SAND2012-9754C

Exceptional service in the national interest

Sandia National Laboratories

Transition from steady to unsteady reaction propagation with variations in bilayer spacing in reactive multilayer foils

Robert Knepper[1], Robert V. Reeves[1], Joel P. McDonald[1], Sara C. Barron[2], Timothy P. Weihs[2], and David P. Adams[1]

1 Sandia National Laboratories, Albuquerque, NM 87185

2 Johns Hopkins University, Department of Materials Science and Engineering, Baltimore, MD 21218

U.S. DEPARTMENT OF ENERGY

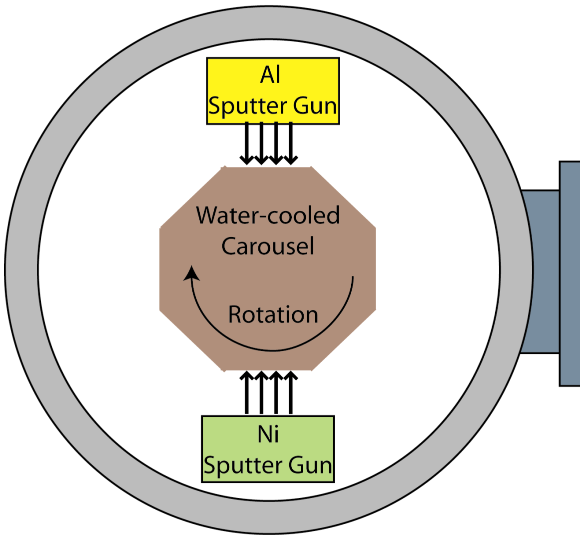
NNSA National Nuclear Security Administration

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000. SAND NO. 2011-XXXXP

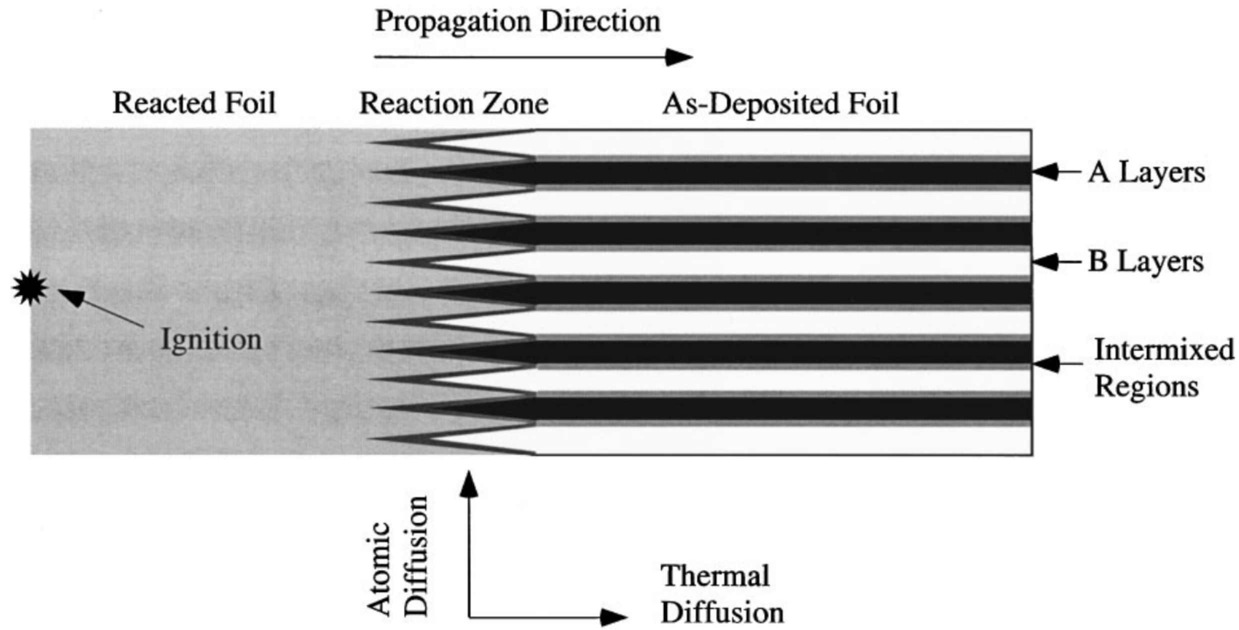
Background

- Sputter-deposited reactive multilayer foils are useful for investigating phenomena related to variations in foil geometry due to their easily controllable and consistent microstructures
- In various material systems, reaction propagation could occur in either a steady or unsteady fashion, but until recently, only one mode had been observed for any given chemistry
  - Steady: Al/Pt, Sc/Au, Y/Au, Y/Cu
  - Unsteady: Sc/Cu, Sc/Ag
- Changes in geometry (bilayer spacing, total thickness, etc.) have been observed to lead to changes in velocity or cause reactions to fail to self-propagate
- This work marks the first time transitions from steady to unsteady propagation have been observed with variations in bilayer spacing

Foil deposition



Top view of sputter chamber



Schematic of a reactive multilayer foil

- Al/Ni and Al/Co multilayer foils deposited by sputtering
  - Power to sputter guns determines composition
  - Carousel rotation rate determines bilayer spacing
  - All foils deposited to a total thickness of 7.5-9.0 microns
- Reactions monitored using high-speed video camera and thermal imaging camera

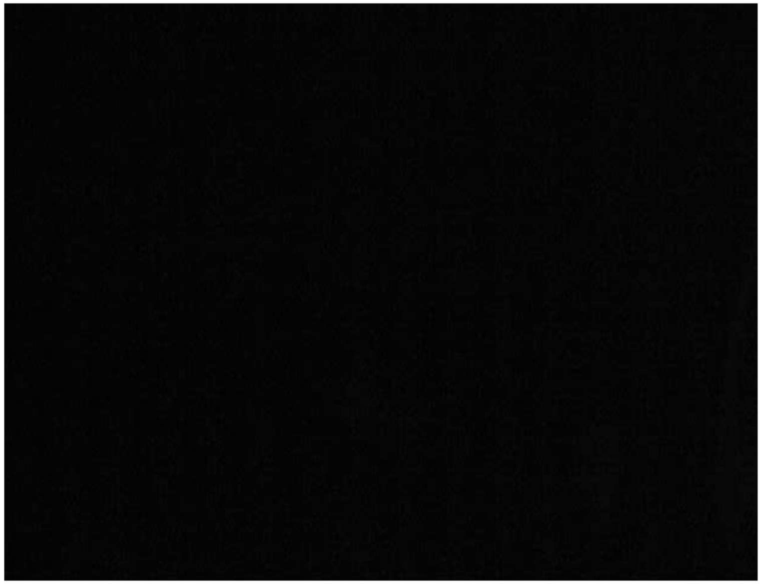
High speed imaging – Al/Co

High speed video of a propagating Al/Co foil with a 33 nm bilayer spacing

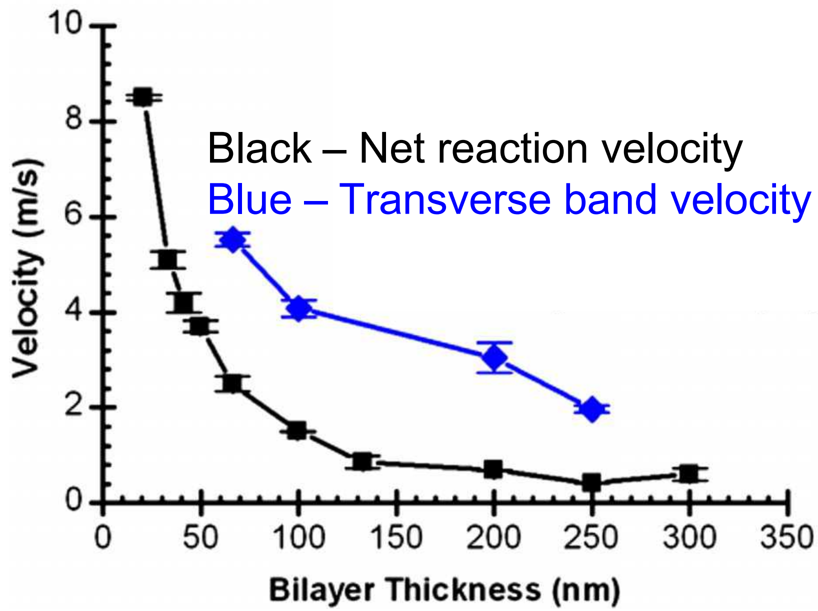
High speed video of a propagating Al/Ni foil with a 125 nm bilayer spacing

- High speed video shows that Al/Co foils with bilayer spacings of ~ 33 nm or less have a steady reaction propagation
- Al/Co foils with bilayer spacings of ~ 125 nm or more propagate in an unsteady manner

High speed imaging – Al/Co



High speed video of a propagating Al/Co foil with a 66 nm bilayer spacing



Reaction velocity vs. bilayer spacing plot for Al/Co foils (McDonald, 2009)

- A transition between steady and unsteady propagation appears to occur at bilayer spacings of ~ 66 nm
  - Previous experiments showed that transverse band width increases with increasing bilayer spacing (McDonald, 2009)
  - Large bands in film with 66 nm bilayer spacing suggests that it is just starting to shift from steady to unsteady propagation
- Transition appears to occur where velocity changes rapidly with bilayer spacing

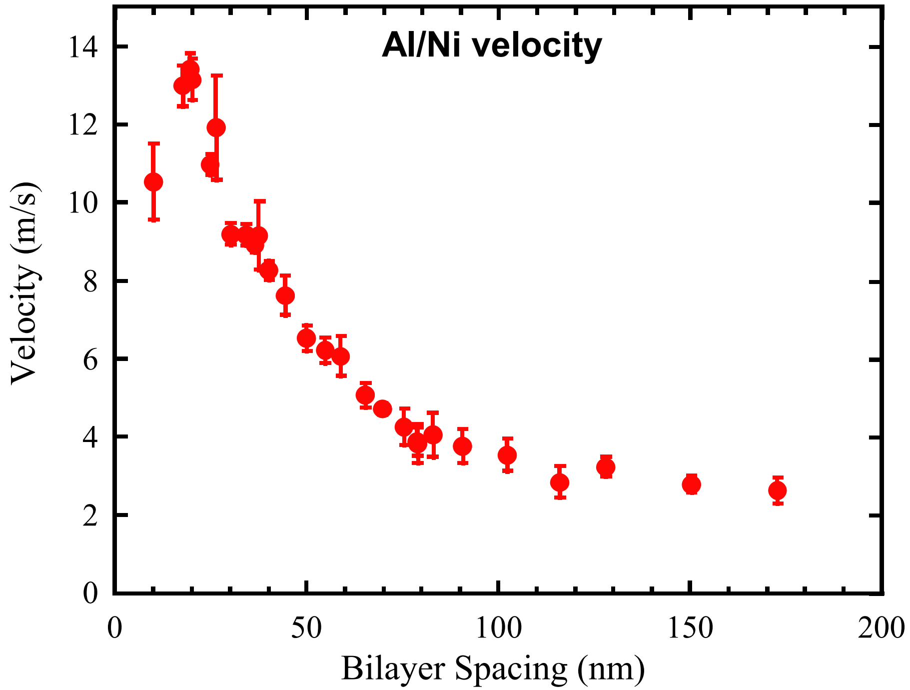
High-speed imaging – Al/Ni

High speed video of a propagating Al/Ni foil with a 100 nm bilayer spacing

High speed video of a propagating Al/Ni foil with a 150 nm bilayer spacing

- High speed video shows that Al/Ni foils with bilayer spacings of ~ 100 nm or less have a steady reaction propagation
- Al/Ni foils with bilayer spacings of ~ 150 nm or more propagate in an unsteady manner

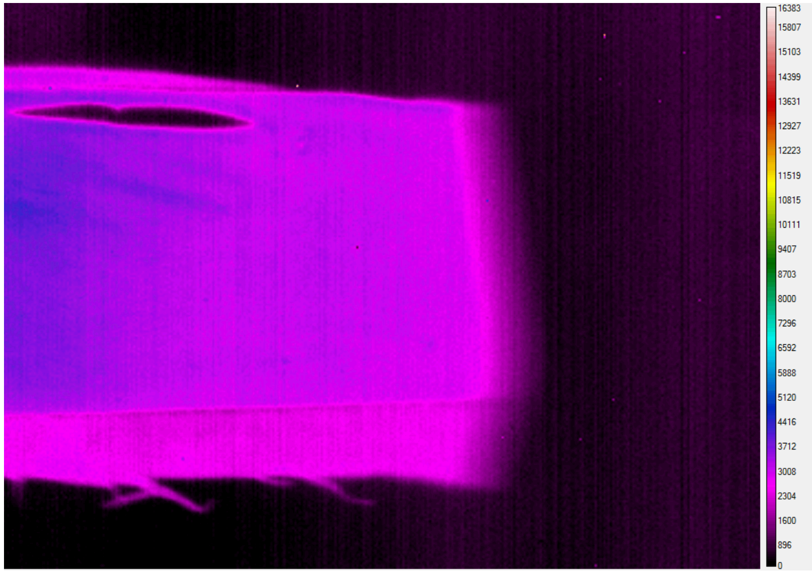
Reaction Velocities – Al/Ni



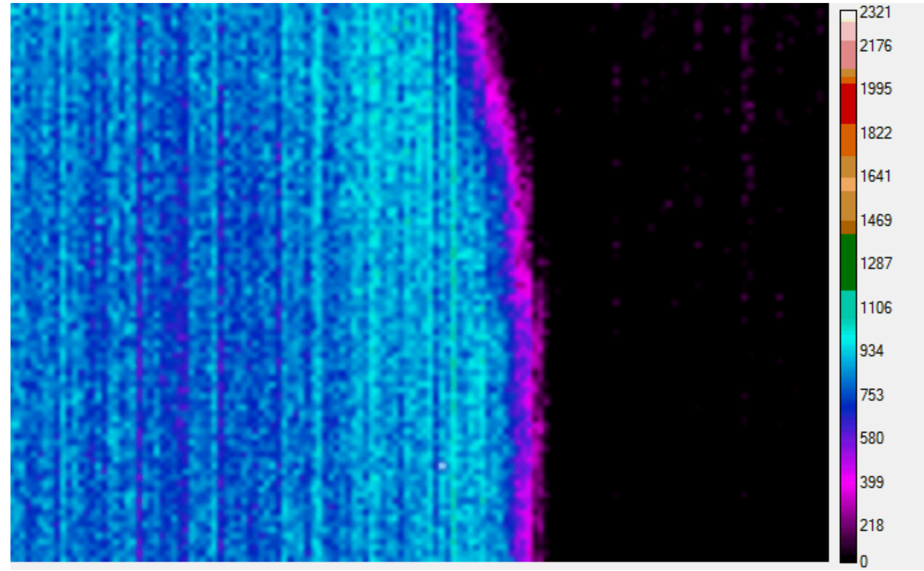
Reaction velocity vs. bilayer spacing plot for Al/Ni foils (Knepper, 2009)

- In contrast to Al/Co foils, the transition from steady to unsteady propagation in Al/Ni foils occurs in a region where reaction velocity changes little with bilayer spacing

Thermal imaging – Al/Ni foils



Thermal image of a propagating Al/Ni foil with a 100 nm bilayer spacing

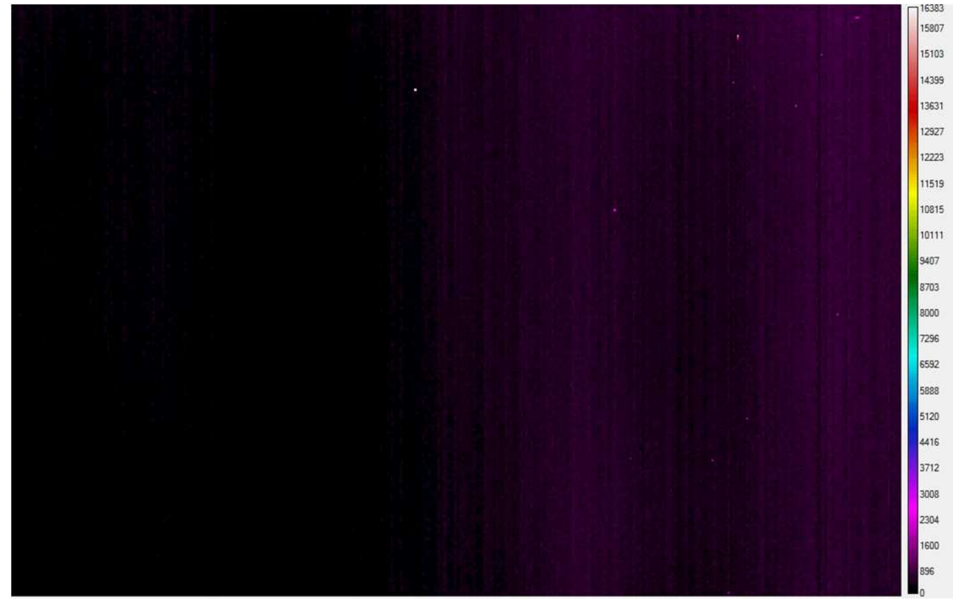


Thermal image of a propagating Al/Ni foil with a 150 nm bilayer spacing

- Reaction propagates too fast for thermal camera to capture more than one image during reaction propagation.
- Details of reaction front structure are smeared out as the front will move ~ 300 μm over the exposure time of ~ 140 ms (transverse band width ~ 100-200 μm in the foil with 150 nm bilayer spacing)

Large Bilayer Spacings – Al/Ni

High speed video of a propagating Al/Ni foil with a 1 micron bilayer spacing

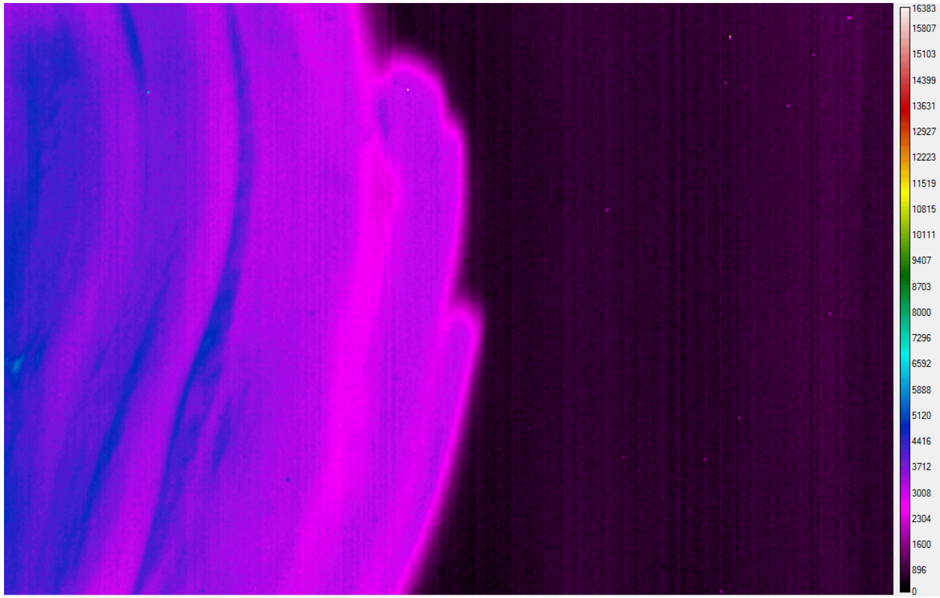


Thermal image sequence of a propagating Al/Ni foil with a 1 micron bilayer spacing

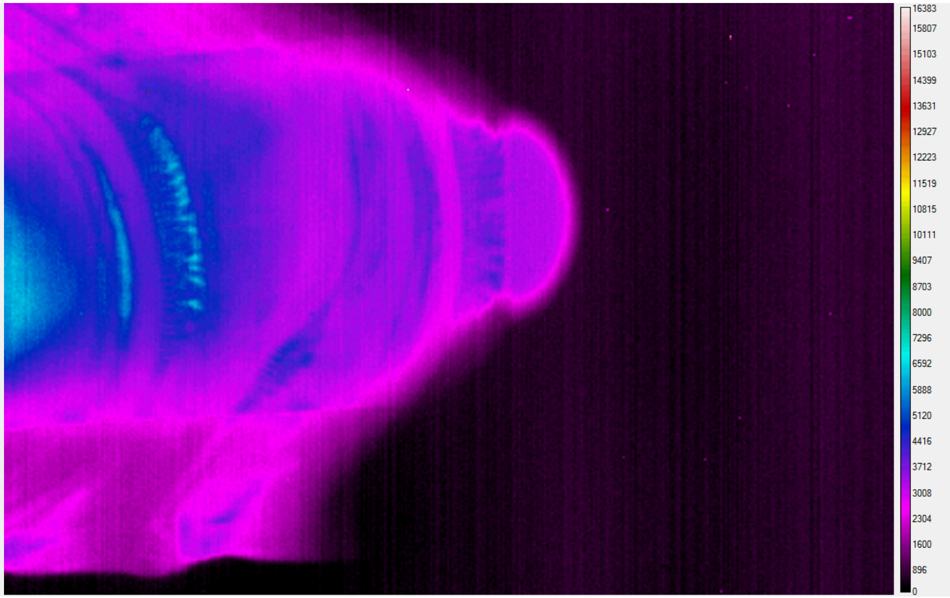
- Larger bilayer spacings lead to slower reaction propagation (~ 0.25 m/s) and larger transverse bands (~ 700 μm)
- Lower spatial and temporal requirements allows thermal camera to capture several images during reaction propagation

Two distinct mechanisms of unstable reaction propagation

Sandia National Laboratories



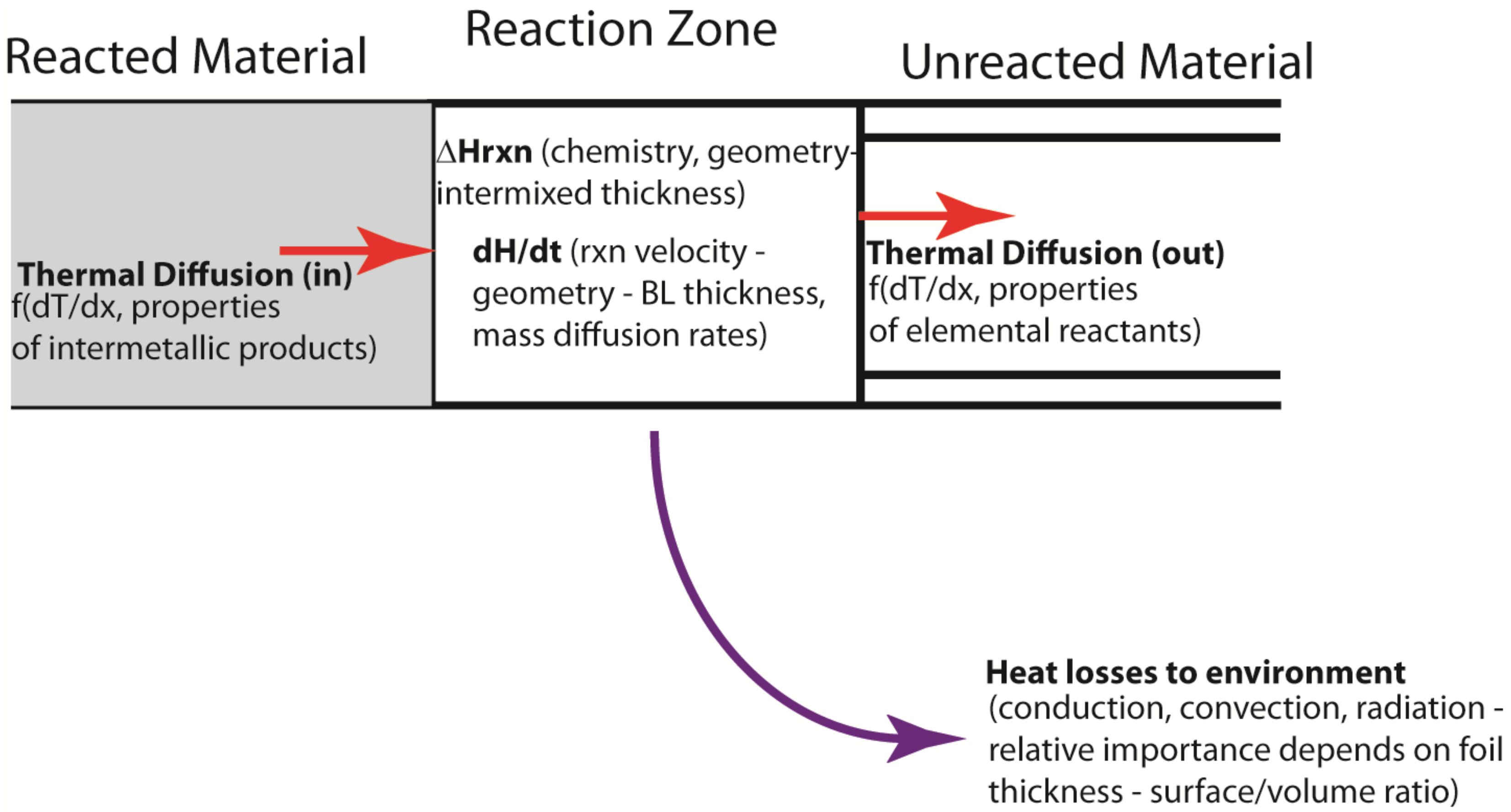
Thermal image of a propagating Al/Ni foil with a 1 micron bilayer spacing showing bands propagating from the edge of a foil



Thermal image of a propagating Al/Ni foil with a 1 micron bilayer spacing showing a band nucleating in the center of a foil

- Bands nucleated at foil edges or where two bands collide (due to localized variations in thermal transport ahead of the reaction front, seen commonly)
- Bands nucleated in the center of the foil after a period of pre-heating (homogeneous nucleation – less common)

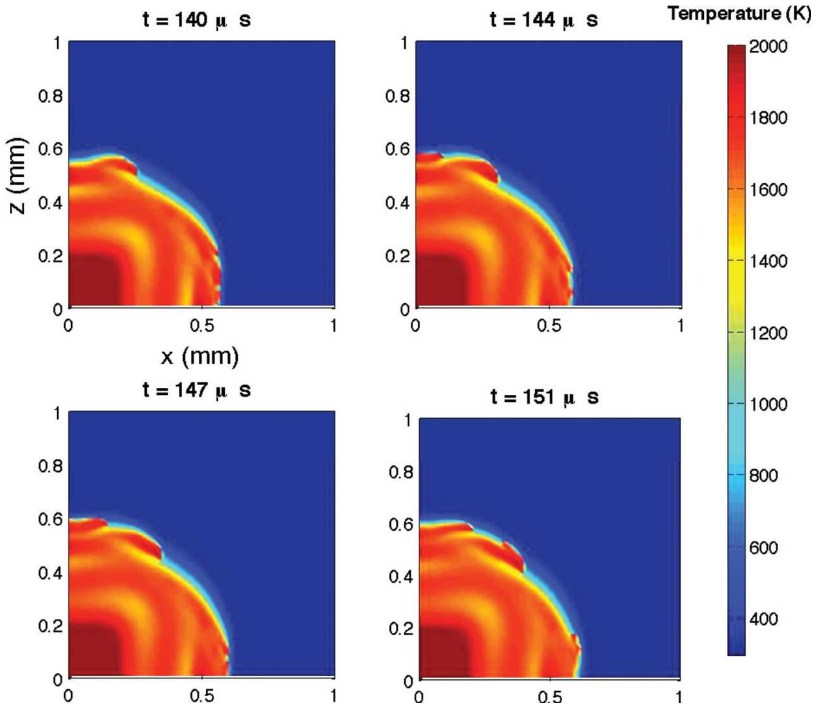
Thermal transport variables



Current models

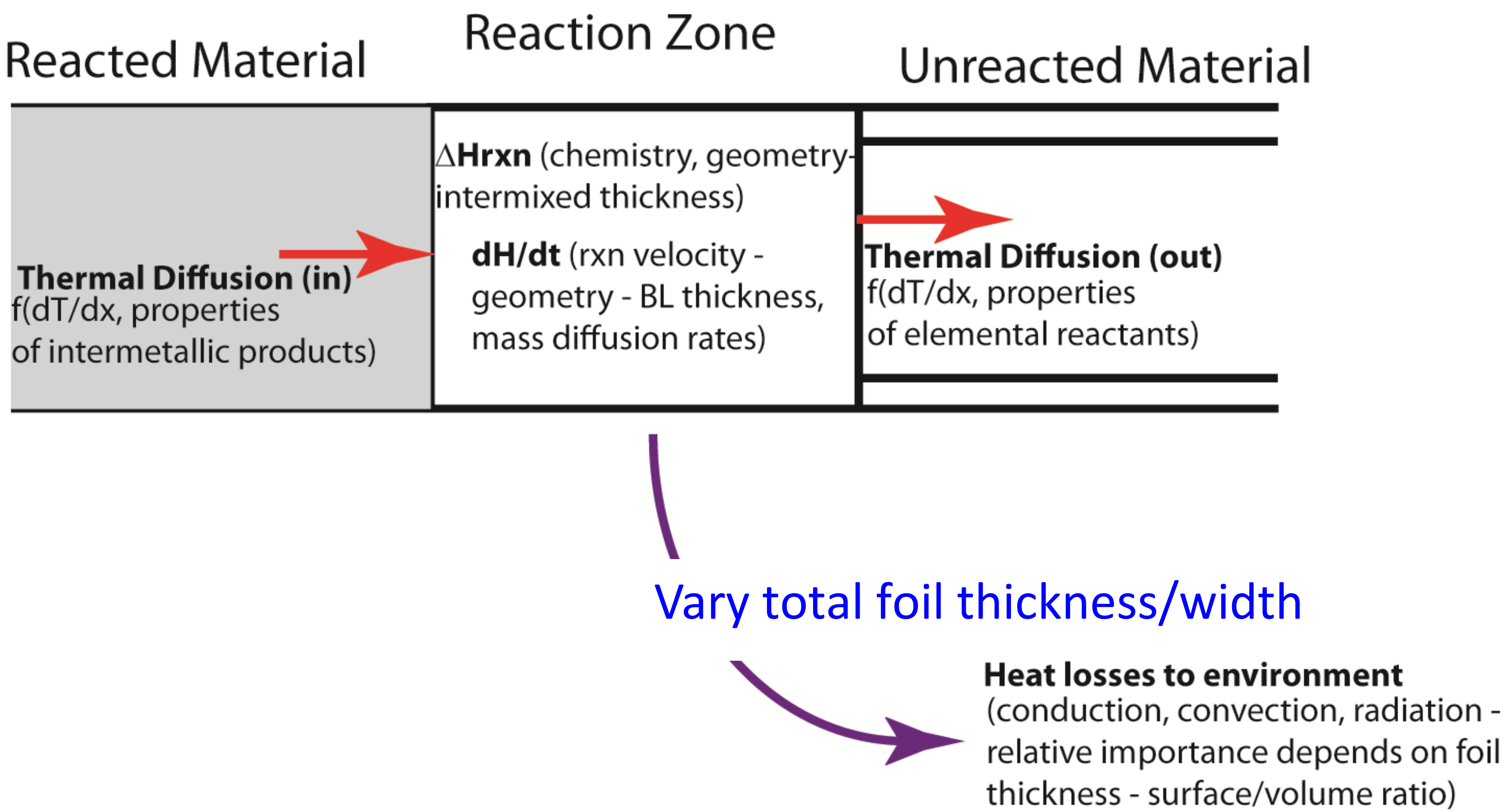
- Transitions between steady and unsteady propagation have been simulated, but current models cannot account for heat losses to the environment or other edge effects (limited to periodic boundary conditions at this point)

- Need experiments to determine the importance of such variables

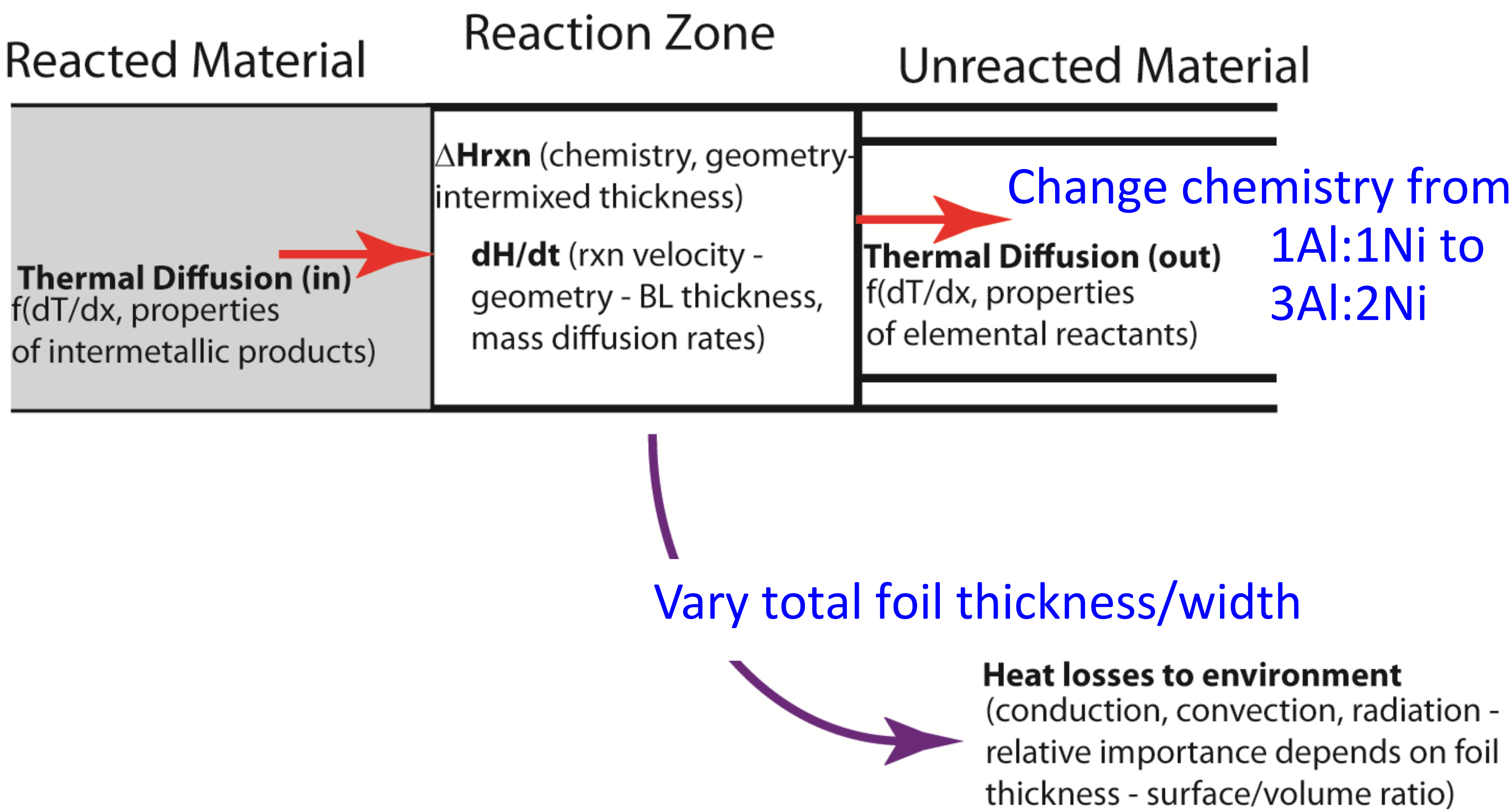


Simulations showing unsteady reaction propagation in Al/Ni multilayer foils (Alawieh, 2011)

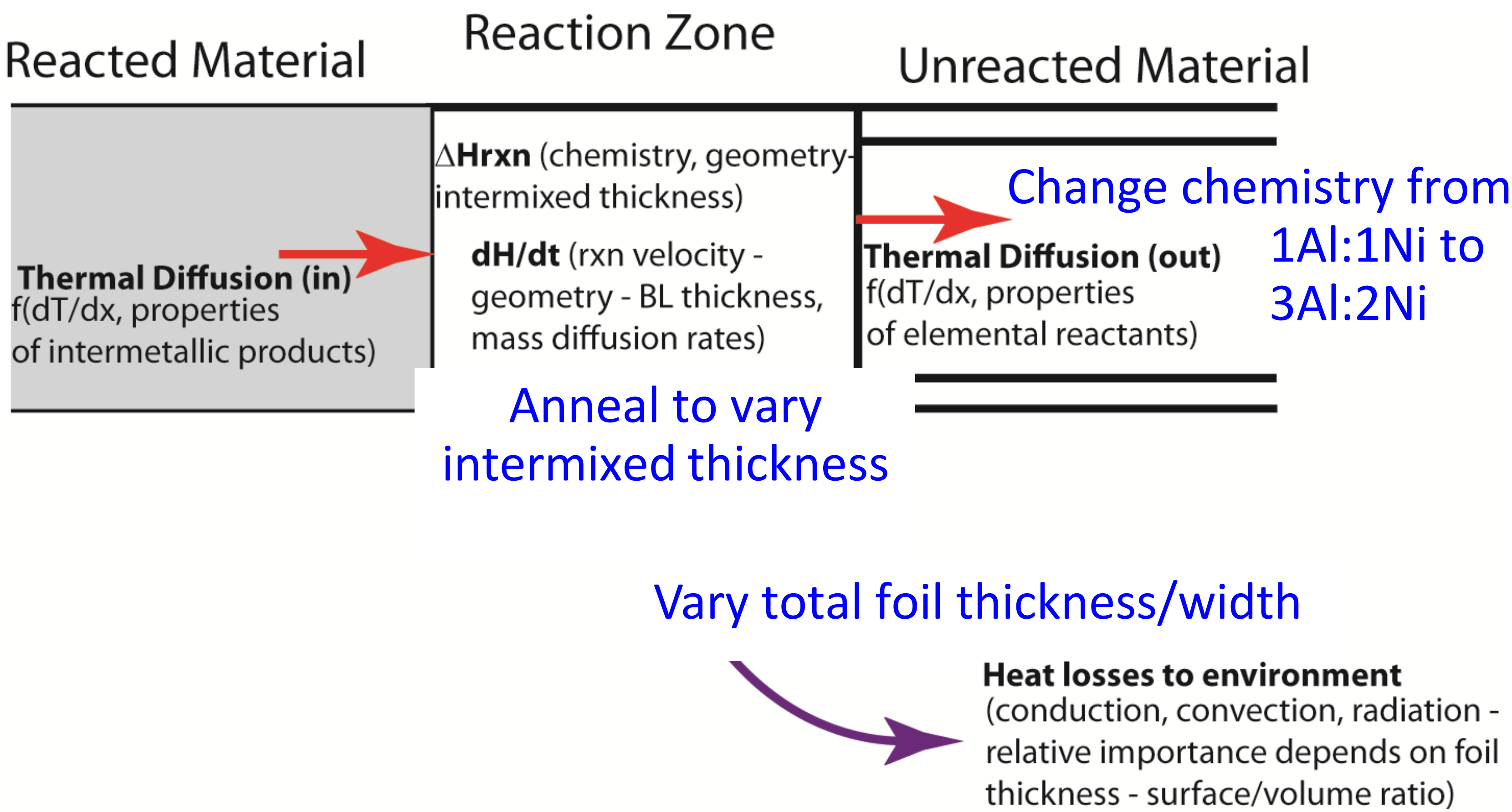
Potential experiments



Potential experiments



Potential experiments



Conclusions

- Transitions from steady to unsteady reaction propagation observed in both Al/Ni and Al/Co multilayer foils with variation in bilayer spacing
- Unsteady reaction arises from a decoupling of chemical reaction from thermal transport phenomena
- Likely involves a complex interplay of thermal transport, mass diffusion, and reaction kinetics
- Several experiments have been suggested that could elucidate the relative importance of some of these factors

Acknowledgements

- Sandia's Laboratory Directed Research and Development Program

- Office of Naval Research (MURI #N00014-07-1-0740)