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Modeling of Pulsed Laser Ignition Dynamics of Nanolaminates

[1]C. D. Yarrington, [1]M. J. Abere, [2]M. Beason, [1]D. P. Adams

[1]Sandia National Labs, Albuquerque, NM

[2]Purdue University Department of Mechanical Engineering, West Lafayette, IN

Abstract

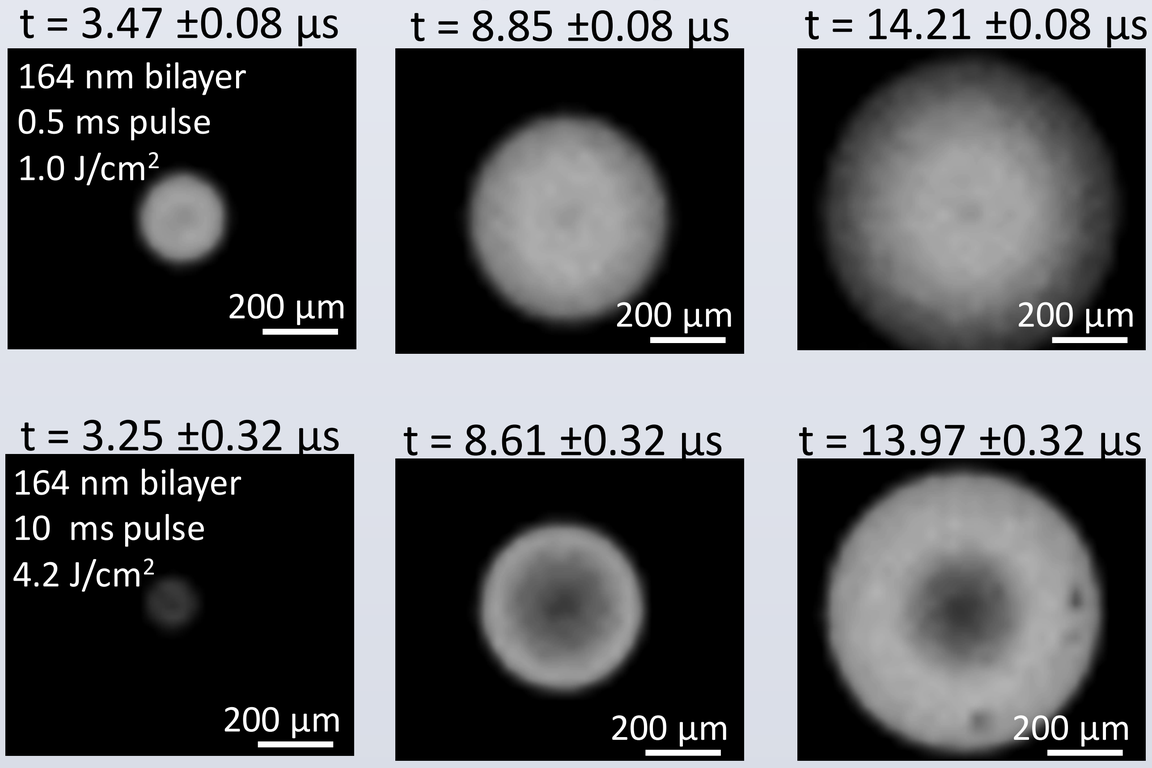
Two reactive multilayer designs consisting of alternating Al and Pt nano-layers of varying bilayer thickness were revisited under more disparate laser ignition conditions. These samples were irradiated by single laser pulses of 10ms and 0.5ms and constant spot size. Observations were carried out with Phantom high-speed cameras. At the longer pulse length of 10ms, a significant temperature deficit developed at the beam center upon ignition. This deficit in temperature was not observed at the smaller pulse duration. These results may serve as indicators of the underlying reaction mechanisms. This experiment was also simulated in the SIERRA/Multiphysics Module Aria [1], a finite element based multiphysics code. A layered Arrhenius diffusion model was chosen to represent product layer growth. The simulation results do predict a temperature deficit for the 10ms pulse case, and no deficit at 0.5ms. Predictions of steady state velocities far from the ignition zone are low however, perhaps indicating the presence of kinetic compensation.

Motivation

As nano-layered reactive materials mature into larger application spaces, deeper understanding of the reaction mechanisms that govern steady, and unsteady behaviors such as spin-waves are sought after. Possible areas that depend on foundational understanding of mechanisms are in creating optimized material designs (material combinations, bi-layer thicknesses, stoichiometries), understanding performance and failure margins, and understanding how these materials age. This joint experimental/modeling effort represents one way that these mechanisms can be studied for ignition.

Laser Ignition Experiments

Al/Pt nanolaminates of a 164 nm equiatomic design were subjected to laser pulses of 0.5 ms and 10 ms duration, with a 530 micron spot size (flat-top beam profile). Images taken at three different times after ignition are shown in Figure 1. A curious result is found in the 10ms pulse shots, where a temperature deficit is seen in the middle of the ignition zone. It is hypothesized that at longer pulses, heat generated from pre-ignition reactions is able to diffuse away from the laser heated region, resulting in the observed temperature deficit.

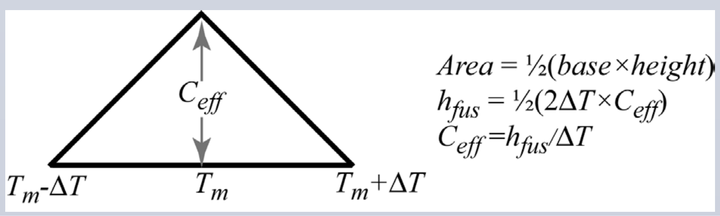


Simulation of Laser Ignition

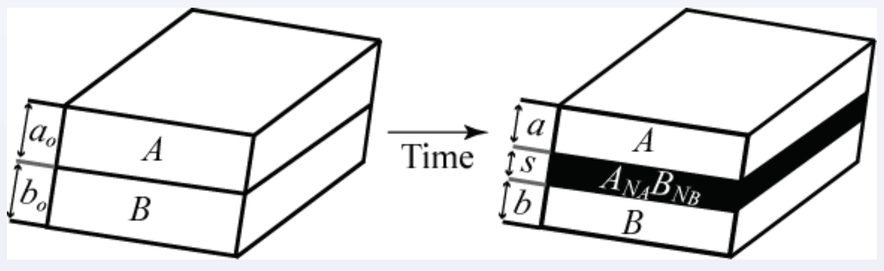
The sample was modeled as a 2d axisymmetric disc, with 400nm square quad elements in the 530µm diameter irradiated zone of the laser spot, and a linear increase of 10x to the outer diameter, which was 10x the laser spot size. In addition to the energy flux in the irradiated zone, radiative boundary conditions were used on the top and bottom surfaces of the sample. The reactive sample was modeled as a continuum, and common mixture rules were used to determine mixture specific heat (mass fraction weighting) and thermal conductivity (volume fraction weighting). Constituent melting was modeled using effective capacitance, whereby the integrated area under a triangular spike in specific heat equals the latent enthalpy of fusion.

$$k_{AlPt} = \frac{M_{w,Al}/\rho_{Al}}{M_{w,Al}/\rho_{Al} + M_{w,Pt}/\rho_{Pt}} k_{Al} + \frac{M_{w,Pt}/\rho_{Pt}}{M_{w,Al}/\rho_{Al} + M_{w,Pt}/\rho_{Pt}} k_{Pt}$$

$$C_{AlPt} = \frac{M_{w,Al}}{M_{w,AlPt}} C_{Al} + \frac{M_{w,Pt}}{M_{w,AlPt}} C_{Pt}$$



Layered Diffusion Model

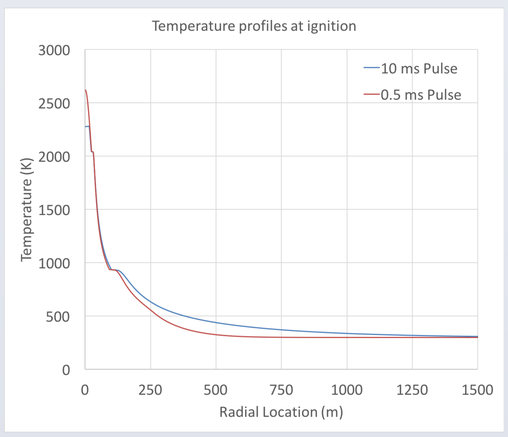


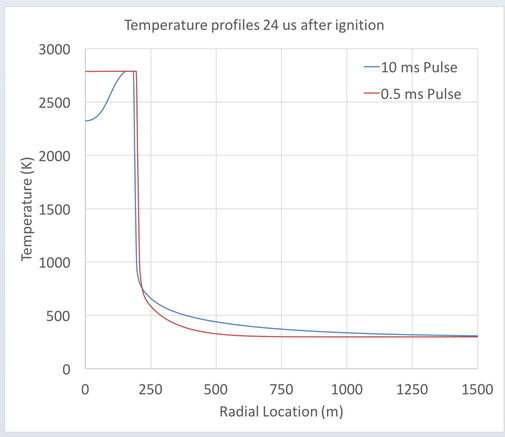
The diffusion limited reaction model implemented in SIERRA was originally derived by Hardt and Phung [2]. This model assumes a constant concentration gradient of unreacted atoms across the product layer. In this case in an Al layer, the concentration in the reactaed layer would start at the initial concentration (or density) of Al, and end at 0 on the other side of the product layer. From these assumptions, a simple derivation results in a model for product layer growth.

$$\frac{ds}{dt} = \frac{Dw}{s} \qquad w = 1 + \frac{b_0}{a_0} \qquad D = D_0 \exp\left(-\frac{E}{RT}\right)$$

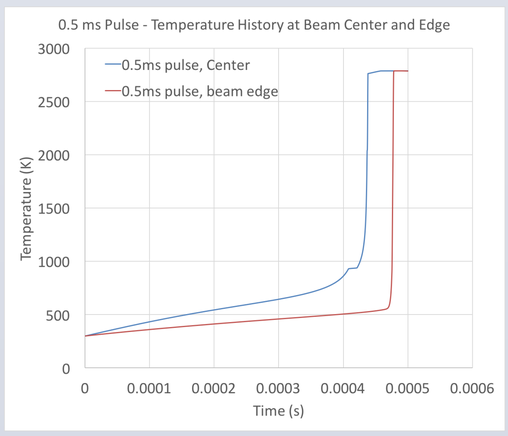
Results

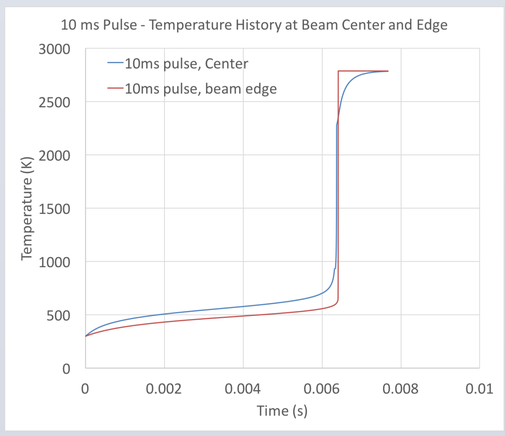
Radial temperature profiles of the simulation results at the moment of ignition show that the temperature at the center peaks at 2280K for the 10 ms pulse, while the 0.5 ms pulse is still increasing. 24 µs after ignition, a clear deficit is seen in the temperature for the 10 ms pulse.





Clear differences are also seen in the temperature time histories at beam center and edge. For the 10ms pulse, the evolution of temperature at the center of the beam is initially higher, but rises only slowly after ignition due to diffusion of heat inwards. For the 0.5ms pulse, temperatures at the center are consistently higher initially, and the max temperature is reach shortly after ignition.





Preliminary work on simulations with 65 nm bilayer designs indicates that this model can be used for a range of bilayer designs, as a change in only the bilayer thickness yields good agreement with experiment.

Steady state velocities are not reported here, because it was found that using the same diffusion kinetic parameters for ignition and steady state wave velocities results in slower velocities than observed in experiment. This may be due to heating rate dependent kinetics, for which kinetic compensation may allow for dual prediction of ignition delay and wave speed. In other experiments, there is indication that aged foils have similar ignition delays but lower velocities. Future work in this area will focus on these questions.

Acknowledgements

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References

1. Notz, P. K.; Subia, S. R.; Hopkins, M. M.; Moffat, H. K.; Noble, D. R.; Okusanya, T. O. *SIERRA Multimechanics Module: Aria User Manual - Version 4.40*; Sandia Technical Report SAND2016-4159; May, 2016.
2. Hardt, A.; Phung, P., Propagation of gasless reactions in solids—I. Analytical study of exothermic intermetallic reaction rates. *Combustion and Flame* **1973**, *21* (1), 77-89.

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