used to support related DARPA, BAMS research efforts.

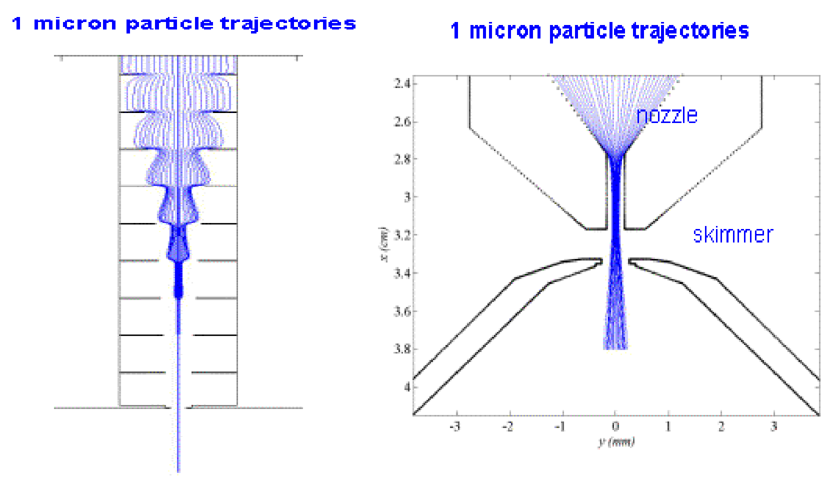


Figure 10: Particle trajectories in "in let" design for Mass Spec.

In addition to above mentioned work, the capability includes the following, species Brownian motion [8,9], the ability to import images of media, and improved force predictions to species response to non-uniform electric fields [10].The key to optimizing these systems is the ability to study and understand the physics governing system performance. Additionally, work was performed to include fast multipole E&M field calculations to enhance prediction of colloidal interactions and external electric field effects on suspended particles.

6.0 Exit Plan

Through Engineering investments this capability is now packaged with a user interface and CAD capabilities and is now known as µLatte 3D. As mentioned above, the particle tracking capability developed under this effort was used on optimization of a Mass Spec. inlet design. Additionally, this capability was used to characterize the response of adhered species to shear fields on an EPD LDRD/ER. These capabilities have made contributions to several pending proposals.

7.0 Summary

This capability is extremely useful. Specifically, enabling the accurate study of fluid flow and particulate transport in complex micro- and nano- environments is very important and has broad application to laboratory needs.

8.0 References

[1] Dieter A. Wolf-Gladrow, "Lattice-Gas Cellular Automata and Lattice Boltzmann Models," Springer-Verlag Berlin Heidelberg 2000

[2] Clague, D.S. and P.J. Cornelius, The hydrodynamic force and torque on a sphere in Poiseuille flow, the *Int. J. Numer. Meth. in Fluids* (2001); 35:55-70

[3] Simulating Particle Transport in Microfluidic Devices, S&TR, Dec 2001.

[4] He, X., Chen, S., and G.D. Doolen, A novel thermal model for the lattice Boltzmann method in incompressible limit. J. Comp. Phys. **146**, 282 (1998).

[5] Phillips, R.J., Deen, W.M., and J.F. Brady, Hindered transport of spherical macromolecules in fibrous membranes and gels. AIChE J. **35**, 11 (1989).

[6] Baron, P.A. & Willeke, K. (eds.), 2001, "Aerosol Measurement: Principles, Techniques, and Applications", John Wiley and Sons, New York.

[7] Karniadakis, G.E. & Beskok, A. 2002, "Micro Flows: Fundamentals and Simulation", Springer-Verlag, New York.

[8] Russel, B., Saville D. and W. Schowalter, Colloidal Dispersions, Cambridge University Press, 1989.

[9] A Random Walk through Time and Space, S&TR, July/Aug 2005.

[10] Enzhu Liang, Rosemary L. Smith, and David S. Clague, "Dielectrophoretic manipulation of finite sized species and the importance of the quadrupolar contribution," Jan 1. Phys. Rev E. 2005