Parallel Reacting Flow Calculations for Chemical Vapor Deposition Reactor Design[1]

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ABSTRACT

The MPSalsa reacting flow code has been developed at Sandia National Laboratories to compute coupled three-dimensional fluid flow and detailed reaction chemistry for modeling reacting flow systems, such as Chemical Vapor Deposition (CVD) reactors. MPSalsa has been developed to take advantage of the tremendous computational speed, memory, and scalability of distributed memory parallel computers. An unstructured finite element discretization allows for the modeling of complex geometries. Complex physics for general mixtures of ideal gases, including thermal diffusion, mixture averaged and true multicomponent diffusion, variable physical properties and both gas and surface reactions can be modeled. In this brief manuscript we discuss the reacting flow model and numerical method and summarize representative calculations using MPSalsa for the CVD growth of Gallium Arsenide in a horizontal reactor with a tilted rotating substrate.

INTRODUCTION

Computer modeling of chemical vapor deposition (CVD) reactors can greatly aid in the understanding, design, and optimization of these complex systems. Modeling is particularly attractive in these systems since the costs of experimentally evaluating many design alternatives can be prohibitively expensive, time consuming, and even dangerous, when working with toxic chemicals like Arsine (AsH_3); however, predictive modeling has not been possible for most systems since the behavior is three-dimensional and governed by complex reaction mechanisms. In addition, CVD reactors often exhibit large thermal gradients, large changes in physical properties over regions of the domain, and significant thermal diffusion for gas mixtures with widely varying molecular weights. As a result, significant simplifications in the models have been made which erode the accuracy of the model's predictions. In this paper, we will demonstrate how the computational resources of massively parallel computers can be exploited to make possible the analysis of models that include coupled fluid flow and detailed reaction chemistry in three-dimensional domains.

Our research effort is aimed at the synthesis of two important research areas: 3D flow and transport modeling of reactors and the simulation of detailed reaction mechanisms.

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Due to the large computational resources needed to tackle such a problem, there are few attempts in the literature. Ouazzani and Rosenberger (1990) and Kleijn and Hoogendoorn (1991) each solved three-dimensional models for reactant transport and GaAs deposition in horizontal reactors. The first paper decoupled the species transport from the fluid mechanics and heat transfer. Both works used solution procedures that require structured grids, so the effect of a rotating disk could not be readily included.

Reacting Flow Model

Considerable effort has gone into the development of the MPSalsa simulation code [Shadid, Moffat, Hutchinson, Hennigan, Devine and Salinger (1996a, 1996b)] to make the CVD reactor model as accurate as possible. Table 1 shows the modeling equations implemented in MPSalsa that were used in this paper. Constituative relations for the Newtonian stress tensor, $\mathbf{T}$, heat flux vector, $\mathbf{q}$, and species mass fluxes, $\mathbf{j}_k$, are based on non-equilibrium statistical mechanical theory of multicomponent, dilute polyatomic gases. The diffusive flux of species includes a mixture averaged approximation and true multicomponent formulation with the effects of thermal diffusion included. All physical properties and reaction rates in the model are allowed to vary as functions of composition and temperature using the CHEMKIN [Kee, Rupley, Meeks and Miller (1996)] and SURFACE CHEMKIN [Coltrin, Kee, Rupley and Meeks (1996)] suites of library routines and the associated TRANLIB [Kee, Dixon-Lewis, Warnatz, Coltrin and Miller (1986)] thermodynamic database. The geometry is discretized using an unstructured finite element mesh, which can flexibly represent complex geometries and allow the placement of finer meshes in areas where steep gradients are expected. For a thorough description of the equations and parallel implementation specifics, the reader is referred to the MPSalsa theory document.

Momentum	$\frac{\partial(\rho\mathbf{u})}{\partial t} + \nabla\bullet(\rho\mathbf{u}\mathbf{u}) - \nabla\bullet\mathbf{T} - \rho\mathbf{g} = 0$
Total Mass	$\frac{\partial\rho}{\partial t} + \nabla\bullet(\rho\mathbf{u}) = 0$
Thermal Energy	$\hat{C}_p\left[\frac{\partial(\rho T)}{\partial t} + \nabla\bullet(\rho\mathbf{u}T)\right] = -\nabla\bullet\mathbf{q} + \sum_{k=1}^{N_s} h_k\nabla\bullet\mathbf{j}_k - \sum_{k=1}^{N_s} h_k W_k\dot{\omega}_k$
Species Mass Fraction for Species k	$\frac{\partial(\rho Y_k)}{\partial t} + \nabla\bullet\rho\mathbf{u}Y_k = -\nabla\bullet\mathbf{j}_k + W_k\dot{\omega}_k, k = 1, 2, \ldots, N_S - 1$

Table 1: Governing Conservation Equations

The solution to the transport-reaction equations is approximated using a Petrov-Galerkin finite element method. All of the variables, $(\mathbf{u}, P, T, \underline{\mathbf{Y}})$, are interpolated with trilinear basis functions within each hexehedral element of the unstructured mesh. A discrete set of residual equations is created by weighting each governing equation by each basis function and integrating over the domain. An additional pressure stabilization term is added to the continuity equation residual to allow for equal order interpolation of $\mathbf{u}$ and P [Hughes, Franca and Balestra (1986) and Tezduyar (1992)].

Numerical Methods

The MPSalsa reacting flows code has been developed to take advantage of the large speed and memory of distributed memory parallel computers so that models involving coupled 3D flows and detailed reaction mechanisms can be solved. Since the transport equations are highly nonlinear, high priority has been given to the robustness of the solution procedure. A fully coupled Newton's method is used to solve the nonlinear problem, where all the equations are solved simultaneously, and all the interactions between the equations and variables are included in the Jacobian matrix. Each processor computes the residual equation and associated rows of the Jacobian matrix for the nodes it owns. Since these calculations are local, they exhibit near perfect scaling as the problem size and number of processors are increased proportionally [Shadid, Hutchinson, Moffat, Hennigan, Hendrickson and Leland (1994)]. In our implementation the matrix elements are fully summed, this method has the advantage of allowing for a large set of general algebraic preconditioners to be used.

At each iteration of the nonlinear solver, the linear system is solved iteratively using the AZTEC library [Hutchinson, Shadid and Tuminaro (1995)] of preconditioned Krylov solvers. The computational core of these methods is a matrix-vector multiplication and the preconditioner, which dominates the cost of our entire solution procedure. In the calculations in this paper, we chose to first scale the matrix by a row-sum approach and use an incomplete LU decomposition as the preconditioner.

RESULTS AND DISCUSSION

In this section we present results of CVD calculations made using MPSalsa on the 1824 computational processor Intel Paragon at Sandia National Laboratories.,The reactor configuration is presented in Figure 1, along with surface elements of the 43,568 element mesh. The reactants enter the reactor through the rectangular region on the left. The bottom of the reactor slopes up at a 9 degree angle, forcing the flow to accelerate. The outlet of the reactor has only one quarter the area of the inlet face. The rectangular susceptor has a disk inset into the middle of it, which spins to average out the deposition rate. Tilting the base of the reactor is designed to offset the decline in deposition rate down the length of the reactor, due to reactant consumption, by accelerating the flow and shrinking the boundary layer over the disk, thereby decreasing mass transfer resistance to the surface.

The example we present is for a horizontal CVD reactor with tilted susceptor and rotating substrate which only admits three-dimensional solutions for the vapor phase deposition of Gallium Arsenide (GaAs) from a mixture of trimethylgallium $GaMe_3$ and Arsine (AsH_2) in excess Hydrogen (H_2) carrier gas. We use the blocking mechanism of Moffat, Killeen and Baucom (1995) which has 4 gas species, 4 surface species undergoing 4 reactions. Steady-state solutions of $432,225$ unknowns were reached directly from a trivial initial guess in 18 minutes on 288 processors of the Intel Paragon, using a restarted GMRES method, with a Krylov subspace dimension of 150 vectors, and a maximum of one restart per linear solve. An ILU decomposition with no overlap between processors, with row-sum scaling is used as the preconditioner.

A representative solution for flow and deposition in the horizontal reactor with tilted susceptor is shown in Figure 2, for an inlet velocity of $V = 30$ cm/sec and disk spin rate $\Omega = 500$ rpm. Streamlines for fluid that enters near the base of the reactor show the effect of the counter-clockwise rotation of the disk -- one path illustrating how fluid can be entrained to make multiple passes over the disk. The shaded contours on the susceptor visualize the gas phase mass fraction of $GaMe_3$ approaching the deposition surface, which is nearly proportional to the deposition rate since this is the limiting reactant. The asymmetry of the contours, which corresponds to non-uniform growth, illustrates the effect of large rotation rates.

This effect is examined in Figure 3, which contains contour plots of instantaneous GaAs deposition in Angstroms/sec at $V = 30$ cm/sec for three different spin rates: $\Omega = 50$, $\Omega = 250$, and $\Omega = 500$ rpm. The surface finite element discretization over

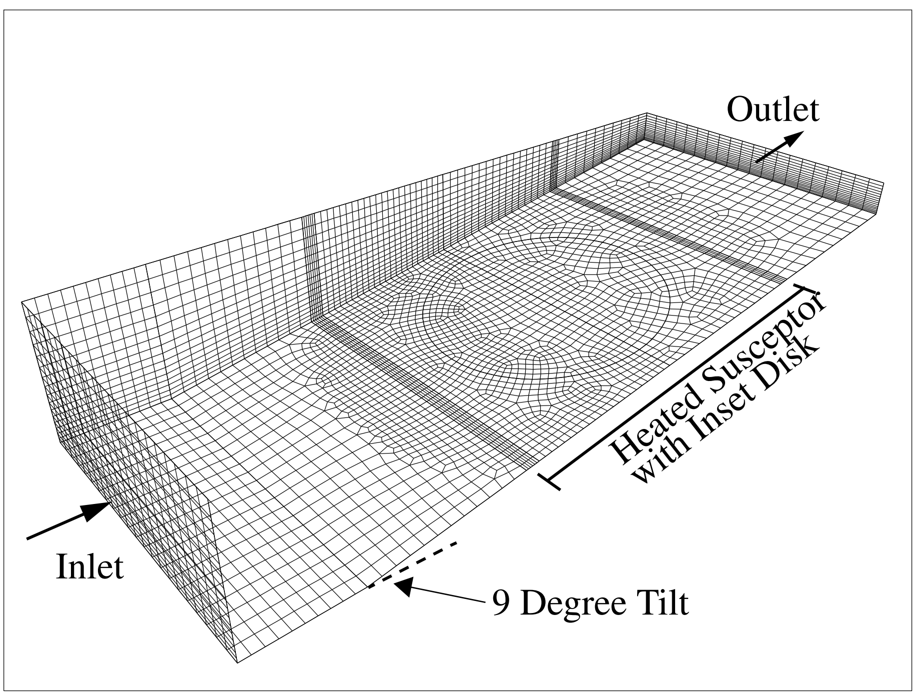

FIGURE 1. Surface elements of the mesh for the horizontal reactor with tilted susceptor.

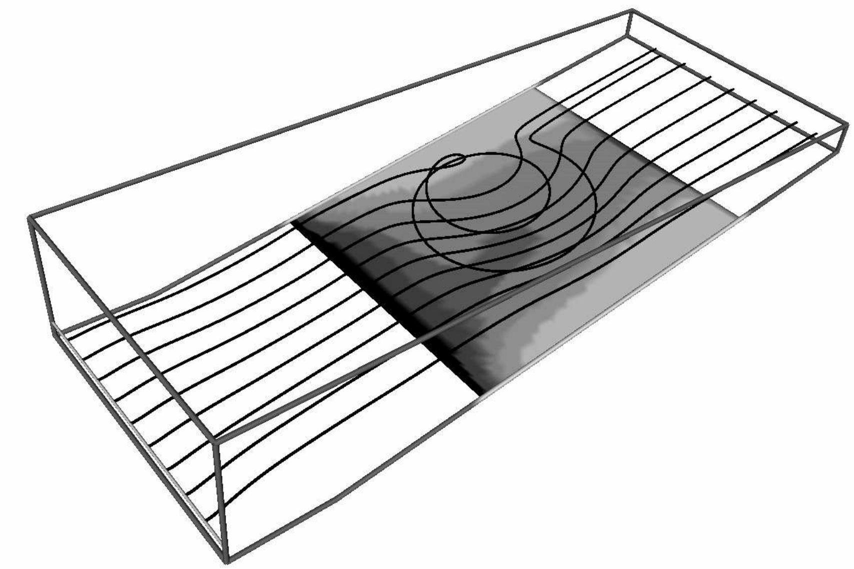

FIGURE 2. Visualization of a solution for GaAs deposition in the horizontal CVD reactor with tilted susceptor, at rotation rate $\Omega = 500$ rpm and inlet velocity $V = 30$ cm/sec.

the disk is also shown. The solutions at the two higher rotation rates clearly show how the spinning disk can skew the deposition; however, the $\Omega = 50$ solution does not exhibit noticeable asymmetry. Deposition rates show decreased deposition near the walls, which is due to the formation of a parabolic flow profile. A more detailed analysis of the effect of spin rate and other parameters is in progress.

CONCLUSIONS

The MPSalsa massively parallel chemically reacting flows code has been developed to solve three-dimensional flows (at low Mach number) coupled with chemical reactions using an unstructured finite element method. Through the use of the CHEMKIN suite of library routines, the gas phase reactions, surface reactions, transport properties, and physical properties are rigorously modeled. Efficient and robust parallel algorithms allow for rapid solution of large systems of equations.

Since steady-state solutions were achieved for our reactor models in about 15 minutes on a fraction of our machine, we have begun detailed analyses of these reactor models with optimization studies over several parameters. Optimization of operating parameters, such as inlet flow rates and disk rotation rates, can aid operators in understanding the responses of the reactor to these parameters. Even more informative will be optimization studies that include design parameters, which can not be economically studied by experiments. Without the investment in efficient and robust methods for parallel solutions, detailed analysis of these models would not be feasible.

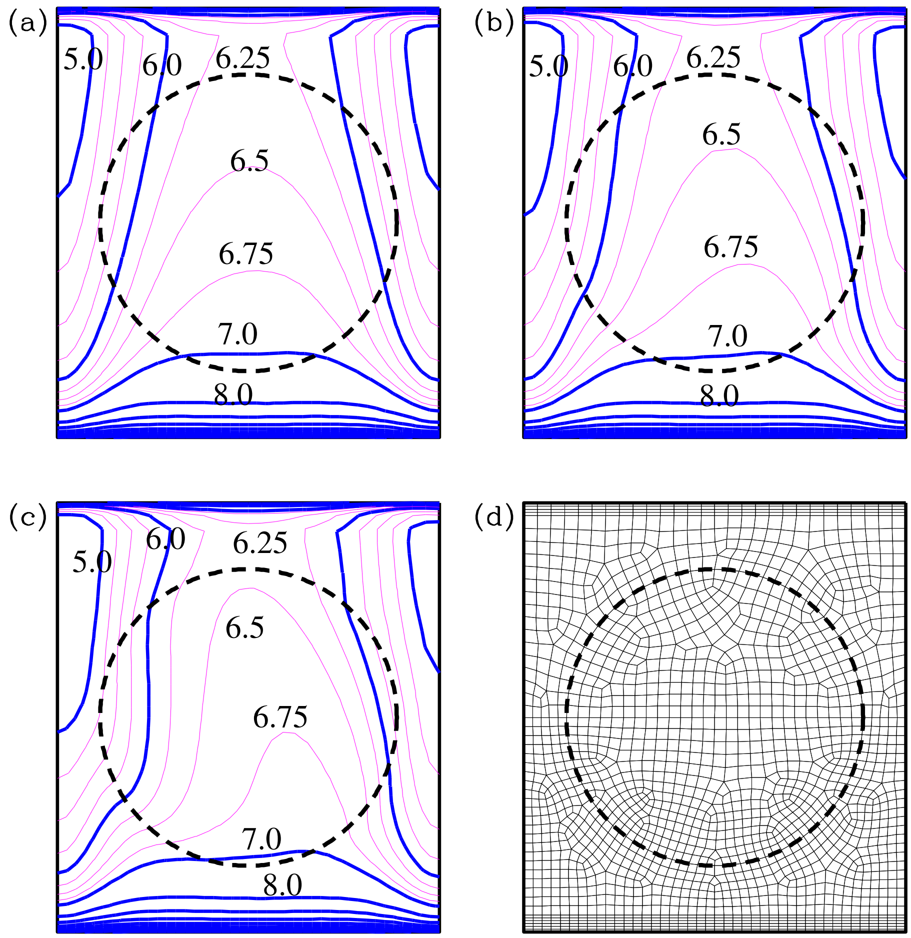

FIGURE 3. Instantaneous contours of GaAs deposition rates over the entire susceptor in Angstroms/sec, (a)-(c), and the surface elements of the finite element discretization, (d). The higher spin rates significantly deform the deposition rates from a symmetric solution.

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