Center for the Simulation of Accidental Fires & Explosions

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1.0 Executive Summary

The Center for the Simulation of Accidental Fires and Explosions (C-SAFE) at the University of Utah is focused on providing state-of-the-art, science-based tools for the numerical simulation of accidental fires and explosions, especially within the context of handling and storage of highly flammable materials. The objective of C-SAFE is to provide a scalable, high-performance system composed of a problem-solving environment in which fundamental chemistry and engineering physics are fully coupled with non-linear solvers, optimization, computational steering, visualization and experimental data verification. The availability of simulations using this system will help to better evaluate the risks and safety issues associated with fires and explosions. Our five-year product, termed Uintah 5.0, will be validated and documented for practical application to accidents involving both hydrocarbon and energetic materials.

Although the ultimate C-SAFE goal is to simulate fires involving a diverse range of accident scenarios including multiple high-energy devices, complex building/surroundings geometries and many fuel sources, the initial efforts during the first five years are focusing on the computation of a specific, well-defined scenario: rapid heating of a container with conventional explosives in a pool fire (e.g., a high energy device involved in an intense jet-fuel fire after an airplane crash).

This large-scale problem requires consideration of fundamental gas and condensed phase chemistry, structural mechanics, turbulent reacting flows, convective and radiative heat transfer, and mass transfer, in a time-accurate, full-physics simulation of accidental fires. This simulation will be expansive enough to include the physical and chemical changes in containment vessels and structures, the mechanical stress and rupture of the container, and the chemistry and physics of organic, metallic and energetic material inside the vessel. We will include deflagration-to-detonation transitions (DDT) of any energetic material in the fire, but the simulation will end when/if detonation occurs. C-SAFE will provide coupling of the micro- and meso-scale contributions to the macroscopic application in order to provide full-physics across the breadth of supporting mechanistic disciplines, and to achieve efficient utilization of ASCI program supercomputers.

We are utilizing a simulation development roadmap (SDRM) consisting of three distinct, sequential steps, which parallel the events in our physical problem: Ignition and Fire Spread, Container Dynamics and High Energy Transformations. A fire or explosion is initiated by an ignition event (which we assume to occur and which is not computed in detail). The extinction of an ignition event or its growth into a large fire is defined by the fire spread computations. The fire can cause the container of HE material to undergo changes, perhaps rupture and, simultaneously or sequentially, the HE material itself can undergo transformations which lead to an explosion. The overall mission is to integrate these computational steps into a coupled fire and explosion system. To fulfill this mission we are drawing on three core disciplines available at the University: molecular fundamentals, computational engineering, and computer science.

We believe the C-SAFE program to have four unique features which have been further amplified during the third year:

- The use of a one mesh / integrated solution approach for coupling the fire with the container and its high energy contents.
The inclusion of first principles molecular dynamics calculations to compute the fundamental chemistry of the high energy materials and other reactants under specific conditions of the problem.

Computational steering and system utilization analysis in conjunction with an advanced problem solving environment.

Integration of experimental testing with actual high energy materials from the beginning of the program.

This report summarizes the progress made during the third year of the program. Significant results have been obtained in each major area as described below.

Software Integration

In Year 3, the C-SAFE team has pursued an integration strategy to guide our product development, establishing a balance of effort and resources directed toward computational issues (parallelization, scaling, etc.) and the development of accurate, full-physics codes.

During the course of the year, we decided to switch from SAMRAI to a much more limited locally-developed code to provide the common computational framework for mesh and solver functionality. Although we found SAMRAI to be an extremely powerful tool, particularly with respect to AMR issues, we felt that its size and complexity could not be justified for our very specific and narrow application.

We have organized our integration efforts at four levels within the Uintah architecture, from the top-level blueprint of the Uintah system, the PSE structure and the solver strategies, to the individual software modules.

- Uintah System: the user interface must provide a unified view of the various simulation codes and databases which comprise a complete scenario simulation, and allow insight into performance of the codes on ASCI supercomputing platforms, so as to relate the data to the initial conditions, parameters, and modules. Moreover, data formats must be consistent across codes and throughout the visualization modules.

- Uintah PSE: within the interactive, computational problem solving environment the user must be allowed a clear visual methodology so that various algorithms - implemented as modules within the PSE - are syntactically simple to assemble into computational units, and semantically meaningful when executed together. Moreover, interactive computational steering must be provided to allow the user to inter-operate with the ongoing simulation.

- One mesh / integrated solvers: in terms of our particular simulation domain - fire spread and container dynamics - we are achieving integration through the Uintah Computational Framework (UCF), a common adaptive mesh refinement package being developed at as part of the C-SAFE effort. This provides us with a built-in capability to share state information between the grid and the particles involved in the simulation.

- Software Integration: Finally, the software from the various steps in the project must work together as a cohesive system; we are assuring this through our four software engineers - one in each step, and in the PSE - who have developed center-wide
coding standards, revision control methods, class browsers, bug reporting and documentation standards.

Firespread SDRM Step Team

Vision

For simulation science to be useful for practical engineering systems analysis such as large-scale fires in the presence of structures, the software tools need to be modular, rely on experimental and theoretical science, and be computationally efficient and scalable.

Mission

The FS team is committed to creating a set of modules for the simulation of fires in the presence of structures that affect the fire and are affected by it. The focus of this set of computational modules is to be able to realistically model the C-SAFE scenario of a fire in a jet-fuel pool that affects a missile inside the fire. To attain this objective, the FS team is committed to creating: 1. a common component architecture that can combine diverse physical, numerical, and computational modules, 2. efficient, scalable solution techniques on large-scale parallel architectures, and 3. accurate full-physics science that utilizes fundamental physical and chemical information from the micro-scale to obtain realistic, macro-scale simulations.

Goals

1. Create a component-based software architecture that is integrated with the UCF and that allows one to freely choose options for CFD, mixing, reaction, radiation, and solvers and combine them with each other and with structural mechanics components in the UCF and that can use the parallelization capabilities of the UCF to perform massively parallel calculations.
2. Develop, implement, test and compare models for Large-Eddy Simulation (LES) for the buoyant turbulent reacting flows that are encountered in pool fires.
3. Formulate, verify, and validate a general, multimaterial algorithm for fires in the presence of structures that incorporates accurate models for inter-material momentum, heat, and mass transfer.
4. Create, implement, verify, and validate submodels for mixing and reaction, including a verified, efficient strategy for tabulation of scalar species and reaction rates.
5. Develop a complete set of chemical kinetic mechanisms for jet fuel surrogate, including mechanisms for soot, using a combination of empirical models, reaction rate databases, and databases of transport and kinetic data generated from fundamental chemistry models.
6. Create an efficient, massively parallel, radiation model for pool fire simulations, with submodels to account for soot properties and spectral radiation.
Research Group Tasks

Component-Based Software Architecture and Large-Scale Parallelization

This effort is primarily concerned with enabling massively parallel simulations of fires in the presence of structures. To this end, the objectives are

1. To create a component-based software architecture in C++ that is integrated within the Uintah Computational Framework (UCF) and that uses lower-level FORTRAN modules for the core computational routines.
2. To allow for flexibility in options for mixing models, reaction models, radiation models, and solvers.
3. To allow for scalable parallelization within the framework of Uintah
4. To use this tool to achieve parallelization and scalability for coupled fire-structure problems on massively parallel machines.

LES Model Development

The primary objective of this endeavor is to create and refine models for turbulence in fires using LES. This involves the implementation of the latest LES models and creation of new ones for turbulent fluxes, scalar mixing and reaction rates. The models can range from simple Smagorinsky models to scale-similarity and dynamic Smagorinsky models. Verification and validation are important aspects of this effort. In particular, data from various sources, including the National Institute for Standards and Technology (NIST) and Sandia National Laboratory, will be used to test and benchmark the models.

Multimaterial Model Development

The objective of this endeavor is to create, implement, verify and validate a multimaterial CFD algorithm for fluid-structure interactions as can be encountered in the CSAFE scenario, within the context of the UCF. The model should be capable of handling both continuous and discrete states of matter, and should be capable of treating arbitrary volume fractions of gas and solid in any control volume.

Chemical Mechanism for Pool Fires

The objectives of this task are:

1. To develop a surrogate for jet fuel. This includes theoretical and experimental (validation) aspects.
2. To develop a chemical kinetic mechanism for the surrogate. This will involve compilation of data in the literature, experimental work, as well as calculations from transition-state-theory.
3. To develop the Chemical Information Management System, a database that serves as a bridge between fundamental chemistry and computational engineering.
4. To develop a soot mechanism. This involves a combination of collecting databases of experimental work of other researchers, assimilating and filling gaps in kinetic mechanisms, using the idea of classes of reactions to create and extend formation mechanisms for soot precursors, developing theories for polymerization, agglomeration, and other mechanisms for size growth, and developing soot oxidation mechanisms.
Submodels for Mixing and Reaction Models

The main objectives in this task are:

1. To develop a systematic methodology for reduction of large chemical mechanisms from hundreds and maybe thousands of degrees of freedom to a few (of the order of 5-10) degrees of freedom, based on a time-scale analysis. This will involve the use and extension of Intrinsic Low-Dimensional Manifold (ILDM) and other similar time-scale based ideas for mechanism reduction.
2. To validate reduced chemical kinetic mechanisms using DNS (Direct Numerical Simulation) of turbulent reacting flows.
3. To develop submodels for effective treatment of turbulent micromixing, including multidimensional beta-PDF (Probability Density Function) models. This will also involve creating efficient calculation procedures, such as in-situ adaptive tabulation (ISAT) for integration over these high-dimensional multivariate distributions.

Parallel Radiation Model

This task will involve:

1. Parallelization of the existing Discrete Ordinates Method (DOM)
2. Investigation and implementation of Monte-Carlo methods for radiative heat transfer.
4. Incorporation of optical properties of soot for correct prediction of emission, absorption and scattering of radiation.
5. Incorporation of spectral effects in radiation.

Fire Spread Year 3 Key Results

Demonstration of Large-Eddy Simulation Pool Fire Calculation.

Hydrocarbon pool fires are characterized by a wide range of continuum length scales (1mm-1km) and corresponding time scales (1fs-1s). This entire range can not be directly computed even on the peta-flop computers yet anticipated for the ASCI program. However, simulating as much of the dynamic behavior of a puffing pool fire is essential to capturing accurate physics for fire and other process affected by fire physics and chemistry such as heat transfer to engulfed and surrounding objects and emissions from the pool fire itself. We have developed and demonstrated a simulation tool that uses LES techniques to resolve the puffing dynamics of a hydrocarbon pool fire.

Software Design and Integration of Fire Physics.

Multidisciplinary simulations require a software design that allows physics-based components to be independently developed and integrated into a complex system simulation. We have designed component-based software that abstracts the complexity of fire physics and chemistry. The software components modularize and encapsulate the functionality found in the multidisciplinary problem of accidental pool fires. This component-based software is integrated with the Uintah Computational Framework to provide the flexibility to couple with structural mechanics and to perform massively
parallel computations required to simulate accidental fire scenarios in the presence of structure.

**Multi-phase, Multimaterial Algorithm Developed**

A general multimaterial formulation has been developed that will accurately cover the full spectrum of fluid-structure interactions where any or all of the materials can be continuous or discrete and can have arbitrary volume fractions ranging from zero to unity. We have compiled a design document describing the theory and algorithm for this multi-phase, multimaterial formulation.

**Multi-physics Linear and Nonlinear Solver Analysis**

Multidisciplinary simulations require solution procedures that efficiently and accurately solve linear components and the coupled non-linear systems problem. We have incorporated the Portable, Extensible Toolkit for Scientific Computation (PETSc) developed at Argonne National Laboratory to test a wide variety of linear and nonlinear solvers for coupling the multi-physics aspects of the fire problem.

**Subgrid-Scale Combustion Model Development**

The chemical and physical processes that are occurring at scales smaller than the resolved mesh in fire simulations require a model. We have developed a statistical probability density function (PDF) model that accounts for subgrid-scale molecular mixing, and a reaction model that reduces the degrees of freedom of the chemical reaction mechanism for fire. These subgrid models are original contributions to fire simulations to allow coupling of the formation and destruction of soot and Polycyclic Aromatic Hydrocarbons to LES fire simulations.

**Soot Formation and Destruction Mechanism Identification**

Soot is a dominant component of hydrocarbon pool fires. The chemical kinetic mechanisms for formation and destruction of soot are important to pool fires as well as most other combustion processes. We have assembled an overall soot formation mechanism starting from heptane fuel as a surrogate hydrocarbon fuel. This mechanism draws on experimental and ab initio approaches to obtaining rate data for the elementary reactions assembled from the literature. The mechanism has been validated by comparison to data from laboratory flames.

**Radiative Heat Transfer Coupled with Fire Simulation**

Radiation is the dominant mode of heat transfer from and within pool fires. We have coupled a discrete-ordinates radiation model to the LES fire simulation to include radiation with soot in a pool fire calculation.

**Fire Simulation Validation**

We have compared fire simulation with experimental data from pool fires at a range of scales including fires up to 20 m in diameter. Puffing frequency, soot volume fraction and radiant flux have been compared. Validation of individual components within the fire simulation has also been performed by comparison of the soot formation mechanism with measurements from laboratory flames.
Container Dynamics SDRM Step Team

Vision
Development of a general continuum mechanics formulation to simulate a wide range of continuum phenomena that accurately represent underlying physical processes.

Mission
The CD team is focused on providing reliable simulation tools to predict large-deformation, multi-physics phenomena resulting from chemical transformation, phase transition, and thermal/mechanical loading of solid materials. Inherent in this mission is commitment to developing physically based submodels for material behavior and the efficient simulation of these processes through massively parallel computation.

Goals
- Develop a comprehensive simulation framework for studying multi-physics solid material behavior. The framework will allow for incorporation of a wide range of processes including:
  - frictional contact
  - large deformations
  - constitutive models that capture behavior of PBX’s
  - explicit fracture
  - Fragmentation
  - chemical transformation
- Prediction of material properties through MD simulations
- Micromechanical analysis of composite PBX material response
- Implementation of CD modules in the C-SAFE PSE
- Couple the solid mechanics simulation tool to a cfd solver allowing tightly coupled fluid-structure interactions
- Demonstrate scalability of tools on massively parallel architecture

Research Tasks:

General MPM Code Development.
The underlying computational approach for our problem is the Material Point Method (MPM). A strong research effort is devoted to tailoring this algorithm to C-SAFE applications. Specific research areas include implicit MPM development, parallelization, and incorporation of material contact. Key Personnel: Guilkey, Schmidt, Bardenhagen, Weiss.

Fracture Mechanics.
Simulations that realistically account for rupture and fragmentation must incorporate accurate treatment of fracture processes. This effort is directed at developing thermodynamic based algorithms for incorporating explicit fracture in the MPM code. Key Personnel: Nairn, Tan.
**Constitutive Modeling.**
Material response for PBX’s should describe rate dependence of loading, hysteresis, effects of damage, and phase transformation, among others. Our constitutive modeling effort involves collaboration with National Laboratory personnel to most accurately describe PBX response. Key Personnel: Bardenhagen.

**Material Property Determination.**
This research is focused on using MD simulations to predict behavior of HMX, polymeric binders, and investigate interface effects between HMX and binder materials. Also involved in this research is development of new algorithms to more efficiently simulate molecular dynamics. Key Personnel: G. Smith, Bedrov, Byutner.

**Micromechanic Analysis.**
PBX’s consist of a high-volume fraction, random arrangement of energetic particles in a polymeric binder. The material properties of the composite material can be greatly affected by particle size volume fraction, size distribution, and internal damage. This research effort is directed at the development of micromechanic models for PBX materials and extending models to include more physical characteristics such as damage and stress bridging. Key Personnel: Adams, Biswajit, Narra.

**Fluid-Structure Interactions.**
This research is concerned with the development of tightly coupled fluid-structure interaction algorithms. Efforts are directed at a general multi-material continuum mechanic formulation that allows for arbitrary constitutive models and equations of state. Key Personnel: Harman, Mell.

**Container Dynamics Year 3 Key Results:**

1. Development and validation of implicit MPM formulation.
2. Incorporation of MPM into new Uintah Computational Framework (UCF) and excellent scaling to 128 processors.
3. Large scale simulations of granular compaction.
4. Demonstration of efficient and accurate frictional contact algorithm.
5. Three-dimension explicit fracture incorporated into MPM.
6. Thermophysical properties for crystalline HMX (a, b, and d phases) for range of temperature and pressures via MD simulations.
7. Thermophysical properties for polymeric binders for range of temperature and pressures via MD simulations.
8. Development of algorithms for incorporating damage and stress bridging in micromechanics analysis.
HE Transformations SDRM Step Team

Vision
The macroscopic behavior of large-scale fire simulation programs should reflect the fundamental behavior of systems at the molecular level.

Mission
The HE Transformation Team is committed to creating a set of computational tools for calculating the fundamental molecular properties of energetic materials that may exist in a fire environment, and to provide a means of bridging extreme scales of length and time to estimate macroscopic behavior of the fire based on molecular fundamentals.

Goals
1. Create a robust set of quantum chemistry tools for calculating molecular properties of energetic materials including physical properties (elastic constants, thermal conductivity and heat capacity as functions of temperature and pressure) and chemical properties (chemical bond dissociation energies, reaction pathways and kinetic constants).
2. Demonstrate the utility of these tools within the framework of the initial C-SAFE simulation scenario (i.e., a steel cylinder filled with polymer-bonded explosive suspended above a hydrocarbon pool fire).
3. Create tools for revealing the effects of condensed phase environment on the rates and mechanisms of reactions of energetic materials.
4. Create C-SAFE computational modules that run in the problem-solving environment (PSE) that describe the sub-grid scale ignition and combustion behavior of energetic materials, based as much as possible on molecular fundamentals derived from the computational tools described above.

Research Group Tasks

Greg Voth Group
This research group is primarily concerned with creating a massively parallel version of "first-principles" molecular dynamics software that is capable of simulating chemical reactions of energetic materials. The parallelism is required to treat large-scale condensed phase systems (many atoms and molecules interacting with each other as solid or liquid). The "first-principles" aspect is required because it is not yet possible to construct analytical potential energy surfaces for large molecules that are capable of describing bond-breaking and bond-making events in many dimensions simultaneously. The "first-principles" approach solves for the energy and forces of the complete quantum mechanical system (all atoms) at each time step in the simulation.

Grant Smith Group
This group is principally concerned with calculating physical properties of energetic materials (e.g., elastic, PVT, thermal conductivity) that do not involve chemical
reactions. This information is required mainly by the Container Dynamics group for determining the physical properties of materials in the simulation. Analytical functional forms for intermolecular potentials are derived from *ab initio* quantum mechanics calculations, including internal degrees of freedom for allowing individual molecules to be flexible in the calculation. Smith’s group has been developing massively parallel classical molecular dynamics code that allows these properties to be calculated for pure materials (e.g., HMX, DMNA and polymer binders). Future development efforts will include the capability of describing the properties of interfacial regions (e.g., HMX/viton) of composite materials.

**Thanh Truong Group**

This group plays a dual role. The HE Transformations step funds this group to perform calculations of gas phase reaction rate constants as a function of temperature. The focus is on a small set of chemical reactions that are important in gas phase flame chemistry of HMX and RDX. This effort includes a code development task to derive estimates of reaction rates for chemically complex systems based on detailed calculations of simpler chemical analogs. The Fire Spread step funds the Truong group for development of a database tool for storing a library of evaluated rate constants for gas phase combustion reactions.

**Merrill Beckstead Group**

This group is working to develop a sub-grid scale module for describing the ignition and combustion behavior of HMX or PBX (particles in the Material Point Method approach of the Container Dynamics group). The focus of this model is to incorporate as much detail as possible from the elementary gas phase flame reactions into a simplified (i.e., computationally fast) module that lives in the Uintah PSE.

**Chuck Wight Group**

This group also has a dual task. The first is to develop reliable means of analyzing laboratory data on thermal decomposition of composite explosives (PBXs) for incorporation into sub-grid scale combustion simulation programs. The intent is to provide an intermediate stage of incorporating experimental information into the C-SAFE simulation while the molecular fundamentals tools are being developed to handle the highly complex chemical environments of composite materials. The second task is to develop a sub-grid scale combustion simulation program. This effort is focused on providing as much detail as possible about the structure of the burning surface and the detailed solid-gas reactions, while using simplified representations of the gas phase chemistry.

**HE Transformations Year 3 Key Results:**

**Massively Parallel Molecular Dynamics (MD) Simulations**

The HE Team has taken a two-pronged approach to calculating macroscopic physical and chemical properties of energetic materials based on molecular level quantum mechanical calculations. For physical properties such as thermal conductivity, elastic moduli, and cohesive strength, we have developed a computational tool for
running static quantum chemistry calculations and transforming the results into classical force field parameters. The parameters are then used in massively parallel MD simulations to determine the macroscopic physical properties as a function of temperature, pressure and density. For chemical properties (rates and mechanisms of condensed phase chemical reactions), the classical MD approach is generally unable to accurately describe the forces associated with bond-breaking and bond-making events. Therefore, we have developed a parallel implementation of "first-principles" MD codes that compute the forces from quantum mechanical calculations at each step of the simulation. The principal benefit of the parallel implementation is the ability to solve very large sparse matrices in the quantum chemistry part of the code.

**Gas Phase Flame Chemistry**

Most of the heat generated by the gas phase flame occurs by chemical reactions that take place in a complex series of isolated binary collisions. For the high explosive HMX, a reasonably complete chemical mechanism for this involves about 50 different types of molecules and 250 different chemical reactions. Many of the rate constants for these reactions are not known accurately, so the HE Team has developed new computational tools for calculating the rate constants as a function of temperature. In a real sense, this task has been to take rate constants that have been developed through fitting of engineering combustion models and validate them against high level kinetic calculations. Several of the rate constants for key reactions in the flame chemistry model have been revised in this way, providing a tighter connection between the overall simulation and the fundamental properties of molecules.

**Particle Combustion Models**

One of the weakest parts of engineering combustion models is the description of condensed phase reactions that take place at the burning surface of energetic materials. The HE Team has taken a dual approach to the development of sub-grid scale calculations of burn rates of individual particles in the MPM part of the large-scale simulation. The sub-grid scale models are required because the resolution required to describe the gas phase flame structure at a burning surface is typically a few microns. However, the computational cost of carrying out the large-scale simulation at this resolution is prohibitive. Therefore, the sub-grid scale model must be able to predict the mass burning rate of each MPM particle as a function of temperature, pressure, chemical state and thermal history. The HE Team has developed four different sub-grid scale programs for this task. The simplest and fastest of these is 1-D, assumes an infinitely thin surface, and uses an equilibrium chemical reaction set. The next model takes account of surface structure in 1-D with a simplified but dynamically varying chemical reaction set to describe the vaporization and gas phase flame structure. The third model incorporates a full chemical reaction set to describe the gas phase flame structure, and the fourth model is a 2-D model that can account for texture of the burning surface. Only the simplest and fastest model will be used to dynamically supply burn rate data to the simulation engine (Uintah Computational Framework), while the others will be used in off-line mode to provide a means to validate the parameters used in the simplified model.
Computer Science

Vision

The year 5 C-SAFE product should be a highly scalable parallel PSE implementation that can be used to simulate the behavior of energetic materials engulfed in fire.

Mission

The Computer Science Team is developing the Uintah Problem Solving Environment (PSE), which includes the specification of the overall Uintah software architecture and incorporation of software parallelization strategies for efficiently utilizing ASCI hardware platforms. This includes the development of visualization, performance analysis tools, and scientific data management tools for large scale simulations.

Goals

1. Problem Solving Environment (PSE): The PSE team has focused its efforts on creating the Uintah Computational Framework (UCF) supporting both MPI and MPI/Thread parallelization. During this effort, the PSE team has guided the integration of the other teams’ codes into the overall Uintah architecture.
2. Visualization: The Uintah visualization tools have been greatly enhanced and include (1) extended particle visualization tools, (2) grid visualization and query methods, (3) hardware based multi-resolution volume rendering, (4) animated streamlines, and (5) to utilize Real Time Ray Tracing (RTRT).
3. Software and Data Management (SDM): Development efforts in C-SAFE scientific data management during the past year have focused on bringing tools to productive use within C-SAFE tasks. This involved three areas: SDM web service, Uintah Blazer, and Validation team combustion research support.
4. Performance Analysis: We are currently working to develop tools that will help track the utilization of processor resources based on the task graph parallelization abstraction.

Research Group Tasks

Chris Johnson Group

This past year, the main efforts of the Problem Solving Environment (PSE) team have been focused on the examination and implementation of the Uintah Computational Framework (UCF). The UCF has been designed and implemented by the PSE team to satisfy three goals: 1) To provide a general framework for massive scale simulations of fluid and particle physics, 2) to maximize parallelization and scalability, and 3) to allow scientists from outside the computer field to have an intuitive method for easily inserting their algorithms into a parallel framework without being bogged down by all the details of parallel programming.
Chuck Hansen Group

The Visualization team has been working on three major areas: (1) step-specific visualization tools, (2) multi-resolution techniques, and (3) visualization data manipulation and retrieval. The step-specific visualization tools include particle methods for the Container Dynamics (CD) step, volume rendering techniques for the Fire-Spread (FS) step, vector visualization methods for both FS and CD, and sphere rendering methods for the real-time ray-tracer which is targeted towards the CD and Molecular Dynamics (MD) steps. This past year has also focused on common infrastructure within the Uintah PSE which applies to all steps as well as step-specific visualization methods.

Gary Lindstrom Group

The Scientific Data Management (SDM) team has established a dedicated web server to support web-based C-SAFE activity, and to provide SDM infrastructure including stronger security, such as digital certificates, as security needs increase. In consultation with the UCF design and implementation team, a Blazer web service was established for the registration, browsing and querying of UCF simulation run files. In consultation with Validation Team, and their combustion research colleagues around the world, another service has been established to provide combustion simulations using the Blazer web server. We have participated in the ASCI CDM effort as well.

Al Davis Group

The key abstraction that the UCF builds upon for parallelization is the task graph. This abstraction aids us in analyzing the UCF’s performance. Using graph analysis techniques, we are able to determine the critical and near-critical paths through the task graph. This allows us to focus our efforts on speeding up the portions of the code that actually create the bottlenecks during execution. We are also working on having the ability to perform visualization of statistical data derived from the task graphs.

Computer Science Year 3 Key Results:

Uintah PSE

To date, we have focused on: continued integration of C-SAFE codes into the Uintah PSE, greater Uintah MPI-style parallelism, coordination of center-wide code and data integration, coordination of center-wide software engineering and development support (CVS, C-SAFE web site, bug tracking, etc.), implementation of the new Uintah architecture to support MPI and shared memory, development of Uintah PSE in accordance with evolving CCA model, and development of time-varying visualizations for Container Dynamics and Fire Spread.

The new PSE architecture has been developed so as to support distributed memory parallelism for scalability, a flexible component model for time-varying simulations, interoperability with other DOE CCA efforts, and integration for the C-SAFE simulation architecture. The parallelization strategy is organized in terms of a master controller which has low bandwidth connections to the control workstation as well as slave controllers which actually control worker processes. A high bandwidth connection is used between the worker processes for both shared memory communication as well as message passing. Thus, the PSE visual interface and visualization tools are
handled locally and heavy computational processes are run remotely on high performance computing platforms.

**Uintah Computational Framework**

The C-SAFE architecture coordination has involved several issues. The design of the Uintah computational framework has required a detailed study. In order to achieve our main goals, it is absolutely essential to provide a common computational framework for the Fire Spread and Container Dynamics codes. Therefore, much of our effort this year has focused on the development of that framework within the Uintah PSE. In addition, we have continued to develop visualization algorithms, performance analysis tools and simulation management support technology.

The status of the Utah Computational Framework is that about 20K lines of new code have been written since October, and although there is still much to do, we are able to run problems that were impossible heretofore. We have run a 16M particle MPM simulation at 7 seconds per timestep on 128 processors. We are currently pursuing integration, larger problems and a larger number of processors.

**Integration of application codes into PSE**

We have identified 4 PSE software integration levels, and are proceeding with both the design of the C-SAFE simulation system, as well as the implementation of the Uintah code based on these levels. They are: Level 1: software produces data for use with PSE visualization or further PSE computation; Level 2: software is wrapped in a PSE module or communicates with the PSE at run time; Level 3: software is designed using PSE infrastructure; Level 4: software is implemented with PSE component model. Another feature of the current PSE architecture is that it allows separation of functionality so that different teams can develop modules independently, as well as providing the shared computational framework (mesh and solvers). At this point, the MPM code is fairly mature within the PSE. The Fire Spread code is in its prototype stage, as is the database and simulation management tools (i.e., Uintah Blazer) are in prototype stage.

**Scalability**

The Uintah PSE scalability challenge stems in large part from the wide range of loads due to AMR, different numbers of particles in different spatial regions, cost of ODE solvers which can be spatially dependent, radiation models, and architectural communication limitations. In order to get a handle on these issues, we have developed a key abstraction: the task graph. Each algorithm defines its required inputs and outputs. These are described per patch by variable names and spatial relations (using ghost cells). Communication is performed at the edges of the graph. Furthermore, we have put a lot of thought into the output requirements of such codes and plan to have output within the mix of processes to be handled by the scheduler. The scheduler decides which task will be executed by which processor and is guided by cost models for computation and communication. The disadvantages of the task graph approach are that an optimal solution is NP-hard (however, good solutions aren’t too hard), creation of the schedule can be costly (only needs to be recomputed at re-gridding or periodically for load...
balance; cost can be amortized over several steps). On the other hand, it accommodates flexible integration needs and workload profiles, offers a mix of static and dynamic load balancing, helps manage complexity of a mixed threads/MPI programming model, and allows pieces to evolve independently.

**Scientific Data Management**

The Scientific Data Management team has developed a combustion database that allows access to combustion models (Fortran code) which can be executed through a web interface. Inputs, outputs and meta-data are stored in a relational database. Results can be graphed using a simple graphing applet. The user can perform side by side comparisons of the different models. This system is in use by the combustion community (using a simple password protection scheme). In terms of large-scale data management, a uniform output database from the Uintah computational framework is under development. An XML-based repository holds the Uintah problem specification, simulation meta-data (e.g., run date, user, machine, etc.), output data, and checkpoint data.
**Validation Step Team**

**Vision**

Effective model development requires validation of individual model components as well as the integrated code, and such validation is carried out at multiple levels of detail.

**Mission**

The Validation team will provide support to the other Center steps in validating key physical aspects of the targeted simulation scenario. We will do this by providing experimental information when such data is not available or is incomplete. We will also provide an interactive bridge between Center steps by validation of coupled physical processes and involving representatives from multiple steps in planning, executing and evaluating results from validation activities. Validation efforts will be increasingly guided by feedback from sensitivity analyses within the Uintah PSE.

**Goals**

Our goals are to provide validation information to the Center effort at four different levels of detail:
1. Fundamental rates and properties
2. Critical tests of key processes in individual steps
3. Pilot-scale tests with JP-8 and surrogate fuel flames, particularly to test processes occurring at interfaces
4. Integrated experiments involving the explosion of cylinders containing HMX-based material

**Research Group Tasks**

**Web-Based Validation**

This validation task addresses the development of an interactive, web-based tool for comparing soot formation mechanisms being developed both within and outside of C-SAFE. The web-based interface is allowing electronic collaboration with key researchers around the world, to produce the best possible soot mechanism. This task is in support of the Fire Spread step and involves the assistance of the Computer Science step.

**Surrogate Formulation**

This task, in support of the Fire Spread step, involves the development of a validated surrogate fuel composition to facilitate the development of a reduced combustion mechanism for the fire code. Jet fuel is comprised of hundreds of compounds, and a simplified representation is required to allow chemical kinetic description. The surrogate formulation is critical, as it provides the basis for full combustion and soot mechanisms being developed under other tasks, and then these detailed mechanisms will then be reduced to allow implementation into the CFD code. Physical properties to be matched are those of greatest influence in pool fires and include boiling rate and sooting tendency.
Soot Concentration Measurements

The efforts in this task provide for validation of the surrogate formulation developed in the previous task. A laser-based absorption/emission technique is being developed in collaboration with SNLL to allow for detailed measurements of soot volume fraction and soot temperature. The technique will allow for comparison of soot formation tendencies between the actual jet fuel and the surrogate fuel, thereby providing validation of the efficacy of the surrogate. This task is in support of the Fire Spread step.

Slot Burner Measurements

The construction and operation of a two-dimensional slot burner is the main component of this task. The burner will be used for surrogate fuel validation, and for validation of the implemented reduced mechanisms for fuel combustion and soot formation. The slot burner provides a simple geometry and laminar flow configuration that will provide data for validation of the reduced mechanisms and for verification of their implementation into the fire code within the Uintah PSE. The additional complications of turbulent flow and free entrainment will be verified by comparison with data taken in the pilot-scale facility and at the Laurence Canyon burn site at SNLA. These efforts are in support of the Fire Spread step.

HMX characterization

This experimental task is being carried out by Chuck Wight’s group and involves the detailed characterization thermogravimetric analysis of the decomposition and reactions of the HMX material as well as the binder material. Samples of the actual material used in the live explosion tests performed at Thiokol have been collected to facilitate their analysis. This task is in support of the HE Transformations step.

Soot Structure Characterization

Ron Pugmire’s group is applying their solid state NMR technique to determine the chemical structure of soot particles formed early in a pool fire flame. One of the significant gaps in the understanding of soot formation is the process of particle inception from large multi-ring aromatic compounds. By understanding the chemical structure of recently-formed soot particles, mechanistic pathways can be hypothesized to provide a linkage between current models for homogeneous gas-phase molecular growth and soot particle formation and growth. Soot samples are being generated using known pure species that comprise the jet fuel surrogate, the jet fuel surrogate and the jet fuel itself to allow comparison of the various soot structures. This task is in support of the Fire Spread step.

Pilot-Scale Pool Fire Tests

This task is in support of the Fire Spread and Container Dynamics steps and involves the construction and operation of a pilot-scale pool fire test facility. This facility will allow for measurements to be used for model validation of temperature distributions, heat flux to walls, heat flux to an immersed container, and generation of soot samples for NMR characterization, under representative pool fire conditions of turbulent flow, with
either confined or unconfined boundaries. Fuels to be utilized will include the jet fuel and its surrogate, as well as individual surrogate species and comparisons will allow validation of the surrogate fuel properties with respect to heat flux and boiling rate under pool fire conditions. This task couples the physical processes of the pool fire with the inclusion of an immersed container (without HE material) and is in support of the Fire Spread and Container Dynamics tasks.

**Thiokol Fast Cook-off Tests**

This experimental effort involves the use of containers filled with PBX 9501 in pool fire or controlled heating experiments at the remote test facilities of Thiokol Propulsion. The purpose of these tests is to provide information on heat transfer through container walls, pressurization inside containers, container break up, the influence of voidages inside the container on the fast cook off of the HE material. Measured quantities include internal and external temperatures, as well as internal pressures, and extensive video records are made of each test to provide greater understanding of how the explosion might be initiated and how they proceed. Heat transfer modeling of the container is identifying key physical processes that must be included in the container dynamics model within the Uintah PSE. This task is in support of the Container Dynamics and HE Transformation steps.

**Validation Year 3 Key Results:**

**Web site for Collaborative Interaction on the Development and Validation of the Soot Model**

A Web site has been developed to allow the group working on the development of the soot model to interact with our collaborators at LLNL, LBL, SNLA, SNLL, UC Berkeley, Polytechnic of Milan, and the University of Naples. The site permits the remote running of the soot models on a local server, thus providing a means for all collaborators work with the same version of mechanisms and models. The site also provides a forum for dialogue on the results of mechanism comparisons.

**Surrogate Fuel Formulation.**

The criterion for the development of a surrogate fuel was that the fuel should reproduce the features of JP-8 important for pool fires. These include the vaporization rate and the radiation characteristics, which are heavily dominated by the soot formation. Oxidation mechanisms and rate constants need to be available for the components of the surrogate mixture and the components should be available at economic prices for testing. A surrogate mixture consisting of a mixture of n-tetradecane, n-dodecane, methylclohexane, i-octane, tetralin, and xylene has been found to represent well the distillation curve and the sooting index of the test JP-8 selected.

**Soot Concentration Measurements**

The equipment has been assembled for the laser absorption measurements and preliminary measurements have been made in a simple flame. The equipment for the emission measurements has been ordered. The student working on this project has visited
SNLL three times this year to work in the laboratory with Chris Shaddix, and has also assisted in the application of the technique with Chris during an outdoor test burn at SNLA.

**Slot Burner Measurements**

The construction of the burner is nearing completion and we anticipate bringing it on line near the end of October or in early November. Initial characterization using the laser diagnostic from the previous task will be accomplished using a simple gaseous hydrocarbon fuel, during which time a technique will be developed for vaporizing the liquid fuels to be used to simulate jet fuel.

**Identification of Soot Precursor Structures.**

One of the less certain aspects of a soot formation model is the transition from vapor to solid. In order to critically test this aspect of the soot model, NMR techniques are being developed to determine the structures of soot precursors and young soots. Within the past year, major progress has been made in the characterization of gas phase soot precursors and soots in the early stages of evolution. The soot precursors and soot samples were produced in (1) a laminar flow drop tube furnace, and (2) a flat flame burner, and five model aromatic compounds were studied over a temperature range of 1250 to 1470 K. Results to date indicate that products observed in the early stages of soot formation reflect the material from which the soot samples are made.

**Pilot-Scale Pool Fire Tests**

The adequacy of the surrogate formulation to represent JP-8 will be tested in a pilot-scale facility capable of conducting up to one-meter-diameter pool fires in a controlled environment. Construction of the facility at the University of Utah off-campus combustion research center has been completed, and preliminary characterization and equipment shakedown has begun using jet fuel pool fires.

**Thiokol Fast Cook-off Tests**

Five fast cook-off tests were performed during the month of October 1999 at Thiokol Propulsion’s Promontory facility. One of the tests was an inert test utilizing barium nitrate, and the other four tests were live tests containing the explosive material PBX-9501. The purpose of this testing campaign was to obtain information on the importance of bonding of the explosive material to the container in which it was housed, and the effect of this bonding on the time to explosion. Additional information was also obtained on the effect of pressure-sealing of the container, reproducibility of results, and the heat-up characteristics of the cylinder arrangement. Heat transfer modeling of the temperature data available from these tests, has indicated the critical nature of debonding on the time required for explosion. Calculations indicate that the dominant resistance to heat transfer into the HE material is due to small air gaps between the cylinder wall and the HE material. These gaps can be present initially due to incomplete bonding of the HE material to the container, and then are exacerbated in later stages due to disparities in the thermal expansion of the steel and the HE material.
One of the key physical observations from these tests was the identification of "ballooning" behavior for the container prior to rupture. The apparent diameter of the container, determined by measuring the fragments after the explosion, increased by as much as 50% prior to the container splitting apart. This observation will provide important feedback to the Container Dynamics group to allow for appropriate constitutive models required for modeling the scenario of a live container in a pool fire. In addition, it became apparent that the variation of potentially important parameters, such as bonding of the explosive material to the container or sealing of the container, are less significant than test-to-test variations due to uncontrolled conditions such as weather.
2.0 Introduction to C-SAFE

Under sponsorship from the ASCI program, the University of Utah has created the Center for the Simulation of Accidental Fires and Explosions (C-SAFE). Its focus is specifically on providing state-of-the-art, science-based tools for the numerical simulation of accidental fires and explosions, especially within the context of handling and storage of highly flammable materials. The primary objective is to provide a system that includes a problem-solving environment in which fundamental chemistry and engineering physics are fully coupled with non-linear solvers, optimization, computational steering, visualization and experimental data verification.

Although the ultimate C-SAFE goal is to simulate fires involving a diverse range of accident scenarios including multiple high-energy devices, complex building/surroundings geometries and many fuel sources, the initial efforts during the first five years of the program will focus on rapid heating of a container with conventional explosives in a pool fire (e.g., a bomb involved in an intense jet-fuel fire after an airplane crash).

Such large-scale problems require consideration of fundamental gas and condensed phase chemistry, structural mechanics, turbulent reacting flows, convective and radiative heat transfer, and mass transfer, in a time-accurate, full-physics simulation of accidental fires. This simulation will be expansive enough to include the physical and chemical changes in containment vessels and structures, the mechanical stress and rupture of the container, and the chemistry and physics of organic, metallic and energetic material inside the vessel. It will include deflagration-to-detonation transitions (DDT) of any energetic material in the fire, but the simulation will end when/if detonation occurs. C-SAFE will provide coupling of the micro-and meso-scale contributions to the macroscopic application in order to provide full-physics across the breadth of supporting mechanistic disciplines, and to achieve efficient utilization of ASCI program supercomputers.

2.1 Simulation Development RoadMap (SDRM)

We are using a Simulation Development Roadmap (Figure 2.1) to define the sequential events which occur in this specific problem as illustrated on the next page. The simulation will begin at the point of ignition. The process consists of three distinct sequential events: Fire Spread, Container Dynamics and High Energy Transformations. These steps describe the processes occurring in a number of different accident scenarios of interest to the center. We assume that an ignition event occurs and then the fire begins to spread. The presence of the fire can cause the container of HE material to undergo changes, perhaps rupture and/or even ignite itself. Simultaneously or sequentially the HE material in the container may undergo transformations which may lead to ignition and explosion. The overall mission is to integrate these computational steps into a coupled fire and explosion system.

Fire Spread

Within C-SAFE the Fire Spread SDRM step team is responsible for providing a validated simulation of a pool fire with structural material involved in the fire. This team has responsibilities for all gas phase computations on the computational mesh, including
the aspects of the simulation dealing with explosive or energetic material that occur in the gas phase on the mesh.

**Container Dynamics**

Responsibilities of the Container Dynamics step to the C-SAFE product is the numerical description of the response of all solid phase material to both thermal and mechanical loading. This includes both structural simulations of "containers" and HE material as well as material property specification of solid phase materials. Simulation methodologies employed for the structural simulation will involve particle Lagrangian techniques (the Material Point Method) over a background Eulerian grid. Material properties for homogeneous phases will be obtained from molecular dynamics simulations while micro-mechanical modeling will be used to develop composite material property behavior and material response models (constitutive relations) for use in the macroscopic simulation. Coupling to the Fire Spread step at the macroscopic level and the HE Transformation step (through subgrid energy conversion models) are key aspects of this step.

**HE Transformations**

The primary responsibility of the HE Transformations SDRM Step is to generate a reliable sub-grid scale model for chemical reactions of energetic materials, taking into account effects of porosity, formation of microcracks and interfacial properties that are not described explicitly by the grid structure itself (which will be generated by the CD
Step). Most of the effort of this step will be put into condensed phase chemistry of energetic materials, but the HE Transformations step also includes gas phase reactions of energetic materials and effects of release of combustible gases into the underlying hydrocarbon fire.

**Applied Mathematics within C-SAFE**

In the last year there has been evolution of the role of the applied mathematics team. In the past we have had a separate division within C-SAFE to focus on applied math concerns. We have eliminated that division this year and placed the individuals within each of the SDRM step teams. This move is consistent with our gradual transition towards a more integrated approach to solving our common problem. In the past we had an applied mathematics team working on simple test problems and guiding the steps in the choice of solvers, preconditioners, time stepping strategies, etc. We have now found it time for an applied math culture to permeate the steps and to be part of the thinking process of the key people within the steps. This move is also consistent with the way we have organized our expertise in the discipline of Computer Science: we have software engineers within the steps who are strongly integrated into the Uintah PSE and Computational Framework development and are helping the rest of their respective teams in absorbing the essential paradigms of large-scale computation, code management, and framework-building into their own work and coding practice. As a result, we have abstracted key members of the former applied mathematics group into the individual steps; these are then responsible for providing advice and solving problems pertaining to the specific concerns of their steps, for making the right connections with key individuals in the broader applied mathematics community, and for making sure that the general direction of the step is consistent with a well-considered applied mathematics base.

We have found this model productive. The entire team is more focused on mathematical concerns that relate to the solvability of the overall problem. These include: robust parallel preconditioners, implicit methods for the full nonlinear coupled problem, efficient linear and non-linear solvers, and boundary matching at the boundaries between disciplines and steps (see sections 3.1.3, 3.2.1 and 3.3.3). Examples of this new applied mathematics culture can be seen widely within the center, from robust linear solvers used by the HE team to the development of implicit MPM and CFD methods to the use of efficient integrators for chemical kinetics. We anticipate that, as we continue to move into the arena of full-physics, multiple-step calculations, the importance of having a strong applied math culture within the steps will become more and more important.

**Efforts supporting all SDRM step teams**

The Computer Science and Validation teams share the responsibility of supporting the three SDRM team efforts. The Computer Science group is developing the Problem Solving Environment (PSE), which includes the specification the overall Uintah software architecture and incorporation of software parallelization strategies for efficiently utilizing ASCI. The development of visualization and performance analysis tools within the PSE are also tasks assigned to this group, as is the support of data management infrastructure, including the development of large-scale simulation management tools.
The primary responsibilities of the Applied Mathematics team are to provide general software libraries, special purpose codes, and expertise for addressing essential computational tasks throughout C-SAFE. These tasks include adaptive mesh refinement, time integration, solution of very large scale linear and nonlinear systems, the associated development of preconditioners and multigrid/multilevel techniques, sensitivity analysis, and stiff solvers.

The Validation team is responsible for providing experimental information on coupled physical processes that will be used by the other C-SAFE steps for model validation. Some of this information will be obtained from existing sources in the scientific literature; however, some will be determined through experimental testing performed by the Validation group.

**Software Integration**

The C-SAFE team has developed an integration strategy to guide our product development, establishing a balance of effort and resources directed toward computational issues (parallelization, scaling, etc.) and the development of accurate, full-physics codes. We have organized our integration efforts at four levels within the Uintah architecture, from the top-level blueprint of the Uintah system, the PSE structure and the solver strategies, to the individual software modules.

- **Uintah System**: the user interface must provide a unified view of the various simulation codes and databases which comprise a complete scenario simulation, and allow insight into performance of the codes on ASCI supercomputing platforms, so as to relate the data to the initial conditions, parameters, and modules. Moreover, data formats must be consistent across codes and throughout the visualization modules.

- **Uintah PSE**: within the interactive, computational problem solving environment the user must be allowed a clear visual methodology so that various algorithms - implemented as modules within the PSE - are syntactically simple to assemble into computational units, and semantically meaningful when executed together. Moreover, interactive computational steering must be provided to allow the user to inter-operate with the ongoing simulation.

- **One mesh / integrated solvers**: in terms of our particular simulation domain - fire spread and container dynamics - we are achieving integration through the Uintah Computational Framework (UCF), a common adaptive mesh refinement package being developed as part of the C-SAFE effort. This provides us with a built-in capability to share state information between the grid and the particles involved in the simulation.

- **Software Integration**: Finally, the software from the various steps in the project must work together as a cohesive system; we are assuring this through our four software engineers - one in each step, and in the PSE - who have developed center-wide coding standards, revision control methods, class browsers, bug reporting and documentation standards.

**2.2 Product Vision**

During the first year of the program, the team carefully defined in great detail the characteristics of the five year product, termed Uintah 5.0. This product vision continues
to serve as the blueprint used to ensure that all members of the team are working toward the same goal and to help prioritize the use of limited resources. This section of the report summarizes the outcome of product visioning efforts.

**Product Concept**

The year 5 C-SAFE product (Uintah 5.0) will be a computational modeling system that can be used to simulate the behavior of energetic materials engulfed in fire. Features of the system will include:

- modular components, including the ability to adopt technologies from other sources;
- accurate representation of fluid-structure interactions through the use of a single computational grid to represent both flames and structures;
- modeling of multiscale phenomena via coupling of the macroscopic fire and container elements with each other and with sub-grid models for the physical and chemical properties of high energy explosives;
- a problem solving environment (PSE) which integrates simulation, data analysis, and visualization that will allow scientists to easily experiment with simulation parameters, explore the impact of alternative models and solution methods, and investigate the solution space of a particular problem scenario.

Uintah 5.0 will be validated by comparison of the results of a simulation scenario that consists of a container of highly energetic material engulfed in a hydrocarbon fire with experimental data obtained from the open literature, the DOE national laboratories, and experiments conducted by the C-SAFE team.

The creation of the primary C-SAFE product will also result in the development and incorporation of new secondary technologies, including:

- performance tuning tools for parallel processor code development
- advances in fire spread modeling
- advances in molecular chemistry
- advances in modeling solid state materials.

**Clients**

The envisioned primary user of Uintah 5.0 will be a small team of scientists and engineers with an interest in the effects of fire on materials. The user will not be a code developer, but may wish to use the PSE to select modules and strategies, possibly including modules developed at the labs. Secondary users may include code developers who will use the system as a testbed for generating difficult problems on which new methods may be evaluated.

Uintah 5.0 will be targeted to run on large-scale parallel systems currently being developed under the auspices of the ASCI PathForward program, although reduced versions will be able to operate on more generally available computers so that the program results will have the potential to be utilized by both the commercial and academic sectors.

**Specific Focus of Release 5.0**

The specific, computational focus of Uintah 5.0 (to be released during the 5th year of the project) will be as follows:
Hydrocarbon Fire
  arbitrary shape and size
  natural gas (via a burner) or light, liquid hydrocarbon fuels (pool fire)
  single or multipoint ignition
  quiescent conditions or specified wind
Container
  Metal construction
  Arbitrary location relative to the fire
  Specified initial temperature distribution
  Arbitrary size and shape
  Both closed and open ended construction
HE Material
  HMX formulations
  Multiple binders including HTPB and Viton

**Individual Modular Components**

Uintah 5.0 will contain the following modular components:
  Fire Spread
  TFANS/LES CFD Model
  finite rate hydrocarbon chemistry
  chemistry/mixing coupling (PDF)
  spectral, DO or Monte Carlo radiation
  soot formation and destruction
Container Dynamics
  general constitutive models
  multimaterial formulation
Material Point Method (MPM)
  rupture and fragmentation
  crack growth
  supporting material property calculations
High Energy Transformations
  supporting electronic structure calculations
  classical and quantum MD simulations
  chemical reaction rate constants
  condensed phase reaction mechanisms
  subgrid scale inhomogenity
Computational Framework
  robust problem solving environment
  visualization
  performance optimization
  simulation and data management
Applied Mathematics
  adaptive mesh refinement
  robust equation solvers
  sensitivity and probability analysis
2.3 Organizational Structure

Twenty key U of U faculty joined with strategically selected faculty from nearby Brigham Young University (1) and Worcester Polytechnic Institute (1) and experimental scientists from Thiokol to create the C-SAFE team. Twenty-four post doctoral fellows/professionals have joined the team as have 18 graduate students and 1 undergraduate student.

The leadership of the C-SAFE center has remained the same since the early days of the proposal preparation. Distinguished Professor David Pershing (who is now the Senior Vice President for Academic Affairs) continues to serve as the Center Director. Professors Tom Henderson, Computer Science; Greg Voth, Chemistry; and Phil Smith, Chemical and Fuels Engineering serve as Associate Directors.

Because of the problem complexity, both the personnel and the tasks are organized into six interdisciplinary teams in a coupled matrix structure as illustrated in Figure 2.2. Each C-SAFE participant (faculty, staff or student) is a member of one of these interdisciplinary teams. Each team is charged with the development of one subset of the overall application. The teams are composed of a step leader, a professional software development engineer (except in the cases of the validation and applied mathematics groups) and faculty, staff and student participants from the various scientific disciplines which are appropriate for the activity of the team. We believe this tightly integrated structure is essential to simultaneously ensure that (1) the common objective of developing a verified, fire and explosive simulation system is attained, and (2) modern scientific/computational techniques are used throughout. Decisions regarding selection of key components to integrate into each step are being based on nonlinear sensitivity analysis and numerical optimization of our overall accidental fire simulation. Our delivered product will be the C-SAFE system that embodies the complete technology for performing integrated and validated simulations of full-physics fires.

The Director, Associate Directors and team leaders form an Executive Committee which provides overall leadership for the center. This committee meets monthly to discuss strategic, administrative and technical issues. It recommends approval of the annual task structure and associated budgets. The group also serves to reinforce cross-disciplinary and cross-step communications by reviewing ongoing efforts on a regular basis.
C-SAFE Team Structure

SDRM-specific Step Teams

Ignition

Team: Fire Spread
Leader: Phil Smith
Software Engineer: Rajesh Rawat
Seshadi Kumar
Key Participants: Thanh Truong, Adel Sarofim

Accidental Detonation

Team: High Energy Transformations
Leader: Chuck Wight
Software Engineer: M. Ovchinnikov
Ap. Mathematician: Kris Sikorski

Team: Container Dynamics
Leader: Pat McMurtry
Software Engineer: John Schmidt
Ap. Mathematician: Ruddy Mell
Key Participants: Dan Adams, John Nairn, Jeff Weiss, Grant Smith

All-SDRM Step Teams

Team: Validation
Leader: Adel Sarofim
Software Engineer: none
Key Participants: Eric Eddings, Ron Pugmire, Thiokol

Team: Computer Science
Leader: Tom Henderson
Software Engineer: Davison de St. Germain
Chris Johnson, Al Davis, Chuck Hansen
Gary Lindstrom, Kris Sikorski

Key Participants:
3.0 Technical Progress Report

3.1 Fire Spread

3.1.1 Background

High heat flux and emissions from accidental fires pose significant human and property hazards. The Fire Spread step of the Strategic Development Roadmap is concerned with modeling the propagation of an ignition event over a pool of liquid hydrocarbon fuel.

These events have the potential of heating the material in the container or rupturing the container itself due to mechanical and/or thermal stresses. Consideration of the fire spread will include situations where the main container has ignited by impact and the subsequent reaction sequence or explosion has ignited the surroundings.

The propagation of a fire or explosion around the HE container depends on the availability of fuel and oxidizer and on the rate of the ensuing reaction. The mixing between the fuel and oxidizer depends on the fluid mechanics of the vapor phase. Often
the fluid flows are highly turbulent, leading to large variations in the length and time scales over which mixing occurs. The complex three-dimensional geometry involved adds to the difficulty in simulating such flows. The chemical reactions involve several thousands of elementary steps and hundreds of major and minor species and intermediates. These reactions are highly exothermic, and the resulting energy transfer occurs through both convection and radiation. Radiation is the dominant mode of heat transfer at high temperatures and is strongly dependent on the absorptive, emissive, and scattering properties of the gas mixture. It is therefore strongly affected by the presence of soot in the gas mixture. The large heat generation from these reactions leads to rapid changes in the properties of both solid and gaseous materials. When the rupture of the container is affected by these events, fragments of the container, which are ejected further, interact with the gas phase. In addition, all these processes are highly coupled. For example, chemical reaction depends on the level of mixing; however, chemical reaction affects the temperature through the amount of heat generated, and this changes the density and thus the level of mixing via fluid flow.

During the previous two years of this project, the Fire Spread (FS) Team demonstrated proof of concept for fire by completing a serial simulation tool of 3-D, large-scale pool fires. The transport processes included Large Eddy Simulation (LES) approximations for fluid dynamics and convection-diffusion scalar transport, and a discrete-ordinates method for radiation. The tool contained models for all essential physico-chemical subgrid scale models including turbulence, scalar mixing, and chemical reactions including soot. It demonstrated implicit and explicit solvers for the coupled non-linear system. It was verified for computational accuracy by several methods.

Figure 3.1.2: Contours of constant CO concentration on a plane through the diameter of a 20 m heptane pool fire simulation. This sequence of eight slices shows the simulation in various times subsequent to ignition.
including the method of fabricated solutions. Preliminary validation of modeling approximations has been initiated including comparisons to data for puffing frequency over a range of four orders of magnitude in pool diameters.

During this third year of the project, work has continued on advancing the physics of the fire simulation tool with sample results shown in Figures 3.1.1 and 3.1.2. Progress made on the advances in the physics and chemistry components are discussed in Section 3.1.4. Our major objectives for this third year have been to:

- Integrate all components of the fire simulation into the Uintah Computational Framework (UCF),
- Couple the fire simulation and computational fluid dynamics (CFD) components with the Material Point Method (MPM) components at the algorithm level to create a coupled multimaterial formulation.
- Resolve applied mathematics issues concern the coupling of the large, multiphysics fire problem.
- Advance the state of the physics and chemistry components of the fire simulation including the LES, subgrid scale turbulence, the chemical reaction and turbulent mixing, and the chemical kinetics for fire including soot.

### 3.1.2 Integration and Parallelization

The fire code that we developed and validated in the first two years, Arches, was a stand-alone, physics-based, serial code. To achieve the goal of developing a generalized framework for large-scale, massively parallel simulations of accidental fires and explosions, our focus in year three was on integrating the fire code in the UCF. Uintah is a component-based, visual problem-solving environment developed by the CS team. Uintah provides the framework for large-scale parallelization for different applications.

**Integration Strategy**

The integration of the Arches in Uintah is built on three principles: 1) Develop different reusable physics-based components that can be used interchangeably and interact with other components, 2) reuse the legacy stand-alone fire code (written in Fortran) as much as possible, and 3) use components developed by third parties, specifically non-linear and linear solvers designed for solving complex-flow problems.

Our first step was to design a generalized, component-based architecture for fire simulations that can be incorporated in any computational framework that provides support for parallelization. We worked closely with the CS team for the software design specifications. At first, we worked on integration with the SAMRAI (Structured Adaptive Mesh Refinement Applications Interface) framework. Later, we took our design, made some minor modifications and integrated it with the UCF. Figure 3.1.3 illustrates different design components and their relationships for fire simulations. These components are designed around real world concepts (such as subgrid scale micromixing as represented by the Mixing Model component), and they encapsulate functionality found in multiphysics problems. Many of the design decisions were necessitated by the parallel-processing paradigm provided by UCF.

After finalizing the design, we added details to the design model to describe and optimize the implementation of the different physics-based, reusable components. These
components fulfill two important features. First, they provide an interface to UCF. An example of how these components interface with UCF for a CFD simulation is shown in Figure 3.1.4.

Boxes in the figure represent Arches components and the oval shapes represent the Data Warehouse (described in the next section) component of UCF. A request to advance simulations in time is passed to Arches Integrator, an Arches component, from the UCF. Arches Integrator reads the data provided by Data Warehouse and uses the Arches Solver component to solve the set of nonlinear equations defined at the time of the problem setup. It also creates a temporary Data Warehouse (referred to as new dw in the figure) to store data computed during the time step simulation. It can be seen that all the Arches components interact with the temporary Data Warehouse during the course of the simulation. At the end of one time step, Arches Integrator writes the data computed for the new time step back to the original Data Warehouse.

Second, the components define and encapsulate a family of algorithms. This design provides the flexibility to use and test different algorithms interchangeably in plug and play fashion. Figure 3.1.5 illustrates the SGSTurbulenceModel component for LES where different
Figure 3.1.4: An example fire simulation

Figure 3.1.5: Subgrid scale turbulence model component of Arches
turbulence models are implemented and can be used interchangeably depending on the problem we are solving.

Code reuse was necessary to minimize the code development process since most of our stand-alone code was already validated. To accomplish this task, we spent significant time modularizing the Fortran subroutines for use in our components.

Realistic fire simulations must account for relevant physical processes such as turbulent reacting flow, convective and radiative heat transfer, multi-phase interactions, and fundamental gas phase chemistry. Representations of these physical processes lead to very large, highly nonlinear, partial differential equations (PDEs). Robust nonlinear and linear solvers for these large-scale nonlinear sets of PDE on massively parallel ASCI platforms are required. To this end, we selected Portable Extensible Toolkit for Scientific Computation (PETSc) to provide general-purpose solvers. PETSc provides a suite of nonlinear and linear scalable solvers for scientific applications modeled using PDEs. [1] PETSc solvers are written to be independent of the underlying data structures that define the problem. We have exploited this feature in the creation of the interface between PETSc and Uintah. The interface provides the capability to manipulate Uintah data structures as the vectors and the sparse matrices that are used by PETSc. The interface also allows us to write custom algorithms that can be used as preconditioners to the PETSc solvers.

Through the Uintah-PETSc interface, we now have access to the full range of solver capabilities provided by PETSc. David Keyes and the applied math team in our step are working closely to test different solvers and to develop customized preconditioners to use with PETSc solvers. Wing Yee, a graduate student in our group, worked with David Keyes this summer on implementing Additive/Multiplicative Schwartz preconditioners with PETSc. Wing will continue to work with David Keyes.

The fluid flow component of the Arches is now integrated into the UCF. An explicit solver is in place that uses the Uintah-PETSc interface for solving the pressure equation in an implicit fashion. The reaction and mixing modules of Arches have already been abstracted into C++ modules, and their incorporation into the UCF is expected to be a short time objective.

**Parallelization Strategy**

The integrated fire code simulates the propagation of an ignition event over a pool of liquid hydrocarbon fuel. To simulate this event, the code uses algorithms that couple turbulent fluid dynamics (CFD), turbulent subgrid mixing and reaction chemistry for fire, and radiative transport. Due to the wide range of physical and chemical length and time scales involved in the simulations, it is necessary to have the flexibility to explore different parallel algorithms. For example, CFD uses large-eddy simulations in which large, energy-carrying length and time scales are resolved and the smaller scales are modeled. A domain decomposition strategy is used to parallelize such a system. While this strategy works well for CFD systems, chemical reactions and turbulent subgrid scale mixing models are functions of state space (chemical species) variables and are independent of the grid. Thus, the domain decomposition strategy used in physical space will not yield the best performance for parallelizing these systems. Instead, we partition our domain in state space to yield good scalability. The same holds true for radiative heat transfer modeling which uses integro-differential radiative transport equations. These
equations can be parallelized efficiently only by dividing the tasks on the basis of different rays that need to be tracked on the whole grid rather than on patches of the grid. The computation of radiative fluxes can be made even more efficient because the spatial grids for predicting radiative fluxes can be much coarser than those for CFD. Thus, the flexibility in maintaining different grids and parallelization strategies is crucial for a fire simulation environment.

UCF is designed to provide the flexibility required to parallelize such a multiphysics environment. Additionally, it hides the complexity of parallel data management from the application developer. UCF uses a component called the Data Warehouse which is tightly coupled with the Uintah environment. The Data Warehouse presents developers with the abstraction of a global, single-assignment memory with automatic data lifetime management and storage reclamation. It manages the distribution of dependent ghost data among relevant processors efficiently while hiding the details from the developer. For parallel CFD computation, UCF uses the components provided by the application developer and assigns them to different processors determined by the Scheduler, a component of UCF, based on the combination of inputs provided by the user and the task of dynamic load balancing. Figure 3.1.6 shows a typical task graph created during the process of solving the pressure equation, one of This process has been divided into two tasks. In the first task, a linear matrix is generated through discretization and the application of boundary conditions. Before the start of this task, each processor has the
data it requires for computing the matrix. In the second task the matrix is solved using PETSc (see previous section). As shown in Figure 3.1.6, the computational domain is divided into four patches, and each patch is associated with a processor. Arrows in the figure indicate data dependencies between the processors. The Data Warehouse uses this task graph to determine dependencies and data transfer.

We are currently working on parallelizing our CFD solver using the Uintah-PETSc interface. Our next step will be to parallelize the integrated fire components, including the mixing and reaction models, on ASCI machines.

### 3.1.3 Multimaterial Formulation

In the first two years, several advances were made in the individual disciplines that comprised C-SAFE. In this past year we identified the fluid-structures interface as a very important area that deserved focused attention. As a result, we identified a person to work on the interface and develop a model for fluid-structure interaction, i.e., a multimaterial formulation. What follows is a summary of the model that has been developed. A detailed account can be found in Kumar (2000).

#### Methodology

This section summarizes the methodology and logic underlying the development of the multimaterial formulation. The multimaterial formulation is designed to work for mixtures of different materials in which the relative amounts of each material can each arbitrarily vary (subject to the constraint that all of space is occupied by some material). The formulation is also designed to account for the variations in observable material response with length scale. If one could observe and resolve all scales of motion in the problem at hand, one could have a model for the smallest scale and that would describe the entire problem. However, practical limitations force one to choose a lower bound on the scales that one can resolve in a calculation. This means that one must have models for material length scales that are both at (and above) the level of the resolved scales and below the level of the resolved scales. In the C-SAFE scenario, one can have material

![Figure 3.1.7: Determination of continuous or discrete state depends on the control volume chosen](image-url)
elements that are relatively large and contiguous, such as a block of metal that comprises the wall of a missile, as well as material elements that are very small, such as fragments that break off or that are formed when an explosion takes place. When the length scale of a material element in a problem is greater than that of the local resolved scale, the material is defined here to be in the continuous state. Likewise, when the length scale of a material element is lesser than that of the local resolved scale, the material is defined here to be in the discrete state. 3.1.7 shows an example of how the definition of such states is dictated by the limits on the calculation.

One therefore needs a model for multiple materials that can treat both continuous and discrete states of each material. Furthermore, the behavior of each material, whether in the continuous or discrete state, is influenced by the presence of other materials. This happens in passive and active ways. The passive way is the dilution effect of other materials on the effective behavior of a material. The active way is the effect of the motion of one material on the motion of another.

We therefore define two field quantities that are fundamental to the discussions that follow. One is the material volume fraction for each material $k$, $\theta_k$, and the other is the degree of discreteness, $\alpha_{k,d}$. The volume fraction, $\theta_k$, is defined as the fraction of the total volume occupied by material $k$. Thus, if the mass of material $k$ per unit volume of the control volume can be defined as the bulk density, $\rho_k$, of material $k$, the volume fraction of material $k$, $\theta_k$, can be written as:

$$\theta_k = \frac{\rho_k}{\rho_k^o},$$

in which $\rho_k^o$ is the density of pure material $k$. The degree of discreteness, $\alpha_{k,d}$, is defined as the fraction of the total material in any control volume that is in the discrete state. Knowledge of these parameters, along with accurate models of the materials as a function of these two parameters, completely specifies the behavior of a material. In what follows, we illustrate this methodology on the momentum and continuity equations.

**Material properties in a mixture as a function of $\theta_k$ and $\alpha_{k,d}$**

The material properties that we are primarily concerned with when we consider the momentum equations are the stresses of the materials (and their divergences). For a pure (idealized) fluid, this material stress can be decomposed as

$$\sigma = -pI + \tau$$

Here $p$ is the thermodynamic pressure in the material and $\tau$ is the deviatoric stress.

For a continuous solid, such a decomposition does not make sense; solid stresses can, in general, be completely anisotropic, and $\sigma$ for a solid is, generally, a six-element symmetric tensor.

For a solid that is discretely distributed within a fluid, the effects of anisotropy cannot be resolved, and so the only resolvable stress that we can observe for a discrete solid is the surrounding pressure, in accordance with conditions of equilibrium. In such a case, one can define a pressure for the discrete solid that is equal to the surrounding fluid pressure. Hence, the stress tensor for a discrete solid can be written as:
With these definitions for the pure materials, we proceed to model the stresses in a multimaterial mixture. The model for the effective continuous-state stress in a solid of the material $i$ that occupies only a fraction $\theta_i$ of the total volume is

\[ \nabla \cdot \sigma_{i,c,eff} = \nabla \cdot (\theta_i \sigma_{i,c}) \]

Similarly, the model for the effective continuous-state stress in a discretely distributed solid of the material $i$ that occupies only a fraction $\theta_i$ of the total volume is

\[ \nabla \cdot \sigma_{i,c,eff} = -\theta_i \nabla p_{i,c} \]

Since only a fraction $\alpha_{i,d}$ of the material is in the discrete state, the effective continuous-state stress divergence of the solid material in a mixture is modeled as a linear combination of 4. and 5.:

\[ \nabla \cdot \sigma_{i,c,eff} = -\alpha_{i,d} \theta_i \nabla p_{i,c} + (1 - \alpha_{i,d}) \nabla \cdot (\theta_i \sigma_{i,c}) \]

Similarly, for a fluid that might be in continuous as well as discrete states (e.g., small unresolved gas bubbles within a solid), the effective continuous-state material stress can be modeled as

\[ \nabla \cdot \sigma_{i,c,eff} = -\theta_i \nabla p_{i,c} + (1 - \alpha_{i,d}) \nabla \cdot (\theta_i \tau_{i,c}) \]

The quantity $p_{i,c}$ that appears in the above equation is the thermodynamic pressure in the fluid material $i$. (For two fluids that share an interface, there is an equilibration condition between the different fluid pressures across their interface. This relationship depends on properties of the interface such as the surface tension between the two fluids. For the purposes of this document, we will not elaborate on these effects, but assume, for convenience, that we are only dealing with a single fluid and a single solid material.) The reason the factor $(1 - \alpha_{i,d})$ appears before the divergence of the deviatoric stress in is that when the fluid is present in the discrete state, it cannot act as a continuum exerting viscous forces over the resolved scale.

Materials can also experience a discrete-state stress. This stress is caused by interactions among the discrete entities of the material. Such a stress is usually negligible in dilute concentrations (low volume fractions) of a material, but if a material is predominantly in the discrete state ($\alpha_{i,d} \cup 1.0$) and is heavily loaded ($\theta_i \cdot 1.0$) then the interactions between the discrete chunks of material $i$ can be significant and can lead to an appreciable stress, denoted by $\sigma_{i,d}$. Models for $\sigma_{i,d}$ can be found in the literature for specific situations (e.g., Gidaspow, 1994).
Multimaterial Governing Equations and Inter-Material Interactions

The governing equations for multimaterial flow follow those of single-material flow very closely, and have terms that account for convection, diffusion, and source terms. The governing equations, in vector form, are:

$$\frac{\partial}{\partial t} (\rho_i^n \theta_i u_i) + \nabla \cdot (\rho_i^n \theta_i u_i)$$

$$= \nabla \cdot \sigma_{i,c,\text{eff}} + \alpha_i d \nabla \cdot \sigma_{i,d} + \sum_{l=1}^{N} K_{il}(u_i - u_l) + \sum_{l=1,i}^{N} \dot{m}_{il} u_l$$

These equations have, on the left side, terms for the transient and convective parts of transport, just like the Navier-Stokes equations (Bird et al., 1960). On the right side, we have the stress-divergence terms, for both the effective continuous-state and discrete-state stresses, followed by the body force term. This is then followed by a modeled term for the inter-material momentum transfer term; this is the active way in which one material is assumed to influence another. This formulation of the inter-material momentum transfer term has been adopted by other researchers in the literature as well (e.g., Harlow and Amsden, 1975; Kashiwa and Rauenzahn, 1994). The final term on the right hand side is the inter-material momentum transfer term due to mass transfer. Both the mass and momentum transfer terms are conserved among materials. In other words, when (8) is summed over all materials, the inter-material interaction terms vanish.

The momentum equations are accompanied by equations representing the conservation of mass, as follows:

$$\frac{\partial}{\partial t} (\rho_i) + \nabla \cdot (\rho_i u_i) = \dot{m}_i = \sum_{l=1,i}^{N} \dot{m}_{il}.$$  

Here $\dot{m}_{il}$ denotes the rate of change of mass from material $l$ to material $i$ per unit volume.

In addition, one also has to solve the equation of state for the fluid,

$$\rho_i^n = \rho_i^n(P_{i,v}).$$

It should also be noted here that the governing equations will also involve an energy equation, which has not been elaborated here for simplicity of presentation. The introduction of the energy equation does not present any major complications in the formulation presented herein; we believe that the major accomplishment shown here is the correct treatment of the momentum fields.

Solution Procedure

As a consequence of the model presented above, the equations for the fluid and solid mechanics are fairly independent. The solution of the solid mechanics equations is done using the mixed Lagrangian-Eulerian formulation (often referred to as an updated Lagrangian method) called the Material Point Method. Since this method is covered in detail elsewhere in this report, we shall not elaborate on it here. The solution of the fluid mechanical equations is effected through the use of an algorithm that derives from the
SIMPLER (Semi-Implicit Method for Pressure-Linked Equations), and is also covered in detail in Kumar (2000). The crux of the SIMPLER method is the combination of the finite-difference (or finite-volume) forms of the momentum and continuity equations of the fluid material to yield an equation for the pressure in the fluid. Thus, we focus below on the coupling between the solid and fluid materials, and the overall coupled solution algorithm.

**Fluid-Solid Coupling**

The coupling between fluid and solid materials occurs through several mechanisms: the volume fraction, $\theta$, the momentum exchange between the fluid and solid materials, the degree of discreteness, $\alpha$, and the reaction terms, $m_d$.

The material-point method (MPM) is a technique that involves marker particles that denote only solid materials. The convection of the solid is thus directly captured in a Lagrangian fashion. As a result, the solid mechanical equations do not have to account for the volume fraction in the left-hand side of (8); the volume fraction of solid is calculated as a consequence of the MPM. The material stress divergence in MPM, however, is calculated from a grid-based Eulerian calculation, and must involve a multiplication involving $\theta$, as shown in (4).

In contrast, the fluid-phase calculations are done purely in an Eulerian reference frame. As a result, the volume fractions, $\theta$, need to be included in the discretized fluid equations. This can lead to a singular coefficient matrix when the fluid calculation is implicit, and a division by zero when the fluid calculation is explicit, when the volume fraction of the fluid phase goes to zero. Thus, the fluid mechanical equations can only be solved for a volume fraction that is finitely (computationally) distinct from zero. In

![Figure 3.1.8: Dynamic Cell Typing for handling continuous solid materials in a fluid-solid situation; the figure on the left shows the real situation; the figure on the right shows the computational approximation (used when, for example, $0_{\text{solid}} \geq 0.999999$). The hatched area is the solid domain, and the grey area represents a solid intrusion.](image-url)
practice, this has been found to be a very small number (of the order of $1 \times 10^{-5}$), but this still entails special treatment when $\theta_i$ is smaller than this number. The procedure adopted here is that when the fluid volume fraction is so small, then the computational cell under consideration is treated as an inert intrusion (see Figure 3.1.8).

This necessitates the use of two different mechanisms for modeling the fluid-solid momentum exchange. In one, the presence of a distinct fluid-solid boundary is acknowledged, and in the other, fluid and solid are assumed to coexist in the same cell. When the solid is treated as an inert intrusion (i.e., there is a distinct fluid-solid boundary), the momentum flux between the two phases is calculated as a function of the relative velocities at the boundary between solid and fluid using turbulent boundary layer approximations. This flux is calculated in the fluid equations as a boundary condition and then passed on to the solid mechanics as an interface condition of the following type:

$$\sigma_{solid} \cdot t_i = \tau_{fluid} \cdot t_i,$$

where $t_i$ is a tangent plane (with two directions $i$) separating the fluid and solid phases.

When the volume fraction of fluid is not too small (as in the scenario in the left side, in which fluid and solid coexist in the same volume) then the effect of the solid is taken into account via the governing equations, through the term $\kappa_{li}(u_i - u_l)$, in which $l$ could be the solid material and $i$ the fluid material. The same term, with the signs reversed, is used in the governing equation for the MPM calculation. In addition, at an interface, the normal components of the stress in the two materials must equal each other, leading to

$$\sigma_{solid} \cdot n = p_{fluid},$$

where $n$ is the normal vector to an interface plane.

The degree of discreteness, $\alpha_{lid}$, is calculated in the MPM method. This is possible because of the cracking algorithm in MPM (see MPM section for details) which allows one to see if a material point is isolated or connected to other material points. By calculating the mass of all material points within a control volume that are isolated and dividing that mass by the total mass of material points within a control volume, the degree of discreteness, $\alpha_{lid}$, can be calculated.

Finally, the reaction terms, $\dot{n}_{lid}$, are calculated as part of the MPM calculation using reaction rate parameters and mechanisms provided by the HE group. These reaction terms are then passed on to the fluid mechanical calculations. One must be careful to distinguish these reaction terms, which are multimaterial reaction terms, from the sources due to chemical reaction that occur in the gas phase. The latter are purely homogeneous reaction terms and play no part in the mass transfer between gas and solid.

**Overall Solution Algorithm**

The overall solution algorithm is presented here for an explicit solution of the governing equations at each time step. This is for non-reacting flow, but the extensions for reacting flow do not change the essentials of the fluid-solid coupling strategy, and so are not given here. Another reason for not showing the extensions is that the energy equations have not been presented in the above equations and discussions.
1. At time $t = 0$, start with initial values of $\theta_f^0, \theta_s^0, \alpha_s^0, u_s^0, u_f^0, \rho_f^0$

2. $t^{n+1} = t^n + \Delta t$

3. Update grid cells in calculation to decide location of inert intrusions

4. Calculate the inter-material momentum exchange terms, $K_{fs}^n(u_s^n - u_f^n)$, for cells other than inert intrusions

5. Calculate momentum fluxes for cells next to inert intrusions

6. Discretize the momentum and continuity equations for the fluid phase, using values at time level $n$

7. Solve fluid pressure equation, using old values for velocities and volume fractions, to get the pressure at the new time step, $p^{n+1}$

8. Calculate the pressure force on solids, using $p^{n+1}$

9. Solve the solid phase momentum equations to get $u_s^{n+1}$

10. Solve the fluid phase momentum equations to get $u_f^{n+1}$

11. Obtain the solid material volume fraction, $\theta_s^{n+1}$, from MPM

12. Obtain the degree of discreteness of the solid phase, $\alpha_s^{n+1}$, from MPM

13. Calculate the fluid volume fraction from $\theta_f^{n+1} = 1 - \theta_s^{n+1}$

14. Solve the equation of state for the fluid, using the new pressure, to get the pure material fluid density at the new time step, $(\rho_f^0)^{n+1}$

15. Calculate the bulk density of the fluid material, $\rho_f^{n+1} = (\rho_f^0)^{n+1} \theta_f^{n+1}$


**3.1.3 Applied Mathematics**

In the past year we have focused our applied mathematics efforts on three major endeavors: interdisciplinary coupling, nonlinear and linear solvers, and adaptive mesh refinement.

**Interdisciplinary Coupling**

The coupling of the FS and CD steps required the development of a new formulation for fluid-structure problems as described in the previous section. Here we focus on the ideas of boundary matching and error reduction using implicit solvers.

The solver presented in section 3.1.3 for the solution of the fluid-structure problem was an explicit solver. While this is an acceptable strategy for a proof-of-concept study of the multimaterial formulation, this strategy has several attendant difficulties. First, since the solution is time-marched once and never corrected, there is no estimate of how accurate the update was. In other words, the residual of the transient problem is never driven to zero. There is the hope that, if small enough time steps are taken, the error in the time advance can be minimized, but one never has a quantitative understanding of the error involved. In addition, since explicit methods are sequential, changing the sequence of operations in an explicit technique can change the answers.
other words, since the operator is split, the solution can change depending on what splitting is chosen.

We believe, therefore, that implicit solvers offer the best hope of solving the coupled nonlinear problem involving both fluid and solid mechanics because they repair the damage of operator splitting by iterating on the solution. To this end, we have created a fire code that can be used in both explicit and implicit modes. We have also collaborated with the CD team to develop an implicit version of the MPM code in addition to the current explicit version. While the first attempt at a coupled solver will use the explicit solver, our eventual goal is to use implicit solvers for the entire coupled problem. The following details a tentative implicit solution strategy for the fluid-structures problem:

1. At time \( t = 0 \), start with initial values of \( \theta_f^0, \theta_s^0, \alpha_{s,d}^0, u_f^0, u_s^0, \rho_f^0 \)
2. \( t^{n+1} = t^n + \Delta t \)
3. \( \phi^{n+1,0} = \phi^n \) for all \( \phi \) (velocities, densities, etc.)
4. \( k = 0 \)
5. Update grid cells in calculation to decide location of inert intrusions
6. Calculate the inter-material momentum exchange terms, \( K_{fs}^{n+1,k} (u_s^{n+1,k} - u_f^{n+1,k}) \), for cells other than inert intrusions
7. Calculate momentum fluxes for cells next to inert intrusions
8. Discretize the momentum and continuity equations for the fluid phase, using the last update of the value at time level \( n + 1 \), viz., \( \phi^{n+1,k} \)
9. Solve fluid pressure equation, using old values for velocities and volume fractions, to get a new update for the pressure at the new time step, \( p^{n+1,k+1} \)
10. Calculate the pressure force on solids, using \( p^{n+1,k+1} \)
11. Solve the solid phase momentum equations to get \( u_s^{n+1,k+1} \)
12. Solve the fluid phase momentum equations to get \( u_f^{n+1,k+1} \)
13. Obtain the solid material volume fraction, \( \theta_s^{n+1,k+1} \), from MPM
14. Obtain the degree of discreteness of the solid phase, \( \alpha_{s,d}^{n+1,k+1} \), from MPM
15. Calculate the fluid volume fraction from \( \theta_f^{n+1,k+1} = 1 - \theta_s^{n+1,k+1} \)
16. Solve the equation of state for the fluid, using the new pressure, to get a new iterate for the pure material fluid density at the new time step, \( \rho_f^{n+1,k+1} \)
17. Calculate the bulk density of the fluid material, \( \rho_f^{n+1,k+1} = (\rho_f^0)_{n+1,k+1} \theta_f^{n+1,k+1} \)
18. Compute residuals for each variable, using \( n + 1, k + 1 \) values.
19. Compute overall residual, \( r^{n+1,k+1} \)
20. If \( \left( \frac{r^{n+1,k+1}}{r^{n+1,0}} \right) \geq \varepsilon \), where \( \varepsilon \) is a specified tolerance, \( k = k + 1 \). go to step 5.
21. \( n = n + 1 \)
22. Return to step 2.
The successful implementation of this algorithm requires that a proper implicit version of the MPM method be formulated and implemented. To this end, two implicit versions of the MPM method have been attempted in the past year. For proper coupling with the CFD algorithm, both algorithms necessitate the resetting of the background grid used in the MPM method after each nonlinear iteration within a time step. One version of the implicit MPM method uses a fixed-point wrapper outside the explicit MPM code, much like the pseudocode shown above, to obtain an implicit version. While this implicit version did not display any speedup relative to the explicit version, it did yield the same answers as the explicit method, making it a potential candidate for inclusion in the above algorithm (in step #11). The other implicit version of the MPM method uses a linearization of the problem to yield a global stiffness matrix and attempts to iterate on the displacement vector in a fixed-point fashion. Again, this method can be used with a resetting of the background grid after each iteration within a time step. More details on this strategy can be found in the section on MPM.

**Nonlinear and Linear Solvers**

The current fire code uses a fixed-point iterative solver (Picard method) for the implicit cycle. While this method has yielded validated results, we want to improve both the robustness and efficiency of the code. Consequently, we are exploring various nonlinear and linear solvers for the code. We are using PETSc, a package developed at Argonne National Laboratory, to test a wide variety of linear and nonlinear solvers. The PETSc suite has been partially integrated with the UCF; as described previously, its suite of linear solvers is currently being used to solve the pressure equation within the explicit fire solver. Wing Yee, a graduate student, will work on linear and nonlinear solver issues within the UCF using PETSc. Assisting us in this endeavor is David Keyes of CASC at LLNL, who is co-advvising Wing on his dissertation. One of the focal points of this work is a study of Newton-Krylov-Schwarz methods as candidates for nonlinear parallel solvers within the context of the problems in C-SAFE.

**Adaptive Mesh Refinement**

The C-SAFE problem spans length scales ranging from about 1 km to about $10^{-6}$ m. There is also a large range in the associated time scales. This variation in length and time scales cannot be resolved using a single grid for the entire problem. The advantageous aspect of the C-SAFE problem is that such a large variation does not exist everywhere in the domain. In particular, while high resolution is needed near the base and in the center of the fire, the edges of the fire can be coarsely resolved. High resolution is also needed to resolve the volume in the vicinity of the container. This problem can be resolved by using Adaptive Mesh Refinement (AMR), whereby we locally refine and/or coarsen the mesh adaptively as needed in the calculation.

To this end, we have been pursuing a collaboration with the CASC group at LLNL (Scott Kohn, Rich Hornung, and Andy Wissink) to prototype an AMR fire code using the same modules that have been used in the UCF. The goal of this collaboration is to leverage the vast experience that the CASC group has with regard to the mathematical algorithms needed for AMR. At present, the modular version of the Arches fire code is being integrated with SAMRAI (the meshing framework for managing a hierarchy of adaptive grids).
3.1.4 Physics and Chemistry

Large Eddy Simulation (LES)

Modeling turbulence in fire simulations is important because reaction rates of the major constituents are controlled by the mixing process. In large scale pool fires, turbulent mixing is induced by thermal instabilities, i.e., the rising of high temperature, low density combustion products makes way for an influx of cool air. This results in the production of baroclinic vorticies and the onset of turbulence.

Since it is prohibitively expensive to perform direct numerical simulation (DNS) of the turbulent fluid motion in practical applications, we must rely on a model to compute the subgrid scale (SGS) stresses and energy dissipation. The filtered Navier-Stokes equations (LES equations), including the SGS stress term that must be modeled, are shown below. Note the tilde implies a filter has been applied to a given variable at the grid level (i.e., it is some average value of that quantity within the mesh resolution). For a discussion of filtering, see the CSAFE 1999 annual report, http://www.csafe.utah.edu/documents/FY99AnnualReport.pdf.

\[
\frac{f\tilde{u}_i}{ft} + \frac{f(\tilde{u}_i\tilde{u}_j)}{fx_j} = -\frac{1}{\rho} f\tilde{p} + \nu \frac{f^2\tilde{u}_i}{fx_i} - \frac{f\tau_{ij}}{fx_j}
\]

The models currently in use can be broken into two major categories, eddy viscosity models and synthetic field models. The former treats the SGS stress tensor in the same way as the viscous stress is treated for a Newtonian fluid as seen in the following equation.

\[
\frac{f\tilde{u}_i}{ft} + \frac{f(\tilde{u}_i\tilde{u}_j)}{fx_j} = -\frac{1}{\rho} f\tilde{p} + (\nu + \nu_t) \frac{f^2\tilde{u}_i}{fx_i} - \frac{f\tau_{ij}}{fx_j}
\]

Overwhelmingly, the most common approach for determining \( \nu_t \) is the Smagorinsky (1963) model, outlined below. Here, \( C_s \) is the Smagorinsky coefficient, experimentally determined to be approximately 0.2 for isotropic turbulence, and \( \Delta \) is the grid filter width.

\[ \tau_{ij} = -2\nu_t \tilde{S}_{ij} \quad \nu_t = (C_s \Delta) |\tilde{S}| = \sqrt{2\tilde{S}_{ij}\tilde{S}_{ij}} \]

Synthetic field models treat the SGS stress based on a mathematical identity used in the derivation of the LES equations. The so-called scale similarity model is written simply as,

\[ \tau_{ij} = \tilde{C}_{sim} (\tilde{u}_i\tilde{u}_j - \tilde{u}_i\tilde{u}_j) \]

The overbar denotes filtering at a test grid level, typically 2\( \Delta \), and \( \tilde{C}_{sim} \) has generally been taken as unity.
Comparison with DNS data reveals that the Smagorinsky model does well at predicting energy dissipation but does a poor job at matching the SGS stresses. The scale similarity model, on the other hand, predicts the stresses accurately, but misses the energy dissipation rate. On this basis, the following mixed model has been proposed.

\[ \tau_{ij} = C_{sim} \left( \overline{u_i u_j} - \overline{u_i} \overline{u_j} \right) - 2(C_{smag} \Delta)^2 \overline{S}^T \overline{S}_{ij} \]

Together with the dynamic determination of the Smagorinsky coefficient discussed below, the mixed model is the current state of the art in LES models (note that \( C_{sim} \) is held constant). For the latest review article on progress in LES of turbulent flows see Meneveau et al. (2000).

One other drawback of the eddy viscosity approach is it that the constant coefficient tends to overpredict stresses in regions of high shear, such as the near wall region. To overcome this drawback, an approach was introduced by Germano et al. (1991) which dynamically computes the coefficient based on information from the resolved flow field. The dynamic approach assumes that the coefficient should be constant, at a given point in space time, over the inertial range of length scales. Again, the test filtering operation is employed, and a new SGS stress tensor is defined at the test scale, \( T_{ij} \).

\[ \tau_{ij} = -2(C_{\Delta} \Delta)^2 \overline{S}^T \overline{S}_{ij} \]

\[ L_{ij} = T_{ij} - \tau_{ij} = \overline{u_i u_j} - \overline{u_i} \overline{u_j} \]

\[ T_{ij} = -2(C_{\Delta} 2\Delta)^2 \overline{S}^T \overline{S}_{ij} \]

\( L_{ij} \) is known as the Germano identity and is computable from the resolved velocity fields. By plugging in the assumed models for \( T_{ij} \) and \( \tau_{ij} \), we can find an approximation for \( C_s \) by minimizing the least squares error (since \( L_{ij} \) is symmetric there are 6 equations and 1 unknown, \( C_s \)).

Our strategy for evaluating LES models includes physical validation, numerical verification, and computational cost analysis. In other words, we may not have the luxury of spending computational time for the current state of the art. We must first establish proof of concept and then add model complexity. Hence, we began by implementing the simplest model, the Smagorinsky model with an implicit filter (for an implicit filter, the discretization of the Navier-Stokes equations implies a filter on the velocity field). The next step is to implement the dynamic procedure on this model. The new subroutines have been written and will soon be implemented in Arches.

We will perform coarse validation of the physics against data taken from a 7.3 cm helium plume and a 10cm and 30cm methanol pool fire experiment performed at NIST (Mell et al. 1996, http://155.99.210.42/~ruddy/WORK/C-SAFE/poolfire.html). A more stringent test of the model’s capabilities will be to compare stresses and energy...
dissipation rates with DNS data. Through our collaboration with Jackie Chen of Sandia Livermore, we have access to a reacting DNS code and will use this code for subgrid validation of momentum and of scalar transport.

Since momentum and scalar transport are highly coupled in turbulent flows, it is logical to seek a class of subgrid models which embodies this fact. The method of fractal interpolation may provide such a link. Shown in Figure 3.1.9 are magnifications of a scalar field from DNS data.

For many years, turbulent structures have been identified as fractal in nature. In fact, the notions of scale similarity used in the SGS stress models are a direct attempt to mimic this behavior. Not until recently (Meneveau et al., 1999), however, have attempts been made to reconstruct a subgrid velocity field using fractal techniques. So far, this task has proven extremely complicated, but the technique shows promise for closing both the momentum and scalar transport equations. We will continue to explore this promising area of research as we add complexity to our LES model.

**Turbulent Mixing and Chemical Reaction**

In our fire simulations, the mixing and reaction models are the bridge between the molecular scales of turbulent micromixing and reaction and the macro scales, resolved on the computational mesh, of fluid motion and scalar transport. The reaction model provides scalar information such as density, temperature, and reaction rates based on the local (subgrid scale) extents of mixing and of reaction. The extents of mixing and reaction, also known as the mixing and reaction variables, are the independent variables in the system. The mixing model filters the scalar information using values obtained from the resolved scales on the grid and a realistic representation of turbulent micromixing.
The mixing model then passes the filtered state-space information to the transport model for use in computing the scalar and momentum fields.

For the third year, we have built on the mixing and reaction model framework we developed in the second year. The framework, written in C++, emphasizes both generality and flexibility. For example, the mixing model requires as input the choice of mixing model type, the values for the mixing and reaction variables and their statistics necessary for the chosen model, and the desired state-space information. It returns to the transport model the filtered state-space information. Any mixing model that fits into this general form can be plugged into the C++ framework and used for the filtering step.

This past year we have worked to improve the computational efficiency of the prescribed filtered probability density function (PDF) mixing model implemented in the second year. Such models are computationally expensive as integration is required to obtain the filtered information. One solution we have applied is to dynamically tabulate the filtered state-space variables as functions of the independent mixing and reaction variables. When the transport model requires a filtered state-space variable, it interpolates from the table using the resolved values of the independent variables. If values needed for the interpolation are not already tabulated, the necessary calculations are performed and the results are added to the tabulation. This method works well for problems with only a few independent variables. However, as the degrees of freedom increase, interpolation becomes very expensive.

Adaptive tabulation with piecewise interpolation reduces the cost and improves the accuracy of multidimensional interpolation. In this scheme, an error analysis is performed on the tabulated value(s) obtained from the piecewise interpolation. If the error is too high, local refinement is performed in the table and the error from a new piecewise interpolation is calculated. Local table refinement continues until the error criteria are met. We have developed an adaptive tabulation algorithm that uses a k-d tree structure and performs piecewise interpolation and local refinement. We are currently developing some error estimation tools.

The equilibrium model we implemented in the second year used either the NASA code or the STANJAN code to compute gas phase equilibrium. This model was coupled with a global soot model to compute soot volume fraction can be computed. Building on the C++ reaction model framework, we have added a new reaction model with finite rate chemistry in the third year.

The new reaction model is based on a kinetic mechanism proposed by Belardini et al. (1996) and has six steps: fuel decomposition to acetylene, acetylene oxidation, and the soot reactions of nucleation, surface growth, carbon oxidation, and agglomeration. We modeled this mechanism with six reaction variables where each variable describes the progress from no reaction to complete reaction of a particular reaction in the mechanism. All state-space variables are computed based on the degree of local mixing (i.e., value of the mixing variable) and the extent of each reaction (i.e., values of the reaction variables). This reaction model has been particularly challenging because the first reaction is highly endothermic and the system of equations is very stiff.

We are currently doing model verification using a plug-flow reactor model. Verification is the process of checking the chosen method of implementation against a standardized method of implementation. In the case of the Belardini reaction model, the standard reaction mechanism implementation in a plug flow reactor (PFR) is to integrate...
the rate equations as they appear in the Belardini reference. In our method of implementation, we obtain the rates by first transforming the problem from temperature and species composition space to the space defined by the independent variables (i.e., the mixing and reaction variables and the enthalpy). Given values for these independent variables, the reaction model then returns the rate equations for integration. The PFR results from both implementations should be the same given identical input and integration parameters. Figure 3.1.10 shows a comparison of PFR results obtained from integration of the reaction rates taken directly from the reference and from integration of the rates returned by the reaction model. Only one set of points can be seen on the C\textsubscript{2}H\textsubscript{2} plot because the results from the two models are very close. The soot particle number density (RHON) results are slightly different but still show good agreement. After further verification, we will perform fire simulations using this new reaction model.

We were able to test the portability and flexibility of our C++ reaction model framework thanks to work performed during the summer by Diem-Phuong Nguyen, the graduate student doing reaction model development. Diem spent the summer at Lawrence Livermore National Laboratory working with Overture, an object-oriented framework for solving systems of partial differential equations. The Overture platform allows one to run simulations in a serial or parallel environment for simple or complex geometries. Diem wrote several C++ modules using Overture classes to couple one of our reaction models to a simple transport model. Based on this experience, she has suggested some improvements to our interface format that we will implement.

The next step in the development of the reaction model is to incorporate into the UCF the modules developed previously by our group (Rawat, 1997) for computing an intrinsic low-dimensional manifold (ILDM) for a large, gas phase reaction mechanism. We will use this ILDM reaction model to reduce the degrees of freedom in the reaction mechanism for the JP-8 surrogate (described in the next section) to one or two and then couple this reduced gas phase chemistry to the four-step soot model of Leung et al. (1991).

An important element of both mixing and reaction model development is model validation. In the past year, we have made significant steps toward obtaining the
required validation data: accurate statistical data at the subgrid scale. Such data cannot be obtained by experimental methods. An alternative is to use data generated by Direct Numerical Simulations (DNS) of representative flow situations. DNS fully resolves all mixing and reaction time scales using highly accurate numerical schemes. DNS results provide spatially and temporally resolved data, which may then be averaged to validate and/or calibrate mixing and reaction models.

We are doing mixing and reaction model validation using DNS in close collaboration with Sandia National Laboratories. Currently, we are involved in improving the Sandia DNS code to more accurately incorporate transport properties of multicomponent reacting mixtures. Transport properties will be evaluated from kinetic theory to provide more accurate values for shear viscosity, bulk viscosity, thermal conductivity, thermal diffusion, and mass diffusion. This will increase the accuracy of the DNS for reacting flows by describing the coupling between mixing and reaction at a higher level of detail. We are also sharing techniques such as ILDM and ISAT to improve the efficiency of chemical reaction calculations in DNS. Increasing the accuracy and efficiency of the Sandia DNS code will allow us to explore the ranges of applicability for various mixing and reaction models.

**Soot Formation and Destruction Mechanisms**

In the past year, we have developed a Simulation Roadmap to define the sequential steps that must occur in the development of our soot model. Figure 3.1.11

![Figure 3.1.11: Soot Model Development Roadmap.](image-url)
shows the Soot Model Development Roadmap (SMDR) we have set up to supply the information for each step in the development process. The successful completion of our mission will require the integration of all the steps in our road map. The area of soot formation and destruction encompasses two steps of the SMDR: gas-phase chemistry and its coupling to the soot chemistry.

**Gas phase chemistry**

The SMDR begins with the definition of surrogate mixtures, which represents the starting point for model validation. Surrogate mixtures representing JP-8 and other fuels have been developed that reproduce the physical parameters and the chemical characteristics that control soot formation. Each of these mixtures uses fuels for which the chemical kinetics are known. Six compounds have been identified for the JP-8 fuel surrogate: iso-octane (i-C8), m-xylene, methylcyclohexane (MCH), tetralin, dodecane (C12H26) and tetradecane (C14H30). In preparation for subsequent work, we compiled the information that is available in the literature on these six compounds.

This past year we have worked with Dr. Westbrook’s group from Lawrence Livermore National Laboratory (LLNL) to develop a reaction mechanism, which could represent the JP-8 surrogate. Using data present in the literature, we set up a preliminary scheme, named C-Safe1, which consists of 180 species and 840 reactions. It is composed of an aromatic submechanism up to the formation of pyrene developed by Marinov et al. (1998) at LLNL, an n-heptane submechanism by El Bakali et al. (1999), and an iso-octane mechanism by Pitsch et al. (1996) and Westbrook et al. (1988). We included n-heptane in the scheme because C7 chemistry is part of the C12 and C14 reaction mechanisms. The LLNL submechanism leading to high aromatics was analyzed last year, and among the schemes present in the literature, it showed the best agreement with the data on the formation of pyrene, considered to be a key intermediate to soot (in particular for the HACA mechanism, see below). We also obtained information on MCH and tetralin from Prof. Glassman’s group (J. Stewart et al., 1998; J. Stewart, 1999) at Princeton University. They performed some studies on supercritical pyrolysis of MCH and tetralin, and proposed possible reaction pathways for some of the major products. Their scheme represents a starting point for further implementations.

The chemistry of C12 and C14 is being developed together with the thermochemistry and the transport data for the new compounds identified. We are working on two codes: one to calculate the thermochemical properties of the new compounds and the other to automatically generate the reaction scheme based on the molecular properties of the compounds.

The newly developed scheme, Csafe1, has been validated in some conditions. Figures 3.1.12 and 3.1.13 report the comparison between modeling results and experimental data for n-heptane and iso-octane premixed flames. The code used for the simulations is PREMIX in Chemkin III (Chemkin, release 3.5). The Csafe1 scheme is able to reproduce the concentration profiles of the fuel, oxidizer, major products and benzene fairly well. The latter is very important since benzene is considered to be the building block for the formation of higher aromatics, and its good prediction represents a key step in the model development.

Using our JP-8 surrogate, Prof. Ranzi’s group at Polytechnic of Milan in Italy has developed a scheme, named HTJP8, which includes reaction submechanisms for all the
Figure 3.1.12: Comparison between modeling results and experimental data for flame 1 (C\textsubscript{7}H\textsubscript{16}-O\textsubscript{2}-N\textsubscript{2}, \( \Phi = 1.9 \)) and flame 2 (C\textsubscript{8}H\textsubscript{18}-O\textsubscript{2}-N\textsubscript{2}, \( \Phi = 1.9 \))

compounds listed in the surrogate formulation. Starting from a detailed C1-C4 kinetic mechanism and assuming analogy rules for similar reactions, the scheme has been extended to heavier species. The resulting kinetic model of the oxidation of hydrocarbons consists of about 200 species and 3,000 reactions (Ranzi et al., 1994; 1995; 2000). The kinetic scheme allows the modeling of the pyrolysis and oxidation of alkanes up to n-tetradecane and aromatics up to coronene. The HTJP8 scheme, developed to run with the Dsmoke code, has been validated for some premixed flames. The computed results for a butadiene flame (Cole et al., 1984) are plotted in Figure 3.1.13 and compared with some of the major compounds experimentally identified. Thermochemical information and transport properties for this mechanism were primarily obtained from the Chemkin thermodynamic database. Unavailable thermodynamic data were estimated using group additivity and difference methods.

All the results are available at the web site http://panda.cs.utah.edu/~validate (see Web-based Validation section). Our efforts during the next year will be to include all the compounds identified in the surrogate in the C\textsubscript{safe1} scheme and to develop the chemistry for those compounds not present in the literature. At the same time, we will validate the
Figure 3.1.13: Comparison of species concentration profiles in a premixed butadiene flame obtained from the HTJP8 scheme and from the experimental data of Cole et al. (1984).

HTJP8 scheme in different operating conditions and compare results with the Csafe1 mechanism and with other reaction schemes.
Soot chemistry

Another step in the SMDR is the coupling of the gas phase chemistry to the soot model. Available soot models can be grouped in two classes. Prof. Frenklach’s group at the University of California at Berkeley developed the first class, the HACA mechanism. The HACA mechanism (Frenklach and Wang, 1994), which stands for (Hydrogen-Abstraction-Acetylene-Addition) involves molecular weight growth to form large soot nuclei followed by reactions of acetylene and polycyclic aromatics with the soot surface. During this year, we have worked with Prof. Frenklach to couple the Csafe1 scheme with the HACA mechanism. Results using n-heptane as the fuel have been obtained for a PSR reactor and are reported in Figure 3.1.14. Qualitatively, the scheme is able to reproduce the trends for soot formation with increasing temperature and increasing fuel/oxidant ratio.

The second class of soot models involves the formation of tars. Tars represent a large portion (75 - 90%) of the organic material that remains unresolved in analyses of the exhaust from power plants, diesel engines and other combustion systems as well as in atmospheric studies. These high molecular mass species are a carbonaceous material that is soluble in dichloromethane (and hence, not soot) but are also not resolvable by gas chromatography. This model was developed by DAnna and Violi (1998) at the University of Naples in Italy. According to this model, the formation of high molecular mass aromatics and soot are just a rearrangement of PAHs. Soot inception consists of the progressive aromatization of the initially less aromatic structures through H-atom abstraction processes. These abstractions create additional double bonds and, consequently, fused aromatic rings in the cluster with the increased extension of aromatic islands inside the structure (Violi et al.,
During this year we have worked with the University of Naples to improve their model in order to determine the kinetic feasibility of the new proposed pathway. Working with Prof. Truong, we performed *ab initio* calculations using the Density Functional Theory (DFT). We have used these calculations to analyze formation pathways for high molecular mass compound growth and have shown why reactions between aromatic moieties are needed to explain recent experimental findings. Figure 3.1.15 shows a sequence of chemical reactions between aromatic compounds (e.g., phenyl) and compounds containing conjugated double bonds (e.g.,acenaphthylene) and structures 1 and 2 represent some of the possible products (Violi, Truong and Sarofim, 2000). In particular, the compound reported as structure 2 has an energy that is 105 Kcal/mol lower in energy than that of the reactants at the B3LYP level, indicating that this is a possible path at combustion conditions (Violi et al., 2000).

The reactions we have studied represent the first propagation step of a sequence that can be easily repeated for the formation of high molecular mass compounds. The distinguishing features of the proposed model lie in the chemical specificity of the routes proposed.

We will focus our efforts next year on improving the *ab initio* calculations by looking for different pathways and by treating the tar formation and surface growth.
mechanisms as parallel pathways to soot. We will try to determine where one or the other
pathway dominates. As a critical test of the pathway to soot, the structures of young soot
will be determined by Nuclear Magnetic Resonance in Prof. Pugmire’s laboratory (see
Validation section of this report).

Computational Chemistry

Truong and co-workers have been developing computational tools for predicting
kinetics of gas phase reactions, including reactions in the JP-8 surrogate mechanism C-
Safe1, from first principles. The challenge of this task is that the combustion of
hydrocarbon fuels often involves a large number of chemical reactions where
experimental or accurate theoretical rate constants are not available. Calculating kinetics
for every reaction from first principles would require significant computational resources
and time and thus is not a suitable approach. We have recently proposed a novel
Reaction Class Transition State Theory (RC-TST) for estimating relative rate constants
for reactions in a given class. This approach for modeling gas phase reaction rates is
based on conventional TST theory and a fundamental concept that all reactions in the
same class have the same reactive moiety and thus have similar potential energy
information along a particular reaction coordinate. The RC-TST theory assumes the
potential energy information and rate constants of the principal reaction (the smallest
reaction in the class) are available. Such conditions can easily be satisfied since the
principal reaction is often small and its potential energy surface can be accurately
calculated at a high level of theory. Relative rate constants for other reactions in the class
can be estimated knowing only the differential reaction energy and barrier height. We
have shown that the differential reaction energy and barrier height can be calculated
efficiently at a low level of theory. Also, no frequency calculation is required for
obtaining the relative rate constants. Thus, the new RC-TST theory demands much fewer
computational resources in comparison to the simple TST theory.

To illustrate the new theory, RC-TST results for the H+ C₃H₈ hydrogen
abstraction reactions are plotted in Figure 3.1.16. Rate constants for abstraction of the

\[ H + C₃H₈ \rightarrow H₂ + C₃H₇ \]

Figure 3.1.16: RC-TST results for the H+ C₃H₈ hydrogen abstraction reactions; H + CH₄
reaction was used as the reference reaction.

primary hydrogen and the secondary hydrogen and the overall reaction are plotted along
with available experimental data. The principal H + CH₄ reaction was used as the reference reaction. Excellent agreement with experimental data was obtained. In fact, we have also successfully applied the new theory to many reactions in the H + H—R class and to other classes of reaction.

Truong and co-workers have also developed a Chemical Information Management System (CIMS) to serve as a main link between fundamental chemistry and computational engineering. CIMS allows for storage, query, mining, and analysis of chemical information that is either from separate calculations within the UNITAH PSE or from the literature. It uses an SQL 95 standard database storage format with a Java graphical user interface, seen in Figure 3.1.17, which works on all operating systems relevant to the UNITAH PSE. CIMS has two components, namely the kinetics and the physical properties. The kinetics component of CIMS is now functional. Currently, CIMS contains kinetic information of more than 1500 gas phase reactions for combustion of hydrocarbons and energetic materials. Literature information will continue to be added to the database. Concurrently, we are improving CIMS and adding new functionality as well as working on the component for physical properties.

Figure 3.1.17: Graphical user interface for Chemical Information Management System.
3.1.6 References


Chemkin Collection III release 3.5, license n. 1283.


3.2 Container Dynamics

The Container Dynamics (CD) research team is responsible for developing computational tools for all solid phase dynamics in the various C-SAFE simulation scenarios. In addition to strong research efforts directed at predicting material response, CD efforts integrate with all research groups in the center. These activities involve integration of HE burn models, coupling between fluid and solid phase mechanics, integration within the Uintah Computation Framework, or UCF (e.g., parallelization, visualization, performance analysis), and development of models to incorporate information obtained from fundamental molecular dynamics (MD) simulation into continuum level simulation.

Efforts in the Container Dynamics group over the past year have been directed at the following:

1. Integration of our basic structural dynamics code (Material Point Method, or MPM) into the Uintah Computational Framework (UCF), including optimization and parallelization efforts.
2. Development and validation of an implicit MPM code
3. Contact algorithm improvements (mechanical and thermal)
4. Algorithms for explicit fracture including thermal effects
5. Constitutive model development for HE materials
6. Micromechanics development for HE materials
7. Material property determination from MD simulations, including new algorithm development.

Although focusing on different discipline areas (computer science, applied math, physical modeling) items 1-4 all are closely tied to the algorithmic development of the general C-SAFE MPM code within the UCF. Our physical modeling of contact and fracture is unique to the material point method with some important advantages (for our applications) over finite element and meshless techniques. While we draw heavily on analogies between finite element techniques and MPM in the development of the implicit algorithm for MPM, this is a new extension of MPM techniques.

Items 5-7 are integrated efforts to provide accurate descriptions of material properties and material response at the continuum level. Our work in MD simulations involves standard approaches for determining material properties, development of new algorithms to more efficiently extract material properties, and strong efforts on developing parallel MD code. Our micromechanics effort is focused on techniques to extract homogenized material properties for an HMX-binder composite while exploring approaches to account for damage and stress bridging in the micromechanical models. Inputs are material properties for individual components obtained from MD simulations. These efforts show up in large-scale simulations in the form of inputs for our constitutive model.
3.2.1 MPM within the UCF

The most significant strategic decision for C-SAFE in year 3 was to change our computational framework for parallelization and integrating code from SAMRAI to our own in-house developed computational framework (the Uintah Computational Framework, or UCF). Motivations for this change included allowing complete local control over framework development, the ability to optimize development for C-SAFE applications, the ability to more efficiently use local parallelization expertise, and to provide a more efficient framework for integrating various C-SAFE components in the overall problem solving environment (PSE). One of the most significant near term benefits of this strategy has been to bring together C-SAFE researchers from all disciplines to work more closely on all aspects of integration. In particular, all disciplines have been intimately involved in the path of the UCF development.

This choice has not been without its challenges. Most significant has been a short-term slow down in the ability to perform very large-scale scientific simulations as a result of efforts directed at the new computational infrastructure. The investment, however, shows significant long-term potential, both for massive parallelization and integration of multi-physics phenomena.

General Status of MPM Multi-Physics Capabilities

MPM, as originally described by Sulsky et al. (1994,1995) represents an evolution of Particle-in-Cell (PIC) methods from the original use of Lagrangian mass points on an Eulerian mesh to reduce mass diffusion (Harlow, 1964). The technique was extended a full particle approach where momentum and energy were attributed to mass particles (Brackbill and Ruppel, 1986). Finally, the Material Point Method was coined to describe the full particle approach developed by Sulsky et al. to include properties of solids, in particular particle stress and history effects. During the past two years in C-SAFE, we have been extending the capabilities of MPM to simulate contact, fracture, and burn processes. In addition we have developed algorithms for static and implicit dynamic formulations of the MPM which extend the class of problems conveniently treated by MPM. To carry out the multi-physics simulations of interest to C-SAFE, the basic description of a particle in MPM has necessarily been extended to include features beyond the general descriptive and mechanical properties. Currently implemented particle properties in the C-SAFE MPM code are listed in the table below.

<table>
<thead>
<tr>
<th>general</th>
<th>$X$, position; $m$, mass; $V$, volume; $\varphi$, material type</th>
</tr>
</thead>
<tbody>
<tr>
<td>mechanical</td>
<td>$\sigma$, stress; $v$, velocity; $F$, external force</td>
</tr>
<tr>
<td>thermal</td>
<td>$\theta$, temperature; $\tau$, temperature gradient; $q$, heating rate</td>
</tr>
<tr>
<td>fracture</td>
<td>$n$, crack facet surface normal; $a$, crack facet size; $G$, energy release rate</td>
</tr>
<tr>
<td>burn</td>
<td>$\dot{m}$, mass burn rate</td>
</tr>
</tbody>
</table>

Table 3.2.1: Currently implemented particle properties for simulating multi-physics phenomena in MPM.
Demonstration Simulation

To demonstrate current code capabilities within the UCF, we have performed a series of simulations investigating the response of granular HE materials to compaction. Simulation of the compaction of granular material provides a test of the current capabilities of the MPM code, makes connections with a number of efforts within the center, and provides the first real opportunity to perform scientific investigations using large-scale computations within the UCF. The behavior of granular material has received a fair amount of attention within the scientific community recently. Because of the rich behavior granular material has been found to exhibit, and the ability to collect data both on the scale of the individual grains and en masse, there is a rich database available for validation. Numerical simulation of granular material response requires accurate modeling of many contacting grains. The simulation of the dynamic compaction of dense granular material further requires accurate modeling of grain deformation and fracture. These capabilities are precisely the strengths of the current state of the C-SAFE solid mechanics code.

It has been found that a prevalent load carrying mechanism in granular material is provided by a small subset of the grains forming highly loaded connected paths of contacting grains, or force chains. Force chains result in a very heterogeneous stress state at the microscale. Dry granular material provides a relatively simple starting point, data for validation, and has application to energetic materials. However, the objective of the granular compaction work is the inclusion of an interstitial material and the simulation of initiation mechanisms in PBXs. PBXs are composed of >90% by volume energetic grains in a relatively weak matrix and there is evidence that force chains occur in PBXs as well. Under weak shock stimulus, a regime characteristic of accidents involving unexpected initiation of high explosives due to impact, there is evidence that the energetic grains fracture in stress chains. It is generally agreed upon that non-shock initiation of PBXs is due to energy localization at the microscale and the development of hot spots. While a number of energy localization mechanisms have been proposed, their relative importance remains to be convincingly determined. The inhomogeneous stress state resulting from the development of force chains may play a role in energy localization by promoting frictional sliding, plastic deformation and/or fracture, all of which will be included in the simulations.

A tool to generate dense packings of spheres with given size distribution using Monte-Carlo techniques was developed to create approximations to the microstructure of PBX 9501 (Dmitry Bedrov, Oleksiy Byutner, Oleksiy Byutner,
Grant Smith). Using a measured distribution of grain sizes for PBX 9501, an 80% packing fraction of 1000 spheres in a 1-mm cube was generated for preliminary calculations (Figure 3.2.1). Much bigger collections of grains will be used in the investigation to provide a better representation of the measured grain size distribution, and to determine the sample size required for statistics representative of an essentially infinite number grains (i.e., statistical information representative of the continuum scale). The material properties for the HMX grains, generated by the molecular dynamics effort, are used in these calculations. The stress wave structure from a preliminary calculation with 1M cells and 6M material points is shown in Figure 3.2.2. Only stressed particles are shown, displaying the non-uniform structure of the stress wave at a point about half way through the packing. A closer look at an interior slice (with the stress rescaled to emphasize stress paths) indicates the development of force chains amongst the smaller particles between large grains, Figure 3.2.3. It remains to be seen if this structure is maintained when the sample size is (dramatically) increased.

**Parallel Performance**

The granular compaction simulation has been run on up to 256 processors at LANL. Preliminary performance is illustrated in Figure 3.2.4. Excellent scaling is observed for up to 128 processors. Jumping across boxes to 256 processors results in severe loss of performance. We have identified several factors that contribute to this as described below.
Initial problems which prevent scaling across boxes

On Blue Mountain, for reasonable size problems (up to 11 Million particles), we have achieved good scaling using up to 128 processors (single box) with complicated multi-material constitutive models. However, we are having a difficult time getting large multi-box runs to complete without triggering an internal MPI error related to heavy message passing across the HIPPI channels. We have been informed by the LANL technical consultants that they are well aware of the problem and are working with SGI to resolve this issue. Although we have achieved some numbers for larger runs (256 processors as shown in Figure 3.2.4), it is difficult to assess how our current implementation will perform in general for multi-box runs. We have, however, identified known inefficiencies with our current implementation and corrective actions that are outlined below.

- Reduction of the amount of data passed between processors. Initial results are based on a simplified model of data communication between patches. In this model, neighboring patches communicated all of the particle data instead of just boundary and ghost cell data. Even with the extra overhead associated with passing non-ghost cell data, we achieved outstanding scaling for up to 128 processors. We suspect that for our simplified data communication algorithm, the extra overhead would incur a significant penalty when communicating between 128 processor boxes. We are working to implement passing only the necessary particle data.

- Implementation of a combined threads/MPI communication protocol. Our initial scaling data is based on a MPI communication protocol (Figure 3.2.4). Although scaling results are very good, this may not be the most efficient communication mechanism. To achieve maximum performance within a box, the ideal is to implement a combined threads/MPI style of communication. Initial results of the combined threads/MPI communication are promising. For up to 16 threads we have seen good. For each 128 processor box, we intend to implement 8 MPI processes with each process having 16 threads.

- Improving the load balancing for non-uniform problems. The results presented are based on simulations with nearly uniform geometric and computational loads. For the multi-
physics simulations, non-uniformity will be the norm, and load balancing must take this into account. As the MPM code is coupled to the CFD code, and the non-uniformity computational loads are apparent, we will work to design a load balancing scheme that attempts to distribute the loads based on both the computational and communication requirements. The component nature of the UCF environment allows for the different communication and load balancing "modules" to be plugged into the overall simulation system without having to change each of the core simulation routines. This greatly simplifies the task of implementing an alternative communication scheduler (MPI vs MPI/threads) and load balancer (round robin vs. non-uniform). This task will not begin until the CFD-MPM coupling is in place and initial data are available to guide the implementation of the non-uniform load balancing module.

Since the UCF is still early in its development we are optimistic regarding scaling over large numbers of processors and will be directing significant resources to this problem. It should also noted that we have increased scalar performance of the MPM code by an order of magnitude in the UCF and our scaling for up to 128 processors is significantly improved over our implementation in place last year.

3.2.2 Development and Validation of an Implicit MPM Code

All MPM simulations demonstrated over the three years of this project have been explicit dynamic simulations. From the early decision to use the material point method for our structural dynamics simulations is was recognized that for many aspects of our simulation scenarios it was essential to develop an implicit and static formulation. There have been efforts directed in this area at other institutions (UNM, LANL) but no published reports have appeared.

Implicit MPM Algorithm

During the past year the development of an implicit MPM formulation has been a high priority in 3.2. and has resulted in successful algorithm development and validation. The approach we have adopted borrows heavily from strategies traditionally used in implicit finite element method (FEM) calculations.

The approach begins with the linearized equations of motion in matrix form (e.g., Bathe, 1996)

\[
K^k \cdot \Delta u^k + M^k \cdot \Delta a^k = F^{ext}_g(t + dt) - F^{int}_g(t + dt) - \Delta M \cdot \Delta a^k - M \cdot \Delta a^k - \Delta M \cdot \Delta a^k - \Delta M \cdot \Delta a^k - M \cdot \Delta a^k
\]

Where \( K^k \) is the stiffness matrix, \( \Delta u^k \) is the iterative increment in the displacement vector, \( F^{ext}_g \) is the external force vector, \( F^{int}_g \) is the internal force vector resulting from the divergence of the material stress, \( M^k \) is the mass matrix, and \( \Delta a^k \) is the acceleration vector. \( k \) is the iteration number, \( t \) the current time, and \( dt \) the increment in time.
Using Newton’s method, the displacement vector $u_g(t+dt)$ is approximated by iterative improvement, based on the estimate for $u_g$ from the previous iteration, $k-1$, and an approximation for $du_g$ obtained by inversion of Eq 3.2.1.

$$u_g^k(t + dt) = u_g^{k-1}(t + dt) + du_g^k$$  \hspace{1cm} (CD.2)

Note that $u_g(t+dt)$ is not the total displacement, but is the displacement from $t$ to $t+dt$. The effects of the total displacement of the material are contained in the positions and total deformation gradient of the particles.

Iteration is repeated until convergence is achieved, as determined from the following criteria:

$$\| du_g^k \| < \varepsilon_d \quad \text{and} \quad \| du_g^k Q_g^k \| < \varepsilon_c$$ \hspace{1cm} (CD.3)

where $Q_g$ is the right hand side of Eq. (3.2.1).

Once the nodal displacements, $v_g^k(t+dt)$ are determined, the trapezoidal rule is used to find the nodal velocities, $v_g(t+dt)$ and accelerations, $a_g(t+dt)$ by:

$$u_g(t + dt) = \frac{dt}{2} (v_g(t) + v_g(t + dt))$$ \hspace{1cm} (CD.4)

$$v_g(t + dt) = v_g(t) + \frac{dt}{2} (a_g(t) + a_g(t + dt))$$ \hspace{1cm} (CD.5)

Eq. (3.2.4) can be solved for $v_g(t+dt)$, and when (3.2.4) and (3.2.5) are combined with (3.2.2), the acceleration for the current iteration $k$ at time $t+dt$ can be approximated in terms of known quantities at time $t$ and estimates at time $t+dt$ from the previous iteration $k-1$:

$$a_g^k(t + dt) = \frac{4}{dt^2} \left( u_g^{k-1}(t + dt) + du_g^k \right) - \frac{4}{dt} v_g(t) - a_g(t)$$ \hspace{1cm} (CD.6)

This value for $a_g^k(t+dt)$ is used in Eq. (3.2.1). Details of the implementation of the computational algorithm can be found in Guilkey and Weiss (2000).

**Implicit Code Demonstration**

An important scenario of interest to our research group is the response of a steel container filled with an energetic material (explosive) subject to a pool fire. The heating of the container leads to phase change of the contents from solid to gas, resulting in a pressurization of the container. While an algorithm to include all of the relevant physics is an eventual goal, a much simplified version used for validation of the implicit code is demonstrated here. Figure 3.2.5 shows a one-quarter symmetry 2-D slice of a cylindrical...
container, represented by material points on the underlying grid. (While the algorithm has been implemented in a fully 3-D code, 2-D example was chosen for ease in showing results.) The cylinder has a density \( \rho = 7.86 \times 10^{-3} \text{ kg/cm}^3 \), bulk modulus \( k = 1.66 \times 10^7 \text{ N/cm}^2 \) and shear modulus \( G = 7.70 \times 10^6 \text{ N/cm}^2 \). Arrows represent loads applied normal to the material points on the inner surface of the container. The integral effect of these loads simulates pressurization of the container. The applied forces are varied to achieve the desired pressure.

For a thin walled cylinder, the resultant hoop stress is given by \( \sigma = pr/t \) where \( r \) is the radius of the container and \( t \) is its thickness. Figure 3.2.6 shows a plot of the theoretical hoop stress solution, as well as the solution obtained by the MPM code. For these results, the MPM code was run in static mode so all inertial contributions to the solution were neglected.

The same problem was solved dynamically with both the implicit and explicit MPM codes. For these, a load curve was used to increase the pressure as a function of time, such that \( p = 61.8 \times 10^6 \text{ N/cm}^2 \). Timestep size for the explicit case was \( 1 \times 10^{-5} \) seconds, and for the implicit case was \( 4 \times 10^{-3} \) seconds. Convergence properties of the algorithm are shown in Figure 3.2.7. Figures 3.2.8 and 3.2.9 show the Von Mises stress distribution for the implicit and explicit solutions, respectively. The particle displacement and stress distribution is similar for the two solutions, but a nearly imperceptible difference in the particle locations has led to the visible difference in the stress distribution. Namely, because of the material motion, certain cells will end up with one particle per cell in one or more of the coordinate directions. When this happens, those particles are subject to a larger stress to maintain the force balance at the surrounding vertices. In these cases,
the slight difference in particle displacements resulted in a different row of cells having one particle per cell in one of the coordinate directions, resulting in the moderately different stress distribution.

This example highlights one of the other advantages of the implicit MPM formulation, besides the capability for larger timesteps. Namely, while explicit MPM does well with highly dynamic problems, it frequently has difficulty solving quasistatic problems with significant strain. In this example, approximately 4% strain was reached in .004 seconds, and the explicit code was able to produce a solution. However, in attempting to reach the same strain in .04 seconds, the explicit algorithm could not achieve a solution. The implicit code was able to reach a converged solution for any of the pressurization rates that were attempted.

3.2.3 Material and Thermal Contact

During the performance of engineering systems, the majority of the loading applied to components is by contact with adjacent components. Component contact loading can often be idealized in simulations of systems operating under design conditions, where these interactions are tightly controlled (e.g., by using bearings, lubrication or joints). Outside of normal operating conditions this is not generally the case, although accurate simulation of the system’s response may be equally important. Under severe loading, large deformation or failure of one component may result in unanticipated contact with other components. The classic example is a car crash. One aspect of the C-SAFE simulation scenario is the destruction of a container filled with explosives. Algorithms designed to handle contact between container and explosive during initial heating, chemical decomposition, and fragmentation are needed.
Inherent in the MPM algorithm is a no slip, or perfect bonding, contact condition between adjacent materials. More importantly, the MPM algorithm provides a convenient framework for applying more general contact conditions. This framework avoids the use of an interface stiffness parameter (as in a penalty formulation), which can be difficult to select for contact between materials with very different stiffness (as for the case of a steel container filled with PBX). A recently developed Coulomb friction contact algorithm has been implemented. The algorithm is computationally efficient, the cost is linear in the number of contacting materials. A separate contact detection step is unnecessary, and a solution is achieved with one sweep through the computational mesh. The algorithm reduces to the no slip condition inherent in the MPM algorithm when interfaces stick.

The frictional contact algorithm as originally implemented possesses qualities making calculations involving large numbers of contacting materials tractable. However, extensive testing indicated occasional problems. The formulation was reviewed and found to violate the explicit stability condition on rare occasions when material point registration on the overlying computational grid met specific conditions. An addition to the algorithm was made to check for violation of the stability criterion and rescale the contact impulse when necessary. The modified algorithm retains the qualities of the original, plus greatly increased robustness.

One of the test problems investigated was that of a sphere, initially at rest, on an inclined plane, under gravity. For a rigid plane and sphere an analytic solution exists corresponding to rolling without slipping. Energies developed during rolling are plotted for the rigid sphere and two deformable sphere simulations in Figure 3.2.10. For the rigid case the total energy is equal to the kinetic energy, and is plotted in black. For the deformable sphere cases the total energy is the sum of the kinetic and strain energies. The first case is shown in blue and corresponds to both sphere and plane having stiffnesses approximately that of natural rubber. The strain energy reflects the regular occurrence of mild collisions, the contact algorithm results in the sphere skipping slightly. The majority of the energy is kinetic for the rubber case, and the total energy is very similar to that for the rigid sphere (The deformable case has larger total energy in part because it is not initially in contact and free falls for a fraction of a second before contacting the plane. There is also some error in the calculation of the strain energy). Although natural rubber is fairly soft, deformability plays a small role and accurate resolution of the geometry provides results similar to the rigid case. To demonstrate the algorithm’s ability to easily handle large deformations and corresponding variation in the contact area, the sphere’s elastic properties were reduced by a factor of 1000, resulting in a material approximating jello. The jello case is depicted in red, and inspection of
the energies reveals the trade off between kinetic and strain energy as the sphere slumps down the plane. Snapshots displaying the initial configuration and deformed configurations at approximately .8 second intervals (indicated with dotted vertical lines in Figure 3.2.10) are shown in Figure 3.2.11. For this case there appears to be some sliding during rolling, and corresponding dissipation of energy.

Algorithms have also been developed for thermal contact between different materials. In MPM based simulations, interactions among particles occurs through background mesh. For the mechanical contact problems as described above, the grid based multi-velocity field is solved and used. For thermal contact problems, the same ideas are applied to solve the grid based multi-temperature field. The same advantages in mechanical contact are realized in thermal contact, namely no interface tracking is required (an advantage over FEM), and no neighboring particles search is necessary (advantage over mesh-free methods, which have gained substantial attention in the computational mechanics community recently).

### 3.2.4 Explicit Fracture in MPM

The goal of our efforts in simulating dynamic fracture is to follow the evolution of a solid body from an initial loading through to fragmentation. This is illustrated schematically in Figure 3.2.12.
Two separate algorithms have been developed for implementing fracture in MPM. In
the year 2 annual report we reported and demonstrated a new triangle boundary description to
track crack growth. However, for fracture and explosions with large number of cracks, the
complex crack interactions are expensive to calculate with the triangle surface boundary
greenometry description. A least squares approximation for the boundary point velocity
interpolation was used and was also computational expensive.

The current UCF parallel implementation is strong in providing particle and grid
structure, but does not support boundary triangle surface parallelization. As a result, we are
using particle-node visibility information to modify the normal shape functions used for field
interpolation. A particle in a cell will not interpolate information to a grid node if a crack
surface blocks the connection.

To accomplish this, the regular shape function between particle \( p \) and node \( n \), \( N^{(n,p)} \), is
modified into a broken cell shape function, \( \tilde{N}^{(n,p)} \), as

\[
\tilde{N}^{(n,p)} = N^{(n,p)} + \sum_m N^{(m,p)} \left[ 1 - V_{\text{vis}}^{(m,p)} \right]
\]

where the visibility function between node \( n \) and particle \( p \), \( V_{\text{vis}}^{(n,p)} \), is defined as

\[
V_{\text{vis}}^{(n,p)} = \begin{cases} 
1 & \text{particle } p \text{ can see node } n \\
0 & \text{particle } p \text{ is blocked from node } n \text{ by crack surface}
\end{cases}
\]

and the total number of the visible nodes to
particle \( p \) is \( N_{\text{vis}}^{(p)} = \sum_n V_{\text{vis}}^{(n,p)} \). This modified
broken cell shape function satisfies the basic
shape function requirements. This is illustrated
schematically in Figure 3.2.13.

Current code capabilities include crack
initiation and propagation along arbitrary paths
through the material points. Thermal effects
around the crack tip and crack propagation
induced heating and thermal stresses are also
included.

**Examples**

We have previously demonstrated the
ability of the MPM with fracture to accurately compute the energy release rate. Our newly
implemented fracture routines are being assessed and validated through cracking in three-
point bending. The three-point bending problem was chosen for validation as there is theory
that predicts when the loading will reach the point to cause fracture in the material. Our
simulations are consistent with the theoretical predictions on this point. A compressible
Mooney-Rivlin hyperelastic constitutive model is used here for the three-point bending material. By analyzing the particle stress tensors computed via MPM, a determination is made where cracks should initiate and in what direction they should propagate. The crack initiation and propagation are simulated by creating a new microcrack surface facet associated with a broken particle that will block the particle-node visibility, thus modifying the shape function and create a numerical microcrack. In addition, in analyzing the crack tip energy release rate, the simulation computes how much heat is transferred from mechanical strain energy around the crack tip zone. Future simulations will include the dynamic temperature field and its contribution to thermal stresses.

The low-resolution simulation shown in Figure 3.2.14 illustrates the basic breaking fracture process. The high-resolution simulation shown in Figure 3.2.15 provides information on crack nucleation and stress concentration at the crack tip. The banding seen in Figure 3.2.15 is caused by the visualization tool, where particles are drawn as filled circles. Void spaces between particles are the source of banding so that the banding is an indication of the particle deformation field. Both simulations were run in 3-D and one slice of the domain is chosen for visualization.

Efforts to integrate fracture processes in the granular compaction simulations is currently underway. As described in section 3.2.1, MPM simulations of granular compaction
can be used to make connections among various center activities (micromechanics, fracture, material response). Effects of microstructure parameters in the compaction simulations such as grain size, intergrain void size, intergrain crystal defects, binder quality, and viscoplastic binder properties all can have direct effects of crack growth. Consolidation of explosive materials to high densities can lead to microcracking. Within the C-SAFE CD group we are currently working to simulate microcracking in granular compaction to study the evolution of microstructural changes in pressed HMX explosives.

### 3.2.5 Constitutive Model Development for HE Materials

The mechanical, thermal and chemical response of plastic bonded explosives (PBXs) is well recognized to be complex and strongly dependent on details of the microstructure. While this has been demonstrated convincingly both experimentally and via numerical simulation, progress toward appropriate continuum constitutive models is only beginning to be made. This is in part due to the composite nature of these materials and the complexities of the polymeric constituents (brittle crystals in a soft semi-crystalline matrix) and their interactions. It is also attributable to general difficulties in modeling polymeric continua, where for the simplest case (amorphous polymers) many viscoelastic constitutive models have been proposed, but no general consensus on an appropriate formulation has emerged.

A number of attempts have been made to model the macroscale response of PBXs. Purely viscoelastic formulations have been used with some success, capturing aspects of rate dependence and cyclic loading hysteresis. Viscoplastic models have been used to capture aspects of permanent deformation, for example that due to creation and elimination of porosity. Maximum load and failure behavior have been modeled with the addition of damage models. A somewhat different tact has been taken with the Statistical Crack Mechanics (SCRAM) approach, where the primary energy localization mechanism is assumed to be frictional sliding in shear cracks, and explicit inclusion of deformation due to fracture provides a complex macroscale response.

Given the attention the development of macroscopic constitutive models of PBXs is currently receiving, the decision was made to select an existing constitutive model and collaborate in its development by exercising the model in scenarios for which the UCF is tailored, particularly cookoff. Optimal selection of an appropriate constitutive model demands attention to a number of considerations. An avenue for connection to micromechanics work is clearly important, as microscale and mesoscale material properties (calculations) are being used to determine macroscale model parameters. Equally important is the incorporation of sufficient complexity in the macroscale modeling to capture gross features of material response important in the simulation scenario, such as maximum load behavior and the accumulation of damage, but not yet available from micromechanics work. Finally, recognizing that PBX constitutive models are relatively early in their development, the robustness of the modeling effort, in the sense of the generality of the formulation and calibration, and the suitability for PBX 9501 (used in the C-SAFE validation effort), both merit attention.

Needless to say, if all the above requirements were met constitutive modeling of PBXs would no longer be a research area. A compromise is found in the ViscoSCRAM model.
ViscoSCRAM is a three-dimensional, finite deformation thermomechanical constitutive formulation which incorporates viscoelastic and fracture deformation mechanisms via simple versions of viscoelasticity and SCRAM. The model has a relatively small number of material parameters, including initial “elastic” moduli, the very first parameter expected to be produced from the micromechanics effort. Other parameters, such as relaxation times, and average crack growth rate, are candidates for more sophisticated coupling between micro and macro modeling efforts. Further, ViscoSCRAM’s tracking of an average crack size provides an avenue for seeding imperfections and initiating macroscopic fracture. ViscoSCRAM has been calibrated for PBX 9501 under uniaxial stress loading over quasistatic and dynamic strain rates. It predicts maximum loads and damage accumulation for these experiments and the effects of model parameters on the predicted peak loads have been established. ViscoSCRAM has been used to predict response for 3-D impact and slow cookoff experiments with good success. We have implemented the algorithm in the UCF MPM code and are in the process of testing and validating the predicted material response.

CD.6 Micromechanics

The effective mechanical properties of a particulate composite with a high volume fraction of reinforcement particles are highly microstructure dependent. Analytical solutions for the effective properties commonly assume a uniform distribution of dispersed particles in a binder to produce a simplified boundary value problem for the mechanical properties of the composite. Energy-based methods, using the calculus of variations, use the same assumptions and provide bounds on the mechanical properties. These methods produce useful benchmarks for the testing of numerical methods that take into account details of the microstructure. As the volume fraction of the reinforcing particulate increases or as the stiffness ratio of the phases increases, however, these bounds on mechanical properties vary greatly and are of limited value. The finite element method provides a robust solution to problems with complex microstructures. But as the microstructure gains complexity the geometry becomes increasingly difficult to mesh, especially for three-dimensional analyses. Further, at least six solutions, each with a different set of prescribed boundary conditions and loadings, are required to obtain the homogenized elastic properties.

Our micromechanics modeling effort in C-SAFE has focused on the Generalized Method of Cells (GMC) approach. Two-dimensional and three-dimensional GMC codes have been written to predict the effective, homogenized properties of HE composites based on known properties of the constituents. Below we focus on progress made in micromechanics modeling during Year 3. First, the two-step homogenization approach we have developed for use in GMC modeling of HE composites is presented. Research to identify the proper number and sizes of representative volume elements (RVEs) for analysis is summarized. A stress bridging model, developed and incorporated into GMC during Year 3 is discussed next. Finally, a methodology developed to model microscale damage within GMC is presented.

For C-SAFE simulations, the particulate composite of interest is a mixture of HE crystal particulate dispersed in a rubbery binder. The validation of our approach will be based on experimental studies on PBX-9501 carried out at the National Laboratories. PBX-9501 is composed of a mixture of HMX crystals and Estane+BDNPA binder. The volume fraction of the crystals is around 92% of the pressed mixture. The crystals vary in size from 5 microns to
500 microns and may be considered to be in a bimodal distribution with the sizes peaking at 10 microns and 170 microns. Figure 3.2.12 (from Skidmore et al., 1998) shows the typical microstructure of these composites, illustrating the randomness in the particle spatial distributions.

As a brief review, the Generalized Method of Cells (GMC) is a micromechanics modeling approach that allows for explicit modeling of the microstructure while permitting a computationally efficient solution. Using GMC, a RVE is discretized into a regular array of subcells. Continuity and equilibrium of subcells are satisfied in an average sense (Bednarcyk and Pindera, 1997). Instead of modeling the geometry of each particle explicitly as with finite elements, the RVE is divided into rectangular parallelepiped shaped cells.

**Two-Step Homogenization Process**

To model HE composites for C-SAFE simulations, a two-step homogenization process has been developed utilizing GMC. First, a RVE of a prescribed size is assumed to exist within the random microstructure. The known size distribution of the particles in the composite is used to generate a random distribution of particles in the RVE. This size distribution of particles may be based on the dry blend characteristics or that of the pressed material. Particles are approximated as either spheres or cubes (circles or squares for 2-D modeling). A structured rectangular grid is overlaid on the RVE producing an array of subcells as shown in Figure 3.2.16. Note that each subcell in the grid may intersect many particles. The volume fraction of particulate matter that occupies each subcell is calculated. Next, a 2 x 2 x 2 GMC calculation is performed for the calculated volume fraction where the assembly of particulate matter in the subcell is replaced by a single particle with the same volume. This GMC calculation yields homogenized material properties within the subcell. Since each subcell within the RVE may have a different volume fraction of particles, each subcell may have a different set of homogenized properties. Thus in the first homogenization step, a random distribution of particles is replaced by a regular array of subcells, each of which may have different material properties. A second GMC homogenization step is then carried out for the entire RVE. In this step, the effective material properties of the RVE are calculated from the array of homogenized subcells using GMC. A comparison of GMC results to those obtained from finite element analyses of the same microstructures have shown that GMC provides accurate estimates of the elastic properties for a fraction of the numerical cost of finite element analysis (Banerjee et al., 2000).

The process of explicitly modeling a randomly generated array of particles in an RVE requires the generation of a random set of locations in Euclidean space where the particles reside at the beginning of the computation. Out initial generation of instances of these random microstructures was done in 2-D with circular particles. However, the difficulties involved in meshing a distribution of...
circular particles. However, the difficulties involved in meshing a distribution of circular/spherical particles with sizes differing by more than an order of magnitude has led to modeling square/cubic particles. Tests have provided evidence that approximating particles in the actual microstructure with squares/cubes is believed to be as valid as circles/spheres. Figure 3.2.17a (from Skidmore et al., 1998) shows an actual microstructure of the composite while the GMC representation is shown in Figure 3.2.17b. Another advantage of modeling square/cubic particles is the added simplicity in validating the GMC analyses using the finite element method.

An important consideration for any explicit micromechanical model of a composite is the selection of an appropriate RVE. This selection is not clear for materials with random microstructures such as shown in Figure 3.2.17. The larger the RVE that is selected, the greater the degree of randomness that may be modeled, but the more computationally expensive the calculation. Additionally, a number of smaller RVEs may be analyzed, each with a different microstructure, and the results from each averaged. There is currently no accepted method for determining the proper size or number of RVEs required to obtain a desired accuracy in material property determination.

**Determination of RVE Size**

To determine the suitable size and/or number of RVEs required a large, 2-D RVE containing approximately 100,000 square particles has been generated. The size distribution of particles available given by Skidmore (1997) was used to generate the random distribution within the RVE. By overlaying a regular subcell array onto the square particle distribution, a discretized mesh is obtained which may be used to compute the effective composite properties using both GMC and finite element analysis. The finite element solution using this large mesh is used as the effective material properties. Using the GMC-based two-step homogenization approach, the proper size and/or number of RVEs may be determined. A series of smaller RVEs are randomly selected from the larger mesh and the composite properties calculated. Solutions from different randomly placed RVEs will start to show large variations in material properties with decreasing RVE size. The smallest RVE size at
which material property variations are acceptably small may be considered an acceptable RVE. Preliminary results show that a 20x20 subcell RVE provides the same elastic properties as a 100x100 subcell RVE. Research is ongoing to confirm this behavior using finite element analysis.

3.2.6 Incorporation of Damage into GMC Analysis

Examination of photomicrographs of PBX-9501 reveals that microcracks are present at the particle/binder interfaces, in the binder material between particles, and in some cases through the particle themselves. Thus, it is important to incorporate effects of microcracks into the microstructure when calculating effective material properties. Microcracks may be modeled in GMC as inter-subcell displacement jumps using springs. The determination of the appropriate spring constants has been facilitated using finite element analyses. A circular particle containing an interface crack was modeled under an applied uniaxial load in the two geometric configurations shown in Figure 3.2.18. The virtual crack closure technique was used to determine the energy required to propagate a crack along the particle/binder interface. Assuming a known value for the critical energy release rate, $G_c$, the applied stress required to propagate an interface crack to a specified length may be determined. In addition, the additional compliance resulting from the interface crack may be determined. Together, the effective stiffness of an interface spring may be calculated for GMC modeling.

Results from finite element analysis show that the critical applied stress to propagate the interface crack is highest initially when the crack length is small as shown in Figure 3.2.19. As the crack length increases the energy release rate decreases, indicating that an interface crack, once formed, will propagate in an unstable manner around the perimeter of the particle. Hence, the effective stiffness of a particulate
composite undergoing interface cracking takes on two values: Up to the point where the critical stress is achieved, the material behaves as a fully bonded composite, while beyond this critical stress level, the effective properties are those of a fully debonded composite. Further analysis has been conducted to include distributions of different sized particles that more closely resemble actual HE composites (right figure in Figure 3.2.18). Results have shown that the energy release rate for a partially debonded large particle may exceed the initial peak value as the crack approaches a neighboring smaller. However, the fully-bonded to fully-debonded scenario appears to be a reasonable approximation, and permits the use of a constant spring constants in GMC analyses to model particle debonding. Thus, the single particle with a single crack methodology may be used to determine the effective interface spring stiffness for use in GMC. This requires a critical force before which the crack is closed that can be obtained from Figure 3.2.20 and a spring stiffness after the crack has opened. A normalized effective stiffness vs. volume fraction plot is shown in Figure 3.2.21. The large effect of the presence of an interface crack can be observed from this plot.

Modeling Stress Bridging Effects in GMC

The effect of stress paths across the RVE due to particles touching each other can be quite significant. For elastic materials, the effective stiffness of the composite can be considerably higher than that of a composite where no such stress paths exist (i.e., stress bridging). Since particles are not modeled explicitly in GMC, such stress bridging effects can be incorporated by an effect that is included in the first homogenization step. Our approach is to quantify the effect of stress bridging using a set of detailed sub-scale models and then to use the parameter studies to generate a surface that represents this effect as shown in Figure 3.2.22. This surface is then used to stiffen the composite response in the event of stress bridging.
3.2.7 Progress in MD Simulations

MD simulations are expected to play an important role in determining thermophysical properties of materials of interest to C-SAFE (HMX and polymeric binders). Year 3 efforts have been directed in three primary areas: general code development, parallelization, and determination of thermophysical properties. Work in this area important to progress in both the CD and HE steps. Details on MD code development and parallelization are discussed under High Energy Transformations. Of specific interest to Container Dynamics are the determination of thermophysical properties of HMX and polymeric binders. These properties are being used in the granular compaction simulations and will be inputs to our GMC micromechanics modeling and constitutive models. Below the results of this effort are summarized.

Thermophysical properties of HMX and Polymeric Binders

Liquid HMX. The PVT properties, viscosity and thermal conductivity were obtained as a function of temperature from MD simulations in the temperature domain 550 K < T < 800 K. The pressure dependence of PVT properties and viscosity has been determined. Results are published in Bedrov et al. (2000a, 2000b)

Crystalline HMX. Three polymorphs (α, β, and δ) has been simulated with flexible molecule force field that was developed based on high level quantum chemistry calculations in Year-2. Cell dimensions predicted from MD simulations are in very good agreement with experimental measurements for all three polymorphs. Elastic coefficients and speed of sound were determined at atmospheric pressure and 300K. For β-HMX the results were compared with recent experimental measurements of our collaborators from LLNL. Very good
agreement was found between simulation prediction and experiment. For \( \beta \)-HMX the pressure and temperature dependence of the cell dimension has been investigated, while for \( \alpha \) and \( \delta \)-HMX cell dimensions as function of temperature were determined. The simulations predicted dependencies are in good agreement with available experimental data. (Figure 3.2.23) Several manuscripts describing the results of crystalline HMX simulations are in preparation (Bedrov et al., 2000c).

**Polymeric Binders.**

PVT and viscoelastic properties of poly(vinylidene fluoride) (PVDF- model compound for Viton) and polybutadiene (PBD) have been predicted from MD simulations. (Figure 3.2.24) Simulations of low molecular weight polymer chains allowed us to extract conformational and dynamical characteristics of the polymeric chains. This information was used to parameterize theoretical models that allowed prediction of viscoelastic properties for high molecular weight (entangled) polymers. The predicted PVT and viscoelastic properties for high molecular weight polymers were found to be in excellent agreement with available experimental data. Our group was the first to predict viscoelastic properties of real, entangled polymer melts using only properties determined from MD simulations. Results of this work are published in: Byutner (1999); Smith, G.D., et al. (1999) Byutner, and Smith (2000a) and Byutner and Smith (2000b).

Figure 3.2.23: HMX coefficients of thermal expansion (CTE). Comparisons are shown with experimental data and previous simulations of Thompson which used a rigid molecular force field. Labels a, b, c, and V are unit cell dimensions (a,b, c) and volume (V). Each HMX polymorph has its own CTE for volume and a different CTE for each dimension in the cell.
Figure 3.2.24: Viscoelastic properties of high molecular weight poly(butadiene).
Simulations of Filled Polymers

We have recently initiated simulations of polymeric melts filled with nano-particles. The influence of particle surface on dynamical and conformational properties of polymeric chains will be investigated as a function of interaction strength between particle surface and polymer chain, temperature, pressure and polymer stiffness. Results of this effort will be presented in subsequent reports.

3.2.8 References


Guilkey, J.E. and Weiss, J.A. An Implicit Time Integration Strategy for use with the Material Point Method, 1st MIT Conference on Computational and Solid Mechanics, June


3.3 HE Transformations

3.3.1 HMX Decomposition Mechanism

Truong and co-workers have been focusing in developing computational tools for predicting accurate kinetics of gas phase chemical reactions. Recent accomplishments include methodologies for calculating kinetics of multi-channel reactions and pressure dependent rate constants. Such properties are known to be important in unimolecular decomposition reactions such as the thermal decomposition of HMX. To calculate the pressure-dependent rate constants and branching ratios of different channels, we employed the Master Equation (ME) method that can be described by

\[
\frac{dP_i}{dt} = [M] \delta E \sum_j (R_{ij} P_j - R_{ji} P_i) - \sum_m k_{im} P_i
\]

where \( P_i \) is the population of reactant molecules at the energy level \( E_i \), \([M]\) is the concentration of bath gas; \( R_{ij} \) is the rate of collision-induced excitation/relaxation from the energy level \( E_j \) to \( E_i \) of the reactant molecule in collision with a bath gas molecule; \( k_{im} \) is the microcanonical rate constants at the energy level \( E_i \) for the channel \( m \). This equation can be converted into a matrix form and solved efficiently.

We have developed an efficient computational methodology for calculating \( k_{im} \) based on the microcanonical variational transition state theory with inclusion of quantum-tunneling effects. The collision energy transfer rate of reactant molecules is based on the exponential-down model. The multi-channel dynamics of the first step in the decomposition of HMX is then examined for the first time using the new tool. Experimentally it is well accepted that the first step in the decomposition of HMX is either by the NO₂ bond fission or HONO elimination channel. The NO₂ bond fission channel was found to be the dominant one.

![Graphs showing pressure dependence of rate constants and branching ratios](image)

Figures 3.3.1 (a) and (b) above show the pressure dependence of the rate constants for both channels and their branching ratios, respectively. We found that both channels exhibit a rather weak pressure dependence. The calculated rate constants agree well with the available experimental high-pressure limit for the NO₂ bond fission channel at 800 K. An important result is that their branching ratio shows a strong temperature dependence. At 800 K, the NO₂
bond fission reaction is faster by a factor of 92 at the standard pressure 1 Torr. However, at 1000 K it is faster than the HONO elimination reaction by only a factor of 9. Since the experiments were often carried out at much lower temperatures (250-400 K), the contribution of the NO₂ bond fission channel is much more dominant and thus its significance is overstated. At temperatures above 1000 K, the HONO elimination channel as well as the C-N bond fission channel proposed by the Voth group would have noticeable contributions and should be considered.

The subsequent steps in the decomposition of HMX are much more complex. We have been exploring the potential surfaces for possible decomposition pathways after the first step. Figure 3.3.2 below shows our current progress in this effort.

We have identified two most probable pathways along the NO₂ bond fission channel in the first step. The energetically most probable pathway is the HMX → A2 → B4 → C1 → D1 → E1 → F1 → G1 → H1 pathway. This pathway yields H₂CNNO₂, N₂O, H₂CO and H₂CN as stable products. This provides an explanation for the experimental observations of noticeable concentrations of N₂O and H₂CO. The slightly higher energy pathway HMX → A2 → B4 → C2 → D2 → E2 yields H₂CNNO₂ and H₂CN. This confirms the previously suggested
mechanism. In the light of our branching ratio results above, we should also explore pathways along the HONO elimination channel particularly those from the B1, B2, and B3 intermediates. This is currently under investigation in our group.

3.3.2 Parallel Quantum Molecular Dynamics

Voth and co-workers have furthered their investigation of the four possible reaction mechanisms for HMX: N-NO₂ bond dissociation, HONO elimination, C-N bond scission of the ring, and concerted ring fission [1]. For these calculations, two levels of theory were used, BLYP/611G** and B3LYP/611G** within density functional theory. Dissociation of the N-NO₂ bond is putatively the initial mechanism of nitramine decomposition in the gas-phase. Our results find the dissociation energy of this mechanism to be 41.8 kcal/mol at the BLYP level and 40.5 kcal/mol at the B3LYP level, which is comparable to experimental results. Three other mechanisms are calculated and found at the BLYP level to be energetically competitive to the N-NO₂ bond dissociation; however, at the B3LYP level these three other mechanisms are energetically less favorable.

One particular note of interest is that for C-N bond dissociation a stable ten-membered ring intermediate is formed when the C-N bond is approximately 0.28 nm and its corresponding energy is found to be only 10.3 kcal/mol (BLYP) and 11.5 kcal/mol (B3LYP) above the ground-state gas-phase HMX molecule. This mechanism would not be reversible in the condensed phase, whereas N-NO₂ bond dissociation may be reversible. The irreversibility of the C-N bond dissociation mechanism suggests that further decomposition of the ring, as shown in Fig 3.3.3, may be propagated. The results of these calculations indicate that condensed-phase effect may change, even reverse, the ordering of the energetics between the two mechanisms. The relatively compact structure of the C-N bond scission transition state may be structurally less confining compared to the drastic extension of the N-N bond that must take place for the N-NO₂ group to dissociate.

3.3.3 Parallel MD Code Development

Quantum Molecular Dynamics

The Voth group has continued their progress on developing a massively parallel self-consistent electronic structure method, FIREBALL, which scales linearly with system size [2,3]. The computational bottleneck in such methods is that the band-structure energy is obtained from the diagonalization of a generalized eigenvalue equation, \( H \Psi = \varepsilon S \Psi \).
However, it’s useful to note that the matrices involved in the diagonalization are very sparse for large systems (i.e., ~5% non-zero elements in a 1000 atom system). In the recent years, several linear-scaling or O(N) algorithms were developed to exploit this scarcity and reduce the O(N^3) scaling attributed to solving the given generalized eigenvalue equation.

There are several classes of linear-scaling approaches which can be used, our method of choice is the variational functional developed by Kim, Mauri, and Galli [4]. This functional is of the form:

\[
\tilde{E} = 2 \sum M_{ij} (H_{ij} - \eta S_{ij}) (2 \partial_{ij} - S_{ij}) + \eta N
\]

which was developed by recognizing that the trace of a matrix is equal to the sum of that matrix’s eigenvalues. Their work finds that this functional yields a global minimum without traps at local minima and is efficient at reducing the number of iterations required for achieving a minimum. We have pursued a collaboration with Giulia Galli of Lawrence Livermore National Laboratory. Her added expertise in the area of linear-scaling electronic structure methods to our development of FIREBALL will better ensure its stability and performance of our chosen linear-scaling approach. In addition to the performance and stability of the functional as demonstrated by Kim, Mauri, and Galli, a massively parallel implementation of a similar functional within a tight-binding code by Itoh, Orderjn, and Martin demonstrated that the method scales linear as both the number of processors and number atoms [5].

In collaboration with Kris Sikorski from Computer Science, a nonlinear optimization algorithm was developed for FIREBALL, which minimizes the given energy functional in parallel. The variables in this optimization problem are the coefficients of quantum wave functions. The energy functional is a polynomial function of fourth degree of these coefficients. It also depends on the entries of the symmetric Hamiltonian and overlap matrices. The optimization code first computes the gradient, that is, the vector of derivatives of the energy functional with respect to each coefficient. The functional structure is such that each component of the gradient depends on coefficients held locally on other processors. Currently, this requires a considerable amount of communication, which will be overcome by implementing a procedure to avoid communication with other processors which do not contribute to the local matrix operations. With the gradient computed, the optimization algorithm determines an optimal step length along the gradient (or some other step direction depending on the gradient). It does so by computing the roots of a third-degree polynomial representing the derivative of the energy functional with respect to the step length. Thus the optimal step length is determined using only one computation instead of multiple tests. Initially, the gradient was used as the step direction (steepest descent) since the second derivative matrix is too large to allow computation of the pure Newton direction. Recently, the algorithm was modified to approximate the Newton direction using the quasi-Newton L-BFGS algorithm.
Our own preliminary efforts indicate that a similar parallel implementation within FIRBALL to that demonstrated by Itoh, Orderjn, and Martin in conjunction with the developed optimization algorithm produces parallel scaling as shown in Figure 3.3.4. This plot shows the time per iteration required to minimize the energy functional under various numbers of processors (test case of 448 atoms). The scaling is nearly perfect up to 128 processors, at which point either the inter-machine communication causes the scaling to drop off or size of the problem is too small for the given number of processors. The number of iterations required for convergence has been somewhat dependent on the number of processors under the steepest-descent method. The L-BFGS method has brought down the number of iterations considerably for some of the smaller tests (by a factor of three in most cases); it remains to be seen whether it will do so for the larger tests.

One difficulty with variational linear-scaling algorithms is the dependency on a good guess for the initial coefficients in order to reduce the convergence rate for the functional. Once the first optimization is obtained, subsequent optimizations can rely on the results for the coefficients of the preceding optimization as an initial guess. Since the atomic configuration does not change drastically between molecular-dynamic steps this guess is found to be quite reliable. However, obtaining the guess for the first optimization step may be a bit problematic. Collaborative efforts with Uwe Stephan (Frauenheim’s group in Germany) have been pursued to implement density projection techniques for improving the accuracy and acceleration of our variational O(N) algorithm [6,7].

The accuracy of the parallel linear-scaling implementation in FIREBALL looks promising. As a preliminary comparison between the linear-scaling algorithm and the exact diagonalization, the Mulliken charges of a single gas-phase HMX molecule were examined. This comparison gives an indirect comparison of the accuracy obtained for the density matrix in the linear-scaling algorithm as compared to the density matrix in the exact diagonalization scheme. A direct comparison is not the best comparison because the exact diagonalization scheme allows all the coefficients of the wave functions to be non-zero, and hence becomes non-sparse, but the level of scarcity of the density matrix in the linear-scaling algorithm is kept fixed. Initial results indicate that the RMS deviation of the Mulliken charges between exact diagonalization and the linear-scaling algorithm is 0.0433 (units of e⁻). Improvements to the density matrix can be made with improvements to the functional and implementation of the density projection technique discussed above, which are underway.
Additionally, the non-parallel version of the FIREBALL code was used to study a single gas-phase HMX molecule for evaluation purposes. Initial results indicate that using a minimal (smallest possible) single numerical (SN) (sp³) basis set and the BLYP exchange-correlation functional, FIREBALL predicts an energy difference of 190 kcal/mol between the boat and chair forms, which understandably affirms that a minimal basis set is insufficient. Many improvements have been made to the code to improve the overall precision of the results obtained. For example, the interpolation scheme has been changed. Improving the basis set to double numerical (DN) (ss*pp*3p*3) reduces the chair/boat energy difference to 6.8 kcal/mol. This energy difference gives the same thermodynamic trend as experimental results and demonstrates the need for an adequate basis set. Plans are underway to further improve precision by increasing the basis set used to DN plus polarization, implement better integration and interpolation grids, finish implementing B3LYP, spin-polarized DFT, and improve precision of data stored in data files. It is important to note that in both cases the correct energetic trend is predicted. A large variety of results are found using several different levels of theory and basis set. Using BLYP/6-311G**, Gaussian predicted that the boat form is 0.81 kcal/mol higher in energy than the chair form. B3LYP increases this to 2.33 kcal/mol. Goddard et. al. report results of 3.5 kcal/mol for B3LYP/6-311G(d,p)//MP2/6-311G(d,p) and 2.5 kcal/mol for B3LYP/6-31G(d) [8]. Some of the advancements planned for future calculations using FIREBALL, such as adding polarization to the basis set, will significantly increase memory and CPU requirements during the computation, thus the need for a linear scaling algorithm is even greater as the demands are increased.

**Classical Molecular Dynamics**

Grant Smith and co-workers have continued development of a massively parallel version of their classical molecular dynamics (MD) code for determining thermophysical properties of HE materials. The code has been generalized to be applicable for simulations of systems with arbitrary number of molecules and molecule types. The originally implemented simplified version of the force decomposition algorithm was restricting the code to be run on the \( P=n^2 \) number of processors, where \( n \) is an integer. The code has since been modified so it can run on any rectangular or square grid \( (P=n*m) \), where \( m \) and \( n \) are integers). This flexibility allows us to optimize the performance of the MD code on the massively parallel architectures by tuning the topology of the nodes for equal node loading and reduced communication for the system of interest.

A multiple time step integrator has been implemented in the parallel version of the MD code. This allows us to apply different time steps for integration of fast and slow degrees of freedom. For most of the systems the multiple time step technique results in speeding up simulations up to 3 times in comparison with single integration time step methods.

Parallel tempering (PT) is a new algorithm, which has been recently developed to speed up the equilibration and sampling of configurational space in MD simulations. The algorithm requires parallel simulation of \( N \) identical systems at series of different temperatures \( (T_1, T_2, T_3, \ldots, T_N) \), with occasional switching the coordinates between the systems. Systems at higher temperatures are able to explore the phase space much faster than the systems at lower temperatures. An appropriate protocol of switching the coordinates between systems allows each system to be simulated at all temperatures \( T_i \). When a system \( i \) is simulated at high temperatures it will travel in phase space much faster than it would at low temperatures. Therefore after some simulation time the system \( i \) will return to its starting
temperature with a significantly different configuration from the original one. The PT algorithm is illustrated below in Figure 3.3.5.

We have implemented the PT method for our MD simulations and are currently investigating its performance on massively parallel architectures. This approach appears to be the only feasible option for simulating estane and other polyurethane systems where specific polymers segments have a strong tendency for segregation. In such systems the conventional MD simulations will fail to reach equilibrium conditions at temperatures of interest because the time scales of the association/dissociation of those aggregates are much longer than the accessible simulation times. The PT method will potentially allow us to overcome this problem.

During last year we have used 133,400 and 39,800 CPU hours on ASCI Blue-Pacific and ASCI Red, respectively.

**Code Development Milestones**

1) The first version of the Force Field Toolkit (FFT1.0) has been developed and validated. The code allows automatic fitting of torsional potential, van-der Waals interactions, partial atomic charges and atomic polarizabilities. The geometry optimization and frequency analysis features were also implemented in FFT1.0.

2) Code for Monte-Carlo (MC) simulations that create highly packed configurations spheres of arbitrary size distribution has been created in our group and is currently used in micro-mechanics and compaction simulations by CD team members. (Note that this project was not in our original milestones).

3) A hybrid MC-MD code for flexible cell simulations (e.g., crystals) should be developed by October 2000.

**3.3.4 Thermophysical Properties**

**Liquid and Crystalline HMX**

The PVT properties, viscosity and thermal conductivity of liquid HMX was obtained as a function of temperature from MD simulations in the temperature domain 550 K < T < 800 K. The pressure dependence of PVT properties and viscosity has been determined. These results are published in references [9,10].
For crystalline HMX, three important polymorphs (α, β, and δ) have been simulated with the use of a flexible molecule force field that was developed based on high level quantum chemistry calculations carried out by our group in the previous year. Cell dimensions predicted from MD simulations are in very good agreement with experimental measurements for all three polymorphs. Elastic coefficients and speed of sound were determined at atmospheric pressure and 300K. For β-HMX the results were compared with recent experimental measurements of our collaborators from LLNL. Very good agreement was found between simulation prediction and experiment. For β-HMX the pressure and temperature dependence of the cell dimension has been investigated, while for α and δ-HMX cell dimensions as function of temperature were determined. The simulations predicted dependencies are in good agreement with available experimental data. Several manuscripts describing the results of crystalline HMX simulations are in preparation.

**Polymeric Binder Properties**

PVT and viscoelastic properties of poly(vinylidene fluoride) (PVDF- a model compound for Viton) and polybutadiene (PBD) have been predicted from MD simulations. Simulations of low molecular weight polymer chains allowed us to extract conformational and dynamical characteristics of the polymeric chains. This information was used to parameterize theoretical models that allowed prediction of viscoelastic properties for high molecular weight (entangled) polymers. The predicted PVT and viscoelastic properties for high molecular weight polymers were found to be in excellent agreement with available experimental data. Our group was the first to predict viscoelastic properties of real, entangled polymer melts using only properties determined from MD simulations. A block-scheme illustrating this algorithm is shown in Figure 3.3.6. Results of this work are published in references [11-14].

**Simulations of Filled Polymers**

G. Smith and co-workers have initiated simulations of polymeric melts filled with nano-particles. The influence of particle surface on dynamical and conformational properties of polymeric chains will be investigated as a function of interaction strength between particle surface and polymer chain, temperature, pressure and polymer stiffness.

### 3.3.5 Kinetics of Composite Explosives: PBX9501

Most high explosives are not pure materials, but rather composite mixtures of organic explosives such as HMX with polymeric binders and plasticizers. A few of the plastic-bonded explosive (PBX) formulations are given below in Table 3.3.1. It is important to recognize that whereas pure HMX is a white crystalline solid, PBX9501 is a pliable material with very

<table>
<thead>
<tr>
<th>Name</th>
<th>% Explosive</th>
<th>Binder</th>
<th>Color</th>
</tr>
</thead>
<tbody>
<tr>
<td>LX-14-0</td>
<td>95.5 HMX</td>
<td>Estane 5702-F1</td>
<td>violet spots on white</td>
</tr>
<tr>
<td>LX-10-0</td>
<td>95.0 HMX</td>
<td>Viton A</td>
<td>blue-green spots on white</td>
</tr>
<tr>
<td>PBX 9501</td>
<td>95.0 HMX</td>
<td>Estane, BDNPA/F</td>
<td>white</td>
</tr>
<tr>
<td>PBX 9011</td>
<td>90.0 HMX</td>
<td>Estane 5702-F1</td>
<td>off-white</td>
</tr>
<tr>
<td>PBX 9604</td>
<td>96.0 RDX</td>
<td>Kel-F 800</td>
<td>beige</td>
</tr>
<tr>
<td>PBX 9010</td>
<td>90.0 RDX</td>
<td>Kel-F 3700</td>
<td>white</td>
</tr>
</tbody>
</table>
different physical characteristics. A photograph of a small sample of PBX9501 is shown below in Figure 3.3.6. While the ultimate goal of the C-SAFE project is to predict the macroscopic behavior of such composite explosives (e.g., PBX 9501) based on molecular fundamentals, it is extremely difficult to predict the importance of chemical reactions that occur between the components.

To address this issue, Wight and co-workers have been investigating techniques for extracting kinetic parameters for solid-gas reactions of composite materials and using them to predict the combustion properties of energetic materials (composite explosives and propellants). Our approach to the general problem of reactions of composite materials has been to develop isoconversional model-free methods of kinetic analysis.[15,16] This approach is not subject to the limitations of breaking the overall reaction down into sequences of series and parallel reactions, each of which has an \( n \)-th-order dependence on concentration or extent of reaction. Rather, the model-free approach makes no assumptions about the functional form of the reaction model and extracts the kinetic parameters as numerical functions of the overall extent of reaction. The two best ways of measuring the extent of reaction experimentally are thermogravimetric analysis (TGA: measurements of conversion to gases vs. time and temperature) or differential scanning calorimetry (DSC: heat generation as a function of time and temperature).

Using this general approach, we have been able to determine the overall kinetic parameters for pure energetic materials such as RDX [17] and ammonium perchlorate [18], as well as pure polymers [19,20]. More recently, we have applied the same techniques to composite explosives and propellants. Specifically, we identified three basic steps in the thermal decomposition of PBX9501: vaporization of the BDNPA/F nitroplasticizer, thermal decomposition of HMX in an estane matrix, and gassification of the remaining estane component and polyamide residue from the HMX reaction.[21] These steps are shown in Figure 3.3.7.

There are two immediate applications of this kinetic information within the framework of the C-SAFE project. The first is to utilize the kinetic parameters directly in sub-grid scale combustion models to predict rates of solid-gas reaction rates in a fire environment (i.e., extrapolation of the laboratory data to the high temperatures of a combustion environment). The Wight group has been developing a 1-D transient sub-grid scale model to utilize model-free kinetic data derived from thermal analysis experiments to predict the rates of solid state reactions during ignition and combustion of composite explosives. This sub-grid scale model will be interfaced to the MPM code developed by the Container Dynamics group in order to

![Figure 3.3.6 - PBX9501](image)

Figure 3.3.6 - PBX9501

![Figure 3.3.7 - TGA curves for PBX9501](image)

Figure 3.3.7 - TGA curves for PBX9501
provide information to the overall simulation about mass burn rates and temperatures of individual material points in the simulation as a function of their thermal and temporal history.

The second application of the isoconversional data analysis is to use the kinetic parameters to predict the shelf life of PBX9501. In this latter application, we have recently predicted that PBX9501 should lost about 30% of its plasticizer component in 4 days at 50°C if kept in an open system from which gases readily escape. Sealed systems that retain the plasticizer should undergo much slower degradation. As far as we know, this is the first kinetic prediction of the shelf-life of PBX9501 derived from thermal analysis experiments, and is only possible because the model-free method of kinetic analysis is able to extract reliable kinetic data without making assumptions about the order of reaction.

3.3.6 Literature References


3.4 Computer Science
The major areas of the Computer Science effort include:

1) Problem Solving Environment (PSE): The PSE team has focused its efforts on creating the Uintah Computational Framework (UCF) supporting both MPI and MPI/Thread parallelization. During this effort, the PSE team has guided the integration of the other teams’ codes into the overall Uintah architecture.

2) Visualization: The Uintah visualization tools have been greatly enhanced and include (1) extended particle visualization tools, (2) grid visualization and query methods, (3) hardware based multi-resolution volume rendering, (4) animated streamlines, and (5) Real Time Ray Tracing (RTRT).

3) Software and Data Management (SDM): Development efforts in C-SAFE scientific data management during the past year have focused on bringing tools to productive use within C-SAFE tasks. This involved three areas: SDM web service, Uintah Blazer, and Validation team combustion research support.

4) Performance Analysis: We are currently working to develop tools that will help track the utilization of processor resources based on the task graph parallelization abstraction.

3.4.1 Problem Solving Environment
This past year, the main efforts of the Problem Solving Environment (PSE) team have been focused on the examination and implementation of the Uintah Computational Framework (UCF). The UCF has been designed and implemented by the PSE team to satisfy three goals: 1) To provide a general framework for massive scale simulations of fluid and particle physics, 2) to maximize parallelization and scalability, and 3) to allow scientists from outside the computer field to have an intuitive method for easily inserting their algorithms into a parallel framework without being bogged down by all the details of parallel programming.

3.4.2 The Uintah Computational Framework
To use the UCF, a scientist need only specify the list of tasks that make up the algorithm, along with the data that each task requires and computes. To simplify the implementation of these tasks, the UCF provides a data warehouse which acts as a global memory abstraction to the actual task implementation. Once the tasks are specified, the UCF distributes them to the computational resources supplied to the simulation. Currently this load balancing is done statically, which has been successful for the relatively uniform problems that we have been simulating to date. However, as the integrated simulation comes together, we will develop a more sophisticated load balancer that will monitor and react to the simulation dynamically.
Figure 3.4.1 A taskgraph for a 4 processor/4 patch MPM simulation. Boxes are tasks, and the arcs show the communication between tasks. Red tasks are the longest running, green are the shortest.
**MPI Version:**

We initially concentrated on developing an MPI version of the Uintah UCF. To date, this version of the UCF runs on 500 processors and scales well up to 128. There are several known inefficiencies in the UCF implementation. For example, we are currently sending all particles of a patch from one MPI process to another, instead of just the ghost cells. These inefficiencies are due to time and personnel constraints and will be addressed in the coming months.

**Mixed MPI/Threads Version:**

In the last month, we began implementing the mixed MPI/Threads version of the UCF. This version of the UCF will help minimize the amount of message passing across the simulation by allowing a number of shared memory threads to execute tasks that work on a common group of patches. Not only does this drastically reduce the number of messages and the size of data being passed, but allows groups of threads to use common shared memory data warehouses.

**Visual Programming Interface and the CCA**

We have implemented a component model that is evolving along with the Common Component Architecture (CCA) specification. While the component model does not yet have its own graphical user interface, it will be implemented in the next year. Currently the UCF is comprised of these main components: the Simulation Controller, the Load Balancer, the Scheduler, an MPM and an ARCHES component, and the Data Archiver Component. Each of the components connects to the Simulation Controller component using CCA ports. The UCF has been designed to allow many different algorithms to be easily incorporated into the system.

**Time Varying Simulations**

The UCF Data Archiver and Data Warehouse components have been developed to support time varying simulations. These two pieces of the UCF allow algorithmic components to process and store data in discrete time steps. They also provide a direct interface, via their CCA ports, to the visualization tools that are being developed to display the immense amounts of data generated per time step by the simulation. Over the next two years we will continue to develop and improve the UCF support for time variance.

**Overall Uintah Architecture**

With the development of the UCF, the PSE team’s role in guiding the other teams efforts has increased dramatically. It has been quite challenging as well as very rewarding to develop an integrated computational framework to support the other scientists at our center. The development of the UCF would not have been nearly as successful without the insights provided by our colleagues with regards to the needs of application driven mechanical and chemical engineers.

**PSE/UCF Platform Ports**

The MPI UCF has been ported to Linux. An SP2 port is in progress.
The next goals include: 1) Optimization of the UCF; 2) Finalization of the mixed MPI/Thread portion of the UCF; 3) Development of a more sophisticated load balancer, and 4) Creation of a Visual Programming GUI.

3.4.3 Visualization:

The Visualization team has been working on three major areas:

- Step-Specific visualization tools
- Multi-resolution techniques
- Visualization data manipulation and retrieval.

The step-specific visualization tools include particle methods for the Container Dynamics (CD) step, volume rendering techniques for the Fire-Spread (FS) step, vector visualization methods for both FS and CD, and sphere rendering methods for the real-time ray-tracer which is targeted towards the CD and Molecular Dynamics (MD) steps. This past year has also focused on common infrastructure within the Uintah PSE which applies to all steps as well as step-specific visualization methods.

The Uintah visualization environment has been expanded to include grid structure visualization, grid variable querying, grid variable tracking, hardware based volume rendering and volume slicing in both standard and multi-resolution forms, animated streamlines, and a series of tensor tools including: component extraction, submatrix extraction eigenvalue and eigenvector extraction. Direct tensor visualization via ellipsoids on particles is being developed.

The visualization tools have been modified to read data directly from the new data archive which serves as the repository for all simulation results. As the form of the data in the data archive matches that used in the simulation code, we should eventually be able to update the visualization tools directly as a simulation progresses.

A data converter which transforms data archive data into RTRT data has been written. This allows us to manipulate millions of particles or molecules in real time. This also allows real time isosurface extraction on grid data hundreds of megabytes in size.

Animated streamlines provide visual cues to the magnitude as well as the direction of flow. Our method includes the transparency decay over time so that slow moving streamlines appear short while faster flow is represented by longer streamlines.

The volume rendering components of Uintah are integrated with particle visualization techniques through the use of 3D texture memory based volume rendering. This method will allow simultaneous visualization of the Arches and MPM code working in the same computational environment.

A colormap generation module has been built that allows the user to manipulate the opacity of a color or series of colors. This proves useful when combined with the texture based tools below.

We have prototyped a multi-resolution based volume renderer. The volume renderer can be used to render a grid variable at full resolution. Or if the dataset is large, specific regions of the data can be resolved, while the remainder is rendered at a lower resolution. Colormap modules that include opacity information can be combined with the volume renderer to effectively remove various values from the visualization.

Texture base cutting planes can be used to cut through grid based variable data much faster than the previous cutting plane modules. These planes also use opacity information to
perform binary opacity. If data is either non-existent or marked as transparent the user is allowed to see through the planes to look at other objects.

3.4.4 Software and data management (SDM)

Establishment of Dedicated Web Server

A server dedicated to web-based C-SAFE SDM support was created. This server (blazer.cs.utah.edu) is a Dell Pentium III with 256MB RAM and a 18GB SCSI Ultra 2 hard drive (soon to be augmented with a second 37 GB drive). Data is stored in a local Microsoft SQL Server relational database management system. An Apache web server uses Java servlets and Java RMI to access the MS SQL Server database. Plain text passwords control access to individual pages, as required, for example to perform administrative functions such as file deletions. Infrastructure is available for stronger security, including digital certificates, as security needs increase.

Blazer Simulation Management System

In consultation with the UCF design and implementation team, a Blazer web service was established for the registration, browsing and querying of UCF simulation run files. Given the diversity of platforms used by UCF developers, the decision was made to use a Perl script for collecting, annotating, archiving (in a .tar file), compressing and uploading files describing a UCF run.

Upon receipt the .tar file associated with a UCF run is stored on the Blazer disk, and the XML configuration file is parsed with its most pertinent fields stored as a record in the SQL Server database. This data can then be browsed, queried, inspected, and downloaded for use by other developers.

The emphasis in this effort is to provide a tool with practical utility to UCF developers, while creating a case study for the exploration of more advanced SDM techniques of computer science research interest in their own right.

As of the writing of this report, the Blazer tool has been used by the UCF developers who have directly contributed to its design. Over the next few weeks the Blazer tool will be disseminated to the UCF development community as a whole, and the design of Blazer will be refined and adapted as dictated by requests, problems and suggestions obtained from this broader user base.

Validation Team Model Web Site

In consultation with Validation Team, and their combustion research colleagues around the world, another service has been established using the Blazer web server. This differs from the Blazer UCF service in several ways:

- The simulation run configuration files and output files are considerably smaller and simpler than are those of Blazer runs. Hence support is provided for form-based creation and editing of configuration files, which is not practical for UCF runs.
- The Validation web server directly contains executable code for several combustion models, contributed by Validation Team colleagues. Execution of
these models is directly performed by the Validation web server, with results captured in the MS SQL Server database.

- A user can directly visualize the results of Validation model runs as two-dimensional graphs, both individually and comparatively, through a menu-driven visualization applet.

**Participation In ASCI CDM Effort**

We have continued our participation in the ASCI Tri-Lab Data Models and Formats by attending the October 1999 DMF Workshop at Los Alamos. At this meeting discussions were held with Mark Miller of LLNL the suitability of SAF for use by UCF developers. After the workshop, a tutorial was given to UCF developers, and the decision was made to discuss specific technical and practical points to determine whether UCF development should be based on SAF. Subsequently, a discussion by conference call was held between LLNL and key UCF developers, and which SAF concepts, objectives and tools were examined in detail, in the context of UCF needs. The general conclusion was that SAF is very attractive to UCF developers conceptually, but the rapid UCF development schedule did not align with the project SAF release dates. Nevertheless, UCF developers, primarily through the SDM group, will continue to track SAF development, to continue exploring the possibility that UCF ultimately can be ported to a future SAF release.

Goals for next year include: 1) the primary goals for the coming year are to transition the Blazer; 2) (and Validation) web services to full scale daily production use; 3) Specific objectives include:

- Extend Blazer to initiate, monitor and control local UCF batch executions
- Extend Blazer to manage remote (e.g., at LANL) UCF batch executions, as well as multi-step executions combining codes from multiple C-SAFE Step Teams.
- Incorporate visualization tools in Blazer, including visualization of rendered results, as well as UCF program definition graphs.
- Provide Blazer support for the UCF program construction process, by establishing a database of UCF modules, queryable by input/output signatures, descriptive annotations, and usages in previous UCF runs.
- Extend Blazer XML querying capabilities, with deeper capture of XML data specific to particular UCF problem definitions.
- Enhance and improve the Validation Team web server, on the basis of international use within the combustion community.
- Renew and update contact with SimTracker team, now that Blazer functional and technological requirements are more concretely know.

**Performance Analysis:**

The key abstraction that the UCF builds upon for parallelization is the task graph. This abstraction aids us in analyzing the UCF’s performance. Using graph analysis techniques, we are able to determine the critical and near-critical paths through the task graph. This allows us to focus our efforts on speeding up the portions of the code that actually create the bottlenecks during execution. We are also working on having the ability to perform visualization of statistical data derived from the task graphs.
3.5 Validation

Validation of the overall model and submodels developed in this program requires either experimental information or fundamentally-based computations for comparison. There is information available in the literature that will be utilized for validation purposes; however, there are critical gaps in this information requiring the performance of additional experimental work. Other DOE programs being carried out at the National Laboratories will provide some of this information. It was recognized, however, that there is a need to provide additional validation information that is specific to the mission of C-SAFE. The University of Utah has provided some leveraging funds to the C-SAFE effort, to support experimental work being performed in the C-SAFE validation program not funded by DOE ASCI resources.

The validation effort is being conducted at four levels of complexity, starting with fundamental rates and properties, progressing through experiments on simple processes, to experiments coupling two and three of the C-SAFE steps. As the experiments increase in complexity, the objectives are targeted more to answering key questions raised during the model development than to validation of the integrated model, which would require an accumulation of detailed information beyond the resources of this program.

3.5.1 First Level of Validation: Fundamental Rates and Properties

Validation during code development is the responsibility of code developers, and investigators within C-SAFE are looking to the scientific literature, ab initio calculations and to the National Laboratories to provide as much information as possible for use in code validation. In specific circumstances where such information is lacking, the validation group is utilizing their resource to address some of these voids. One area of great technical importance to the successful simulation of accidental fires and explosions is the formation of soot. This problem presents a significant technical challenge, and success will be dependent upon not only the efforts within C-SAFE, but the successful integration and coordination of efforts by researchers at C-SAFE, the National Laboratories and greater academia. A team comprised of key individuals in the area of soot mechanism development has been constructed to address the problem of soot formation from complex liquid hydrocarbon fuels; specifically, the jet fuel JP-8.

We are using a Simulation Roadmap to define the sequential steps that occur in the development of the soot model, and the Soot Model Development Roadmap (SMDR) was shown previously in the Fire Spread Section as Figure 3.1.11. The SMDR shows the paths that have been set up to supply required information for each step in the development process, and there are key validation contributions that fit into that process as discussed below.

Kinetic mechanisms for soot formation

According to the development roadmap, this area consists mainly of two parts: gas-phase chemistry and its coupling to the soot chemistry. The effort in this area is funded under the Fire Spread task and the details are given in that section of this report. As many individuals have been working in this area, it became clear that a coordinated effort between the many researchers might prove to be most effective in obtaining the best soot formation mechanism possible. Thus, the C-SAFE Validation group has taken the role of coordinating
this effort and is hosting an electronic interaction using the assistance of Computer Scientists within C-SAFE as described in the following section.

**Web-based validation**

To provide a transparent validation effort we worked with Prof. Gary Lindstrom from the Computer Science Department (see in the SMDR - web-based validation step) and we explored the possibility of using Web accessible methods for information exchanges.

During this year we have set up the C-SAFE validation web page for the soot model development with information from four of the major groups working on soot formation mechanisms in order to select and critically evaluate the reference mechanism developed. The groups are: Dr. Westbrook at LLNL, Prof. Frenklach at University of California, Prof. D Alessio at the University of Naples, Italy and Prof. Ranzi at the University of Milan, Italy. The URL is http://panda.cs.utah.edu/~validate. Two codes (Chemkin-Premix and Dsmoke) and several reaction mechanisms are now available on our website. The Dsmoke software, developed by Ranzi’s group at the Politechnic of Milan, Italy, in addition to the well-known Chemkin code, allow for the analysis of different combustion systems, PSR, shock tube, premixed flames, etc.

Several reaction mechanisms are also available on the web site: LLNL’s aromatic mechanism, Frenklach group’s HACA mechanism, Ranzi’s kinetic scheme, n-heptane and iso-octane chemistry. The web site also permits comparison with databases in the literature (e.g., www.ecs.umass.edu/MBMS/), and will be supplemented by the data gathered as part of the C-SAFE project. We have also set up a database with the data collected in the literature for some premixed flames. The modeling results obtained by our group are reported on the web site in order to get comments and suggestions from the other groups (see Discussion page at the validation URL). Until now, the Web page has been limited in access to those who are involved in the collaboration, but our goal is to open up the site to all individuals interested in this topic. To do this, we are working with Prof. Lindstrom and Prof. Ranzi to develop an interface for both the codes to allow the user an easy access.

**3.5.2 Second Level of Validation: Experiments on Key Processes**

Efforts in this area are focusing on experiments to address specific questions or voids in the information available in the literature. The primary areas of research include surrogate fuel formulation to simulate JP-8; characterization of young soot structures to facilitate mechanism development; the development of a laser-based absorption/emission experiment to measure soot properties for surrogate validation, and the construction of a laminar-flow slot burner for use in conjunction with the laser measurements; and also thermogravimetric studies of the decomposition of various HMX/binder formulations. A summary of the progress in each of these areas will be given below, with the exception of the HMX studies, which will be given in the section on High Energy materials.

**Progress in Jet Fuel Surrogate Formulation**

The complexity of real jet fuels makes it very difficult to directly simulate their combustion behavior in fires. Thus, a surrogate that has a simple and clearly defined composition is necessary to quantitatively describe the physics and chemistry in the CFD fire simulation. The surrogate mixture must be able to reproduce the important physical and
chemical parameters of interest in predicting heat transfer from a pool fire to an immersed container. A preliminary study on surrogate formulations was completed in FY98, where several ternary mixtures were studied. Results of these studies showed that a ternary or four-component system was not sufficient to accurately simulate the boiling point curve of the real jet fuel. The ternary system also restricted the selection of aromatic compounds, which in turn led to a poor match with sooting index, another key parameter required for the surrogate.

Surrogate mixtures with more than four components were tested this year. Mixtures with six components were found to demonstrate good agreement with the jet fuel sample and also to provide enough flexibility to adjust the surrogate composition for special needs. Four surrogate formulations are shown in Table 3.5.1.

Table 3.5.1 Composition of surrogates (vol.%)

<table>
<thead>
<tr>
<th>Schulz</th>
<th>Hex-1</th>
<th>Hex-2</th>
<th>Hex-4</th>
<th>Hex-5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isooctane</td>
<td>Isooctane(^a), 10</td>
<td>Isooctane(^a), 5</td>
<td>Isooctane(^a), 2</td>
<td>Isononane(^e), 5</td>
</tr>
<tr>
<td>MCH(^b)</td>
<td>MCH(^b), 20</td>
<td>MCH(^b), 5</td>
<td>MCH(^b), 5</td>
<td>Cis-decalin, 20</td>
</tr>
<tr>
<td>m-xylene</td>
<td>m-xylene, 15</td>
<td>Toluene, 20</td>
<td>m-xylene, 20</td>
<td>m-xylene, 10</td>
</tr>
<tr>
<td>Cylooctane</td>
<td>Dodecane, 30</td>
<td>Decane, 25</td>
<td>Cis-decalin, 18</td>
<td>Tetralin, 15</td>
</tr>
<tr>
<td>TMB(^d)</td>
<td>Tetralin, 5</td>
<td>Dodecane, 25</td>
<td>Dodecane, 30</td>
<td>Dodecane, 25</td>
</tr>
<tr>
<td>Decane, 16</td>
<td>Tetradecane, 20</td>
<td>Tetradecane, 20</td>
<td>Tetradecane, 25</td>
<td>Tetradecane, 25</td>
</tr>
</tbody>
</table>

Note: \(^a\) 2,2,4-trimethylpentane, \(^b\) b-methylcyclohexane, \(^c\) 1,2,4,5-tetramethylbenzene, \(^d\) 1-methylnaphthalene, \(^e\) 2,2,5-trimethylhexane

The boiling point curves of surrogate and real jet fuel are shown in Figure 3.5.1. WPAFB designates a real military jet fuel from Wright-Patterson Air Force Base simulated by Schulz\(^1\) with a 12-component surrogate mixture. The four CSAFE surrogates have almost the same distillation curve over the range 50-100\(^v\%)\%; however, the differences in the first 50\(^v\%)\% of the boiling range is significant. Compared with WPAFB, Hex-5 has a better match than Schulz\'s formula in the first 80\(^v\%)\%. At the high end, however, the curve is a bit lower than both Schulz\'s surrogate and WPAFB because no components heavier than tetradecane were included in Hex-5. Hex-4 is almost the same as Schulz\'s surrogate in the first 80 \(^v\%) with half as many components. The low initial boiling point (IBP) of Hex-1 is due to the high percentage of light components such as isooctane and methyl-cyclohexane. When reducing the percentage of these lighter compounds, a much higher IBP was found as indicated by Hex-2.

The results of estimating soot index and flash point for the surrogate mixtures are summarized in Table 3.5.2. The comparison of smoke points of different fuels is shown in
Figure 3.5.2. To simulate JP-8, a compromise must be made between high flash point and low soot index. In the 5 surrogates, only Hex-5 has qualified flash point. Hex-4 and Schulz’s surrogate behave similarly for volatility and soot formation. Hex-1, and 2 are too volatile to be a good surrogate for jet fuel. A slight composition adjustment will be necessary for Hex-5 to meet the soot index; for example, the tetralin percentage can be decreased in Hex-5 to more closely simulate the soot index.

Table 3.5.2 properties of surrogates

<table>
<thead>
<tr>
<th></th>
<th>Schulz</th>
<th>Hex-1</th>
<th>Hex-2</th>
<th>Hex-4</th>
<th>Hex-5</th>
<th>ASTM</th>
</tr>
</thead>
<tbody>
<tr>
<td>TSI</td>
<td>18.0</td>
<td>15.6</td>
<td>13.4</td>
<td>17.2</td>
<td>20.0</td>
<td>&lt;19.1</td>
</tr>
<tr>
<td>Flash Point, C</td>
<td>22.49</td>
<td>-10.8</td>
<td>12.39</td>
<td>21.70</td>
<td>40.3</td>
<td>&gt;38</td>
</tr>
<tr>
<td>Latent Heat, kJ/kg</td>
<td>364.3</td>
<td>367.4</td>
<td>367.2</td>
<td>365.5</td>
<td>369.8</td>
<td>--</td>
</tr>
<tr>
<td>Combustion Heat, MJ/kg</td>
<td>43.25</td>
<td>43.2</td>
<td>43.3</td>
<td>43.09</td>
<td>42.8</td>
<td>&gt;42.8</td>
</tr>
</tbody>
</table>

It was determined that a six-component surrogate provides sufficient flexibility to simulate the major properties of interest to pool fires of real jet fuels. Both laminar and turbulent flame experiments will be carried out with the different surrogate fuels and individual components that comprise these fuels. The differences in soot volume fraction and structure will be monitored using the soot probe and NMR as described below.

Figure 3.5.1. Comparison of boiling point curves for an actual jet fuel (WPAFB), a 12-component.
3.5.3 Progress in the Characterization of Soot Structures

Understanding the soot formation process is central to the development of a valid model of pool fires. The largest uncertainty in the soot formation mechanism proposed by different investigators is in the gas-to-particle conversion step. In order to guide the model development, we are taking advantage of the specialized capabilities in solid state nuclear magnetic resonance (NMR) spectroscopy at the University of Utah to identify the chemical structure of soot and soot precursors. The past year has been devoted to producing suitable samples and refining NMR techniques to identify the average chemical structural features of soot particles in the early stages of formation. These experimental data have been augmented by theoretical calculations that will be used to identify molecular structures for which experimental data are not presently available. The progress of Prof. Pugmire and his group is briefly summarized below.

Because of its noninvasive and non-destructive nature, nuclear magnetic resonance spectroscopy has emerged as a major tool for characterizing complex structures and dynamics. These experiments have been especially important for studying coal structure, reactivity, and pyrolysis products\(^1\) and these data provided the critical information used to develop the

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**References**

chemical percolation devolatilization model (CPD)\(^2\) which is now widely used to model combustion processes in industry and to model polymer thermal degradation at Sandia National Laboratory\(^3\).

Within the past year major progress has been made in the characterization of gas phase soot precursors and soots in the early stages of evolution. The soot precursors and soot samples were produced in (1) a drop tube furnace and (2) a flat flame burner under conditions where the temperature and residence times could be controlled. The NMR soot characterization experiments have taken advantage of earlier work supported by NSF and DOE. These experiments utilize a one-dimensional (1D) \(^{13}\text{C}\) CPMAS NMR analysis procedure developed in the Utah laboratory for studying coal and coal pyrolysis products. A series of NMR experiments provides data that can be used to determine an average carbon skeletal structure, including average aromatic carbon cluster size, average number of size of attachments per cluster, and average mass of the connecting bridges between the ring structures. Samples have been produced utilizing selected model compounds under conditions specified in Table 3.5.3.

The conditions utilized for the model compound studies were determined by a careful series of pyrolysis experiments utilizing tars produced from an Illinois #6 coal

<table>
<thead>
<tr>
<th>Model compound</th>
<th>Pyrolysis Temperature(s) (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>anthracene(^a)</td>
<td>1250, 1300, 1400</td>
</tr>
<tr>
<td>biphenyl(^b)</td>
<td>1365, 1410, 1470</td>
</tr>
<tr>
<td>pyrene(^b)</td>
<td>1410, 1460</td>
</tr>
<tr>
<td>phenanthrene(^b)</td>
<td>1440</td>
</tr>
<tr>
<td>acenaphthylene/acenaphthene(^bc)</td>
<td>1350</td>
</tr>
</tbody>
</table>

\(^a\)Samples prepared in the University of Utah drop tube furnace.
\(^b\)Samples prepared with flat flame burner at BYU.
\(^c\)Samples prepared from 80/20 mixture of acenaphthylene/acenaphthene

by means of a flat flame burner designed so that the temperature could be controlled within the range that would not produce highly graphitized material. The NMR spectra of the coal tar samples are presented in Figure 3.5.3. It is clear from the NMR data that the tar/soot structure consists of moderate size aromatic structure at temperatures below 1500 K. At higher temperatures, large aromatic clusters are evident and other measurements of physical properties indicate that significant changes occur in the magnetic susceptibility, dielectric constant, and conductivity of samples produced at temperatures >1500 K. These data indicated the temperature range most favorable for studying soot production in its formative stage where chemical information can be obtained which can be incorporated into modeling schemes.

In Figure 3.5.4 one notes that the anthracene soot samples are dominated by unreacted anthracene at 1250 and 1300 K with only small amounts of pyrolysis products present. However, the proton NMR relaxation data indicates that a substantial amount of stable free radicals are present in the pyrolysis products. Even at a temperature of 1400 K a small amount of unreacted starting material still exists. These data demonstrate the resonance stabilization

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of linear PAH’s. Such stabilization is not observed in the structural isomer phenanthrene. While the NMR data on all the samples in Table 3.5.3 have not been fully analyzed at this time, it is clear that products observed in the early stages of soot formation reflect the material from which the soot samples are made. An example can be clearly seen in Figure 3.5.5. The average structural parameters indicate that ring opening reactions occur in biphenyl at relatively low temperatures (e.g., 1365 K, where aliphatic carbons represent 7% of all carbons present), followed by ring condensation reactions at temperatures above 1400 K. Pyrolysis of pyrene under similar conditions show little evidence for ring opening reactions (again due to resonance stabilization of the aromatic ring system) and yields polymerized pyrene as well as larger PAH materials.  

In complex PAH’s, 1D experimental techniques cannot always identify subtle features that are necessary to characterize complex hydrocarbons systems. The key architectural features of PAH’s are found in the types of bridgehead and substituted carbons that form during ring condensation. Different types of aromatic bridgehead carbons (e.g., inner vs. outer bridgehead carbons in planar aromatic systems, condensation of six- and five-member rings that give rise to curvature in aromatic ring systems such as fullerenes and fullerene precursors) and substituted carbons can be identified by the unique features of the principal values of the $^{13}$C chemical shift tensors. The chemical shift principal values can be obtained by means of 2D NMR experiments such as FIREMAT which was developed at the University of Utah.  

The shift tensor data base presently available in the Utah laboratory on 1-5 ring PAH model compounds will be supplemented by studies of selected model compounds that are relevant to soot models. We recognize that much of the material encountered in soot samples will derive from molecules for which no experimental data is available. Fortunately, computational tools now exist for computing the shift tensor values with a high degree of accuracy (e.g., 2-3 ppm) if the molecular structure (particularly C-H bond distances) has been accurately determined. By using MM techniques for geometry optimization in the Utah database of ~30 PAH’s, the theoretical shift tensor principal values for these compounds can be determined with reasonable accuracy (see Figure 3.5.6). By employing cluster theory, it is possible to classify a database containing 305 sets of shift tensor principal values into seven classifications (based on presently available data) of aromatic structures in PAH’s. Hence, it appears to be feasible to use theory to assist in identifying the structural types that are observed in soot samples but for which no model compound data has been obtained. The general classification of the types of PAH carbons are given in Figure 3.5.7.

The major objective of our NMR experiments is to obtain data on initial soot formation and early growth. The research plan involves: 1) Investigation of soot samples derived from carefully selected model compounds (both solids and liquids) obtained at early to intermediate stages in the soot evolution cycle, following initial radical formation and prior to the agglomeration stage; 2) Data on aromatic ring structure growth will be used to assist in modeling the overall growth of soot from incipient formation to a relatively early stage just prior to the formation of the large amorphous carbons that constitute the final stage of soot agglomeration. Experiments will include 1D and 2D techniques with a focus on refining the latter in order to take advantage of the rich amount of information available from the $^{13}$C chemical shift tensor data; 3) The shift tensor data base presently available in the Utah laboratory on 1-5 ring PAH model compounds will be supplemented by studies of selected model compounds (funded by other agencies) that are relevant to soot models; 4) Quantum mechanical calculations will be employed to assist in correlation of experimental data with theory, and where necessary, to interpret the experimental data when the experimental data base is found to be deficient.
References


5. Winans, R. E. and Pugmire, R. J., unpublished high resolution mass spectroscopy data.

Figure 3.5.3. $^{13}$C NMR data of coal and tar/soot samples produced at various temperatures in a flat flame burner. Numbers on right of spectra indicate average number of aromatic carbons/cluster

Figure 3.5.4. $^{13}$C NMR spectra of tar/soot samples produced in a drop tube furnace.

**Biphenyl Soots**

Figure 3.5.5. Average structural information (bar graphs) derived from $^{13}$C NMR data obtained on soot/tar samples of biphenyl and pyrene. Average cluster size (aromatic carbons per aromatic cluster) and number of cross links are given for the model compound as well as for each tar/soot sample studied. Molecular structures are suggestive of the types of materials that are present in the various tar/soot samples

**Pyrene Soots**
3.5.4 Progress in the Measurement of Soot Concentrations

A water-cooled fiber optic probe will be designed and built in order to obtain an in situ measurement of a pilot-scale pool fire. Prof. Jay Gore of Purdue University suggested the original design for the probe, and the probe is being built in collaboration with Sandia National Laboratory in Livermore (SNLL). Decreasing the overall size of the instrument minimizes the intrusiveness of the probe inside the flame. The method investigated in this research is line of sight measurement, thus inhomogeneities along the path length need to be considered. In the two-color pyrometry measurement, the measured temperatures will be sensitive to $\ln(\varepsilon)$, making them biased toward the high temperature regions. Also, the conventional two-color method, rely on the assumption that the particles are small enough to
be in the Rayleigh regime, were absorption dominates over scattering. It has been proposed that the methodology be expanded such that the scattering will be measured in unison with both the absorption and emission.

The absorption measurement experiment will utilize a 5 KHz modulated 635 nm red diode laser beam that gets attenuated in a flame with a given path length. This line of site measurement of monochromatic absorption is related to an effective soot volume fraction. The volume fraction through absorption is obtained using Beer’s law given the assumption of soot particle diameter being below the Rayleigh limit.

The soot volume fraction by emission and temperature are acquired by using two-color pyrometry. Two Avalanche Photodiode Detectors (APD) at 635 nm and 1000 nm measure the monochromatic emissivity directly. The intensity radiated from the flame is equated using Planck’s black body formula and corrected for sooting flame emissivity. Both the temperature and soot volume fraction are obtained using Dalzell and Sarofim’s values for the optical properties of soot for a specified wavelength. However, one-color pyrometry will also be utilized to validate the temperature calculation without utilizing index of refraction. A slot burner built by SNLL will be employed to produce a two-dimensional flame that is reasonably uniform in the longitudinal direction. Two walls adjacent to the flame stabilize the Acetylene and Air mixtures. The equivalence ratio is manipulated such that the conditions inside the flame could vary from optically thin to optically thick.

3.5.5 Third Level of Validation: JP-8 Pool Fire Flames Experiments

The C-SAFE pool fire facility was designed and constructed during the summer of 2000. It will provide experimental data needed to understand and validate the C-SAFE numerical simulations of accidental fires and explosions.

A 0.30 m-diameter pan is used for burning aviation fuels (jet A and JP-8). The burner is water-cooled and is mounted rigidly approximately 0.50 m above ground level. It was designed to operate in both batch and continuous experiments. Thermocouples are placed at different distances relative to the burner lip, which provide a temperature profile of the fuel in the liquid phase. As is shown in Figure 3.5.8, the facility has an adjustable exhaust system to remove combustion gases and soot from the enclosure. Walls and base made of incombustible board are used to vary the boundary conditions.

Figure 3.5.8. Schematic and photograph of the C-SAFE pool fire facility
Height and flame shapes are obtained using a high-speed video camera (8000 frames per second) and still photographs. Radiative and total heat transfer are measured at different locations by using a gas-purged, water-cooled radiometer and a water-cooled heat flux transducer respectively. Both are Gardon-type gauges and were provided by Vatell Corporation. Heat fluxes to the containers will also be deduced from transient temperature measurements as described below.

3.5.6 Fourth Level of Validation: Experiments on Cylinders Containing Explosives

Experiments involving explosive materials immersed in flames were conducted during the month of October 1999 at Thiokol Propulsion’s Promontory facility, Utah facility. The tests were designed to determine the boundary conditions between the HE materials and the container. The concern was that the bonding of the HE material to the container could not be controlled well and could have a significant impact on the modeling of the heat transfer.

The tests consisted of one inert cook-off using 2.86 kg of barium nitrate and four explosive material cook-offs using 4.3 kg of PBX-9501. The materials were contained in a 4-in diameter x 12-in long steel pipe with threaded end caps. Twelve thermocouples were attached to the outside of the cylinder and the other thermocouples and the pressure transducer were placed inside the cylinder at the interface between the material and the cylinder wall. An IR video camera, four standard video cameras, and a high-speed video camera were used to record the events.

As shown in Figure 3.5.9a, the cylindrical container was supported from an A-frame over chains with six propane burners positioned beneath. Propane burners were used in these preliminary tests instead of a jet fuel pool fire to provide a relatively transparent flame and thus avoid obscuring the camera views of the container during heating and explosion.

![Figure 3.5.9. Photographs of test site (a) and container configuration (b).](image)

The first test used a mixture of barium nitrate/Pentek, which served as an inert formulation. The material did not explode, but burned with smoke exiting the cylinder for almost 10 minutes. Tests 2 and 3 were conducted to the same conditions using PBX-9501. The material was not bonded to the cylinder wall and no effort was made to seal the threads on the end caps. From the video record of the two tests, it was evident that gases were
escaping from the threads in the end caps and igniting just prior to the explosion. In Test 4, an attempt was made to seal the end caps to prevent escape of the hot gases. The explosive material was not bonded to the cylinder wall for this test. For Test 5, the threaded end caps were sealed. The explosive material was also bonded to the cylinder wall using the same binder from the PBX 9501 formulation.

In all four live tests, the material exploded but did not detonate, and unburned material was scattered around the test site. The general appearance of the fragments indicated that the end caps came apart from the cylinder, and the cylinder itself split open for all tests. The cylinder did not split open at the welded seam present in the steel pipes; rather, the location for rupture was always at some other location and often directly opposite of the weld. One of the key physical observations from these tests was the identification of ballooning behavior for the container prior to rupture. The attempts to bond the HE material to the container did not show an effect. As will be shown in the following section a gap appears to form between the HE material and the wall. This has a large impact on the heat transfer and needs to be treated in the models.

A transient heat transfer model was developed to predict the measured temperatures and the heat flux at the interface between the material and the cylinder wall. The model describes that there was a significant resistance to the heat flow at the interface mainly due to the fact that the thermocouples were not welded to the metal case and so they could have been displaced from the surface by the thermal expansion of both steel and PBX 9501. For all 4 tests, the contact resistance lies between 0.05 and 0.09 m²*K/Watt. As is shown in Figure 3.5.10, a 0.07 m²*K/Watt contact resistance is enough to drop the temperature from an average of 900 to 330 K. This contact resistance is equivalent to a 1-3 mm-air gap.

Figure 3.5.10. Comparison of predicted and measured temperatures (Test 5).

Duhamel's superposition technique was used to obtain the heat flux at the interface using the measured temperatures. For these experiments, Duhamel's analysis gives:

\[
q = -K \cdot \frac{\partial \theta(0,t)}{\partial t} = \frac{K}{\sqrt{\pi} \cdot \alpha} \cdot \left[ \frac{T(t) - T_0}{\sqrt{t}} \right] + \frac{1}{2} \int_0^t \frac{T(t) - T(s)}{\sqrt{(t-s)^3}} \cdot ds \tag{1}
\]
The heat flux for each test and the experimental times to ignition were compared to the ignition model of Beckstead (1994) as shown in Figure 3.5.11.

![Comparison of calculated and measured ignition times of HMX (Beckstead, 1994).](image)

Figure 3.5.11. Comparison of calculated and measured ignition times of HMX (Beckstead, 1994).

**References**


4.0 Center-wide Discussion

4.1 ASCI Computer Resources

During the last year, C-SAFE made use of the ASCI Computing Platforms as the various parallel codes developed by the center pursued better scaling results. C-SAFE participants continued production work, developed new parallel simulation capabilities, and conducted scalability studies. This activity is summarized in the following sections. Since much of this work was performed on multiple platforms, details of actual usage are organized by area of activity rather than by platform.

Molecular Dynamics

Members of the High Energy Transformations group investigated physical properties of liquid HMX and viton binder using up to 1024 processors on ASCI Red and ASCI Blue Pacific. This work was based on a parallel molecular dynamics code that was developed from scratch during the past year. The code employs a particle-mesh Ewald technique for efficient calculation of long-range electrostatic interactions, and achieved efficient parallelization through use of a force decomposition algorithm.

Other members of the High Energy Transformations group used ASCI Blue Mountain to advance their study of large molecular systems within a quantum mechanical framework. Several implementation issues had to be addressed in order to achieve their long term objective of considering systems of thousands of molecules. A computational bottleneck encountered at initialization was overcome by employing a combination of shared and distributed memory parallelism, reducing the time for this computationally intense phase of the calculation from several days to several hours. A two-step implementation strategy to parallelize the remainder of the computation was also initiated. In the first step, an LAPACK diagonalization routine was replaced by its parallel counterpart from ScaLAPACK. However this approach does not scale well with the number of molecules, and other approaches that better exploit the inherent sparseness of the problem are being considered. Nonetheless computations that were performed in this first phase will provide useful baseline results against which future calculations can be compared. A nonlinear optimization code has been developed which runs and scales efficiently up to 1024 processors. During the last year we have used 133,400 and 39,800 CPU hours on ASCI Blue Pacific and ASCI Red, respectively.

Structural Dynamics

During the past year, members of the Container Dynamics group continued to improve a parallel version of the Material Point Method. The parallelization of a Uintah PSE implementation was accomplished within the Uintah Computational Framework. The performance of this code was evaluated on up to 1024 processors on ASCI Blue Mountain running a 16 million particle simulation. Efforts to incorporate more realistic physics, and to improve the performance of the code, are underway.

Parallel Mixing and Reaction in Fire Simulations

Radiative heat transfer and mixing dominate the execution time of fire simulations. The Fire Spread team began to address these bottlenecks by developing parallel schemes for
mixing and reaction. They achieved good scalability on up to 128 processors on ASCI Blue Pacific through use of a dynamic tabulation strategy and MPI.

### 4.2 Interactions with National Laboratories

**Sandia-CA and Sandia-NM Fire Group**

A major component of the fire simulation is the description of the soot/smoke formation and oxidation processes and their effect on the radiative field for heat transfer calculations. The thermal radiation from the soot is the dominant hazard posed by fire. Sandia and the University of Utah have organized a soot and radiation working group to explore the state-of-the-art, to divide tasks, to share results between the two ASCI programs, and to leverage resources from non-ASCI funded programs. They formally meet twice a year and invite leading researchers in the field from institutions around the world to participate in the working group. They informally exchange results throughout the year. At this stage in the working group, they have identified key gaps in the technology and divided tasks to fill these gaps. The University of Utah is tasked with bringing together fundamental chemistry-based soot kinetic mechanisms to be applicable across the wide range of temperature and equivalence ratios characteristic of open pool fires. Sandia-CA is tasked with developing new laser-based diagnostic tools to withstand the harsh environments of large pool fires and collect soot data from real flames. Sandia-NM is tasked with designing new experiments for full-scale controlled pool fires, providing measurements that will identify mechanistic behavior and collect data for validation of the numerical simulations. The intent of the soot working group is to provide models for soot formation/destruction and soot radiative properties that will contribute to a validated fire simulation. The soot model will be used by both the university and the national laboratory for their fire simulations and will be available to external researchers interested in soot chemistry and properties.

**LANL T-14 group**

We have continued a collaborative relationship with Dr. Thomas Sewell of Los Alamos National Laboratory. Dr. Sewell has calculated some bulk properties of explosives using model interactions between molecules, while holding rigid the internal molecular degrees of freedom. He is thus able to study systems containing many molecules. In complementary work, we can also calculate some bulk properties using the first-principles density functional theory (DFT) capabilities, which we have developed.

Through our collaborative efforts, we have begun to perform calculations to understand the condensed-phase of HMX. As a preliminary investigation, we have mapped out the energetic profile of HMX as a function of the lattice vectors with constrained lattice angles. The product of our collaboration has resulted in a manuscript describing these preliminary results, which has been accepted for publication. This collaborative effort will help us to make progress for future studies to understand the reaction mechanisms that exist in the condensed-phase of HMX.

**LLNL**

The greatest difficulty in developing a massively parallel electronic structure method, which scales linearly with system size, is to ensure that the algorithms employed are well
conditioned. Our more recent goals are to pursue the necessary requirements for employing
the latest technology for our approach in obtaining a massively parallel electronic-structure
method. To further this goal, we have pursued a collaborative effort with Giulia Galli of
Lawrence Livermore National Laboratory. She is well established in the electronic-structure
community not only as an expert in linear-scaling techniques, but electronic-structure methods
in general. Informal discussions are ongoing, but our hope is that we will develop a strong
collaboration here in order to achieve a high quality product not only for C-SAFE, but for the
electronic-structure community in general.

**Common Component Architecture (CCA) Forum**

The Common Component Architecture (CCA) Forum is a group of researchers from
national labs and academic institutions committed to defining a standard Component
Architecture for High Performance Computing. The C-SAFE project encounters many of the
same challenges as other large computational projects in the National Laboratories, namely
the complexities of large-scale parallel simulation software. A recent CCA publication stated
that "[The CCA] research stems from a growing recognition that the scientific community
needs to better manage the complexity of multidisciplinary simulations and better address
scalable performance issues on parallel and distributed architectures. Driving forces are the
need for fast connections among components that perform numerically intensive work and for
parallel collective interactions among components that use multiple processes or threads." The
University of Utah PSE group has been working to address many of these same issues,
and is an active participant in the CCA forum.

We have helped to define the current CCA component model. We are currently
implementing the next generation Uintah PSE based on this model as expressed through the
evolving CCA specification. When complete, Uintah components will be able to interoperate
with other CCA compliant components from national laboratories and other academic
institutions. We will continue to work with the CCA forum to complete and improve this
model, based on our experience with implementing large-scale parallel simulations within the
Uintah PSE.

**LLNL — Structured AMR Approaches**

Members of the CASC group at LLNL have been developing structured AMR
algorithms. We have had significant input from them in developing the UCF for C-SAFE.
Rich Hornung and Scott Kohn of the SAMRAI group at CASC have visited the University of
Utah several times, and C-SAFE members have spent extended periods of time at LLNL.
Michael Pernice also visited CASC several times during the year to discuss the design on
implicit solvers on grid hierarchies.

Based on knowledge C-SAFE has gained from this collaboration, we have developed a
more narrowly focused AMR framework implemented within the Uintah PSE. We are
continuing to work with LLNL/CASC at the algorithmic level. Michael Pernice from C-SAFE
was working closely with LLNL on implicit time-stepping and nonlinear solution methods, as
well as adaptive methods for radiation transport problems (he has now joined LANL). We
look forward to continuing this collaboration and comparing/contrasting our approaches.
LANL group

Collaboration with LANL personnel has been key in the development of a multi-material CFD solver and in the design for coupling CFD to MPM. Todd Harman (C-SAFE) spent one week at LANL this summer working on this implementation with Kashiwa. We have also collaborated with Todd Williams with respect to micromechanical modeling on HMX. He has exchanged visits with Dan Adams (C-SAFE) between Los Alamos and Utah during Year 2. Other work with LANL involves interactions between Scott Bardenhagen (C-SAFE) and Brackbill (T-3) on new contact algorithms implemented into MPM and between Bardenhagen and Joel Bennett on constitutive modeling of PBXs.

Former C-SAFE students and staff who have accepted employment at the National Laboratories

Phil Sutton, M.S. Computer Science, accepted a permanent/full-time position at Lawrence Livermore National Lab with the Visualization group.
Stephan Domino, Ph.D. Chemical & Fuels Engineering accepted a Post-doctoral Fellowship at Sandia beginning summer 2000.
Mike Pernice, Ph.D., staff member from the Center for High Performance Computing, accepted a full-time position at Los Alamos National Laboratory, beginning September 2000.

1999 summer visitors (1 undergraduate, one Post-doctoral Fellow
2000 summer visitors (3 graduate students, one senior staff member)

C-SAFE — DOE National Laboratory Collaborations Summary

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4.3 Interactions with other ASCI-related Centers

Interactions with other ASAP centers

The CSAFE participation in the ASCI Alliance Centers meeting in Pasadena allowed us to better understand the common issues facing the Alliance centers. The summary presentations about each center gave greater insight into where technical similarities exist, as well as the computational challenges facing each center. Furthermore, the breakout discussion sessions on the topics of multi-physics coupling and validation, scalability, and software integration focused on the basic difficulties involved in each center’s project.

ASCI Frameworks Workshop

Chris Johnson (Utah) and Rick Stevens (Argonne) hosted the ASCI Frameworks Workshop that met Jan 10-12, 2000 at the University of Utah. This combined CS PSE developers and application code developers from both the DOE national labs and the Alliance centers. In addition, the CCA Forum is an ongoing collaboration, and a recent meeting was held at Utah.

Advanced Visualization Technology Center (AVTC)

The DOE Advanced Visualization Technology Center (AVTC) is a partnership of university research groups and DOE laboratories (initially Argonne National Laboratory, Los Alamos National Laboratory, and the University of Utah, but soon to include other researchers as well). The goal of the AVTC is for the research and development of breakthrough technologies that will enable the visualization, storage and manipulation of large-scale datasets produced from multiteraflops-class supercomputers and to help design and implement Data and Visualization Corridors (DVC) within the ASCI National Labs. The DVCs will combine high-performance visual environments with large-scale storage and data management systems to ultimately allow a user to explore, analyze, compare, and share the results of billion-zone computations.

The interaction between AVTC and CSAFE has been focused on how the CSAFE simulation steering environment can be utilized in an immersive setting. Specifically, using the computational steering infrastructure (Uintah PSE) developed for CSAFE, the AVTC is exploring immersive interfaces, such as the Immersive Workbench, for interaction with running simulations. Furthermore, the AVTC is investigating other large-scale visualization methods and techniques that could be used by CSAFE researchers. As the AVTC develops such advanced visualization capabilities, they will be made available for use by CSAFE scientists.

ASCI-PALS at Northern Arizona University

The University of Utah, Lawrence Livermore National Laboratories and IBM have formed a partnership with the Multicultural Engineering Program in the College of Engineering and Technology at Northern Arizona University to establish a pilot pipeline program for Native Americans and other underrepresented minorities that supports education and ultimately career entry related to ASCI programmatic goals. This program leverages off an existing effort at NAU, which currently ranks seventh in Native Americans enrolled in academic institutions. Entitled ASCI-Pathways Leading to Success or ASCI-PALS, it
supports a series of activities, including Academic Excellence workshops, college transition programs, career-readiness seminars, and research experiences for undergraduates.

Two C-SAFE faculty gave seminars on C-SAFE activities and 6 Northern Arizona University students visited the University of Utah in the spring. One ASCI-PALS project directly involves C-SAFE in that it involves the development of a parallel ray tracing code on a small IBM SP system.

In addition to these interactions, the ASCI-PALS program has been leveraged as part of an NSF Educational Innovation grant that began in the Fall 1999. This project involves the development of educational modules to be used in various scientific and engineering courses at the University of Utah, Worcester Polytechnic Institute and Northern Arizona University.