

Experimental and Numerical Investigations of Thermal Ignition of a Phase Changing Energetic Material

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ABSTRACT

Fortuitous exposure to high temperatures initiates reaction in energetic materials and possibilities of such event are of great concern in terms of the safe and controlled usage of explosive devices. Experimental and numerical investigations on time to explosion and location of ignition of a phase changing polymer bonded explosive material (80 per cent RDX and 20 per cent binder), contained in a metallic confinement subjected to controlled temperature build-up on its surface, are presented. An experimental setup was developed in which the polymer bonded explosive material filled in a cylindrical confinement was provided with a precise control of surface heating rate. Temperature at various radial locations was monitored till ignition. A computational model for solving two dimensional unsteady heat transfer with phase change and heat generation due to multi-step chemical reaction was developed. This model was implemented using a custom field function in the framework of a finite volume method based standard commercial solver. Numerical study could simulate the transient heat conduction, the melting pattern of the explosive within the charge and also the thermal runaway. Computed values of temperature evolution at various radial locations and the time to ignition were closely agreeing with those measured in experiment. Results are helpful both in predicting the possibility of thermal ignition during accidents as well as for the design of safety systems.

Keywords: Thermal ignition, cookoff, explosive safety, reactive heat flow simulations, explosive device

NOMENCLATURE

E_i	Activation energy
C_p	Specific heat
H	Sensible enthalpy
h_{ref}	Reference enthalpy
k	Thermal conductivity
S	Chemical source term
T	Temperature
t	Time
β	liquid fraction

1. INTRODUCTION

An energetic material in confinement may absorb heat energy from surrounding warm environment and can undergo chemical decomposition and explosion. This phenomenon is often termed as thermal cookoff and such eventualities are of great concern among explosives' research community. Therefore reliable data for time to ignition and location of ignition are essential for safeguarding the ammunitions. Various theoretical, experimental, and simulation studies have been reported in the past few decades exploring the physics of thermal ignition of explosives¹⁻⁵. Physics of thermal ignition involves combination of many coupled processes such as heat transfer, chemical reaction, deformation, phase change, etc.

In accident scenarios, the heat from surrounding hot ambient is carried in to the exterior of the container of explosive device by radiation and convection and then conducted into the explosive material through its outer confinement case. Thereafter the explosive charge undergoes the process of thermal decomposition initiated by such energy additions. Cookoff can be classified as fast cookoff or slow cookoff based on the nature of imparting heat. A fast cookoff may happen due to sudden heating by surrounding pool of fire whereas a slow cookoff is often triggered by a continuous heating process by warm ambient. According to NATO Standardisation Agency (NSA) guidelines, Allied Ordnance Publication (AOP-39-STANAG 4439), temperature build up of 1 K to 30 K per hour (due to fire in an adjacent magazine, store or vehicle) forms the baseline threat range for slow cookoff. Experimentation as well as modelling and simulation of thermal ignition of energetic material are quite helpful to ascertain the possibility of occurrence of ignition in accident scenarios. Such analyses are also helpful in evaluating the extent of damage of energetic material due to heating, the influence of geometry of confinement in time to explosion, and the nature of reaction violence. Nowadays, the design of safety mechanisms such as vents are being developed based on the data obtained from simulation of heating phase of the cookoff.

Objective of the present work is to simulate thermal ignition process of a phase changing polymer bonded explosive material in a cylindrical confinement subjected to

rapid heating conditions corresponding to the fast cook off. A phase changing polymer bonded explosive which contains 80 per cent RDX and 20 per cent polyurethane was used in the present experimental study. This is prepared by hand mixing in small quantities and then followed by curing in an oven. An experiment set up was designed and fabricated to perform the controlled heating conditions. A PID ramp rate controller was used to achieve a controlled heating rate on surface. Location of initiation of thermal runaway depends on the rate of heat applied on confinement's surface. Therefore the thermal response of energetic material is measured at various radial locations till ignition.

The numerical simulation of cook off explosion is quite challenging due to the widely differing time scales corresponding to the modelling of heat transfer and chemical reaction. In addition to this, energetic material undergo phase change during heating and self sustained reaction pools (hot spots) are being developed at isolated locations which finally leads to the thermal runaway. A computational model for solving heat transfer with phase change and chemical reaction was developed and implemented in the framework of a standard commercial package (Ansys FLUENT) using a user defined function developed for this purpose. The governing equations for unsteady heat transfer with phase change and chemical reaction have been solved using finite volume method. reaction modelling for the thermal decomposition of RDX was carried out using a three step Arrhenius model chemical kinetics. Heat generation effect was implemented using a user custom field function and solved along with the unsteady heat transfer solver. Boundary condition corresponding to the rapid heating conditions on surfaces was implemented using a user defined function to update temperature on surface based on the prescribed heating rate. Post processing of the simulation data provides pattern of melting and temporal evolution of temperature gradients in reactive material.

2. LITERATURE REVIEW

Thermal explosion of energetic materials is a complex phenomenon comprising of a series of physiochemical process and its characteristic are dependent on heating rate, sample geometry, physical and chemical properties of the material, strength of the radial and axial confinement, etc. The level of vigor that explosion may exhibit depend on the aforementioned aspects. Therefore experimental confirmation is essential in establishing the nature cookoff and its outcome for different ambient heating conditions. Experiments are often performed with preset nature of confinement and boundary heating to ascertain the predictability of computational tools used to simulate the response of munitions with similar geometry undergoing cookoff. Wardell⁷, *et al.* carried out a scaled thermal explosion experiment (STEX) by providing uniform heating for well defined and controlled boundary conditions. Temperature history was obtained at different internal and external locations. Radial and axial strains in the vessel wall were also measured, even at the onset of explosion, so as to quantify reaction violence. Chidester⁸, *et al.* carried out large-scale annular cookoff (LSAC) tests investigate the symmetry and violence of a cookoff event arising from slow, symmetric

heating of a moderately heavily confined annular test assembly filled with polymer bonded explosive. Kaneshige⁹, *et al.* studied the temperature field inside the energetic material, as it approaches ignition, using the Sandia instrumented thermal ignition (SITI) apparatus. Sandusky and Chambers¹⁰ could provide temperature, pressure, and volume information until the onset of reaction using variable confinement cookoff test (VCCT). Cheese¹¹, *et al.* used electrically heated tube test (EHTT) to study the effect of different thermal profiles during slow/fast cookoff on the violence of reaction.

The theoretical and numerical analysis are capable of bringing more insight of the physio-chemical process during cookoff. Shouman² and Merzhanov³ had given comprehensive reviews of conventional thermal explosion theory and modelling. There are inherent limitations for carrying out numerical simulations due to the presence of multi-scale processes involved in this phenomenon as well as nonlinearities arising due to temperature and species dependency of properties and physical state. An advanced Arbitrarily Lagrangian-Eulerian solver (ALE3D) developed by Lawrence Livermore National Laboratory forms a single platform for coupled heat transfer, mechanical and chemical simulations, capable of handling realistic boundary conditions. Yoh and McClelland⁵ performed modelling of heating, ignition, and explosion of PBX in STEX geometry using this solver. Asante⁶ recently simulated the thermal behaviour, including thermal decomposition, for TNT contained in actual ammunition geometries using CFD codes. TOPAZ2D is a two dimensional implicit finite element heat transfer solver developed by Lawrence Livermore National Laboratory. Chidester⁸, *et al.* used TOPAZ with appropriate chemical kinetic decomposition models for simulating heating phase of cookoff process. Kaneshige⁹, *et al.* performed heat transfer computations using COYOTE, a multidimensional, nonlinear heat conduction solver developed by Sandia National Laboratories. With advent of multi-physics solvers and accurate burn rate models and data^{12,13}, computational methods have been emerged as synergistically complementing tool for explosive safety experiments.

3. MODELLING OF REACTIVE HEAT FLOW

Computational model for the establishment of thermal ignition process of RDX are based on the following assumptions:

- Heat conduction is only present in explosives for both solid and liquid phases. Heat convection is assumed negligible due to absence of bulk fluid motion.
- The confinement wall, except the heating surface, is treated as adiabatic.
- Self-heating explosive reaction follows the Arrhenius reaction model.

The energy conservation equation for reactive heat flow in phase changing energetic material can be written as¹⁴

$$\frac{\partial}{\partial t}(\rho H) = \nabla \cdot (k \nabla T) + S_q \quad (1)$$

where k is a phase averaged conductivity worked out based on the linear variation of conductivity with volume fractions of individual phases. Enthalpy of material is computed as the sum

of the sensible enthalpy and the latent heat

$$H = \left(h_{ref} + \int_{T_{ref}}^T C_p dT \right) + \beta L \quad (2)$$

h_{ref} is reference enthalpy at a reference temperature T_{ref} and β is the liquid which can be computed as

$$\beta = 0 \text{ if } T < T_{solidus}$$

$$\beta = 1 \text{ if } T > T_{solidus}$$

$$\beta = \frac{T - T_{solidus}}{T_{Liquidus} - T_{Solidus}}$$

Thermal ignition is initiated by exothermic chemical reactions and this will accelerate if the rate of heat generation is exceeding the heat dissipation from the confinement. Inclusion of heat generation due to chemical reaction source term of energy equation is done using Tarver and McGuire Model¹⁵



Where A represents RDX, B represents $H_2C=N-NO_2$, C represents (CH_2O+N_2O) or $(HCN+HNO_2)$, and D represents the final products. Kinetics properties are given in Table 1. These chemical reactions follow the Arrhenius equation and the net heat production during the all the three reaction process is computed as

$$S_q = \sum_{i=1}^3 \rho_i Q_i Z_i \exp\left(-\frac{E_i}{RT}\right) \quad (4)$$

The Finite volume method (FVM) based solution of the present problem is obtained by integration of energy conservation Eqn. (1) for each control volume i as

$$\int_t^{t+dt} \left(\int_{CV} \frac{d(\rho H_i)}{dt} dV_i \right) dt = \int_t^{t+dt} \left(\int_{CV} \nabla \cdot (k \nabla T) dV_i \right) dt + \int_t^{t+dt} \left(\int_{CV} S_q dV_i \right) dt \quad (5)$$

Boundary of the confinement is heated in such a way that it results in a steady increase of temperature on its surface corresponding to a fast cookoff scenario. Boundary condition corresponding to this unsteady temperature increase is implemented in the present finite volume solver using a user defined function. A custom field function has been developed

Table 1. Kinetic parameters for thermal decomposition of RDX based PBX¹⁶

Reaction step	E (kJ/mol)	Z (S ⁻¹)	Q (kJ/kg)
$A \rightarrow B$	197.2	5.76×10^{19}	-418
$r_1 = \rho_A \times Z_1 \times e^{(-E_1/RT)}$			
$B \rightarrow 2C$	184.338	4.740×10^{17}	1254
$r_2 = \rho_B \times Z_2 \times e^{(-E_2/RT)}$			
$C \rightarrow D$	142.8	1.586×10^{15}	5016
$r_3 = \rho_C^2 \times Z_3 \times e^{(-E_3/RT)}$			

for computing heat generation based on Tarver and McGuire reaction model. A flow chart for the implementation of user defined function and custom field function is given in Fig. 1.

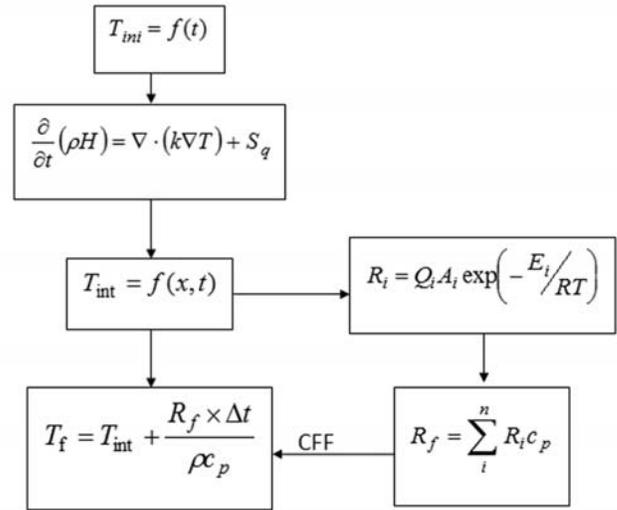


Figure 1. Flow chart for the implementation of user defined function and custom field function.

4. EXPERIMENTAL STUDY OF THERMAL IGNITION

Often the cook-off phenomena is experimentally simulated in laboratories with the different test conditions by making use of heat sources providing conductive, convective, radiative heat fluxes or a combinations of these. Energetic material used for the present study is a PBX (Polymer bonded Explosive) which contains 80 per cent RDX and 20 per cent polyurethane. The material sample is prepared manually by mixing in very small quantities. It is directly charged to the test confinement by compacting the powder gently. This is carried out with utmost care by experts with non-sparking tools in a room provided with electrostatic dissipative tile flooring. Later this sample is cured by heating in an oven for 5 days at 60 °C in an isolated location. For safety, the periodic calibration of this special purpose oven is ensured and all the possibility of shock loading of charge is eliminated during the entire process of preparation.

In present study, tests were carried out using pure conductive heat source using electric heater wire wrapped around the surface of a cylindrical confinement which provides temperature build up rate of 0.3 K/s. A PID ramp rate controller is used for realizing this prescribed temperature buildup rate. A thyristor supplies DC power input to the heater and controller ensures the surface temperature to be within $\pm 3K$ of programmed value. A data acquisition system was used to capture the unsteady temperature measured at various radial locations. A block diagram and photograph of the power supply, heat control and data acquisition system of test set-up are given in the Fig. 2.

The thermal ignition experiment was conducted on a newly designed apparatus solely to perform ignition study of energetic material. Apparatus consisting of a pair of rigid metallic fixtures to hold the cylindrical confinement horizontally as

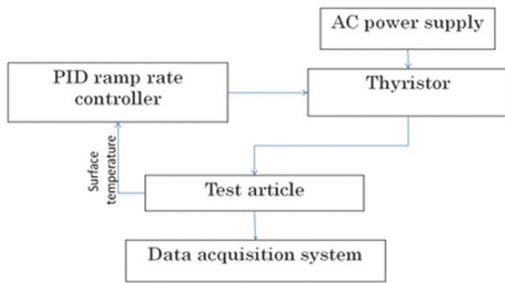


Figure 2. Block diagram of power supply, control and data acquisition system during heating trial.

shown in Fig. 3. The cylindrical confinement used to house the explosive is made of stainless steel 304 has a length of 80 mm and 14 mm diameter. Two end caps of outer diameter 25 mm and length 30 mm with (M20 × 1.5 mm internal thread) was fastened to this cylindrical confinement. These end caps has a through internal hole of 3 mm diameter to vent the gases and two identical aluminum sheets of 0.1mm thick were used to cover this to reduce the heat losses. Entire fixture assembly was installed in an explosion test pit following all necessary safety measures. Five K type thermocouples were used in the charge at different depths (axially at different locations) as shown in Fig. 3. Heating trials, without filling energetic material, for verification of required temperature buildup were performed. PID controller could maintain the required rate of 0.3 K/s based on the feedback of surface temperature in heating trials as well as in actual tests.

Major objectives of the experiments were to find the ignition time and the location of ignition point within the confinement. Temperature profiles provided by five thermocouples at various radial locations are given in Fig. 4.

Temperature plots shows the similar trend in temperature rise till melting starts. The non- uniform temperature distribution across the charge during the heating is evident from these temperature profiles . Thereafter the slopes of temperature profiles in the melt region in explosive becomes more flat. Temperature profile corresponding to 0.5 mm depth comes on the top which shows the rapid heating a difference of 18 K to 22 K was observed between internal point to external points resulted by uneven heat diffusion in energetic material. Detailed analysis of the data from temperature profile corresponding to 0.5 mm depth shows that ignition happened near to the surface of the confinement at 673.7 s and reaction started at 488.2 K.

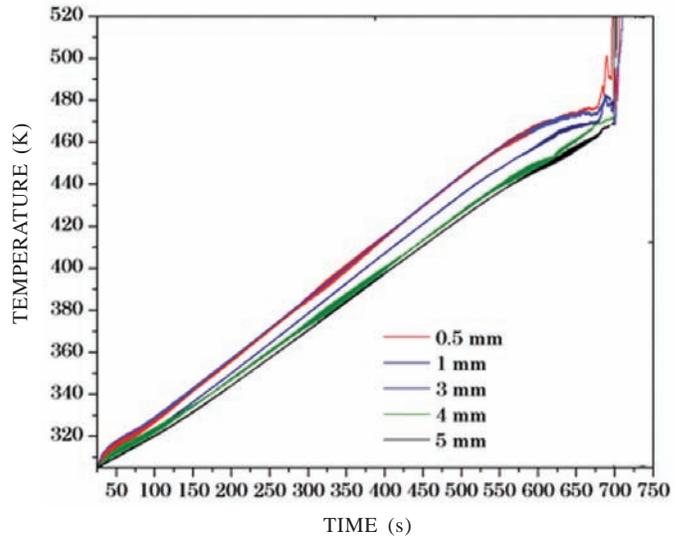


Figure 4. Temperature-time profiles at various radial locations

Photograph of post test article is given in Fig. 5, which has no indications of violent explosion. This indicates that due to the heating rate of 0.3 K/s resulted in the deflagration of energetic material which conforms to the results reported for fast cookoff tests.

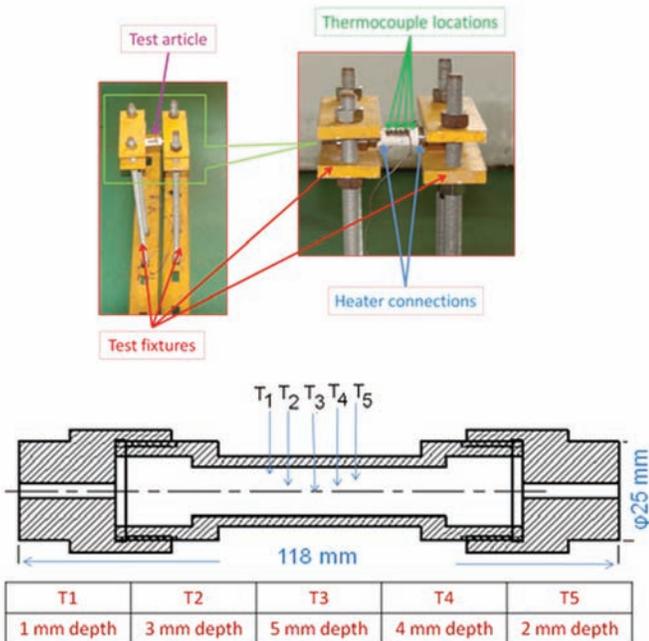


Figure 3. Fixture used for the experiment and details of thermocouple location.



Figure 5. Photograph of post test specimen.

5. NUMERICAL SIMULATION OF THERMAL IGNITION OF PHASE CHANGING ENERGETIC MATERIAL

Coupled unsteady heat transfer, phase change and chemical reaction simulation is carried out to study the thermal ignition of RDX. Objectives of these simulations include the

estimation the ignition time, locations of ignition and transient temperature response at various locations within the domain till ignition.

A two dimensional axisymmetric computational domain was chosen for simulating thermal ignition in cylindrical confinement containing explosive material. Numerical simulation has been carried out for a quarter of the actual geometry of the same test article chosen for the experiment. It is assumed that the radial and axial heat fluxes are predominant than that of heat flux in tangential direction in the actual object. Details of dimensions, boundary conditions applied for the simulation and a sample grid used for the analysis is given in Fig. 6. Details of the test conditions used for computational study are summarised in Table 2.

Table 2. Details of test object and conditions

Geometry	Confinement cylinder of total length 118 mm and maximum diameter 25 mm at the thread end.
Confinement material	Stain less steel
Energetic material	RDX
Properties of RDX	Density- 1800 Kg/m ³
	Specific heat -2000 J/kg K
	Thermal conductivity - 0.213 W/m K
	Enthalpy of fusion - 220 J/kg
Heating rate	0.3 K/s on 40 mm long lateral surface
	Initial temperature 300 K

5.1 Grid Independence Study and Validation

A grid independence study has been carried out by choosing control volume side lengths of 0.83 mm, 0.41 mm, 0.178 mm, and 0.08 mm in energetic material area in view of development of large temperature gradients in heated wall. Grid independent of the results has been checked with temperature profile for 0.8 mm depth. Figure 7(a) shows progressive grid refinement carried out to check independence of results and Fig. 7(b) gives the temperature profile at a location 0.8 mm below the heating surface. Grid independent temperature has been obtained for the second level of grid refinement onwards (0.41 mm to 0.08 mm) profile during the heating phase; whereas the post ignition temperature profile shows a minor deviation.

Figure 8 shows the comparison of experimentally measured temperature with that obtained in the simulation. Temperature build-up obtained in simulation for pre-ignition period is in good agreement that measured during the experiment. The sharp temperature build-up at the onset of ignition was not captured in the same manner that in the experiment. The simulated temperature profile provides reasonable agreement of predicted ignition time (670 s) and temperature (490 K) with that captured during the experiment. Deviation in post ignition temperature profile is attributed by the prescribed exponent term in in reaction model. Ignition time and temperature were worked out based on rate of temperature rise beyond that can be achieved by heat diffusion.

5.2 Pattern of Melt Front Advancement and Subsequent Ignition

Reactive heat flow simulation of phase changing energetic material is carried out by solving governing equations of

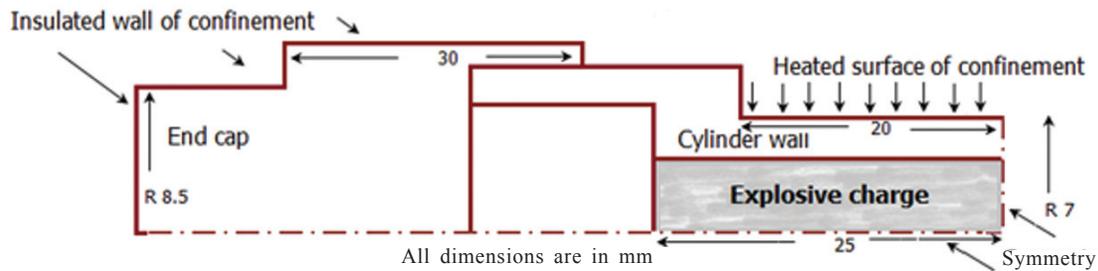


Figure 6. Computational domain and boundary conditions.

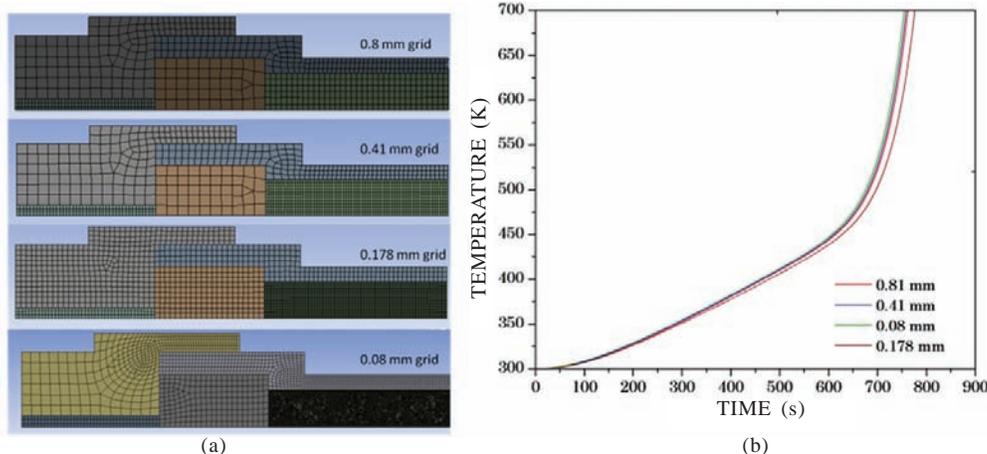


Figure 7. Grid independence study (a) Grid refinement (b) Temperature build-up.

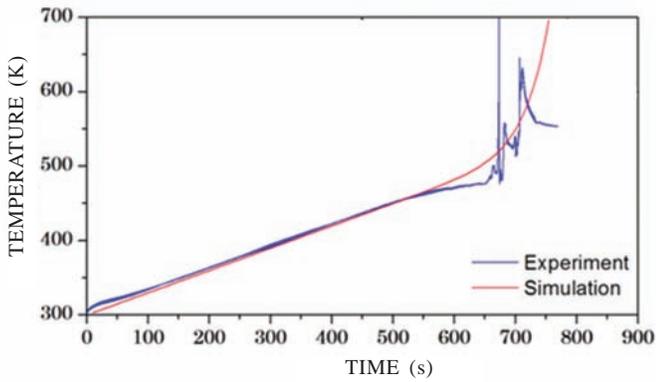


Figure 8. Comparison of temperature build-up in experiment and simulation.

unsteady two-dimensional heat conduction equations with chemical heat source terms. This could portray the evolution of the temperature inside the explosive with the advancement of time due to heating provided on its surface. The melting phenomena can be tracked by temperature-time curve, which shows a plateau region during the melting phase. Temperature build-up was continuously monitored at various radial locations and the advancement of melt front towards axis of the cylindrical charge is shown in Fig. 9. Explosive was initially in the solid phase at the ambient temperature. Analysis has been performed for constant heating rate of 0.3 K/s condition. Phase change starts on the inner surface of the confinement and later spreads inside the explosive container. Figure 10 shows the field plot of liquid fraction (ratio of the liquid phase volume to the total volume). Slow progressive advancement of melt front in to the solid mass can be observed during the initial phase of heating due to its low thermal diffusivity. Thereafter melt front advances quickly in to the core before ignition due to local heat release by reaction.

Field plot of temperature with time progress is given in Fig. 11. Heat given on lateral surface of the confinement spreads also to the region near end fastening. Hence it results in a differential heating in the charge. Slow progress of heat

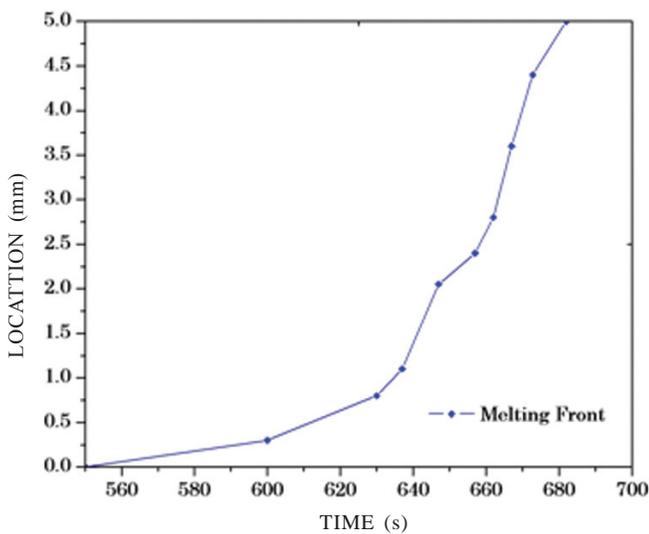


Figure 9. Melt front advancement in to the core of cylindrical charge.

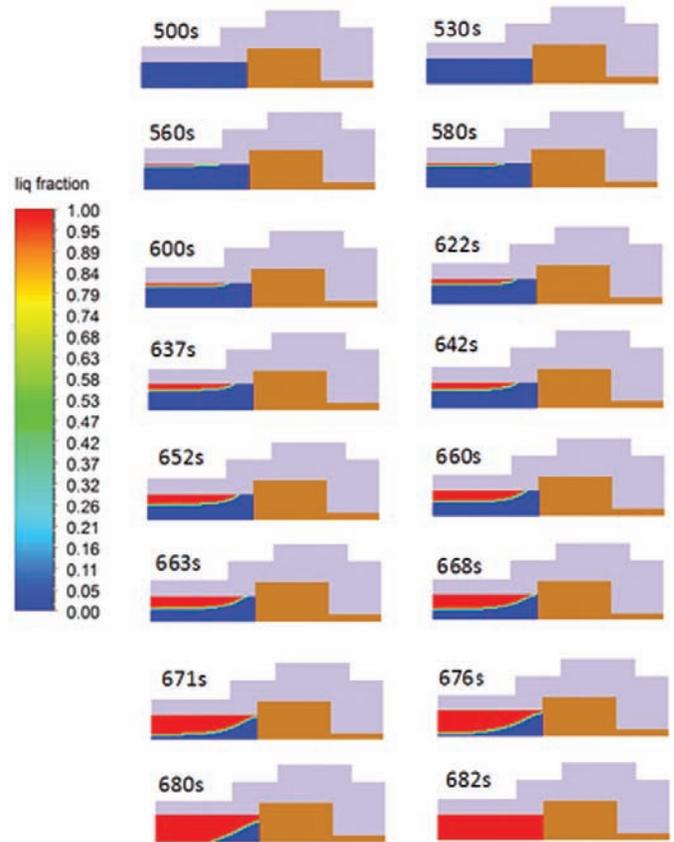


Figure 10. Field plot of liquid fraction with time progress.

diffusion into the charge is observed till ignition and thereafter it shoots up immediately due to the thermal runaway reactions. The field plots confirm that reactions are initiated near the boundary at the onset of ignition. This observation conforms to the present experimental findings as well as the boundary surface ignition reported in other experimental studies on fast cookoff.

6. CONCLUSIONS

Experimental investigations on time to explosion and location of ignition of a phase changing polymer bonded explosive material contained in cylindrical confinement with controlled temperature build up has been carried out. In support of this, a computational model for solving heat transfer with phase change and chemical reaction have been developed and implemented in the framework of a standard FVM based solver using a user defined function. Following are the conclusions based on these studies.

PID ramp rate controller used in the experiment could maintain the desired heating rate (0.3 K/s) on surface corresponding to a typical fast cookoff scenario. Though the temperature profiles provided by five thermocouples at various radial locations showed similar steady temperature build-up, the uneven temperature distribution across the charge during the heating is evident from these temperature profiles. The temperature recorded from thermocouple placed at 0.5 mm depth shows that ignition happened initially near to the surface of the confinement at 673.7 s where in reaction is initiated at 488.2 K. Fast heating rate provided on surface resulted in

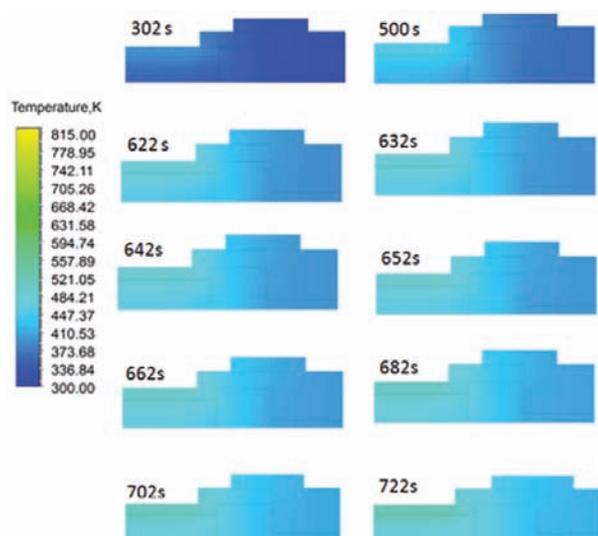


Figure 11. Field plot of temperature with time progress.

ignition near surface. Post test inspection also indicated no signs of violent explosion.

Coupled unsteady heat transfer, phase change and chemical reaction simulations could reveal the pattern of melt front advancement. The pattern of melting of explosive material found to vary with the local heat release by reactions. Numerical predictions of temperature buildup at various radial locations during the heating phase were closely agreeing with that measured in experiment. Simulations also confirmed that the ignition occurred near to heating surface in the confinement.

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