Development and Validation of a Chemical Reaction Solver coupled to the FLASH code for combustion applications

**N. Attal ^a, P. Ramaprabhu ^{a*}, J. Hossain ^b, V. Karkhanis ^a, M. Uddin ^a, J. R. Gord ^c
and S. Roy ^d**

^a Department of Mechanical Engineering and Engineering Sciences, University of North
Carolina at Charlotte, NC-28223-0001, USA

^b Department of Mechanical and Aerospace Engineering, University of Central Florida,
Orlando, FL-32816, USA

^c Air Force Research Laboratory, Wright-Patterson Air Force Base, OH-45433, USA

^d Spectral Energies LLC, 5100 Springfield Street, Suite 301, Dayton, OH-45431, USA

*Corresponding author. Phone: +1 704 687 7557

E-mail address: pramapra@uncc.edu

ABSTRACT

We report on modifications to the widely used astrophysical code, FLASH (Fryxell, Olson et al. 2000) that enable accurate simulations of chemically reacting flows with heat addition. The enhancements to FLASH include the implementation of extensive Hydrogen-air and Methane-air chemistry through multiple, detailed mechanisms (Smooke 1991; Katta and Roquemore 1995; Mueller, Kim et al. 1999; Billet 2005), accomplished by building on the existing infrastructure of nuclear reaction network solvers. The chemical reaction network is represented as a system of coupled ODEs, that are then solved either through the Kaps-Rentrop (Rosenbrok) method (Kaps and Rentrop 1979) or the Bader-Deuflhard method (Bader and

Deuflhard 1983), supplemented by a sparse matrix package for solving linear systems of equations. Furthermore, an existing gamma-law equation of state solver was modified to describe multiple species, each with temperature-dependent properties necessary for realistic simulations of combustion applications. The calculation of temperature-dependent transport properties of constituent species is accomplished through a comprehensive expansion of the materials database. We take advantage of the capability in FLASH to handle the diffusion of heat, mass, and momentum either through an update of the fluxes of each quantity across cell faces, or by directly solving a diffusion equation for each, and the relative merits of each approach for reacting flows are discussed. The capabilities of the modified tool are extensive and in some instances unique, and are documented in detail, along with numerical properties. We also present results from validation of the above capabilities through comparison with analytical solutions, and published numerical and experimental data of chemically reacting flows. Our validation cases include comparison of temporal evolution of species and temperature in a well stirred reactor, comparison of adiabatic flame temperature data, advection of reacting and non-reacting 1D fronts, 2D laminar premixed Methane-Air flame in a Bunsen burner configuration, shock-driven combustion of an initially circular hydrogen bubble, and a reacting Richtmyer-Meshkov instability not studied previously.

Keywords: Implicit Large Eddy Simulations, code development and validation, Shock-flame interaction, reacting Richtmyer-Meshkov instability, laminar flame.

1. INTRODUCTION

The most common source of energy generation remains the combustion of hydrocarbon-based fuels. As the demand for high efficiency combustion processes coupled with reduced pollutants grows, the design complexity of such devices increases. High-fidelity numerical simulations offer a reliable, non-invasive approach for designing and optimizing such devices involving complex flow patterns and multi-physics phenomena. While the advent of massively parallel computing has helped realize large-scale simulations of high Reynolds number flows in idealized geometries, several challenges persist in calculating reacting flows in realistic settings. For instance, Large Eddy Simulations require careful tuning of inherent model constants and parameters based on the type of flow studied, and thus need to be calibrated using experimental results or computationally expensive Direct Numerical Simulations (DNS). An alternative that has garnered attention recently is the so-called Implicit Large Eddy Simulations (ILES) approach, where scalar and energy dissipation is modeled through the cell-averaged numerical mixing resulting from a finite grid. Such methods are thus ‘model-free’, but their suitability to reacting flows where temperature-dependent transport properties often determine details of the flame properties, is yet to be explored in detail. In this article, we have described the development and integration of a chemical reaction suite with the massively parallel FLASH[1] code, originally developed by the FLASH center for Computational Sciences at the University of Chicago. FLASH is a hydrodynamic solver based on the Piecewise Parabolic Method[8] (PPM), and is capable of performing both as a Direct Numerical Simulation (DNS) and an Implicit Large Eddy Simulation (ILES) solver. Thus, the modified code allows a direct comparison of the accuracy and performance of the ILES paradigm with the DNS approach when applied to reacting flows with heat addition.

In the DNS limit, diffusive transport is computed exactly in FLASH, and is used to update momentum and energy fluxes at every timestep. In contrast, the ILES mode takes advantage of

numerical dissipation in so-called ‘under resolved’ simulations to perform low pass filtering of subgrid modes. Thus, the elimination of an explicit subgrid model makes such schemes attractive for simulation of turbulent flows, as they occur in realistic engineering applications. ILES schemes have been successfully applied to a wide range of challenging problems, including turbulent free shear flow [9], homogeneous compressible turbulence [10] and decaying supersonic turbulence [11]. Our objectives in this work are threefold:

- (i) to extend the capabilities of FLASH to simulate a wide array of chemically reacting flows with heat addition, relevant to combustion applications,
- (ii) to perform extensive validation of the modifications to FLASH, and to present these results as a resource for future validation, model refinement and benchmarking efforts, and
- (iii) where applicable, to compare the performance of ILES and DNS numerical approaches to chemically reacting flows.

The extension to FLASH will render it capable of accurately describing (a) H_2 -Air and CH_4 -Air chemistry, (b) temperature-dependent transport properties relevant to the heat addition process in a reactive flow, (c) multi-species equation of state (EOS) with temperature-dependent adiabatic indices, and (d) diffusion solvers that allow for a range of Lewis/Prandtl/Schmidt numbers. These capabilities are summarized in table 1.

When combined with the existing capabilities present in FLASH, the modified solver is expected to find use in investigating a wide range of combustion-related phenomena. The ability to describe reactive flows enables the investigation of the role of chemical timescales, effect of heat addition and flame-induced density variations on a wide variety of laminar and turbulent flows. The existing shock capturing ability in FLASH when combined with the chemical reaction solver will enable simulations of phenomena such as detonation, deflagration, and transition

between the two states, which are important to industrial safety as well as the design of scramjet and pulse detonation engines. FLASH is also equipped with Adaptive Mesh Refinement (AMR), a feature that enables selective refinement of the computational grid, conditioned on local gradients of the primary variables. The data from such simulations can be used for refining and validating turbulent combustion models. Finally, the availability of multiple advection schemes, such as PPM, MUSCL-Hancock and WENO5 within FLASH can be exploited to study their applicability and performance in chemically reacting flow problems.

In this article, we have validated the fully compressible flow code FLASH using several chemically reacting, compressible flow problems, including some cases in which shock waves are present. The rest of this document is organized as follows: In § 2, a brief description of the original FLASH code and its capabilities is presented. In § 3, we describe in detail the modifications we have made to FLASH that enable reactive flow computations, including H_2 -Air chemistry described by three distinct mechanisms, CH_4 -Air chemistry, a comprehensive expansion of the materials database to include temperature-dependent transport properties, an expansion of the EOS suite to include temperature-dependent adiabatic indices for multicomponent mixtures, and extension of the implicit diffusion solver to handle viscous and mass diffusion. § 4 describes in detail, the suite of validation cases performed including 1D advection of reacting and non-reacting fronts, CH_4 -Air jet flames, shock-driven combustion of a Hydrogen bubble and a reacting H_2 - O_2 , single mode, Richtmyer-Meshkov instability (RMI). Some conclusions and opportunities for future work are presented in § 5.

2. FLASH CODE DESCRIPTION

FLASH was developed by the FLASH center for computational sciences at the University of Chicago to study astrophysical flow problems, and is an open-source, multi-physics simulation software. FLASH is highly parallel, designed for high-performance CFD

applications and uses the Message Passing Interface (MPI) protocol for efficient inter-processor communication.

FLASH has a directory structure organization with component groups called ‘units’, which are organized according to their functionality. Adaptive Mesh Refinement (AMR) is implemented through PARAMESH[12], and is dynamically achieved by conditioning the mesh refinement on second order derivatives of the condition variables. In the following sections, we introduce some of the existing units in FLASH, relevant to compressible flow simulations. These units are *Hydro*, *EOS*, *Burn* and *Diffuse*. For a detailed description of the architecture of FLASH and the numerical methods therein, we refer the reader to [13].

2.1 Hydro Unit:

The Hydro unit in FLASH solves the compressible Euler equations (1) – (3), written in conservative form[13]:

$$\frac{d\rho}{dt} + \nabla \cdot (\rho V) = 0, \quad (1)$$

$$\frac{d\rho V}{dt} + \nabla \cdot (\rho V V) + \nabla P = \rho g, \quad (2)$$

$$\frac{d\rho E}{dt} + \nabla \cdot [(\rho E + P)V] = \rho V \cdot g \quad (3)$$

where g , ρ , V , P and E are the gravitational acceleration, density, velocity, pressure and the total energy per unit mass, respectively. The internal energy (e) is obtained separately from equation (4),

$$\frac{d\rho e}{dt} + \nabla \cdot [(\rho e + P)V] - V \cdot \nabla P = 0. \quad (4)$$

Finally, pressure is updated using an ideal gas equation of state (EOS) described in §2.2. In the case of a multi-species simulation, an advection equation for each species i in the system is solved,

$$\frac{\partial \rho Y_i}{\partial t} + \nabla \cdot (\rho Y_i V) = 0 \quad (5)$$

where Y_i is the mass fraction of the i^{th} species.

To solve the Euler equations, FLASH is equipped with both a directionally split and unsplit algorithm for two types of meshes, a uniform grid and an adaptively refined grid in Cartesian, cylindrical, spherical and polar coordinates. The default numerical method used in FLASH is the directionally split Piecewise Parabolic Method (PPM[8]), an extension to second order of the first order Godunov[14] method.

2.2 EOS Unit:

The equation of state (EOS) unit calculates the thermodynamic properties of an ideal, *gamma-law* fluid. The gamma-law EOS is applicable to a single fluid with a constant adiabatic index (γ), using equations (6) – (8) to calculate the thermodynamic properties:

$$P = (\gamma - 1)\rho e, \quad (6)$$

$$P = \frac{N_a k \rho T}{A}, \quad (7)$$

$$e = \frac{N_a k T}{A(\gamma-1)}, \quad (8)$$

where N_a , k , A and T are the Avogadro number, Boltzmann constant, atomic mass of the fluid and temperature. In the case of a multi-component system containing N species, the average atomic mass (A_{avg}) and adiabatic index (γ_{avg}) of the fluid mixture are calculated using equations (9) and (10), respectively:

$$\frac{1}{A_{avg}} = \sum_i \frac{Y_i}{A_i}, \quad (9)$$

$$\gamma_{avg} = 1 + \left(A_{avg} \sum_i^N A_i \frac{Y_i}{\gamma_i - 1} \right)^{-1}. \quad (10)$$

In § 3.1, we describe the extension of this module to handle temperature-dependent specific heats, necessary for modeling combustion flows.

2.3 Burn Module:

The *Burn module* in FLASH was designed to solve nuclear reaction networks, where the evolution of an individual species is governed by [13],

$$\frac{dX_i}{dt} = R_i. \quad (11)$$

Here, X_i and R_i are the molar fraction and the net rate of change of molar fraction of i^{th} species due to all the reactions in the reaction network. For multiple species, equation (11) represents a set of stiff, coupled ordinary differential equations, which are solved using Kaps-Rentrop (Rosenbrok)[6] or Bader-Deuflhard[7] time integration methods, supplemented by an MA28[15] linear algebra package. We have modified this module to solve a system of stiff equations arising from typical chemical reaction mechanisms (§3.4).

2.4 Diffusion Unit:

Diffusion effects in FLASH are incorporated through the inclusion of either an explicit flux-based solver or a general implicit diffusion equation solver. The flux-based diffusion solver is an indirect method to account for diffusion effects, that does not operate on the primary variables, instead updating the fluxes due to diffusion heat, mass and momentum [13]. In contrast, the implicit solver computes the solution to the general diffusion equation from which the primary variables are directly obtained, so that the solution does not require inputs from other units. The diffusion of variable f governed by equation (12) can be solved with an implicit time-stepping in directionally split or unsplit mode,

$$A \frac{\partial f}{\partial t} + C = \nabla \cdot B \nabla f + D, \quad (12)$$

where A , B , C and D can be parameters with spatial variations. The current implementation of the implicit solver in FLASH solves for thermal diffusion, which we have extended in § 3.2 to handle mass and momentum diffusive transport.

3. DEVELOPMENT OF CHEMICAL REACTION SOLVERS AND AUXILIARY MODULES

In this section, we describe the additional capabilities we have developed to enable combustion flow simulations using FLASH. These capabilities include a temperature-dependent,

multi-species equation of state (EOS) (§ 3.1), extension of the implicit solver to handle mass and viscous diffusion (§ 3.2), modifications to the Burn module to handle H₂-Air and CH₄-Air reaction mechanisms (§ 3.4), and an expansion of the materials database (§ 3.3) (temperature-dependent transport and thermodynamic properties) to include species commonly involved in the reaction mechanisms.

3.1 Temperature-dependent Equation of State:

The equation of state (EOS) module described in §2.2 was expanded to include multiple species with temperature-dependent properties to enable FLASH to solve chemically reacting flows with heat addition. The extended EOS can be applied to temperatures ranging from 298K - 5000K, and requires 10 coefficients for each species in the mixture to calculate thermodynamic properties. The existing EOS unit in FLASH [13] can be invoked in three modes according to the input variable pairs supplied in each case viz., *Density-Temperature* (ρ, T), *Density-Pressure* (ρ, P), and *Density-Internal Energy* (ρ, e).

Density-Temperature (ρ, T) mode:

In this mode, the EOS routine first calculates the specific heat at constant pressure (Cp_i) for each species i in the mixture according to [16],

$$\frac{Cp_i}{R} = a_1 + a_2 T + a_3 T^2 + a_4 T^3 + a_5 T^4, \quad T \geq 1000K, \quad (13)$$

$$\frac{Cp_i}{R} = a_8 + a_9 T + a_{10} T^2 + a_{11} T^3 + a_{12} T^4, \quad T < 1000K, \quad (14)$$

where R is the universal gas constant and $a_1 - a_5$ and $a_8 - a_{12}$ are thermodynamic polynomial coefficients of the i^{th} species in the mixture for $T \geq 1000$ K and $T < 1000$ K respectively. Note that coefficients a_6, a_7, a_{13} and a_{14} are used to evaluate additional thermodynamic properties by other modules (e.g. Burn) and will be discussed in §3.4. The adiabatic index γ_i at temperature T corresponding to each species is then computed using

$$\gamma_i = \frac{c_{p_i}}{c_{p_i-R}}, \quad (15)$$

while the mixture adiabatic index γ_{avg} is obtained from

$$\gamma_{avg} = 1 + \left(A_{avg} \sum_i^N A_i \frac{\gamma_i}{\gamma_i - 1} \right)^{-1}. \quad (16)$$

Finally, the pressure (P) and internal energy (e) are then computed using:

$$P = \frac{R \rho T}{A_{avg}}, \quad (17)$$

$$e = \frac{R T}{A_{avg}(\gamma_{avg} - 1)}. \quad (18)$$

Density–Pressure (ρ , P) mode:

When (ρ , P) are supplied as input variables, the temperature of the mixture is first computed using equation (17), followed by the mixture adiabatic index obtained from equations (13) – (16), and internal energy is finally computed from equation (18).

Density–Internal Energy (ρ , e) mode:

Here, (ρ , e) are provided as inputs, while equation (18) is solved iteratively to calculate temperature using the Newton–Raphson root finding algorithm. The solver requires an initial temperature guess, maximum number of iterations, and a tolerance for the absolute allowable error, which are specified in an input file with default values of 1001.0 K, 50 and 1.0e-8 K, respectively. Pressure is then calculated using equation (17). We found that the temperature-dependent multi-species EOS suffered no significant degradation in performance compared to the single-species unit, an important consideration in a reactive flow simulation where the EOS unit could be called multiple times over a single Hydro timestep.

We validate the multi-species, EOS unit was validated by computing temperature and adiabatic indices for species at different temperatures, and comparing with data compiled by NIST[17]. In these tests, the EOS unit was invoked in the *Density–Pressure* (ρ, P) and *Density–Internal Energy* (ρ, e) modes for nitrogen, hydrogen, and oxygen with the computed gas properties compared with highly accurate experimental or quantum mechanics simulation data from various sources[17]. This verifies the implementation of the multi-species EOS unit, as well as the implementation of the Newton-Raphson method used to solve for temperature in equation (18).

3.2 Implicit mass and viscous diffusion equation solvers:

Diffusion is critical to reactive flows (laminar or turbulent), since it determines abundances of reactants across a flame front, and thus the ultimate reaction rate. However, with temperature variations from heat addition, the transport properties governing the diffusive process may also change significantly, and must be modeled accurately. In this section, we discuss specific modifications to FLASH and the diffusion solvers therein that enable accurate description of temperature-dependent diffusive transport associated with heat, mass and momentum. Note that FLASH is equipped with a separate ‘flux-based’ diffusion solver (§ 2.4) that updates the diffusive fluxes in each direction. In addition, an implicit solver for a general diffusion equation is available in FLASH, and described in § 2.4. Currently, the implicit solver is used to solve the thermal diffusion equation (§ 2.4). We have extended the use of this solver to compute solutions to the mass and viscous diffusion equations, using an operator splitting approach. Equation (12) is the general form of the diffusion equation solved by the implicit solver in FLASH. For example, mass diffusion of species i may then be written as

$$\rho \frac{\partial Y_i}{\partial t} = \nabla \cdot (\rho D_i \nabla Y_i), \quad (19)$$

so that Y_i represents the mass fraction of species i , ρ is the mixture density and D_i is the species diffusivity, computed from equations described in §3.2. Thus, for the mass diffusion equation an implicit solver is used, during which FLASH freezes the density field in eq. (19). Similarly, viscous diffusion effects are solved by rewriting equation (12)

$$\rho \frac{\partial V}{\partial t} = \nabla \cdot (\mu \nabla V), \quad (20)$$

where μ is the temperature-dependent dynamic viscosity of the mixture. Typically, the temperature-dependent viscosity and mass-diffusion coefficients of species i are specified, while the thermal conductivity is constrained through a constant Lewis number (Le_i) assumption. The implementation of these solution methods was verified by comparing the axial velocity decay of a non-reacting, axisymmetric laminar jet with the analytical solution from [18] and the DNS results of [19] as well as the temporal decay of a potential vortex with analytical solutions from [20].

3.3 Temperature-dependent material properties database:

The material properties database in FLASH was expanded to accommodate temperature-dependent thermodynamic or transport properties of a single- or multi-species system. In reacting flows with significant temperature changes, the material and thermodynamic properties of species can be strong functions of temperature. Since FLASH is organized in to distinct modules, these properties are calculated by individual units in the course of a reactive flow computation. Thus, a central database of coefficients accessible to all units is required to calculate these properties at any given temperature, and has been implemented.

At initialization, a multi-species unit [13] stores several attributes of constituent species required in the course of a simulation. These include the names of the species, the molecular weights, the enthalpies of formation, the temperature-dependent adiabatic indices, the curve-fit polynomial coefficients for thermodynamic[16] and transport[21] properties.

The EOS unit described in §3.1 calculates the thermodynamic properties, while the transport properties (conductivity, viscosity and mass diffusion coefficients) are calculated in separate *material properties* units described below.

Viscosity:

The pure species dynamic viscosity (μ_i) in Poise is calculated using a curve fit polynomial[21] with 8 coefficients ($b_1 - b_8$) for each species. The coefficients are applied based on the temperature, and according to

$$\ln \mu_i = b_1 \ln T + \frac{b_2}{T} + \frac{b_3}{T^2} + b_4, \quad T < 1000\text{K}, \quad (21)$$

$$\ln \mu_i = b_5 \ln T + \frac{b_6}{T} + \frac{b_7}{T^2} + b_8, \quad T \geq 1000\text{K}. \quad (22)$$

The coefficients required in these calculations are accessed from the central database described above. The multi-species mixture viscosity (μ_{mix}) can then be calculated by a combination averaging formula[22] such as equation (23)

$$\mu_{mix} = \frac{1}{2} \left[\sum_{i=1}^N \mu_i X_i + \left(\sum_{i=1}^N \frac{X_i}{\mu_i} \right)^{-1} \right], \quad (23)$$

or by using a semi-empirical formula[23]

$$\mu_{mix} = \sum_{i=1}^N \frac{\mu_i X_i}{\sum_{j=1}^N X_j \Phi_{ij}} \quad \text{and} \quad \Phi_{ij} = \frac{1}{\sqrt{8}} \left(1 + \frac{A_i}{A_j} \right)^{-\frac{1}{2}} \left(1 + \left(\frac{\mu_i}{\mu_j} \right)^{\frac{1}{2}} \left(\frac{A_i}{A_j} \right)^{\frac{1}{4}} \right)^2. \quad (24)$$

Thermal Conductivity:

The pure species conductivity (λ_i) in W/cm-K is computed from a curve fit polynomial [21] with coefficients ($c_1 - c_8$) for each species given by,

$$\ln \lambda_i = c_1 \ln T + \frac{c_2}{T} + \frac{c_3}{T^2} + c_4, \quad T < 1000\text{K}, \quad (25)$$

$$\ln \lambda_i = c_5 \ln T + \frac{c_6}{T} + \frac{c_7}{T^2} + c_8, \quad T \geq 1000\text{K}. \quad (26)$$

The multi-species mixture conductivity (λ_{mix}) is then obtained from the combination averaging formula[22]

$$\lambda_{mix} = \frac{1}{2} \left[\sum_{i=1}^N \lambda_i X_i + \left(\sum_{i=1}^N \frac{X_i}{\lambda_i} \right)^{-1} \right]. \quad (27)$$

Alternately, the mixture conductivity could be obtained from a constant Lewis number assumption:

$$\lambda_{mix} = \rho Le_i D_{im} c_p. \quad (28)$$

Here, Le_i and D_{im} are the Lewis number and mass diffusion coefficient of the i^{th} species in a mixture containing N species. Note that when λ_{mix} is obtained using equation (28), the mass diffusion coefficients D_{im} have been calculated independently from equation (30) below.

Mass diffusion:

For a given Lewis number, the mass diffusion coefficient of the i^{th} species in the mixture (D_{im}) can be calculated using,

$$D_{im} = \frac{\lambda_{mix}}{\rho Le_i c_p}, \quad (29)$$

where λ_{mix} is independently available from equations (25 - 27). We have also implemented the option to calculate the mass diffusion coefficient independently without making assumptions on the Lewis number. This is achieved by assuming the mass diffusion coefficient D_{im} of each species i in the mixture (m) to be equal to that of a binary gas system $A-B$ (D_{AB}), where A and B can be any major species pair. Then, the binary diffusion coefficient D_{AB} of the system is calculated using the Chapman and Enskog solution[24] to the Boltzmann equation,

$$D_{AB} = \frac{0.00266 T^{1.5}}{P M_{AB}^{0.5} \sigma_{AB}^2 \Omega_D}, \quad (30(a))$$

$$M_{AB} = 2 \left[\frac{1}{A_A} + \frac{1}{A_B} \right]^{-1}, \quad (30(b))$$

$$\sigma_{AB} = (\sigma_A + \sigma_B), \quad (30(c))$$

where σ_{AB} is the characteristic length of the intermolecular forces, Ω_D is the diffusion collision integral, σ_A and σ_B are the Lennard-Jones lengths of species A and B , and A_A and A_B are molecular weights of species A and B . Our current implementation in FLASH includes the following binary gas systems: H_2 - O_2 , N_2 - O_2 or CH_4 - O_2 .

3.4 Burn Module:

We describe the implementation of H_2 -Air and CH_4 -Air reaction kinetics to the existing *Burn* module in FLASH. A general reaction mechanism involving N species and m reactions can be represented as:

$$\sum_{k=1}^N v_{kj}' M_k \Leftrightarrow \sum_{k=1}^N v_{kj}'' M_k, \quad (31)$$

where v_{kj}' and v_{kj}'' are the stoichiometric molar coefficients of species labeled M_k in the j^{th} reversible reaction. Thus, the net rate (q_k) of formation or consumption of the k^{th} species is the sum of contributions from all reactions in the reaction involving that species, and written as

$$q_k = \sum_{j=1}^m q_{kj}, \quad (32(a))$$

where

$$q_{kj} = (v_{kj}'' - v_{kj}') \left(k_{fj} \prod_{k=1}^N C_{M_k}^{v_{kj}'} - k_{bj} \prod_{k=1}^N C_{M_k}^{v_{kj}''} \right). \quad (32(b))$$

In equation (32 (b)), q_{kj} is the rate of formation (or consumption) of the k^{th} species in reaction j , k_{fj} and k_{bj} are the corresponding forward and backward reaction rates, while C_{M_k} is the concentration of the k^{th} species. For most reaction mechanisms, the forward reaction rates are given in the form of an Arrhenius equation,

$$k_{fj} = AT^\alpha e^{\left(-\frac{E_a}{RT}\right)}, \quad (33)$$

where α , A and E_a are the temperature index, pre-exponential factor and activation energy respectively. To compute the backward reaction rates, the concentration-based reaction equilibrium constant (K_c) is first evaluated from

$$K_c = \left(\frac{RT}{P}\right)^{-\Delta v} e^{\left(\frac{\Delta G_T}{RT}\right)}. \quad (34)$$

Here, Δv is the change in mole number (between products and reactants), while ΔG_T represents the corresponding change in the Gibbs free energy. The Gibbs free energy for each species is calculated according to [25]

$$\frac{G_T}{RT} = a_1(1 - \ln T) - \frac{a_2 T}{2} - \frac{a_3 T^2}{6} - \frac{a_4 T^3}{12} - \frac{a_5 T^4}{20} + \frac{a_6}{T} - a_7, \quad T \geq 1000K \quad (35 (a))$$

$$\frac{G_T}{RT} = a_8(1 - \ln T) - \frac{a_9 T}{2} - \frac{a_{10} T^2}{6} - \frac{a_{11} T^3}{12} - \frac{a_{12} T^4}{20} + \frac{a_{13}}{T} - a_{14}, \quad T < 1000K \quad (35 (b))$$

where $a_1 - a_{14}$ are the polynomial coefficients [16] from a curve fit of the thermodynamic data for a given species. From the equilibrium constant K_c , the backward reaction rates can be inferred:

$$k_{bj} = \frac{k_{fj}}{K_c}. \quad (36)$$

From equations (32), it is clear that a reaction mechanism involving N species could be formulated as a system of N coupled ordinary differential equations. The resulting ODE system

is stiff, owing to large variations (several orders of magnitude) in the reaction rates and the corresponding eigenvalues. We use two implicit ODE integrators already available in FLASH viz., the Bader-Deuflhard [7] and Kaps-Rentrop [6] schemes (along with linear algebra package MA28[15]) to solve the stiff system of equations represented by the general equation (32 (a)). For both methods, the energy generated (e_g) during a time advancement Δt is calculated using

$$e_g = \sum_i^N \frac{h_{f,i}}{A_i} (Y_{i,t} - Y_{i,t+\Delta t}), \quad (37)$$

where $h_{f,i}$ is the heat of formation, $Y_{i,t}$ and $Y_{i,t+\Delta t}$ are the mass fractions of the i^{th} species before and after the time advancement respectively.

We have implemented three reaction mechanisms for H₂-Air combustion and one for CH₄-Air combustion in FLASH. The 9-species, 19 reaction H₂-Air mechanism of Billet[2] is reduced from the detailed acetylene combustion mechanism of [26], and was used [2] to study the interaction of acoustic waves with circular and spherical hydrogen diffusion flames and reactive shock-bubble interactions. We have also implemented the 9-species, 19-step reversible reaction mechanism of Mueller et al.[3] as adapted by [27] to simulate premixed H₂-Air vortex rings. The third-body efficiencies (M) for all the reactions in this mechanism were taken from [3]. Both mechanisms exclude NO_x chemistry important for industrial high-temperature burners. Finally, we have also included the 11-species, 20-step reversible reaction mechanism of Katta and Roquemore[4], which was originally used to study the tip opening phenomena in premixed H₂-Air Bunsen burner flames. The calculated burning velocities[4] from this mechanism were in excellent agreement with [28] for equivalence ratios less than 2.5, while higher equivalence ratios resulted in underpredicted values of the burning velocities. Note that in our implementation of this mechanism, the third body efficiency (M) for all the species was assumed to be unity.

To describe CH₄-Air chemistry, we use the reduced reaction mechanism of Smooke [5], the simplicity of which makes it attractive for turbulent flame calculations. Gordon et al.[29] studied the auto-ignition characteristics of different methane combustion mechanisms and found higher values of ignition delays for the Smooke mechanism[5] compared to more detailed mechanisms. However, Hermanns et al.[30] computed the laminar burning velocities using reaction mechanisms of varying complexity, ranging from 16 - 127 species and 25 - 1207 reactions, and found the burning velocity from the Smooke mechanism[5] to be in agreement with data from more complex mechanisms. We have implemented the reduced mechanism of Smooke[5] as adapted by Hermanns et al.[30] with the rate constant for the 10th reaction (forward) from Chen[31]. A second version is also included in FLASH, with the rate constant for this critical reaction set to the value from the widely available GRI-MECH[16] database. We also studied the performance of the original mechanism, and the modified mechanisms in predicting the ignition delay in a well-stirred reactor. We found that when the modified mechanisms were used, the ignition delay is significantly reduced and is closer to expected values for high ignition temperatures whose results are presented elsewhere[32].

Testing of the Burn module and the implementation of the reaction mechanisms were carried out using two simulations described in this section: (i) comparison of the adiabatic flame temperature in a well-stirred reactor with data from [33] and (ii) comparison of the temporal evolution of temperature and mass fraction of species in a well stirred reactor for H₂-Air mixtures with data from [27].

Adiabatic flame temperature calculation:

The maximum flame temperature achieved by combustion of a fuel and oxidizer mixture at equilibrium is the adiabatic flame temperature (T_{ad}) and is widely used as a metric to validate reaction mechanisms in simulation codes. To test our development of the chemical reaction

suite, we performed simulations of a well-stirred reactor at constant pressure. Note that in FLASH, each cell in the computational domain is effectively a constant-volume, constant-mass reactor. Thus, heat addition due to combustion is accompanied by an increase in pressure. Therefore, to model a well-stirred reactor, we update the density in the cell after each timestep with the value from EOS, thereby forcing the pressure to remain constant.

A 1D computational domain was initialized with a homogeneous mixture of hydrogen and air ($Y_{H_2} = 0.029$, $Y_{O_2} = 0.233$ and $Y_{N_2} = 1 - Y_{H_2} - Y_{O_2}$) at atmospheric pressure and a temperature $T = 1500K$, above the auto-ignition temperature. The mixture is allowed to burn to equilibrium, governed by the reaction mechanism given in [2]. The temporal evolution of temperature in a well-stirred reactor is shown in figure 1(a), where the adiabatic flame temperature is inferred as the saturation temperature near the end of the simulation. The calculation was repeated for fuel-air mixtures at different equivalence ratios (ϕ), and the adiabatic flame temperatures from the FLASH simulations compare well with data from CERFACS [33] in figure 1(b).

Temporal evolution in a well-stirred reactor:

The evolution of a homogeneous, H_2 -Air mixture in a well-stirred reactor was computed using the mechanisms detailed in [2] and [27]. A constant pressure reactor condition is enforced using the approach discussed above. Figures 2 (a) and (b) show the temporal evolution of hydrogen mass fraction and temperature from FLASH compared to the results of [27], from a well stirred reactor simulation at 1atm pressure and initialized with a stoichiometric mixture of hydrogen and air at 1200K. Note that the simulations were performed with both time-integration methods described in §2.4 with identical results. As expected, the results from the mechanism in [27] are in better agreement with their DNS data. Both mechanisms accurately predict the

initiation of combustion inferred by the temperature rise (or equivalently the consumption of hydrogen) in the reactor beginning at $t \sim 30\mu s$.

4. VALIDATION OF GAS KINETIC REACTION SOLVER

We now present results from a suite of validation cases that test various aspects of the numerical methods described in §2 and §3, as well as demonstrate some unique capabilities of the modified FLASH code. Furthermore, we hope the data presented here can also be used as a benchmark for other experimental and numerical validation efforts. We compare our simulations with published experimental and numerical results, including the advection of reacting (§ 4.2) and non-reacting (§ 4.1) 1-D fronts, the advection of a 2D circular temperature spot in a directional background flow (§ 4.1), laminar premixed flames in a Bunsen burner configuration (§ 4.3), shock-driven combustion of an initially circular bubble (§ 4.4) and preliminary data from a 2D reacting, RMI flow (§ 4.5).

4.1 Advection of a 1D non-reacting H_2-O_2 front:

In the absence of chemical reactions, the advection of an inert material front tests the ability of hydrodynamic solvers in transporting a sharp interface with minimal losses and without spurious oscillations or overshoots and undershoots. FLASH employs an operator splitting approach, with the so-called HYDRO unit solving the Euler equations (§2.1), while diffusion effects are implemented through a call to the flux-based or implicit diffuse unit (We refer to this as the DNS approach). Alternately, when the Diffuse unit is turned off, the use of a finite grid implies cell-averaged numerical mixing, so that an effective diffusion could be attributed to the numerical mixing process. We investigate the relative merits of each approach in solving a simple convection-diffusion problem of an inert front.

When shock capturing schemes are used to advect a sharp material discontinuity, significant pressure oscillations that travel away from the interface are often observed [13, 32, 34]. Such oscillations can grow over time resulting in distortions of the contact discontinuity,

incorrect advection velocities, and temperature overshoots and undershoots [34]. Since chemical kinetics is a strong function of temperature, the presence of such oscillations may lead to incorrect combustion features such as the flame speed, the flame length and species mass fractions. By repeating the calculation with and without diffusion, we have ensured the reacting and non-reacting fronts are advected with the correct velocity and without overshoots and undershoots, and thus not affected by the modifications to FLASH.

A non-reacting H₂-O₂ front with a sharp interface in temperature and mass fraction was advected with a velocity (*U*) of 2000 cm/s. The computational domain was initialized with a uniform pressure of 1 bar while the initial hydrogen mass fraction profiles (*Y_{H2}*) and mixture density (*ρ_{mix}*) were specified by equations (38) and (39) respectively [34]:

$$Y_{H_2} = \frac{1}{2} [1 + \tanh(C(\frac{l}{2} - |x - x_0|))], \quad \text{for } x \geq x_0, \quad (38(a))$$

$$Y_{H_2} = 1.0, \quad \text{for } x < x_0, \quad (38(b))$$

$$\rho_{mix} = \left(\frac{Y_{H_2}}{\rho_{H_2}} + \frac{1-Y_{H_2}}{\rho_{O_2}} \right)^{-1}. \quad (39)$$

In equation (38 (a)), *C* determines the sharpness of the front and was specified to be 80 cm⁻¹, while *x₀* and *l* specify the location of the center and the width of the hydrogen plateau respectively. For all the cases reported here, the value of *l* in equation (38 (a)) was chosen to be 0.6 cm, while *x₀* = 3.3 cm was used. The temperature-dependent, multi-species equation of state (§3.3) was used to initialize the rest of the thermodynamic properties in FLASH. Table 2 lists the 1D, non-reacting simulations performed using FLASH, categorized by the choice of diffusion implementation (implicit, flux-based, numerical). We compare our results with [34], who describe using Direct Numerical Simulation (DNS) the advection of a non-reacting H₂-O₂ front with initial temperatures of 1000K and 2000K respectively. The DNS approach of Billet and Abgrall[34] allowed for non-unity values of Lewis numbers of the species as well as the inclusion

of Soret effects, while the interface was modeled as a stationary front. In our simulations, we compute a moving front, assume a Lewis number of one for conductivity calculation from the mass diffusion coefficient (equation (28)), and neglect Soret effects. For the cases with numerical diffusion of transport properties, the Lewis number is not defined or may be nominally interpreted to be unity.

Figure 3 is a plot of profiles of Y_{H_2} for cases 1 – 3 (Table 2) from FLASH compared with DNS[34] at $t = 400\mu s$. For cases 2 - 3, the interface position and thickness (defined as the x - distance over which Y_{H_2} varies from 1% to 99%) were observed to be in good agreement with the DNS[34] results in figure 3. For case 1 in the absence of mass diffusion, PPM maintains the integrity of the initial profile as expected by steepening the contact discontinuity. The interface shape for this case shows a slight staircase-like structure ($x \sim 4.4$ cm) which may be attributed to the PPM algorithm interpreting an under-resolved, steep (but smooth) density gradient as a contact discontinuity [13]. In such instances, the contact steepening algorithm restricts the spreading of the discontinuity by representing it as a series of local contact discontinuities. When the effects of diffusion are included, either through an implicit solver of the diffusion equation (case 2), or through a consistent update of the transport fluxes (case 3), the late time profiles exhibit diffusive broadening, comparable to the DNS results of [34]. The slight differences in the shapes of mass fraction profiles for cases 2 and 3 are attributable to differences seen in their temperature profiles. Overall, case 2 shows slightly better agreement with DNS of [34], compared to the flux-based method in case 3.

To test the directionally split hydrodynamic solver in FLASH, we performed simulations of a 2D circular hydrogen ring immersed in oxygen, at 1 atm pressure and advected at a constant velocity. The velocity of advection was specified using the x - (U) and y - (V) components, while all other conditions were identical to the 1D case described above. We found

that symmetry was well preserved to late times for all the cases in table 2 when advection was along ($V = 0$) or at an angle ($V \neq 0$) to the x-axis.

4.2 Advection of a 1D reacting H₂-O₂ fronts:

The simulations described in §4.1 were repeated with the burn module activated, so that chemical reactions between species and heat release are enabled. FLASH simulations of 1D reacting fronts described in this section serve to validate the integration of the Burn module with the Hydro module, the implementation of H₂-O₂ reaction chemistry, as well as the expanded diffusion units. We compare the results from FLASH with that of the double-flux method described in [34].

The problem setup includes an H₂-O₂ material interface, with pressure, temperature and velocity as specified in §4.1. H₂ mass fraction is initially specified by equation (38 (a)) for $0 < x < \infty$, with $x_0 = 3.7$ cm and $l = 0.6$ cm, while the mixture density is given by equation (39). In this configuration, two material interfaces are present, each of thickness 0.07 cm and centered at 3.4 cm and 4.0 cm. Table 3 lists the 1D reacting simulations performed using FLASH. Note that the mass diffusion coefficients for all the species were assumed to be equal to that of the binary diffusion coefficient of the H₂-O₂ system. For cases 5 - 6, we assume $Le = 1$, and calculate the thermal conductivity from the mass diffusion coefficient of the binary H₂-O₂ system. The mixture viscosity was calculated using equation (23), the semi-empirical formula from [23].

Figures 4 (a) - (d) are comparisons of our results from case 4 at $t = 100$ μ s with [34], who included diffusion effects in their double-flux simulations. In figure 4 (a), we plot the temperature on a stretched scale ($T/2000$), while the mass fraction of H₂O is shown in figure 4 (b). The leading and trailing fronts are advected with the correct velocity, and without oscillations. The maximum temperature (~ 3200 K) and its location from FLASH are in good agreement with data from [34], while the flame remains sharp owing to the negligible numerical diffusion in PPM. The

temperature associated with the trailing front exhibits localized oscillations, whereas in the leading front such features are absent. These differences in the late time behavior of the initially symmetric temperature fronts are explained by the direction-dependent oscillations inherent in PPM, and have been reported elsewhere[32] to appear in the absence of any physical diffusion. In figure 4 (c), the oxygen mass fraction profile from case 4 reveals portions of the interface that remain unburnt (at $x = 3.6$ cm and 4.2 cm). The mass fraction of fuel across the interface changes from 0 to 1 (figure 4 (d)), while the equivalence ratio ($\varphi = \frac{Y_{H_2}/Y_{O_2}}{(Y_{H_2}/Y_{O_2})|_{stoich}}$) varies from 0 to ∞ across this region, leaving the mixture too rich (lean) at certain locations across the interface for combustion to take place. Since the contact steepening in FLASH results in the formation of staircase-like structures across the interface, this causes the local equivalence ratio to change. The equivalence ratio determines the combustibility of the mixture, resulting in the observed asymmetries in O_2 mass fractions in figure 4 (c). Note that the discrepancies in H_2O profiles between FLASH and [34] are attributed to differences in the reaction mechanisms employed; H_2 - O_2 combustion in FLASH is described through the 19-step, detailed mechanism of [2] involving nine species, while [34] uses a global 2-step, 4 species description.

In figure 5, we compare results from the flux-based implementation of mass, thermal and momentum diffusion in FLASH (case 5) with data from [34]. The interface location and velocities are in good agreement with DNS results [34], while the oscillatory behavior observed in figure 4 is not seen here. Furthermore, the inclusion of mass diffusion results in complete burning of the material interface, with no spikes of unburnt material. The temperature profile is in agreement with [34], although a small undershoot in temperature is observed at $x = 4.4$ cm and 3.4 cm at the leading and trailing interfaces respectively. We attribute these undershoots to the pressure and velocity oscillations observed in PPM, and reinforced as diffusive fluxes in this method. Furthermore, in our simulations, no assumption on the Prandtl number is made, in contrast to Billet and Abgrall[34] who assume $Pr = 0.4$ to infer a thermal conductivity. Since the simulations

in [34] had $D_{heat} > D_{momentum}$, oscillations in the temperature profile may have been further stabilized.

Figure 6 is a comparison of mass fractions and temperature data from FLASH with diffusion implemented through an implicit solver (case 6) with the results of [34] at $t = 100 \mu s$. Note that this simulation was performed at a resolution of $\Delta x = 0.01$ cm, less than the corresponding values of Δx for cases 4 and 5 (Table 3), but still maintains symmetry without oscillations. Thus, the inclusion of diffusion effects through the implicit solver combines the efficiency of PPM with the stability of the diffusion operator, thereby suppressing oscillations at moderate zoning. Furthermore, unlike case 5, the temperature profile exhibits no undershoots in figure 6.

4.3 2D axisymmetric CH₄-Air laminar flames:

To validate our implementation of CH₄-Air chemistry, we present results from computations of a 2D, axisymmetric, CH₄-Air laminar, premixed jet flame, and compare our results with the experiments of Nguyen et al. [35]. The experimental setup [35] involved a Bunsen-burner with a honeycomb matrix at the inlet, with an inlet diameter for the fuel nozzle of 1.7cm, while the surrounding air co-flow had a diameter of 7.0 cm. In the experiments, the honeycomb matrix and the co-flow jet were used to stabilize the flame. Flame properties were measured using Raman-Rayleigh scattering and laser induced fluorescence (LIF) at equivalence ratios of 1.38, 1.52 and 1.7. Radial profiles of mole fractions (H₂, O₂, N₂, H₂O, CO₂, CO, CH₄, NO and OH) and the temperature were reported at three axial locations for a flame equivalence ratio of 1.38. Centerline (axial) measurements of temperature and species for all three flames were also reported in [35]. Numerical simulations of these flames were reported in Zhou et. al[36], who used the 16-species, 32-step mechanism of [37] with temperature-dependent transport properties on a 2D, axisymmetric cylindrical coordinate system. Note that

Zhou et. al[36] included gravity effects in their simulations, while making no assumptions on the Lewis number.

The computational domain and boundary conditions used in the FLASH simulations are schematically represented in figure 7. We have used the directionally-split, PPM solver in FLASH coupled with the implicit diffusion solver in a 2D, axisymmetric, cylindrical ($r - z$) domain with a uniform mesh spacing of ($\Delta r = 0.05\text{cm}$, $\Delta z = 0.1\text{cm}$). The mixture viscosity is calculated using the semi-empirical formula (equation (24)), while mass diffusion coefficients for all the species were assumed to be equal to that of the binary diffusion coefficient of an $\text{N}_2 - \text{O}_2$ system. The mixture thermal conductivity was calculated by assuming the Lewis number to be unity. We neglect the effect of gravity (buoyancy) in our simulations, an assumption that may be justified by the inclusion of the honeycomb at the inlet in the experiments to shield the flame from buoyancy effects. The computational domain was initialized everywhere with a uniform axial velocity equal to that of the co-flow velocity ($U_2 = 38\text{cm/s}$) with air at 300K and 1 atm pressure. Ignition was achieved by providing a rectangular hotspot (1650K) above the inlet at $0 < z \leq 0.75R$. Table 4 summarizes parameters for the FLASH simulations including the flame equivalence ratios and flow rates (in standard liters per minute (slm)) of methane and air chosen to match the experimental conditions. For all the simulations in table 4, we have used a 16-species, 25-step CH_4 -Air reversible reaction mechanism with modified rate constants for the 10th reaction[30]. Note that unlike the mechanism employed in the simulations of [36], we neglect NO and N while including CH_3O and H_2O_2 . In our simulations, all flames achieve steady state and a typical temperature distribution is shown in figure 8 (case 9).

The axial profiles of N_2 , CH_4 and O_2 from case 7 are in good agreement with experimental measurements (figure 9 (a)). Outside the combustion zone, where the fuel is completely consumed ($z > 5\text{ cm}$), the oxygen mole fraction increases to its value in air (~ 0.21) due to diffusion from the co-flow stream, a process dictated by the values of mass diffusion

coefficients. Thus, differences in X_{O_2} (up to 80%, far away from the inlet) are observed between the experiments and the FLASH simulations, which assumed mass diffusion of all species to be approximated by a $N_2 - O_2$ binary system. Since we assume $Le = 1$, this assumption also affects the thermal conductivity calculation, resulting in the above differences in the predicted quantities away from the combustion zone ($z > 5$ cm). In figure 9 (b), axial profiles of the temperature and mole fractions of the major products (CO_2 and H_2O) are in good agreement with the experimental measurements of [35], while differences are observed outside the combustion zone.

Radial profiles ($z = 2.1$ cm) of reactant (CH_4 , O_2 , N_2) mole fractions from case 7 are compared with experimental data in figure 10 (a), while corresponding product (CO_2 , H_2O) and temperature profiles are shown in figure 10 (b). We find radial profiles of X_{CH_4} , X_{O_2} and X_{N_2} are in good agreement with data from [35], with assumptions made on transport properties having little impact. Similarly, the product and temperature distributions from our simulations agree well with experimental data in figure 10 (b).

In figures 11 - 12 (a) and (b), we show the radial distributions of mole fractions and temperature from our simulations of case 7, and the corresponding experimental results[35] at $z = 5.0$ and 9.0 cm respectively. The drop in the centerline temperature observed in the FLASH data near $r = 0$ is due to a 1 cm discrepancy (20%) between the observed cone length in our simulations and the experiments of [35]. Thus, the r -slice at $z = 5$ cm falls just outside the flame cone in the experiments, while this region characterizes the preheat zone in our simulations. These differences in the cone length are due to the use of a simplified chemical kinetics model in FLASH (that does not include NO_x chemistry), and mass diffusion modeled as that of a binary N_2-O_2 system.

Our results from case 8 are compared with experimental data in figure 13 (a) - (b), with axial profiles of reactants shown in figure 13 (a), while the corresponding product and temperature profiles are displayed in figure 13 (b). Once again data for X_{CH_4} , X_{O_2} and X_{N_2} are in good agreement with experiments. Similarly, X_{CO_2} and $X_{\text{H}_2\text{O}}$ are in good agreement with data from [35] (figure 13 (b)), particularly inside the combustion zone. For $z > 6$ cm, X_{O_2} is over predicted by 50%, likely due to the assumption of $Le = 1$ in the simulations. Similarly, X_{CO_2} and $X_{\text{H}_2\text{O}}$ undershoot experimental data in this region by 40% and 35% respectively, due to the reasons discussed above (figure 9 (a)).

Finally, results from case 9 are plotted alongside experimental data in figure 14 (a) and (b). The plots show much of the same trends observed in figures 9 - 13. However, the axial location of the combustion zone (visible by the sudden rise of temperature for example) is underpredicted in our simulations. Note that the simulations of Zhou et. al [36] also underpredict the combustion zone by approximately the same margin seen in figure 14 and a detailed comparison of FLASH results with Zhou et. al[36] is presented elsewhere [32].

4.4 Shock-bubble combustion:

In this section, we demonstrate the shock-handling capability of the PPM method in FLASH by computing the shock-driven combustion of a 2D bubble. The problem specification is as described in [38] who investigated the impact of including volume viscosity on the bubble evolution. The problem setup is schematically presented in figure 15 which shows the computational domain initialized with a circular hydrogen bubble/cylinder, while a Mach 2 planar shock in air is located at 0.7cm. Ahead of the shock, the ambient velocity (U), pressure and temperature are $1.24\text{e}5$ cm/s, 1000K and 1atm, while the shocked conditions are $4.35\text{e}4$ cm/s, 1565K and 4.45 atm respectively. The mass fraction of the H_2 bubble is specified by[38]

$$Y_{\text{H}_2} = \frac{1}{2} [1 + \tanh(\frac{r_c - r}{c})], \quad r = \sqrt{((x - x_0)^2 + (y - y_0)^2)}, \quad (40)$$

where C determines the sharpness of the interface, r_c is the radius of the bubble, and x_0 and y_0 specify the location of its center. In our calculations, we specified $r_c = 0.28$ cm, the origin at $(0.4, 0)$ cm, while C was chosen to be $3e-3$ cm⁻¹ resulting in an initial interface thickness of $\sim 2.5e-2$ cm according to [38]. The choice of these parameters results in an initial separation of 0.2 mm between the shock and the H₂ bubble as the shock is initially held stationary, while the bubble traverses to the right in figure 15. Upon interaction, the transmitted shock acquires a finite velocity, and moves toward the outflow boundary at $x = 3$ cm.

Table 5 describes the shock-bubble simulations performed with FLASH, and categorized by the choice of diffusion implementation, and the use of Adaptive Mesh Refinement (AMR). The calculations were performed on a computational domain with 20×10 blocks, where each block consists of 8×8 cells. For case 10, the AMR mesh ranged from a minimum value of 1 to a maximum of 4, with an equivalent maximum uniform mesh resolution of 1280×640 (see [13] for details of this calculation). For cases 11 (a)-(b), since the implicit solver was not compatible with adaptive meshing, a uniform mesh refinement of 1280×640 was used. The H₂-O₂ combustion chemistry is described with the aid of the detailed 9-species, 19-step reversible reaction mechanism of [2], while the viscosity and thermal conductivity were calculated using the combination averaging formulas (equations (23) and (27)) for both cases. Note that for case 11 (b), mass diffusion coefficient for all the species is assumed to be equal to binary diffusion coefficient for N₂-O₂.

In figures 16 (a) and (b) we show line contours of pressure superposed on the H₂ mass fraction (grayscale) at $t = 1.5\mu s$ for cases 10 and 11(a) respectively. At this early time, the H₂ bubble has collided with the incident shock, and is compressed by it. The shock interaction with the density interface produces transmitted, reflected and refracted pressure waves seen in the figure. The pressure and H₂ mass fraction contours from cases 10 and 11(a) are shown at time $t = 3.5\mu s$ in figures 17 (a) and (b) respectively, where the initial stages of baroclinic vortex

formation is evident. Figure 18 is a comparison of the axial distribution of pressure along the axis of symmetry of the bubble from cases 10, 11 (a)-(b) with the simulation results of [38] at $t = 3.5 \mu\text{s}$ and show our simulations using the flux-based (case 10) and implicit (cases 11) diffusion methods are in good agreement with [38]. The dash-dot line in figure 18 shows results from a simulation performed with the mass diffusion coefficient equal to $\text{N}_2\text{-O}_2$ binary system for all the species. The x-profile for pressure from this case is in good agreement with the DNS of [38] and our simulations without mass diffusion (case 11 (a)). Note that unlike [38], our simulations for cases 10 and 11(a) did not explicitly include mass diffusion, so that the numerical scheme (PPM) maintains the sharp material interfaces. Also, the preservation of the sharp interfaces renders them susceptible to hydrodynamic instabilities such as Richtmyer-Meshkov and Rayleigh-Taylor instabilities.

At even later times, a complex shock structure accompanied by the breakdown of the H_2 bubble is visible in figures 19 (a) - (d), which shows contours of density and product (H_2O) mass fraction respectively. At $t = 10 \mu\text{s}$ (figure 19 (a)), two counter rotating vortices have saturated and punctuate the structure of the initially circular H_2 bubble. At this stage, combustion is confined in a thin rib that connects the kidney-shaped vortex structures, evident by the presence of H_2O along the ridge. This is a diffusion zone as the fuel here is flanked by air on both sides, thus forming a double diffusion flame. By $t = 20 \mu\text{s}$ (figure 19 (b)), the vortices display more small scale structure leading to increased mixing across the local material interface. The induced velocities due to the vortices deform the bubble, leading to an ellipsoidal structure. The enhanced mixing boosts combustion within the vortex structures, as the mass fraction of H_2O intensifies within the vortex cores. These trends continue to late times, and are observed in figures 19 (c) ($t = 30 \mu\text{s}$) and 19 (d) ($t = 40 \mu\text{s}$). In these realizations, the combustion along the fuel ridge connecting the vortices has extinguished, while intensifying within the vortex cores

under the influence of small-scale mixing. These features are qualitatively similar to the results reported by [38].

4.5 Reacting Richtmyer-Meshkov Instability:

When a shock wave interacts with a perturbed material interface separating gases of different densities, perturbations at the interface evolve according to the Richtmyer-Meshkov instability (RMI). When the fluids separated by the (diffuse) interface are H_2 and O_2 , the interfacial diffusion and shock-driven mixing lead to combustion. RMI has been studied traditionally from the perspective of inertial confinement fusion[39] (ICF) and astrophysical phenomena such as type IA supernovae [40]. Such an interaction of interfacial burning and mixing due to instability might occur in many practical combustion systems, including for example internal combustion engines[41, 42], where shock interactions with pockets of dispersed fuel across a material interface is possible. While shock interactions with premixed flames have been investigated numerically [43] and analytically [44], we are not aware of studies of RMI in the context of *non-premixed* flames. Shock-initiated combustion within a spherical bubble has been studied numerically [2, 38] and experimentally [45], where the focus of the study was ignition through shock focusing and turbulent breakdown of the bubble.

We consider the interaction of a Mach 1.2 shock with a sinusoidally perturbed diffuse/continuous H_2 - O_2 interface. The problem configuration is represented schematically in figure 20, where the shock initially located at $x = x_s$, travels from H_2 to O_2 across the interface. The initial interface location x_i is defined according to,

$$x_i = x_s + \frac{\Delta}{2} + h_0 \left[1 - \cos\left(\frac{2\pi y}{\lambda}\right) \right], \quad (41)$$

where λ is the wavelength, h_0 is the initial amplitude of the interface and Δ is the thickness of the interface defined as the distance between points where the mixture fraction Z varies from ε to $1 - \varepsilon$ with $\varepsilon = 1.0\text{e-}5$. We define the mixture fraction (Z) as,

$$Z = \frac{8 Y_{H_2} - Y_{O_2} + 1}{9} \quad (42)$$

where Y_{H_2} and Y_{O_2} are mass fractions of H_2 and O_2 respectively. Across the interface, $Z(x)$ is assumed to be an error function with thickness twice the initial amplitude (h_0), while $x|_{Z=0.5}$ is taken to be the instantaneous interface location (x_i). The use of a mixture fraction to describe the flow is advantageous, since unlike the individual species mass fractions which can suffer from local discontinuities, the mixture fraction remains monotonic across the interface, thus providing a reliable metric to compare reacting and non-reacting RMI.

The initial density difference between the gases on either side of the interface may be parameterized through the Atwood number (A_t) defined here as

$$A_t = \frac{\rho_{O_2} - \rho_{H_2}}{\rho_{O_2} + \rho_{H_2}}, \quad (43)$$

where ρ_{H_2} and ρ_{O_2} are the densities of H_2 (light) and O_2 (heavy) respectively. For initial values of $\rho_{H_2} = 8.0\text{e-}5 \text{ gm/cm}^3$ and $\rho_{O_2} = 2.4\text{e-}4 \text{ gm/cm}^3$, $A_t \sim 0.5$. The shock was specified with a pressure ratio of ~ 1.51 or $Ma = 1.2$. The initial temperatures in regions II and III in figure 20 were specified to be 300 K and 1625 K respectively. The computations were performed in 2D, in a numerical shock tube with an aspect ratio of 6, employing a uniform grid of resolution $256/\lambda$ in x and y directions, with $\lambda = 6 \text{ cm}$ in equation (41). Outflow and reflecting boundary conditions were enforced at $x = 0$ and $x = 6 \lambda$, while the lateral walls were modeled as periodic boundaries. The reflecting wall at $x = 6 \lambda$ ensures the transmitted shock rebounds from this surface, and impinges on the interface a second time leading to greater mixing, a phenomenon referred to as

reshock. Finally, no explicit diffusion was employed, as the simulations were performed in ILES mode.

For $A_t > 0$, the interface amplitude is expected to grow linearly according to [46],

$$V_0 = k U A_t h_0, \quad (44)$$

where $k = 2\pi/\lambda$ is the wave number, $h_0 = 0.2/k$ is the initial amplitude, $A_t = 0.5$ is the Atwood number and $U \sim 3.0e4$ cm/s is the interface velocity calculated using the Rankine-Hugoniot conditions (see for example [47]). Figure 21 is a plot of mass fraction of H_2O , scaled density ($\rho^* = \frac{\rho - \rho_{H_2}}{\rho_{O_2} - \rho_{H_2}}$) and temperature ($T^* = \frac{T - T_{H_2}}{T_{O_2} - T_{H_2}}$) realized at non-dimensional times (kV_0t) of (a) 0.0 (b) 1.6 (c) 2.0 (d) 3.0, (e) 4.0 and (f) 8.0 where $V_0 \approx 2500$ cm/s from equation (44). The solid line in each figure represents the iso-contour of 50% of the mixture fraction (Z_{50}). The incident shock compresses the interface to post shock amplitude (based on Z_{50}) $\sim 0.8 h_0$ by $kV_0t \sim 1.2e-2$, following which the interface amplitude grows. By $kV_0t = 0.4$, the maximum temperature across the interface is ~ 3250 K comparable to the adiabatic flame temperature of an $H_2 - O_2$ interface ~ 3150 K. The compression of the interface due to shock reduces the interface thickness and appears to increase the mass flow rate of fuel into the flame, leading to the high levels of heat addition. In the classical nonreacting RMI, when the modal amplitude violates $kh < 1$, higher harmonics saturate the growth rate (V_0) marked by the appearance of ‘bubbles’ of light fluid penetrating the heavy, while jets of the heavy fluid are referred to as ‘spikes’. In the reacting RMI presented here, by $kV_0t = 1.6$ (figure 21 (b)), the bubble and spike fronts appear with the bubbles forming a rounded surface, while spikes are slightly elongated. At this time, the reflected shock of strength $Ma \sim 1.25$ travelling from O_2 to H_2 interacts with the interface but in the heavy to light configuration corresponding to $A_t \sim -0.51$. As a result, the interface first undergoes an indirect phase inversion, following which it continues to grow with a negative growth rate, since $A_t < 0$ in equation (44). Mixing is enhanced by the activation of

higher harmonics and small-scale corrugations visible in figures 21 (c) and (d). At late times figures 21 (e) – (f), significant small-scale structure is present leading to widespread mixing throughout the flame brush. The site of such aggressive mixing is visible in figure 21 (f) through the presence of the product of combustion (H_2O).

To characterize the effect of RMI on the flame we define a (1) mixing layer width based on the mixture fraction and (2) an integrated heat release rate (HRR). The mixing layer width (W) is an integral measure of the extent of penetration of one fluid in to another, and is defined as

$$W / \int_0^\infty 4 \langle Z(1 - Z) \rangle dx \quad (45)$$

where the prefactor within the integral ensures the integrand evaluates exactly to the width when a triangular waveform for Z is assumed. The integral heat release rate is defined according to,

$$C / \int_0^\infty \frac{\langle e_g \rangle}{\Delta t} dx \quad (46)$$

where e_g is the heat added per unit mass (equation (37)) due to combustion over a simulation timestep (Δt), and $\langle \cdot \rangle$ indicates planar averaging along the y - direction. Obviously, C is large where products of combustion are formed, and $C \rightarrow 0$ in the pure fluids or in the regions where combustion has already proceeded to completion. The integral heat release rate C thus serves as a measure of the extent of heat addition, and the associated combustion efficiency.

The integral mix width W was computed from equation (45) with Z obtained from our 2D simulations described above, and is shown in figure 22 (a) where it is plotted against the non-dimensional time kV_0t . The evolution of W shows two distinct stages corresponding to initial RMI growth from the incident shock ($0 \leq kV_0t \leq 1.6$), and the post-reshock growth of the mixing layer ($kV_0t > 1.6$). While there is a phase inversion of the original signal (seen in figures 21 (a) –

(f)), the integral measure W is insensitive to this transition. In figure 22 (b), we show the time evolution of $\frac{C(t)}{C(t=t_s)}$, where t_s is the time of arrival of the first incident shock at the interface. Thus at $t = t_s$, the scaled heat release rate reaches its maximum value as the shock compression leads to burning of H_2 present along the monotonic, diffuse interface. However, this peak is short-lived, and appears over a time scale comparable to the shock-interaction time, following which the integral from equation (46) is vanishingly small. Following reshock ($kV_0t = 1.6$), the heat release rate reaches a finite value ($\sim 30\%$ of the initial peak), and is sustained through the remainder of the simulation due to the presence of small-scale structures which have grown out of higher harmonics present at the time of reshock. A gradual decay is visible for $kV_0t > 4$, and is expected, since the presence of combustion products (figure 21 (f)) provides a barrier to further diffusion and chemical reactions between H_2/O_2 on either side. These preliminary results of the reshocked reacting RMI suggest the combustion efficiency could be altered by RMI, and merits further study.

4.6 Parallel Performance Studies:

We performed a detailed performance analysis to determine the impact of code modifications described here on the parallel performance and scalability of FLASH. The problem setup used for this study is the same as that described in § 4.5, except outflow boundary conditions were employed at $x = 12\lambda$ in our numerical shock tube. The simulations were evolved till a scaled time $kV_0t = 1$ and repeated for different values of N_{proc} , where N_{proc} is the number of parallel processors involved in the calculation. In our scaling study, we maintain a uniform grid resolution of 256 zones/ λ in x and y directions, thus ensuring constant total work, while the work per processor is allowed to vary. For the scaling study, the I/O unit in FLASH responsible for writing checkpoint and plot files was deactivated. Figure 23 is a plot of execution time to completion plotted against N_{proc} for FLASH and an *ideal* simulation for which the

execution time is in perfect inverse proportionality to N_{proc} . We find linear parallel scaling for $N_{\text{proc}} < 1000$ above which the execution time for FLASH diverges slightly from the ideal, leading us to conclude the parallel performance of FLASH does not suffer significantly as a result of the modifications described here.

5. SUMMARY AND CONCLUSIONS

We have described modifications to the compressible flow code FLASH that allow for accurate description of chemically reacting flows with heat addition. In this article, we have reported in detail several simulations intended as validation cases to test the added capabilities. Our development work includes the addition of a chemistry library with detailed reaction mechanisms to describe H_2 -Air and CH_4 -Air chemistry, extension of the materials database to handle temperature-dependent transport properties, extension of the equation of state module to handle temperature-dependent adiabatic indices of mixtures, extension of the diffusion module to include diffusion of mass and momentum, and modification of existing ODE integrators to solve for Arrhenius reaction rates in a chemical reaction network. Through the development process, we have verified the massively parallel performance is not compromised.

For all the test cases reported in this paper, the reacting flow setup coupled to FLASH produces results in good agreement with published experimental and numerical data. FLASH simulations of an advecting, 1D front (non-reacting and reacting) produce solutions free of oscillations, using the flux-based or implicit diffusion methods, while directional symmetry is preserved in the corresponding 2D flow. A 2D Methane-Air Bunsen flame was also simulated in cylindrical coordinates, and the results are in good agreement with experimental [35] data. These simulations were performed in DNS mode with the inclusion of diffusion effects through the methods described earlier, and serve to validate the compressible code in a low-speed flow. The modified version of FLASH is also well suited to describe high-speed, chemically reacting flows, including flows where shocks are present and might interact with a flame. We have

reported two such cases, namely an H_2 bubble in air with shock impingement, and a reacting RMI flow subject to an incident and reflected reshock. In both cases, combustion across an initial interface is initiated by shock compression, while the reshock in the RMI flow produces significant small-scale mixing accompanied by product formation through the flame brush region.

We close with a brief discussion of potential future research directions using the enhanced software. While we have reported on examples of reacting flow calculation using the ILES approach[48], further detailed investigations examining the limits and capabilities of this approach to such flows is planned. In particular, a detailed characterization of the effect of numerical diffusion on properties of the solution is a necessary first step, before such methods can gain serious consideration in the combustion community. FLASH provides a convenient platform to make this evaluation, since the same code can be operated in both ILES and DNS modes. Similarly, the availability of multiple advection schemes in FLASH render it a suitable test bed to evaluate and characterize the accuracy of such schemes when applied to reacting flows. While all of the simulations described here used PPM, FLASH is also equipped with MUSCL-Hancock, WENO5 schemes, and we have planned studies examining how these methods perform when applied to reacting flows with sharp interfaces and shocks. Finally, we plan to implement so-called characteristic outflow boundary conditions appropriate for such multi-component reactive flows[49].

ACKNOWLEDGEMENTS:

The work reported in this article was supported in part by the (U.S.) Air Force Research Laboratory under Contract No. FA8650-10-C-2008. N.A. was partially supported by a scholarship grant from Energy Production and Infrastructure Center (EPIC) at UNC Charlotte. FLASH was developed in part by the DOE-sponsored ASC/Alliance Center for Astrophysical

Thermonuclear Flashes at the University of Chicago. This manuscript has been cleared for public release (No. 88ABW-2013-1795).

The authors are grateful to Professor Krishnan Mahesh for allowing his data to be included for comparison in figures 2 (a) and (b).

APPENDIX: CONVERGENCE STUDY

In this section, we present results from grid convergence studies performed in 1D and 2D using the modified version of FLASH. In 1D, we demonstrate convergence for the reacting, H_2-O_2 advection front described in § 4.2 (case 6), for $\Delta x = 0.04, 0.02, 0.01$ and 0.005 cm. In figures A.1 (a) – (c), we plot the x-profiles of Temperature at $t = 25, 50$ and $100 \mu s$ respectively. The corresponding time evolution of the peak temperature is shown in figure A.2 for all four simulations at different Δx . The oscillations observed in the coarse grid simulations appear to be numerical, and are absent in the highly resolved simulations. In figures A.1 – A.2, good convergence is observed for the highest resolution simulations with $\Delta x \leq 0.01$.

The grid convergence test was extended to 2D for a single mode chemically reacting Richtmyer-Meshkov instability (problem statement described in § 4.5). The number of zones spanning a single perturbation wavelength was systematically varied through $64/\lambda, 128/\lambda, 256/\lambda$ and $512/\lambda$, where λ is the wavelength associated with the initial perturbation. Figure A.3 is a plot of the mix width W (from eq. 45) against scaled time ($kV_0 t$) for the four resolutions employed, and shows convergence of the mix width as the grid resolution is increased. Slight variations are observed at late times ($kV_0 t > 3$), due to the appearance of small-scale, secondary instabilities which retain some grid dependence, while the amplitudes of penetration of the mixing layer have converged with respect to the grid resolution.

REFERENCES

- [1] Fryxell B, Olson K, Ricker P, Timmes F, Zingale M, Lamb D, et al. FLASH: An adaptive mesh hydrodynamics code for modeling astrophysical thermonuclear flashes. *Astrophys J Suppl Ser.* 2000;131:273.
- [2] Billet G. Improvement of convective concentration fluxes in a one step reactive flow solver. *J Comput Phys.* 2005;204:319-52.
- [3] Mueller M, Kim T, Yetter R, Dryer F. Flow reactor studies and kinetic modeling of the H₂/O₂ reaction. *Int J Chem Kinet.* 1999;31:113-25.
- [4] Katta V, Roquemore W. Numerical studies on the structure of two-dimensional H₂/air premixed jet flame. *Combust Flame.* 1995;102:21-40.
- [5] Smooke MD. Reduced Kinetic Mechanisms and Asymptotic Approximations for Methane-Air Flames: A Topical Volume. *Lecture Notes in Physics.* 1991;384.
- [6] Kaps P, Rentrop P. Generalized Runge-Kutta methods of order four with stepsize control for stiff ordinary differential equations. *Numerische Mathematik.* 1979;33:55-68.
- [7] Bader G, Deuflhard P. A semi-implicit mid-point rule for stiff systems of ordinary differential equations. *Numerische Mathematik.* 1983;41:373-98.
- [8] Colella P, Woodward PR. The piecewise parabolic method (PPM) for gas-dynamical simulations. *J Comput Phys.* 1984;54:174-201.
- [9] Fureby C, Grinstein F. Monotonically integrated large eddy simulation of free shear flows. *AIAA J.* 1999;37:544-56.
- [10] Thornber B, Mosedale A, Drikakis D. On the implicit large eddy simulations of homogeneous decaying turbulence. *J Comput Phys.* 2007;226:1902-29.
- [11] Porter D, Pouquet A, Woodward P. Three-dimensional supersonic homogeneous turbulence: A numerical study. *Phys Rev Lett.* 1992;68:3156-9.
- [12] MacNeice P, Olson KM, Mobarri C, de Fainchtein R, Packer C. PARAMESH: A parallel adaptive mesh refinement community toolkit. *Comput Phys Commun.* 2000;126:330-54.
- [13] FLASH ASC. Flash user's guide. Chicago: University of Chicago; 2010.
- [14] Godunov SK. A difference method for numerical calculation of discontinuous solutions of the equations of hydrodynamics. *Matematicheskii Sbornik.* 1959;89:271-306.
- [15] Duff IS, Erisman AM, Reid JK. Direct methods for sparse matrices: Clarendon Press Oxford; 1986.
- [16] Smith GP, Golden DM, Frenklach M, Moriarty NW, Eiteneer B, Goldenberg M, et al. GRI-Mech 3.0. URL: http://www.me.berkeley.edu/gri_mech. 1999.
- [17] Linstrom PJ, Mallard WG. The NIST Chemistry WebBook: A chemical data resource on the internet. *J Chem Eng Data.* 2001;46:1059-63.
- [18] Schlichting H, Gersten K. Boundary-layer theory: Springer Verlag; 2000.
- [19] Babu PC, Mahesh K. Upstream entrainment in numerical simulations of spatially evolving round jets. *Phys Fluids.* 2004;16.
- [20] Lugt HJ. Introduction to vortex theory: Vortex Flow Press Potomac; 1996.
- [21] McBride BJ, Gordon S, Reno MA. Coefficients Thermodynamic Properties for Calculating and Transport Species of Individual. NASA Tech Memo. 1993;4513.
- [22] Mathur S, Tondon P, Saxena S. Thermal conductivity of binary, ternary and quaternary mixtures of rare gases. *Mol Phys.* 1967;12:569-79.
- [23] Wilke C. A viscosity equation for gas mixtures. *J Chem Phys.* 1950;18:517.

- [24] Poling BE, Prausnitz JM, John Paul OC. The properties of gases and liquids: McGraw-Hill New York; 2001.
- [25] Burcat A, Ruscic B, Laboratory AN. Third millenium ideal gas and condensed phase thermochemical database for combustion with updates from active thermochemical tables: Argonne National Laboratory Argonne, IL; 2005.
- [26] Miller JA, Mitchell RE, Smooke MD, Kee RJ. Toward a comprehensive chemical kinetic mechanism for the oxidation of acetylene: Comparison of model predictions with results from flame and shock tube experiments*. Elsevier; 1982. p. 181-96.
- [27] Doom J, Mahesh K. Direct numerical simulation of auto-ignition of a hydrogen vortex ring reacting with hot air. *Combust Flame*. 2009;156:813-25.
- [28] Takahashi F, Mizomoto M, Ikai S, Veziroglu T. Nuclear energy/synthetic fuels. *Alternative energy sources III* (Nejat Veziroglu T, Ed). 1983;5:447-57.
- [29] Gordon RL, Masri AR, Pope SB, Goldin GM. Transport budgets in turbulent lifted flames of methane autoigniting in a vitiated co-flow. *Combust Flame*. 2007;151:495-511.
- [30] Hermanns RTE, Kortendijk J, Bastiaans R, de Goey L. Laminar burning velocities of methane-hydrogen-air mixtures: Universal Press; 2007.
- [31] Chen CP. Structure and Stabilization Mechanism of Microjet Methane Diffusion Flames [PhD]. Tainan, Taiwan: National Chen-Kung University; 2007.
- [32] Attal N. Development, Verification and Validation of a chemical reaction solver coupled to the Piecewise Parabolic Method [Masters]. Charlotte: University of North Carolina; 2012.
- [33] CERFACS. Adiabatic Flame Temperature Calculator. URL: <http://elearningcerfacsfr/combustion/tools/adiabaticflametemperature/index.php>. 2012.
- [34] Billet G, Abgrall R. An adaptive shock-capturing algorithm for solving unsteady reactive flows. *Comput Fluids*. 2003;32:1473-95.
- [35] Nguyen Q, Dibble R, Carter C, Fiechtner G, Barlow R. Raman-LIF measurements of temperature, major species, OH, and NO in a methane-air Bunsen flame. *Combust Flame*. 1996;105:499-510.
- [36] Zhou X, Brenner G, Weber T, Durst F. Finite-rate chemistry in modelling of two-dimensional jet premixed CH₄/air flame. *Int J Heat Mass Transfer*. 1999;42:1757-73.
- [37] Tsatsaronis G. Vereinfachung des Methan/Sauerstoff Reaktionsmechanismus, *Brenn. Wärme-Kraft*. 1978;30:249-55.
- [38] Billet G, Giovangigli V, de Gassowski G. Impact of volume viscosity on a shock–hydrogen-bubble interaction. *Combust Theory Modell*. 2008;12:221-48.
- [39] Nakai S, Takabe H. Principles of inertial confinement fusion-physics of implosion and the concept of inertial fusion energy. *Rep Prog Phys*. 1996;59:1071.
- [40] Arnett WD. Supernova theory and supernova 1987A. *Astrophys J*. 1987;319:136-42.
- [41] Yavuz I, Celik I, McMillian M. Knock Prediction in Reciprocating Gas-Engines Using Detailed Chemical Kinetics. SAE Technical Paper; 2001.
- [42] Sivabalakrishnan R, Jegadheesan C. Study of Knocking Effect in Compression Ignition Engine with Hydrogen as a Secondary Fuel. *Chinese Journal of Engineering*. 2014;2014.
- [43] Khokhlov AM, Oran ES, Chtchelkanova AY, Wheeler JC. Interaction of a shock with a sinusoidally perturbed flame. *Combust Flame*. 1999;117:99-116.
- [44] Massa L, Jha P. Linear analysis of the Richtmyer-Meshkov instability in shock-flame interactions. *Phys Fluids*. 2012;24:056101.
- [45] Haehn N, Ranjan D, Weber C, Oakley J, Rothamer D, Bonazza R. Reacting shock bubble interaction. *Combust Flame*. 2012;159:1339-50.
- [46] Richtmyer RD. Taylor instability in shock acceleration of compressible fluids. *Commun Pure Appl Math*. 1960;13:297-319.

- [47] Mikaelian KO. Freeze-out and the effect of compressibility in the Richtmyer–Meshkov instability. *Phys Fluids*. 1994;6:356.
- [48] Grinstein FF, Len G. Margolin,, Rider WJ. *Implicit Large Eddy Simulation*: Cambridge University Press; 2007.
- [49] Baum M, Poinso T, Thévenin D. Accurate boundary conditions for multicomponent reactive flows. *J Comput Phys*. 1995;116:247-61.

TABLE 1: Summary of capabilities added to FLASH for simulating chemically reacting flows with heat addition.

TABLE 2: FLASH simulations of the advection of a 1D, non-reacting, H_2 - O_2 front.

TABLE 3: FLASH simulations of the advection of a 1D, reacting, H_2 - O_2 front.

TABLE 4: Conditions for FLASH simulations of premixed CH_4 -Air bunsen flame.

TABLE 5: Shock-bubble combustion simulations performed using FLASH.

FIGURE 1: (a) Temporal evolution of temperature from a well-stirred reactor simulation using reaction mechanism of [2] and (b) adiabatic flame temperatures from FLASH using the Hydrogen-Air mechanism from [3] compared with data from CERFACS[33].

FIGURE 2: FLASH results of evolution of (a) mass fraction of hydrogen and (b) temperature in a well-stirred reactor using the H_2 - O_2 mechanisms of Billet[2] and Mueller[3] as adapted by [27] compared with the DNS results of Doom and Mahesh[27].

FIGURE 3: Hydrogen mass fraction profiles for cases 1 - 3 alongside the DNS of [34] at $t = 400 \mu s$.

FIGURE 4: Profiles of (a) temperature, (b) Y_{H_2O} , (c) Y_{O_2} and (d) Y_{H_2} from case 4 alongside the results from the double-flux simulations [34] at $t = 100 \mu s$.

FIGURE 5: Mass fraction profiles (H_2 , O_2 , and H_2O) and temperature from case 5 alongside the results from the double-flux simulations of [34] at $t = 100\mu s$.

FIGURE 6: Mass fraction profiles (H_2 , O_2 , and H_2O) and temperature from case 6 alongside the results from the double-flux simulations of [34] at $t = 100\mu s$.

FIGURE 7: Schematic of the computational domain for a 2D CH_4 -Air bunsen flame (cases 7 – 9).

FIGURE 8: Temperature contour plot achieved at steady-state from simulation of a premixed CH_4 -Air flame at an equivalence ratio of 1.7 (case 9).

FIGURE 9: Steady-state centerline (axial) profiles of (a) mole fractions of CH_4 , O_2 and N_2 , and (b) mole fractions of CO_2 , H_2O and temperature from case 7. Data is compared with experiments of [35].

FIGURE 10: Steady-state radial profiles of (a) mole fractions of CH_4 , O_2 and N_2 , and (b) mole fractions of CO_2 , H_2O and temperature at $z = 2.1$ cm from case 7. Data is compared with experiments of [35].

FIGURE 11: Steady-state radial profiles of (a) mole fractions of CH_4 , O_2 and N_2 , and (b) mole fractions of CO_2 , H_2O and temperature at $z = 5.0$ cm from case 7. Data is compared with experiments of [35].

FIGURE 12: Steady-state radial profiles of (a) mole fractions of CH_4 , O_2 and N_2 , and (b) mole fractions of CO_2 , H_2O and temperature at $z = 9.0$ cm from case 7. Data is compared with experiments of [35].

FIGURE 13: Steady-state centerline (axial) profiles of (a) mole fractions of CH_4 , O_2 and N_2 , and (b) mole fractions of CO_2 , H_2O and temperature from case 8. Data is compared with experiments of [35].

FIGURE 14: Steady-state centerline (axial) profiles of (a) mole fractions of CH_4 , O_2 and N_2 , and (b) mole fractions of CO_2 , H_2O and temperature from case 9. Data is compared with experiments of [35].

FIGURE 15: Schematic of the initial configuration and boundary conditions for the reacting shock-bubble simulations.

FIGURE 16: Line contours of pressure (1 – 7.37 atm) superposed on H_2 mass fraction (grayscale) at $1.5\mu\text{s}$ for (a) case 10 (flux-based diffusion) and (b) case 11 (implicit diffusion).

FIGURE 17: Line contours of pressure (1 – 7.37 atm) superposed on H_2 mass fraction (grayscale) at $3.5\mu\text{s}$ for (a) case 10 (flux-based diffusion) and (b) case 11 (implicit diffusion).

FIGURE 18: Pressure across $y = 0.0\text{ cm}$ at $t = 3.5\mu\text{s}$ for cases 10 (flux-based diffusion) and 11 (implicit diffusion) compared with results of [38].

FIGURE 19: Contours of density in gm/cm^3 (top) and H_2O mass fraction (bottom) at $t =$ (a) $10\mu\text{s}$ (b) $20\mu\text{s}$ (c) $30\mu\text{s}$ and (d) $40\mu\text{s}$.

FIGURE 20: Schematic of the RMI problem setup showing initial location of the shock (x_s) and a single mode, diffuse $\text{H}_2\text{-O}_2$ interface.

FIGURE 21: Time evolution of (top to bottom) the scaled density ρ^* , H_2O mass fraction and the scaled temperature T^* contours from 2D simulations of a perturbed, reacting interface with $\frac{\Delta}{h_0} = 2$, and an incident Mach 1.2 shock. The images are realized at $kV_0t =$ (a) 0, (b) 1.6, (c) 2.0, (d) 3.0, (e) 4.0 and (f) 8.0. Reshock occurs at $kV_0t = 1.6$

FIGURE 22: (a) Mixing layer width (W) and (b) integral heat release rate (C) normalized by its value at shock arrival time (t_s) plotted against scaled time (kV_0t).

FIGURE 23: Plot of execution times (t_{exec}) for reacting RMI against number of processors (N_{proc}). The solid line represents the trendline from an ideal simulation.

FIGURE A.1: Temperature profiles realized at $t =$ (a) 25, (b) 50 and (c) 100 μs observed in numerical simulations of advection of a 1D reacting $\text{H}_2\text{-O}_2$ front using $\Delta x = 0.04, 0.02 \text{ cm}, 0.01 \text{ cm}$ and 0.005 cm .

FIGURE A.2: Time evolution of peak temperatures from a 1D $\text{H}_2\text{-O}_2$ reacting front from numerical simulations with $\Delta x = 0.04, 0.02 \text{ cm}, 0.01 \text{ cm}$ and 0.005 cm .

FIGURE A.3: Mix width (W) plotted against scaled time (kV_0t) from simulations of a single-mode Richtmyer-Meshkov Instability at grid resolutions of 64 zones/ λ , 128 zones/ λ , 256 zones/ λ and 512 zones/ λ .

Table 1

Relevant Physics	Existing	Added
Reaction Kinetics	Nuclear reaction networks	H ₂ -Air and CH ₄ -Air
Multi-species Equation of State	Temperature independent	Temperature dependent
Thermal Diffusion	Flux-based Implicit solver	-
Viscous Diffusion	Flux-based	Extended Implicit solver
Mass Diffusion	Flux-based	Extended Implicit solver
Temperature dependent Material properties	-	Species involved in combustion of H ₂ & CH ₄ (Thermodynamic & Transport)

Table 2

CASE	Mass Diffusion	Thermal Diffusion	Viscous Diffusion	Lewis no. Le	Δx cm
1	Numerical	Numerical	Numerical	-NA-	0.0025
2	Implicit	Implicit	Implicit	1	0.01
3	Flux-based	Flux-based	Flux-based	1	0.0025

Table 3

CASE	Mass Diffusion	Thermal Diffusion	Viscous Diffusion	Lewis no. Le	Δx cm
4	Numerical	Numerical	Numerical	-NA-	0.0025
5	Flux-based	Flux-based	Flux-based	1	0.0025
6	Implicit	Implicit	Implicit	1	0.01

Table 4

CASE	Equivalence Ratio	CH ₄ (slm)	Air (slm)	Inlet Velocity(U_1) cm/s
7	1.38	0.97	6.69	56
8	1.52	1.05	6.63	56
9	1.70	1.17	6.57	57

Table 5

CASE	Δx Cm	Δy Cm	CFL	AMR	T_{H_2} (K)	Mass Diffusion	Thermal Diffusion	Viscous Diffusion
10	2.34e-3	2.34e-3	0.25	YES	1000	Numerical	Flux- based	Flux- based
11 (a)	2.34e-3	2.34e-3	0.25	NO	1000	Numerical	Implicit	Implicit
11 (b)	2.34e-3	2.34e-3	0.25	NO	1000	Implicit	Implicit	Implicit

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure 13

Figure 14

Figure 15

Figure 16

Figure 17

Figure 18

Figure 19

Figure 20

Figure 21

Figure 22

Figure 23

Figure A1

Figure A2

Figure A3