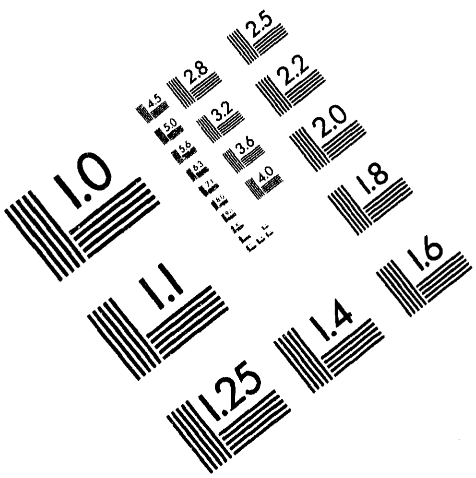

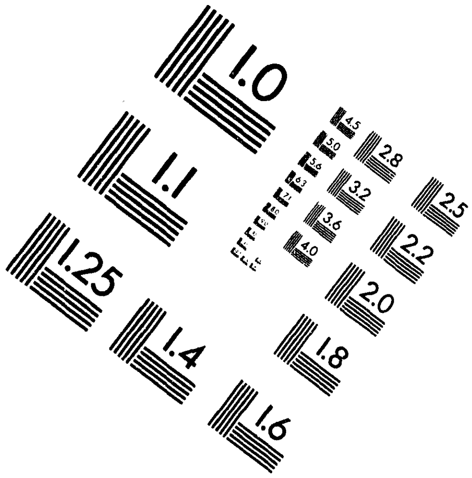

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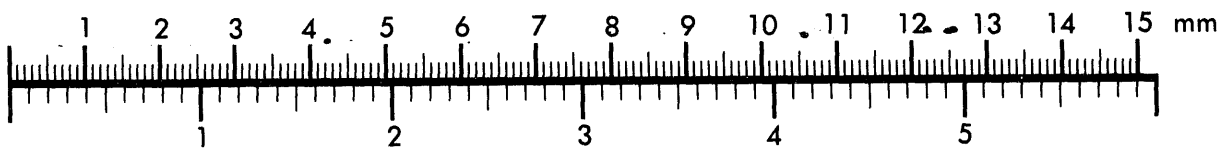

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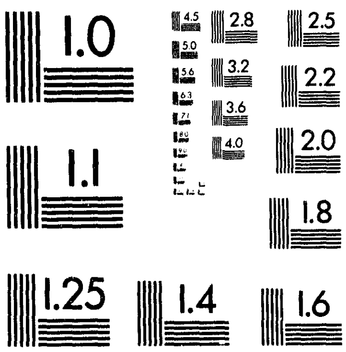

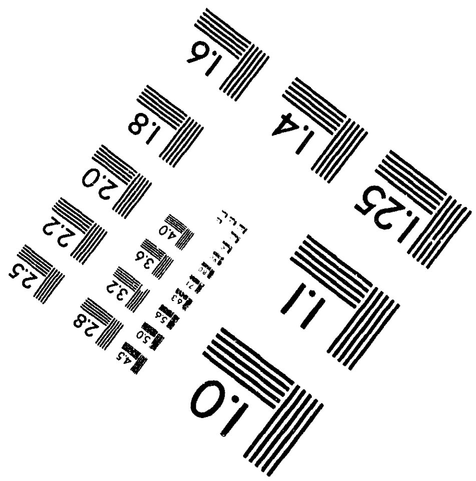

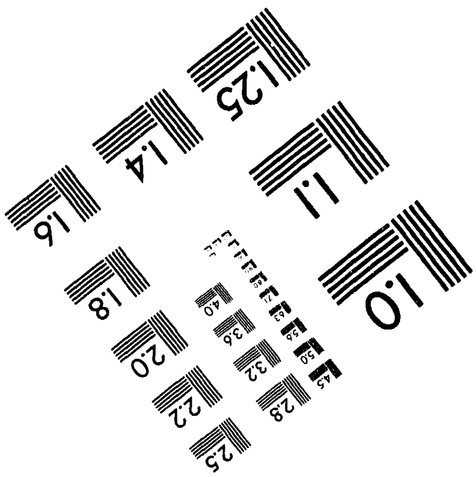

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MULTIDIMENSIONAL THERMAL-CHEMICAL COOKOFF MODELING

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Abstract

Multidimensional thermal/chemical modeling is an essential step in the development of a predictive capability for cookoff of energetic materials in systems subjected to abnormal thermal environments. COYOTE II is a state-of-the-art two- and three-dimensional finite element code for the solution of heat conduction problems including surface-to-surface thermal radiation heat transfer and decomposition chemistry. Multistep finite rate chemistry is incorporated into COYOTE II using an operator-splitting methodology; rate equations are solved element-by-element with a modified matrix-free stiff solver, CHEMEQ. COYOTE II is purposely designed with a user-oriented input structure compatible with the database, the pre-processing mesh generation, and the post-processing tools for data visualization shared with other engineering analysis codes available at Sandia National Laboratories. As demonstrated in a companion paper (See Ref. [1]), decomposition during cookoff in a confined or semi-confined system leads to significant mechanical behavior. Although mechanical effect are not presently considered in COYOTE II, the formalism for including mechanics in multidimensions is under development.

COYOTE II cookoff calculations are presented which demonstrate its capabilities. First, reactive heat transfer is solved simulating a proposed NAWC benchmark cookoff experiment, in which a partially filled container of propellant is heated in an oven. Model predictions for varied heating conditions indicate that the location of ignition occurs at an interior location during slow heating and at a higher heating condition nearer the heated boundaries. As a demonstration of reactive heat transfer in a complex three-dimensional geometry, the thermal field in a rocket motor, a star-grained propellant in a rubber-lined stainless steel casing, is modeled using boundary conditions typifying a fire source.

Research in multidimensional adaptive meshing methods for finite elements is currently an active area in numerical modeling. A two-dimensional adaptive meshing algorithm, based on redistribution of interpolation error, has been studied and implemented in an unstructured thermal/chemical code. An example calculation of the heat transfer in a component containing an energetic material, subjected to a moving heat source, is solved using adaptive meshing. This work is intended to assess adaptive meshing strategies to be implemented in COYOTE II.

1. INTRODUCTION

Prediction of physical phenomena associated with the hazards of cookoff is an important aspect of systems surety analysis. To assess the onset and violence of reaction of energetic materials, it is necessary to address the combined effects of thermal, chemical, and mechanical behavior. Considerations such as geometry, boundary conditions, sliding surfaces/gaps, material strength, and material deformation all combine to make the majority of these problems multidimensional. The finite element method is a well suited numerical approach for analyses of such complex coupled phenomena.

Traditional approaches in finite element analysis have been decoupled, *i.e.* the stress field has been resolved as a separate calculation given a specified thermal state. However, a fully-coupled multidimensional thermal/chemical/mechanical analysis capability is currently in development at Sandia National Laboratories (SNL). This effort builds upon existing capabilities using state-of-the-art software for mesh generation, input/output, results visualization, and shared databases. Translator software is available for transferring and/or interpolating element data from thermal analysis tools to mechanical ones. Many finite element analysis codes at SNL make use of these software modules, avoiding duplication of effort. Future work is aimed at a highly modular code structure which sequentially

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calls various routines in a time step to time step consistent manner, and updates all material and meshing routines with revised properties and boundary conditions resulting from a coupled response. This approach will also accommodate large deformations and material insertion and/or deletion.

In a parallel effort, the coupling of thermal, chemical, and mechanical behavior has been studied in one dimension ([1] and [2]). A constitutive material model describing the elastic-plastic mechanical behavior of reactive energetic material has been developed for integration of thermal/chemical behavior with mechanical response. Insights toward incorporating mechanics with multidimensional heat transfer analysis has been gained from these fundamental studies.

To model cookoff as a multidimensional problem, the COYOTE II thermal analysis program has been selected as the Sandia modeling platform. Like other finite element programs, COYOTE II utilizes the Galerkin finite element method to solve multidimensional problems of heat conduction, radiation, and chemistry. In contrast to the LLNL TOPAZ heat conduction codes ([3] and [4]),* numerical modules and solution routines have been tailored for problems with large numbers of unstructured elements. Furthermore, COYOTE II can solve either two-dimensional or three-dimensional problems with the same software.

Chemistry has been introduced into COYOTE II using an operator splitting methodology. The chemical kinetics assembly routines and a modified stiff solver were ported directly into COYOTE II from the fully coupled one dimensional thermal-chemistry solver, XCHEM-1D [5]. A significant achievement in modeling large thermal radiation problems has also been included with the incorporation of CHAPARRAL [6] into COYOTE II. CHAPARRAL is a program which solves enclosure radiation heat transfer problems. In the following sections, an overview of COYOTE II is provided and demonstrative examples are shown which typify heat transfer with chemistry.

Based on prior studies in one dimension using XCHEM-1D, substantial improvements in computational efficiency and numerical accuracy can be gained using adaptive meshing methods. Unfortunately, the methods have not been completely developed in multidimensions, and have not been implemented in automatic finite element meshing routines. In this paper, a two-dimensional adaptive method based on refinement and redistribution is introduced, which has been implemented in a multidimensional extension of XCHEM. Details of this method are discussed and an example calculation of heat transfer in a cylindrical confinement containing energetic material is presented.

2. DESCRIPTION OF COYOTE II

COYOTE II† [7] is a two-dimensional and three-dimensional finite element computer program designed for multidimensional analysis of nonlinear heat conduction problems. In addition to solving standard thermal diffusion problems, COYOTE II includes the effect of phase change/ablation, condensed phase chemistry and surface-to-surface radiation. Material addition or deletion is included through the use of the finite element "birth/death" capability. Internal boundary effects, like gaps at material interfaces, are incorporated as "master" and "slave" sides of contact slide surfaces. Material properties can be temperature and/or species dependent and either isotropic or orthotropic. A wide variety of boundary conditions are supported in COYOTE II and pre- and post-processing file formats are used which permit integration with existing meshing and graphics visualization programs.

Heat transfer in reactive energetic materials is modeled by the numerical solution of the generalized heat conduction equation given as:

$$\rho c \frac{\partial T}{\partial t} = \frac{\partial}{\partial x_i}\left(k_{ij} \frac{\partial T}{\partial x_i} \right) + Q(T, N) \tag{1}$$

where ρ is material density, c is the specific heat, k_{ij} is the thermal conductivity tensor, Q is the volumetric heat source, t is time, x_i is the spatial coordinate, T is temperature, and N is the species solution vector. Additional differential equations, given by Equation (2), defines a set of reaction rate equations, neglecting advection and diffusion:

$$\frac{dN}{dt} = R(T, N) \ . \tag{2}$$

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* TOPAZ is a LLNL finite element heat conduction program; chemistry is currently only modeled in the 2D version of this code.

† The Roman numeral "II" indicates it is a second generation code introducing a three dimensional simulation capability.

In COYOTE II, prior to the application of the Galerkin finite element method [8], the physical domain is broken into discrete elements. COYOTE II uses an extensive element library including triangles, quadrilaterals, hexahedrons, prisms, tetrahedrons, bars, and shells. Several general purpose mesh generation programs for finite element analysis at SNL provide mesh connectivity, material pointers and boundary condition specifications compatible with COYOTE II. For example, FASTQ [9] is an interactive two-dimensional automatic mesh generation program which includes a paving algorithm. Paving provides an optimized element mesh by dividing the domain into subregions, meshing the subregions and then smoothly joining them. Three-dimensional finite element meshes are created by mapping (*i.e.* translating or rotating surfaces) using the program GEN3D [10]. Other meshing programs include GJOIN [11] which merges separate meshed regions into a single domain, and GREPOS [12] which repositions a finite element mesh by translating, rotating, or exploding two-dimensional or three-dimensional meshes.

Having established a discrete element mesh, a set of finite element basis functions is defined for each element which are a functional representation of the independent variables. COYOTE II allows linear or quadratic basis functions. Equation (1) is approximated as a residual, *i.e.* all terms are collected on one side, multiplied by an appropriate weighting basis function, and integrated over the entire domain. The resulting Galerkin approximation yields a set of nonlinear equations given in matrix form as:

$$M(T)\dot{T} + K(T)T = F_Q(T, N) \tag{3}$$

where $M(T)$ is the mass matrix, $K(T)$ is the stiffness matrix and F_Q is the forcing function vector. Information concerning chemistry variables are contained in the forcing vector *at the Gauss quadrature integration points.* Rate equations for the chemistry variables are defined at these locations (in contrast to nodes) to avoid ambiguities associated with elements adjoining material interfaces.

The time derivatives in Equations (3) are discretized in COYOTE II using one of several user-specified numerical options including first order backward Euler, a second order trapezoidal method, or a first order explicit method. The resulting discretized equations are then linearized using a Picard method or a Newton method producing a set of linear first order equations of the form, $Ax = b$, where A is the Jacobian matrix, x is the unknown vector representing nodal temperature, and b is the residual vector.

Chemical reaction is introduced through the forcing vector. An operator-splitting methodology is used which first advances the temperature in time with all chemistry variables remaining unchanged. Then, the second operator fixes the temperature field and the species rate equations are advanced in time using a modification of CHEMEQ [13], which is a matrix-free hybrid stiff solver. Following the second operator, the endothermic and/or exothermic heat sources are re-evaluated as input for the next time step. Adaptive time stepping is based on time scales relevant for both the heat transfer and the chemistry [14]. Because the solver for the chemistry does not have a large startup overhead, the system of reaction equations are efficiently solved element-by-element at Gauss integration points.

A unique capability of COYOTE II is the means to determine view factors for large problems involving surface-to-surface radiation heat transfer. Thermal radiation from a fire source is an example of an abnormal thermal environment for surety and weapon safety analyses. The code, CHAPARRAL [6], uses either a hemicube algorithm [15] or FACET [16], which has been made into a subroutine library, to compute view factors. For large three-dimensional problems, the hemicube algorithm has been shown to be a computationally efficient method for radiation heat transfer analysis.

Although the assembly of the set of linear equations is computationally expensive, a large fraction of computation time is spent in obtaining the solution of $Ax = b$. The inverse of the Jacobian matrix directly determines the solution vector, and thus direct matrix methods have been traditionally used in finite element analysis. Unfortunately, direct methods become prohibitively expensive in terms of both cpu time and computer memory, especially in multi-dimensional simulations requiring many elements. In COYOTE II, the direct matrix solvers have been replaced by iterative methods using the conjugate gradient approach [17]. This substantially improves the performance of the code with no compromise in accuracy. Conjugate gradient algorithms are among the most robust and fastest methods currently available. Further, these methods require far less memory than direct methods. Because finite element simulations in three-dimensional routinely require 50,000 or more elements, this is a critical feature.

3. COYOTE II RESULTS

Three demonstrative COYOTE II calculations for two different geometries are presented in this work. The first example is a simulation of an experiment proposed by NAWC as a benchmark for cookoff analyses. The experiment, shown in Figure 1, consists of an open aluminum cylinder filled with a propellant material. A fan and external heaters impose both convective and radiative boundary conditions on the problem. Convective and radiative boundary condition parameters used in the COYOTE II calculation are given in Figure 1. For simplicity, a single step global reaction mechanism of the type $A \rightarrow B$ was used. Chemical rate parameters are also given in Figure 1. Well-known thermal properties are used for the aluminum. Two calculation were performed using this geometry. In the first calculation, a uniform ambient temperature of 455K was imposed on the three-dimensional geometry. This problem was chosen to examine two-dimensional (symmetrical) heating. In the second calculation, the reference temperature for the boundary conditions was varied 145 K over a 90° azimuthal direction (from 455 to 600 K), representative of a local hot spot in the oven. This calculation is fully three-dimensional. Figure 2A shows the mesh used in these calculations. Note that all hexagonal elements are used, even in the center of the cylinder; thus, no degenerate elements are present.

Figures 2B-2D and Figure 3 depict the results of these two simulations. Figure 2B displays the symmetric boundary condition calculation at 215 seconds. The aluminum shell is the hottest part of the component and uniform in temperature because of its high thermal conductivity, and chemistry is not yet significant. In Figure 2C, at 2649 seconds, the propellant begins to react, and propellant material at the top and bottom supplements the heat transfer with chemical effects. Finally, at about 2746 seconds, in Figure 2D, ignition occurs near the corners at the bottom of the propellant. This intermediate cookoff event results in an ignition point located in the interior of the propellant near the cup edge. The inertial confinement provided by the hot propellant is likely to cause a violent response.

Figure 3A clearly depicts the three-dimensional character of the second calculation using this same geometry at a time of 985 seconds with boundary conditions representative of nonuniform heating. Approximately 90 seconds later, at a time of 1076 seconds, ignition occurs at the top of the unconfined propellant surface. This event is likely to be less violent and surface burning is expected to occur. Thus, small changes in boundary conditions can lead to a dramatically different response of the system.

The second geometry simulated with COYOTE II is shown in Figure 4. This geometry approximates a rocket motor with a propellant containing a star-shaped bore. The outer layer represents a steel casing with an intermediate rubber liner between the steel and propellant. The geometry contains approximately 70,000 hexagonal finite elements, which is a typical number of elements for three-dimensional problems. The propellant is assumed to be a mix of nitrocellulose, HMX, and aluminum. A three-step reaction is used for the HMX, and a two-step reaction is used for the nitrocellulose, requiring five species as dependent species variables. The reaction mechanism and physi-

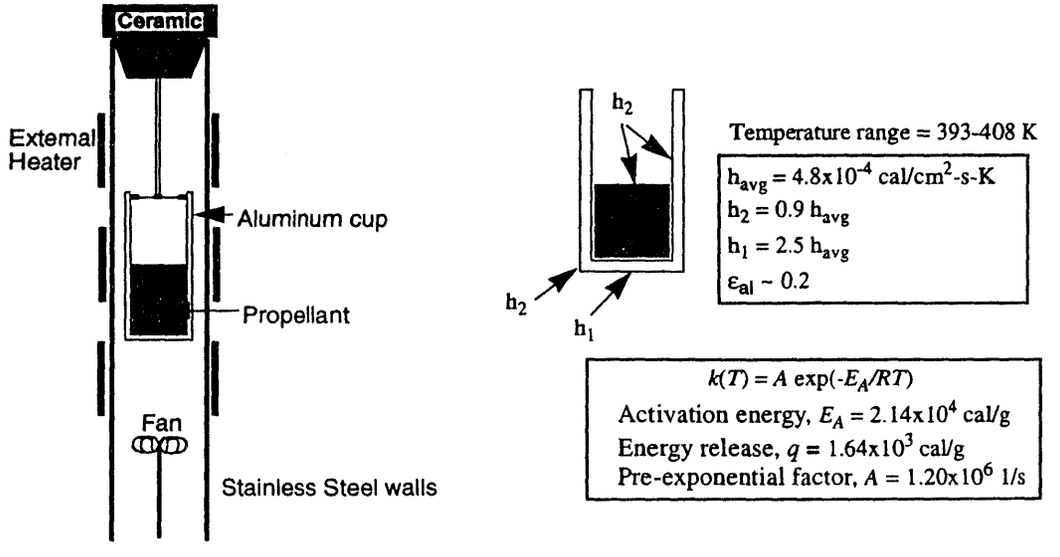

Figure 1. Geometry and boundary conditions for a demonstrative COYOTE II simulation.

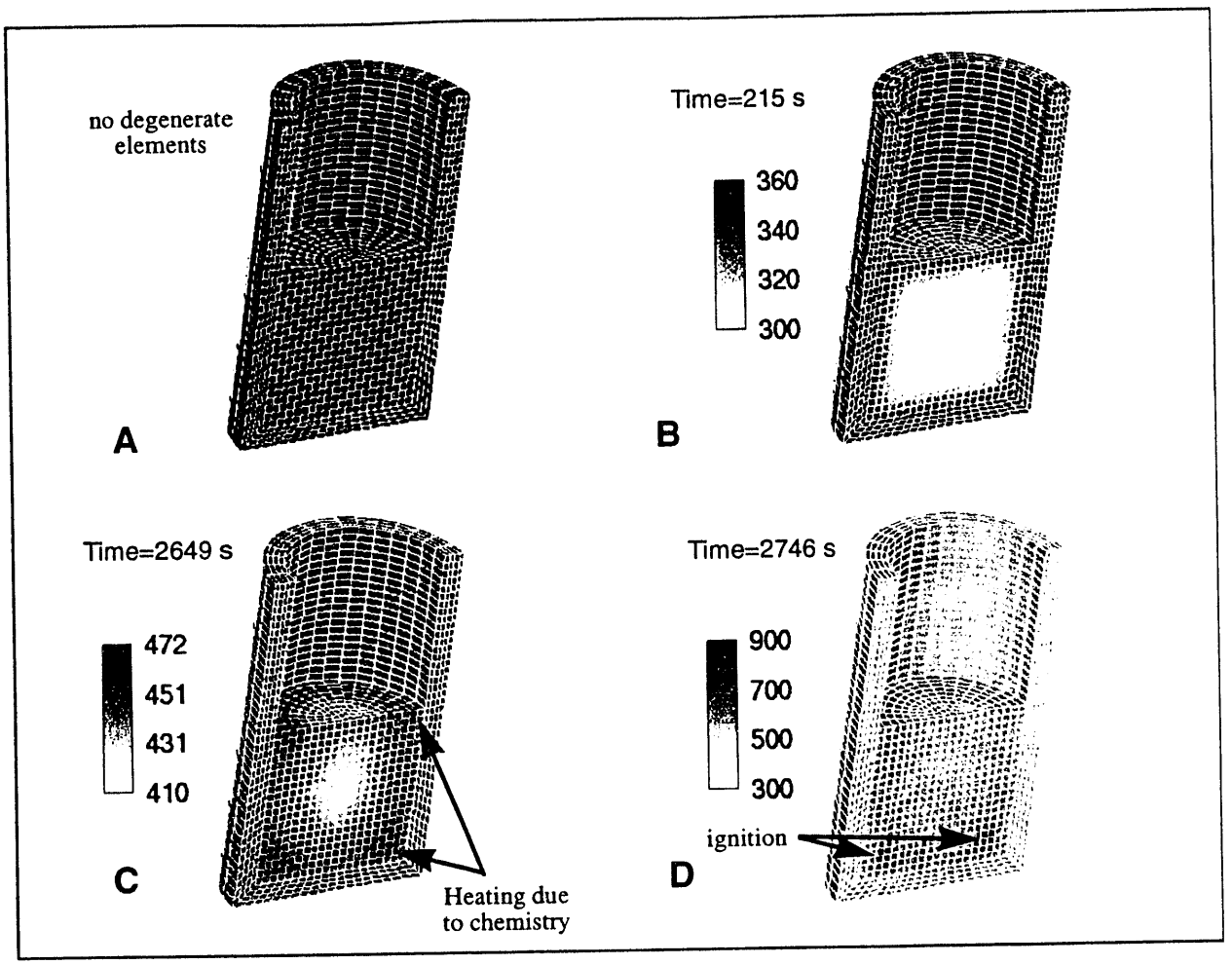

Figure 2. COYOTE II calculation on a propellant (EM) with open case using 2D boundary conditions.

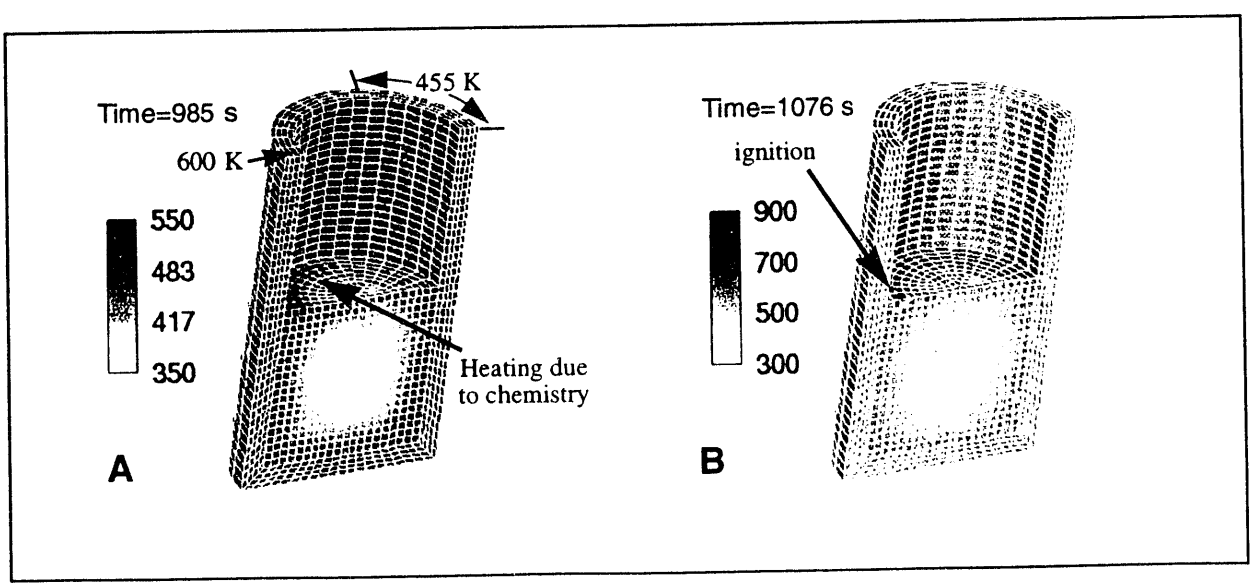

Figure 3. COYOTE II calculation on a propellant with open case using 3D boundary conditions.

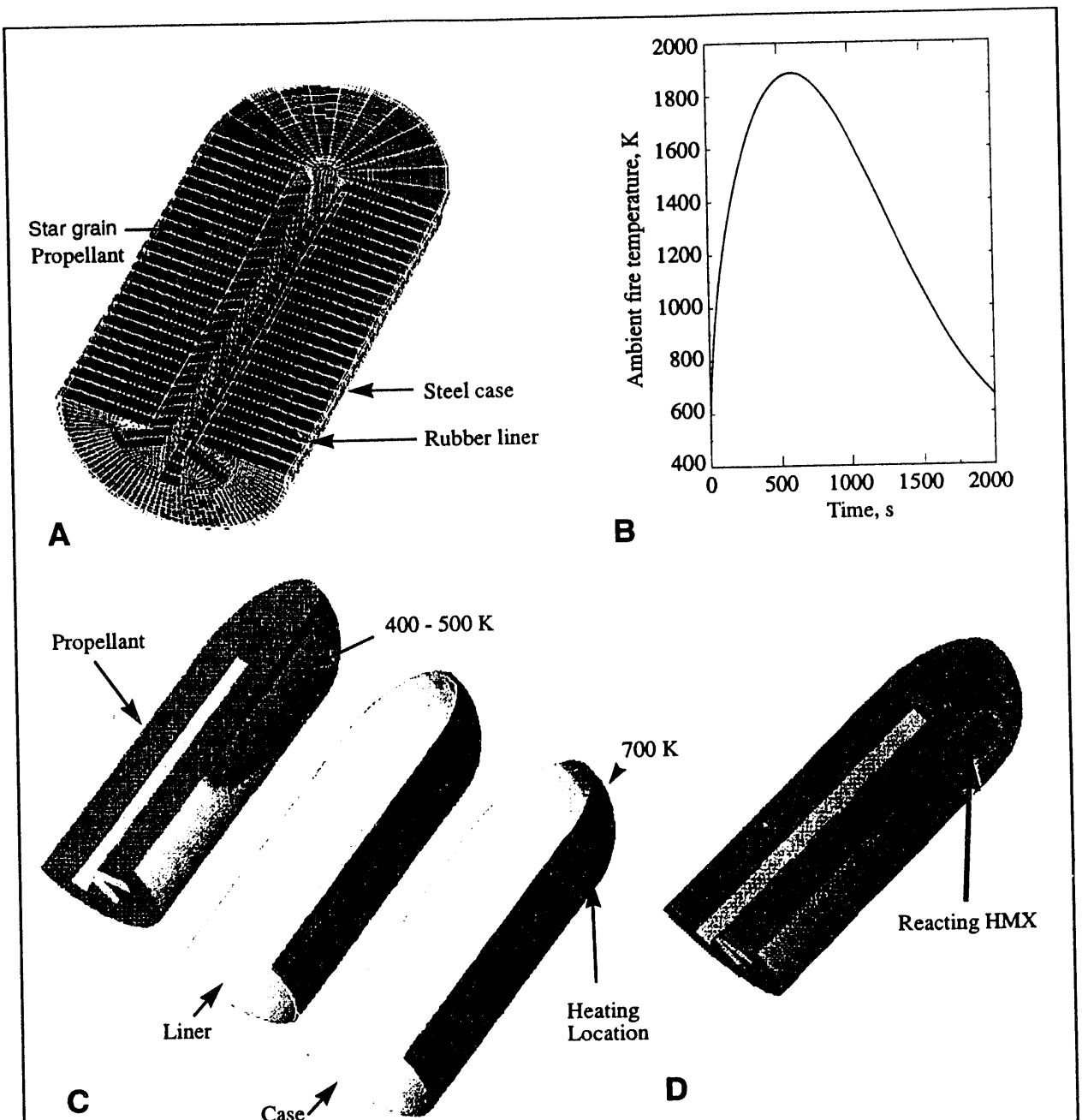

Figure 4. COYOTE II results for a star-shaped rocket motor showing A) the 3D geometry B) the boundary temperature C) the temperature contours at 815 seconds and D) the HMX species contour at 815 seconds.

cal properties for these ingredients can be found in Reference [5]. The steel casing was subjected to a transient convective boundary condition approximating exposure to a hydrocarbon fuel fire for 15 minutes. The calculation was taken out to 14 minutes. This calculation demonstrates that realistic geometries, boundary conditions, and reactive and/or inert materials can be modeled with COYOTE II.

Figures 2 and 3 are depicted in gray tones (for publication purposes); however, normal visualization and interpretation of results are usually rendered in full color. The numerical values in Figures 3 and 4 correspond to a level of gray tone contrast, and as such are qualitative only, *e.g.*, the upper temperature in Figure 2D was 1670 K, and in Figure 3B it was 2303 K.

4. DESCRIPTION OF ADAPTIVE REFINEMENT

Irrespective of the solution method, multidimensional simulations require one to make decisions on the physical mesh dimension as part of the discretization approximation. Often, these decisions are constrained by available computer memory and/or computational time, and the resulting mesh may be coarser than truly desired. These coarse calculations can be inaccurate because the physics occurring on smaller spatial scales may not be effectively captured. Numerical solutions involving chemistry are particularly vulnerable to insufficient resolution problems because chemistry and conduction heat transfer exhibit both time and length scales greatly disparate from one another. In one-dimensional numerical simulation using XCHEM-1D [5], the spatial resolution problem is overcome using an adaptive refinement gridding technique. Multidimensional simulations, with their large number of elements requiring more cpu, can potentially benefit from adaptive meshing.

The development of adaptive refinement methods for multidimensional finite element simulation is currently an active research area. A variety of strategies have been studied [18] including techniques based on spatial mesh (h-refinement), mesh motion (r-refinement) and spatial order enrichment (p-refinement). We present a two-dimensional h-refinement technique which extends the work of Benner and Baer [19] as implemented in the XCHEM-1D method-of-lines solver. This approach has proven to be very robust because grid entanglement problems cannot occur.

In this h-refinement adaptive method, a feedback scheme is formed whereby the previous timestep's solution on a coarse discretization produces posteriori estimates of local error of piecewise bilinear interpolation. These errors control mesh refinement such that finer meshes are created in regions where a prescribed error tolerance is exceeded. Similarly, when these error are smaller than an upper tolerance, elements are recombined to eliminate fine mesh in regions not requiring them.

Figure 4 shows a representation of multilevel adaptivity. Initially, a coarse set of quadrilateral elements is formed on a base level for refinement (level 0). Each element is evaluated and, if refinement is necessary, the element is bisectioned into four sub-elements which, in turn, are re-evaluated for possible additional bisection. At the first refinement level the base element is split into four isomorphic sub-elements. At level 2 refinement, the quadrilateral sub-element corresponds to a level 1 sub-element bisectioned into four smaller sub-elements. Refinement continues until the local error meets the required tolerance or a specified maximum level of refinement has been reached. To preserve smoothness in approximating the dependent variables, an addition constraint is imposed such that neighboring elements do not differ by more than one level. Because all elements are isomorphic subsets, either added or deleted, no degenerate elements appear and mesh entanglement is eliminated.

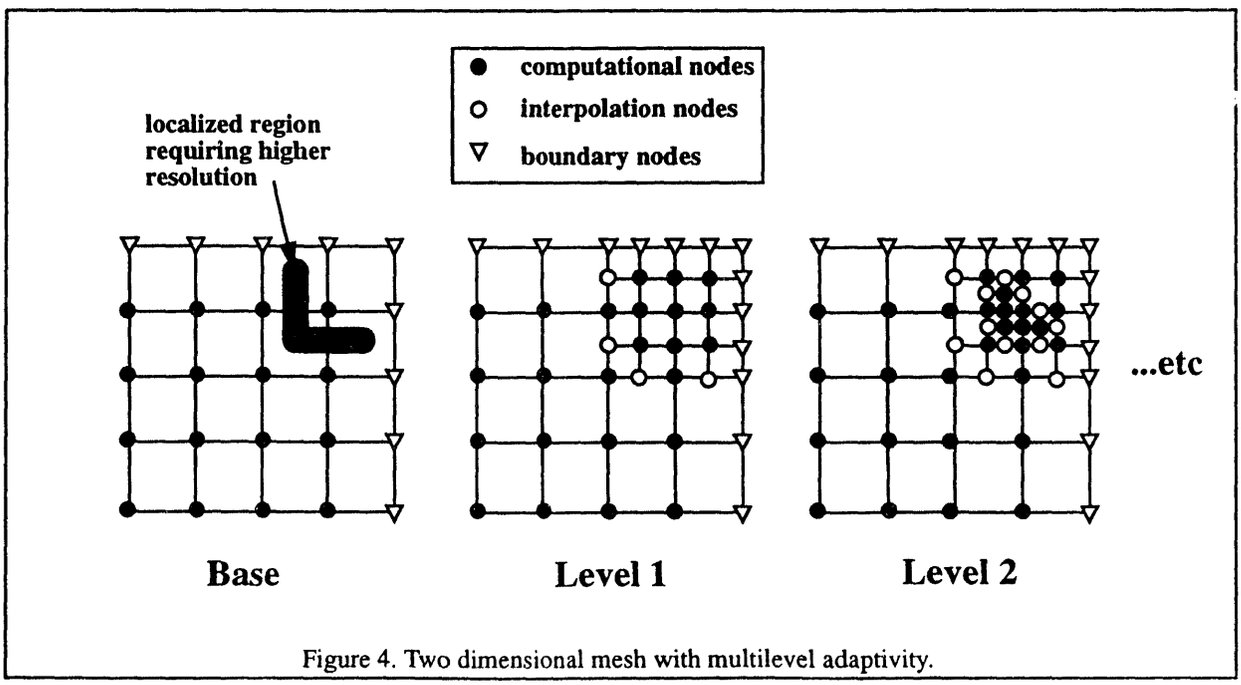

Figure 4. Two dimensional mesh with multilevel adaptivity.

Following a refinement step, the unstructured finite element mesh is reassembled to minimize the bandwidth of the connectivity matrix. Additional nodes are interpolated and sorted from nodes created at boundaries. At the boundary points, boundary condition flags are set and internal constraints are introduced at interpolation nodes.

5. 2D ADAPTIVE MESH REFINEMENT RESULTS

The aforementioned two-dimensional adaptive algorithm is incorporated in a two-dimensional extension of XCHEM-1D [5]. This research code presently utilizes only quadrilateral meshing in a single region of space. However, it does employ an unstructured mesh typical of finite element methods. Among the important issues examined in this study include: the efficient assembly of nodal fields and the incorporation of boundary and internal constraints at interpolations points.

As an example calculation with multidimensional adaptivity, the thermal/chemical field in a cylindrical geometry is predicted. Dynamic remeshing is demonstrated as a localized moving heat source of a finite extent at 700 K provides a variable boundary condition on the outer surface of a cylindrical aluminum confinement. At all other boundaries, a zero flux condition is imposed. The cylinder has an outer radius of 2 cm and an inner radius of 1 cm. The heat source moves toward a region containing a fixed, embedded circular region containing RDX. The calculation is carried out to 108 seconds, at which time the RDX ignites.

Figure 5 displays three time planes as the heat source moves towards the energetic material. At each time plane, the isotherms are shown in gray scale and the corresponding mesh is displayed as a mirrored image of the component below the contours of the thermal field. The temperature scale between the second and third time plane applies to all three plots. The mesh density follows the moving heat source as anticipated. Figure 6 shows details of the adaptive refinement in the region surrounding the RDX once heat transfer in this area becomes significant. Eight levels of refinement are used in this simulation and, as the reaction of RDX begins to occur, dynamic meshing follows the evolution of the decomposition.

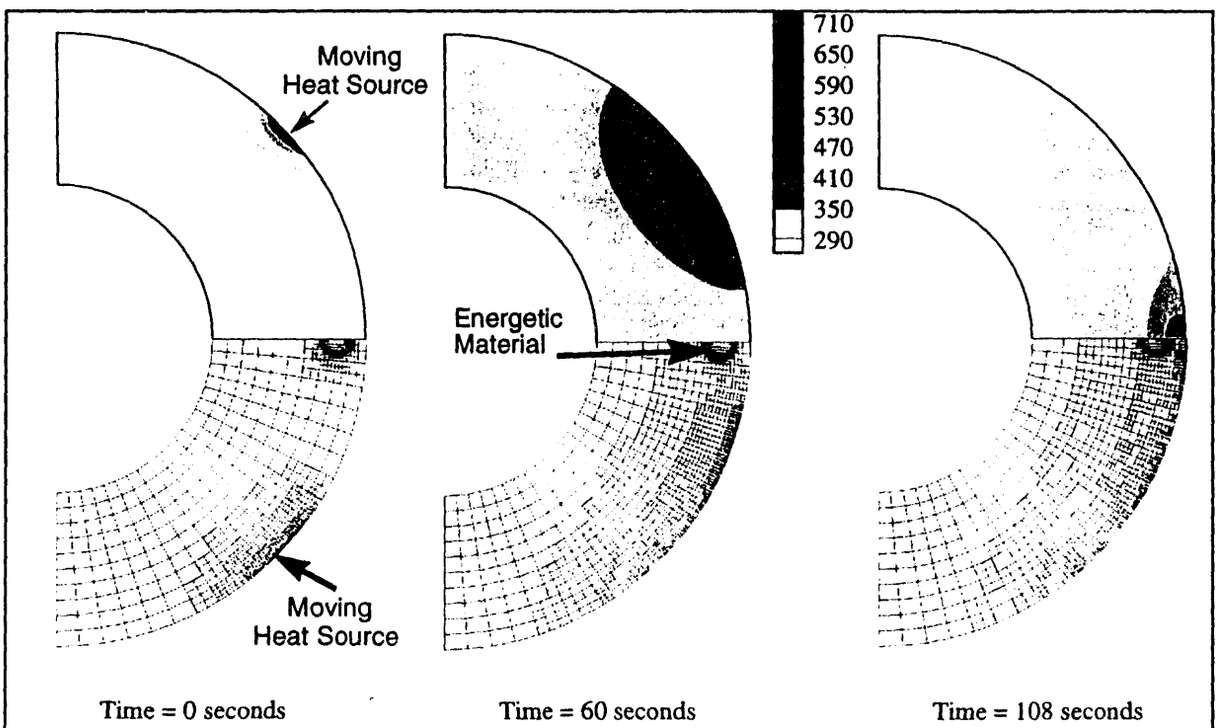

Figure 5. XCHEM-2D temperature contours with corresponding time-dependent refined meshes at three times for a moving heat source on a cylinder.

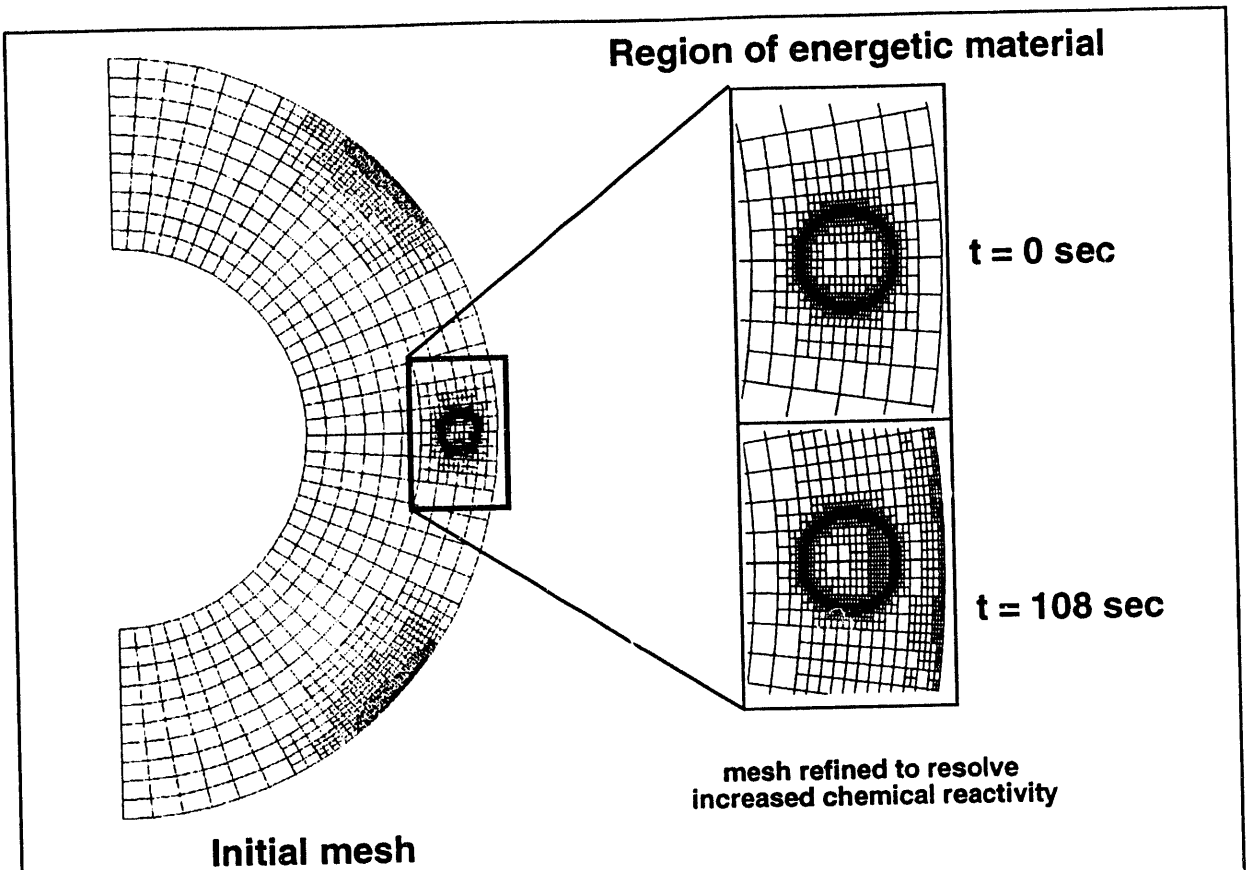

Figure 6. Magnification of the region of energetic material displaying detailed refinement during reaction.

This simulation shows that adaptive meshing can be incorporated into finite element analyses to follow multiple dynamic physical events while avoiding grid entanglement difficulties. Future work will extend this adaptive work to be incorporated into COYOTE II.

6. SUMMARY AND CONCLUSIONS

This paper describes COYOTE II, a thermal conduction heat transfer code which includes thermal radiation heat transfer and condensed phase chemistry. A single version of COYOTE II is applicable to either two-dimensional or three-dimensional geometries. A number of features make COYOTE II a unique tool in performing multi-dimensional cookoff including: a conjugate gradient iterative solver capable of solving problems with a large number of elements; dynamic time stepping; the hemicube view factor routines capable of treating a large number of surfaces; a stiff solver applied to the chemistry equations; and the availability of a large number of user-friendly pre- and post-processing support software.

Two example three-dimensional problems are solved using COYOTE II. The first problem is an unconfined propellant in an aluminum container; the second simulated a rocket motor with a star-grained propellant. The first calculation used 5560 elements and required approximately 2100 seconds of cpu time on a Sun SPARCstation 10 workstation. The rocket motor contained approximately 70,000 elements and demonstrates that realistic geometries, boundary conditions, and any number of reactive material with arbitrary chemical kinetics can be modeled with COYOTE II.

A two-dimensional adaptive meshing algorithm is described. This approach produces accurate results with a minimum of elements, which in turn reduces computer memory and cpu. A demonstration calculation of a moving heat source on a cylindrical component containing reactive material is presented which outlined the salient advantages of adaptive meshing, including accommodating unstructured element analysis free from entanglement prob-

lems. Further, it is noted that this approach is virtually automatic, requiring very little input from the user, and no user interaction during the actual calculation.

In a companion paper [20], a one-dimensional code, TREX, which fully couples thermal, chemical, and mechanical phenomena is described. Future work includes placing this constitutive model in three-dimensional mechanics codes to model reacting materials, and then coupling COYOTE II to mechanics codes. Future studies associated with the adaptive meshing will examine the incorporation of meshes with differing element types (*i.e.* triangular elements mixed with quadrilateral elements), adaptivity in multi-regions, and adaptivity in three-dimensional space.

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