

Direct mechanical ignition of reactive materials for improved safety and simplicity

1. Overview/Abstract:

1.1 Problem Statement

The ability to directly ignite reactive materials through shock loading or by applying a shear stress in the material is an unutilized idea. Unfortunately, the knowledge to predictably describe the response of reactive materials to mechanical loads is unavailable. The current understanding of mechanical ignition does not adequately describe the mechanisms governing mechanical ignition.

We propose to identify and characterize the mechanisms leading to ignition in reactive nanolaminates (Ni/Al and Ti/2B) through dynamic mechanical loading. Ignition limits will be probed by varying stress wave strength, orientation, microstructure, and material properties. The physicochemical alterations leading to ignition will be characterized. Reactive materials with tailored mechanical ignition characteristics will then be engineered, allowing integration of mechanically-ignited reactive materials into components and realization of safety and performance improvements.

1.2 Creative and Innovative Nature of R&D

This work is proposed because of the need for novel reactive materials with tailored mechanical ignition characteristics and the innovative testing method. Testing will isolate relevant mechanical loading conditions including planar impact, mixed compression/shear, and high shear. The research of this proposal will provide a quantified understanding of the microstructural changes that occur in dynamically-loaded reactive materials, which parameters govern those changes, and how to use those characteristics to tailor ignition behavior. With this data, we will develop engineered reactive materials with tunable mechanical ignition sensitivity and generate analytical models to describe their behavior. These new areas of study will lead to future innovation in application and modeling of the materials.

2. Proposed R&D

2.1 Technical Approach and Leading Edge Nature of Work:

We propose to study the mechanisms that govern direct mechanical ignition by focusing on the length scales that are most affected by high-rate mechanical inputs. This will be accomplished using a single, tabletop test apparatus based on a high-power, pulsed laser source. A laser with variable energy output will permit a wide range of test conditions, ensuring that real-world requirements are addressed. Several distinct sample setups will be utilized in order to input high-rate mechanical load of controlled magnitude and type into the experimental foils. The foil materials to be investigated are Ni/Al, a well-characterized, ductile/ductile system, and Ti/2B, a ductile/brittle system that possesses an extremely high energy density.

A unique aspect to this proposal is that modification of the sample design allows access a wide variety of transient stress states resulting from planar shock loading, ramped compression, combined compression/shear loading, or high-shear conditions. Several iterations of the basic test apparatus are shown in Fig. 1. The basic setup, typical of that used in studies of planar shock inputs¹⁻⁵, is shown in Fig. 1A. In this iteration, the pulse from a laser ablates an epoxy layer bonded to a metal foil. The nascent plasma expands rapidly and causes a metal disc to be ejected, forming the flyer plate. The flyer plate accelerates with the plasma expansion before striking the test foil and inputting a planar shock in the material. The shock magnitude can be tuned by adjusting the laser fluence, which modifies the velocity of the flyer plate. The flyer plate material can also be selected to define the shock wave. Figure 1B depicts the other end of the loading spectrum to be studied. In this setup, the solid metal film is replaced by hard, micron-scale particles. Again, the laser pulse ablates the base layer of material. The plasma expansion accelerates the particles into the test foil, causing extreme shear in the test material. Combined compression/shear states will be investigated using laser-induced flyer plates, as in Fig. 1C. It is known that when an anisotropic crystal, like y-cut quartz, receives a planar shock input, the crystal will output both a compression wave and a shear wave.⁶ By depositing the reactive laminate onto an anisotropic crystal and impacting the crystal with the flyer plate, the transmitted stress waves from the crystal will load the test sample in both stress states. The shear/compression ratio can be adjusted by varying the

crystal's surface condition⁷. Finally, high-rate loading using a ramped, shock-less compression, can be investigated by growing the test foil on a thick metal substrate and using the laser to ablate the substrate, as in Fig. 1D. The rise time of the compression wave can be controlled by varying the laser fluence in order to prevent shock wave formation.

The wide range of loading conditions, ranging from planar shock compression to high-shear conditions effectively demonstrate the behavior of the test samples when responding to loading directions from normal to (planar shock) or perpendicular (high shear) to the laminate. This will provide a map of system response to direction, which is important when designing a device that monitors either omni-directional or uni-directional inputs.

The laser setup has advantages over more traditional experimental methods for testing these types of materials. First, the laser approach uses small samples, typically less than 1 mm in diameter, so many test samples can be loaded onto a single stage to increase throughput. Since the material being tested will be sputter-deposited nanolaminates with reactant periodicity much smaller than the test sample (10's of nanometers vs. millimeters), the test is representative of bulk behavior even with the small sample size. The simple changeover between tests is also an advantage. A gas gun can require a day of preparation between shots, but the laser only requires shifting the sample stage to expose a new sample. This reduces cost and also increases throughput. Finally, the adjustability of a laser setup is advantageous. The laser fluence can be precisely varied to create a wide range of loading rates and magnitudes. Also, with simple modifications to the experimental setup, the stress state can be varied between compression and shear. Thus, this system is a uniquely flexible and efficient means for testing the dynamic behavior of materials.

The basic experimental regime will adhere to the following steps for each type of experiment: threshold determination, sub-threshold testing, and analysis. Threshold determination for reaction initiation is critical for determining the mechanical loading levels at which a reaction is initiated and the variability of this event. The sub-threshold testing will provide samples to analyze that will describe the critical microstructural modifications that occur during loading that lead to the reaction threshold conditions. The analysis step will be used to investigate these modified microstructures through standard analysis techniques, like electron microscopy and Auger electron spectroscopy. It is important to understand that ignition in heterogeneous reactive materials is not simply a thermodynamic state. Instead, it is dependent on the process history. Because of this, sub-threshold testing is critical for determination of the physical and chemical processes that occur during mechanical stimulation but prior to ignition. By systematically varying process inputs, like stress state, mechanical properties of the test material, orientation, and design parameters, we will determine which parameters more strongly affect the ignition process and how to exploit those parameters to engineer a desired mechanical ignition response. For example, for additional sensitivity, hard, brittle interlayer inclusions, either in the form of particles or a thin oxide layer, could be added to enhance mixing and presumably increasing ignition sensitivity.

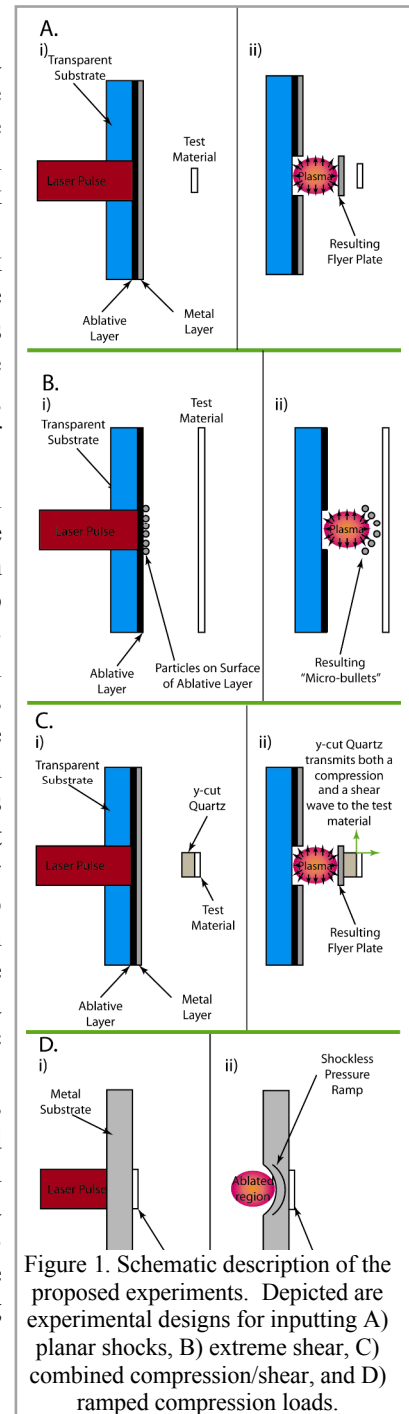


Figure 1. Schematic description of the proposed experiments. Depicted are experimental designs for inputting A) planar shocks, B) extreme shear, C) combined compression/shear, and D) ramped compression loads.

Next, we will develop analytical models to describe mechanical ignition. Equations describing system behavior, from microstructural changes that occur during mechanical equilibration to requirements for mechanically-induced ignition of the materials will be developed. Equations of state, determined through the impedance matching technique, will relate stress wave speed to particle velocity, and to pressure/temperature changes in the material through the Rankine-Hugoniot equations. Our goal will be to create equations that relate these input parameters to critical values and rates of defect formation/domain reduction/ internal energy changes in the test material to describe ignition conditions.

Finally, utilizing the newly gained understanding of microstructural modification that occurs during dynamic loading, we will develop materials with prescribed ignition characteristics and utilize those materials in a generic ignition device.

2.2 Relationship to Prior and Other On-Going Work:

While basic, phenomenological work has been previously performed studying mechanical ignition of heterogeneous reactive materials, the proposed work moves well beyond their scope by describing the process through a parametric experimental study and then using the resulting description to engineer reactive materials with tailored sensitivities. Most previous experimental studies are focused on powder systems under planar shocks.⁸⁻¹⁰ By solely addressing planar shocks, they address loading situations that are easier to interpret analytically but are atypical of real world systems, like omnidirectional failsafe/weaklinks or igniters for a thermal battery. In addition to planar shock compression, we will investigate high-shear and combined shear/compression effects, which are more akin to real-world conditions. Shear conditions have been studied qualitatively in powder systems¹¹⁻¹³, and using numerical models¹⁴, but no systematic study of the ignition mechanisms of reactive nanolaminates has been undertaken. This proposed work would provide a real working knowledge of these mechanisms, allowing reliable use of reactive laminates in novel engineering applications.

2.3 Key R&D Goals, Objectives, and Project Milestones:

2.3.1. Goal and Success Measure

The overall goal for this proposed research is to demonstrate reactive materials with tailored ignition sensitivity to mechanical input. The experimental work will establish a fundamental understanding of the physicochemical mechanisms by which mechanical inputs modify the microstructure of heterogeneous reactive materials, how these mechanisms are affected by design parameters and intrinsic material properties, and a description of the ignitable material state. The project success will be gauged by our use of this knowledge to build reactive materials with tunable mechanical ignition sensitivities.

2.3.2. Key Objectives and Milestones

Objective/Milestone	Completion Date
Complete initial list of reactive systems and downselection	11/01/2013
Validation of experimental setup and procedure	
Design/installation of pulsed laser based experimental setup	02/01/2014
Installation and verification of velocimetry instruments	05/01/2014
Parametric characterization of produced flyer plates	07/01/2014
Deposition of reactive materials with tailored sensitivity	
Deposition of Ni/Al and Ti/B multilayers with multiple periodicities	06/01/2014
Design and deposition of new materials having shear enhancers	04/01/2016
Verification of tailored mechanical ignition thresholds	08/01/2016
Planar shock initiation of reactive materials	
Reaction threshold determination	09/01/2014
Sub-critical testing and analysis of microstructural changes	03/01/2015
Isentropic ramping (direct ablation)	
Reaction threshold determination	02/01/2015
Sub-critical testing and analysis of microstructural changes	06/01/2015
Combined shear/compression initiation	

Reaction threshold determination	03/01/2015
Sub-critical testing and analysis of microstructural changes	09/01/2015
High shear initiation of reactive materials	
Reaction threshold determination	09/01/2015
Sub-critical testing and analysis of microstructural changes	12/30/2015
Model development	
Description of microstructural/thermal requirements for ignition	08/01/2016
Description of microstructural changes due to mechanical inputs	08/01/2016

2.4 Technical Risk:

- Interactions between transient stress inputs and heterogeneous materials is extremely complex and depends on many different experimental parameters
 - The test setup allows high throughput, experimental flexibility, and inexpensive testing capability. This will allow us to perform the necessary amount of tests, while varying individual parameters in the test materials to avoid characterizing “lumped” parameter effects. Also, the test materials are able to be precisely grown using current sputter deposition technologies, which will provide control over individual material properties.
- Ignition characteristics may be insensitive to the experimental parameters that we vary in testing, preventing use of those variables in producing tailored reactive materials
 - Additional means of modifying response through design additions like high-hardness inclusions, like nano-alumina, to enhance shear and encourage mixing have been identified and are feasible.
- Environmental conditions, e.g. electrostatic charge and humidity, could affect ignition threshold.
 - We will rigorously control the environmental test conditions. Test samples will be electrically shorted to the flyer plate generator during testing. Humidity levels will be controlled using a commercially-available environmental test chamber.

3. Resources

3.1 Key Research Team Members:

Name	Role
Robert V. Reeves	PI, reactive materials, dynamic behavior of materials
David P. Adams	Thin film growth, laser ablation

3.2 Qualifications of the Team to Perform This Work:

R.V. Reeves, PhD has 6+ years of experience working with energetic materials. He also has significant experience studying the effect of high-rate mechanical insults on energetic material ignition.

4. Strategic Alignment and Potential Benefit

4.1 Anticipated Outputs:

The most important output of this project is a working knowledge of the structural and material characteristics that govern mechanical ignition of heterogeneous reactive materials. The developed materials will lead to improvements in components that could increase performance, and enhance the safety and reliability of devices containing reactive materials. The test data and developed analytical models will benefit numerical modeling efforts for prediction of component behavior and design, which in turn will reduce testing and future component development costs. Finally, the fundamental research accomplishments will be of interest to researchers in both the broader scientific community. We expect to place several peer-reviewed journal articles in high-impact journals, presentations at national conferences, and at least one invited presentation.

4.2 Leveraging Results:

We anticipate external communications through the production of peer-reviewed journal articles in highly-regarded journals. External presentations at national meetings are also planned.

5. References

- ¹H. Fujiwara, K. E. Brown, and D. D. Dlott, in *Shock Compression of Condensed Matter*, Nashville, TN, edited by M. Elert, M. D. Furnish, W. W. Anderson, W. G. Proud, and W. T. Butler (American Inst. Phys., 2010), p. 1317.
- ²M. W. Greenaway, M. J. Gifford, W. G. Proud, J. E. Field, and S. G. Goveas, in *Shock Compression of Condensed Matter*, Atlanta, GA, 24-29 June, edited by M. D. Furnish, N. N. Thadhani, and Y. Horie (American Inst. Phys., 2002), p. 1035.
- ³M. W. Greenaway, W. G. Proud, J. E. Field, and S. G. Goveas, A Laser-Accelerated Flyer System, *International Journal of Impact Engineering* 29, 10.1016/j.iimpeng.2003.09.027, (2003).
- ⁴S. C. Kelly, S. Barron, N. Thadhani, and T. P. Weihs, in *Shock Compression of Condensed Matter*, Chicago, IL, 26 Jun - 1 Jul 2011, edited by M. L. Elert, W. T. Buttler, J. P. Borg, J. L. Jordan, and T. J. Vogler (American Inst. Phys., 2012), p. 599.
- ⁵K. E. Brown, W. L. Shaw, X. Zheng, and D. D. Dlott, Simplified Laser-Driven Flyer Plates for Shock Compression Science, *Review of Scientific Instruments* 83, (2012).
- ⁶L. C. Chhabildas and J. W. Swegle, Dynamic Pressure-Shear Loading of Materials Using Anisotropic Crystals, *J. Appl. Phys.* 51, 10.1063/1.328312, (1980).
- ⁷J. N. Johnson, Shock Propagation Produced by Planar Impact in Linearly Elastic Anisotropic Media, *J. Appl. Phys.* 42, 10.1063/1.1659974, (1971).
- ⁸D. Eakins and N. N. Thadhani, Shock-Induced Reaction in a Flake Nickel Plus Spherical Aluminum Powder Mixture, *J. Appl. Phys.* 100, 10.1063/1.2396797, (2006).
- ⁹D. E. Eakins and N. N. Thadhani, The Shock-Densification Behavior of Three Distinct Ni+Al Powder Mixtures, *Appl. Phys. Lett.* 92, 10.1063/1.2896653, (2008).
- ¹⁰N. N. Thadhani, Shock-Induced and Shock-Assisted Solid-State Chemical-Reactions in Powder Mixtures, *J. Appl. Phys.* 76, 10.1063/1.357624, (1994).
- ¹¹R. V. Reeves, *Control of Ignition and Reaction Behavior in Gasless Reactive Systems Via Microstructural Modification*, Ph.D. Thesis, Purdue University, (2011).
- ¹²R. V. Reeves, A. S. Mukasyan, and S. F. Son, Thermal and Impact Reaction Initiation in Ni/Al Heterogeneous Reactive Systems, *J. Phys. Chem. C* 114, 10.1021/jp104686z, (2010).
- ¹³R. V. Reeves, A. M. Mukasyan, and S. F. Son, Transition from Impact-Induced Thermal Runaway to Prompt Mechanochemical Explosion in Nanoscaled Ni/Al Reactive Systems, *Propellants Explos. Pyrotech.* In Press, 10.1002/prop.201200193, (2013).
- ¹⁴P. E. Specht, N. N. Thadhani, and T. P. Weihs, Configurational Effects on Shock Wave Propagation in Ni-Al Multilayer Composites, *J. Appl. Phys.* 111, 10.1063/1.3702867, (2012).