SAND2011-7155C

Stability of propagating reactions in exothermic multilayer foils

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2011 Rio Grande Symposium on Advanced Materials
3 October 2011

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Combustion Synthesis of Materials

- Seen as a cost effective method for producing near net size parts from intermetallics, refractories, and ceramics [Holt]
- Can include solid-solid and solid gas reactions [Holt]
- Can be weakly or strongly exothermic
- Weak systems require preheat – Volume Combustion
- Strong systems exhibit Self-Propagating, High-temperature Synthesis (SHS) [Varma]

Holt, J. B. and Dunmead, S. D., Annual Review of Materials Science 21, 305 1991.

A. Varma; A. S. Rogachev; A. S. Mukasyan; S. Hwang, *Combustion Synthesis of Advanced Materials: Principles and Applications*. Elsevier Science and Technology Books: Amsterdam, 1998; Vol. 24, p 416.

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SHS: Basic Process Characteristics

- Ti/2B: T_{ad} = 3190 K, Q = -5.52 kJ/g (21.61 kJ/cc)
- Al/Co: T_{ad} = 1912 K, Q = -1.28 kJ/g (6.65 kJ/cc) [Fischer]
- To compare, TNT Q = -6.22 kJ/cc [Yaws]
- Propagation rates in mixed powders vary from 0.1 cm/s to 10 cm/s [Varma]
- Use of multilayered thin films allows for precise control of diffusion distances, controlled geometry, and clean interfaces between reactants.
- Propagation rates up to ~90 m/s [Adams]

Fischer, S.H., Grubelich, M.C., SAND98-1176C
C. L. Yaws, *Yaws' Handbook of Therm. and Phys.Properties of Chem. Compounds*. Knovel: 2003.
Varma; A. S. Rogachev; A. S. Mukasyan; S. Hwang, *Combustion Synthesis of Advanced Materials: Principles and Applications*. Elsevier Science and Technology Books: 1998; Vol.

Exothermic Thin Films

RVR1

- Used for joining, coatings, etc. [Wang]
  - Require uniform conversion to product for good results
- Unstable reaction propagation witnessed in many films, like Ni/Ti, Al/Co, Ni/Al, Sc/Cu, and Sc/Ag [Mcdonald]
- Could lead to quenching, incomplete conversion, poor joining performance
- Has been related to bilayer thickness [McDonald], and interlayer heat transfer in the samples [Alaweih]
- Better understanding of causes of instability necessary for utilization

Wang, J., Besnoin, E., Knio, O.M., Weihs, T.P., J. Appl. Phys. 97, 7 2005
McDonald, J.P., Hodges, V.C., Jones, E.D., Adams, D.P. Appl. Phys. Letters, 94, 034102, 2009
Alawieh, L., Knio, O. M., and Weihs, T. P., Journal of Applied Physics 110 2011.

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RVR1 Need to add spin movies or pictures to this page
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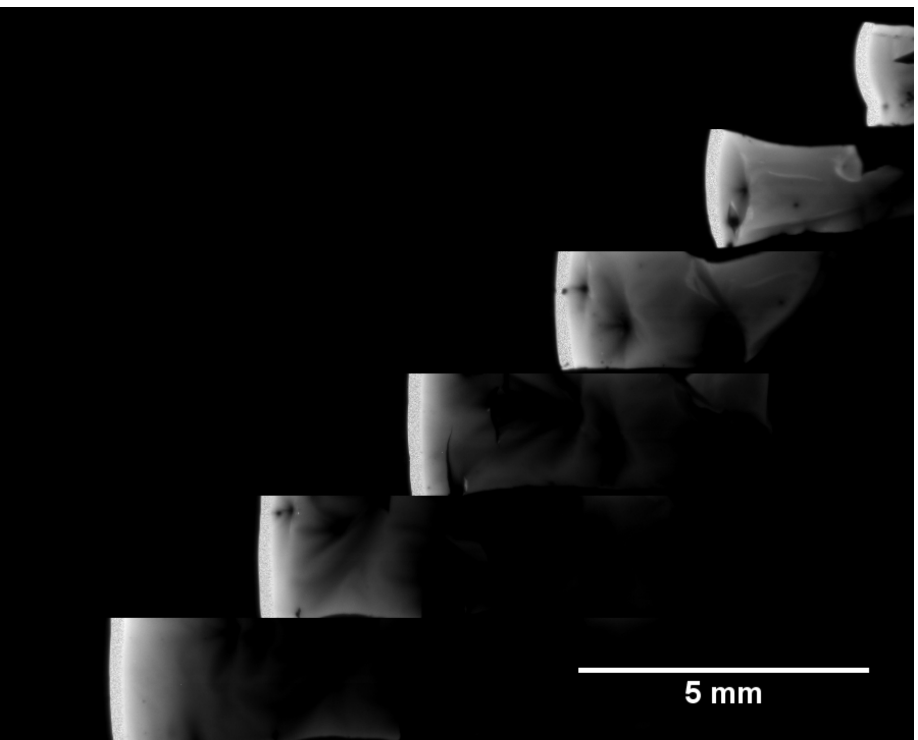
RVR8

Ti/2B Exothermic Foils – Stable SHS wave

- Highly exothermic SHS systems
- Does not exhibit spin for bilayer thicknesses from 9.3 to 53 nm

RVR3



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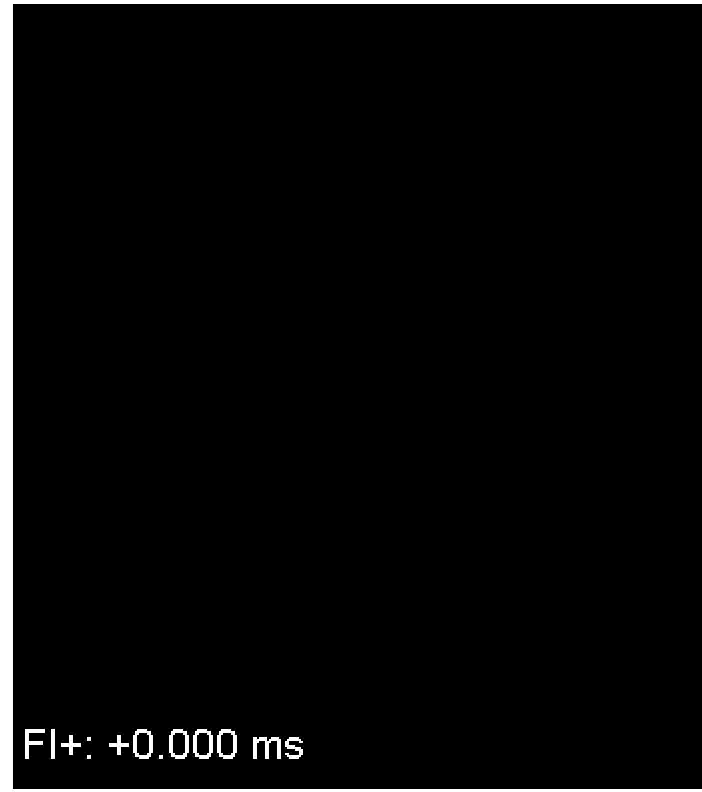
RVR3 Image of velocity vs thickness
rvreeve, 9/23/2011

RVR8 Velocity plot vs bilayer
rvreeve, 9/26/2011

RVR4

Co/Al Exothermic Foils

- Exhibit spin over wide range of bilayer thicknesses
- Transverse wave speed increases with decreasing bilayer
- Transverse wave thickness decreases with decreasing bilayer
- Thin bilayer designs (21 nm) propagate steadily in SHS mode



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RVR4 Needs
Movie or images of spin
plot of vel. vs. bilayer
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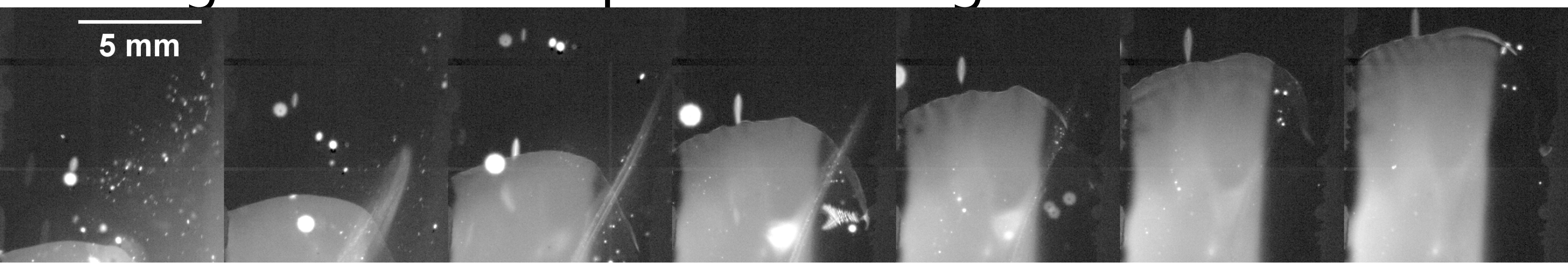
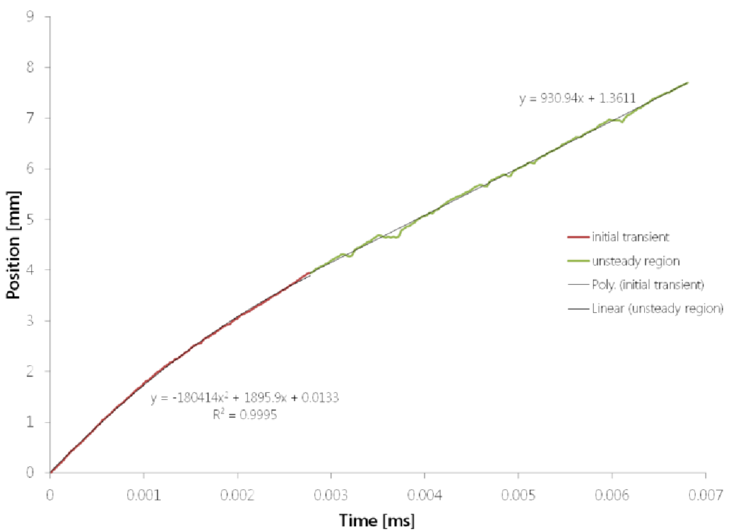
RVR5

Ni/Ti Exothermic Foils

- Ni/Ti: Q = -0.638 kJ/g
- Transitions from steady propagation near the ignition site to spin far from ignition



5 mm thick NiTi foil, 30 nm bilayer thickness slows from 1.9 m/s to 0.9 m/s with quasi steady spin propagation



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RVR5 Need
Movie or frames of spin
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Perspective

- Ti/2B – Very exothermic, no spin
- Al/Co – Moderately exothermic, spin over wide range of designs
- Ni/Ti – Low exothermic, spin common
- Is instability in the SHS wave based solely on exothermicity?
- Why is Ni/Ti able to propagate with such weak exothermicity?

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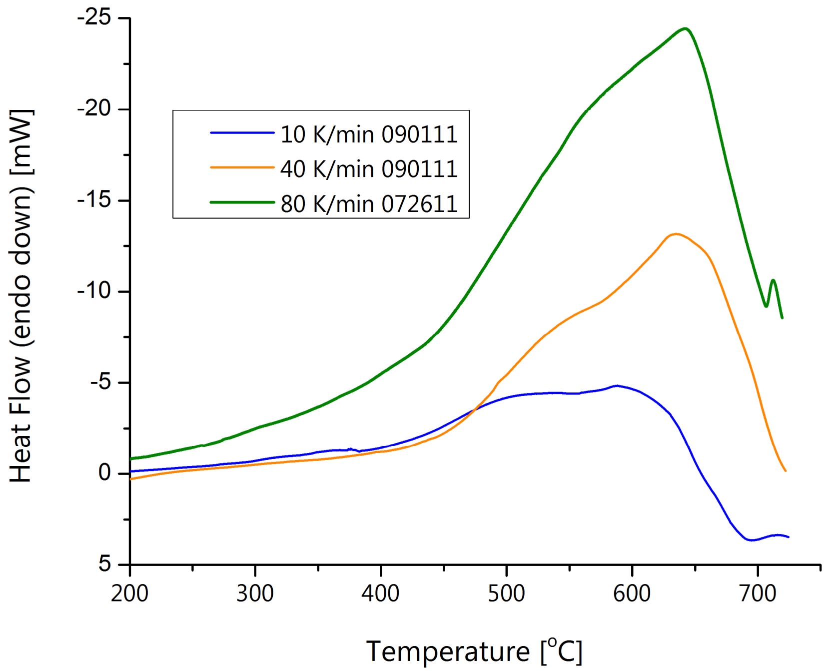
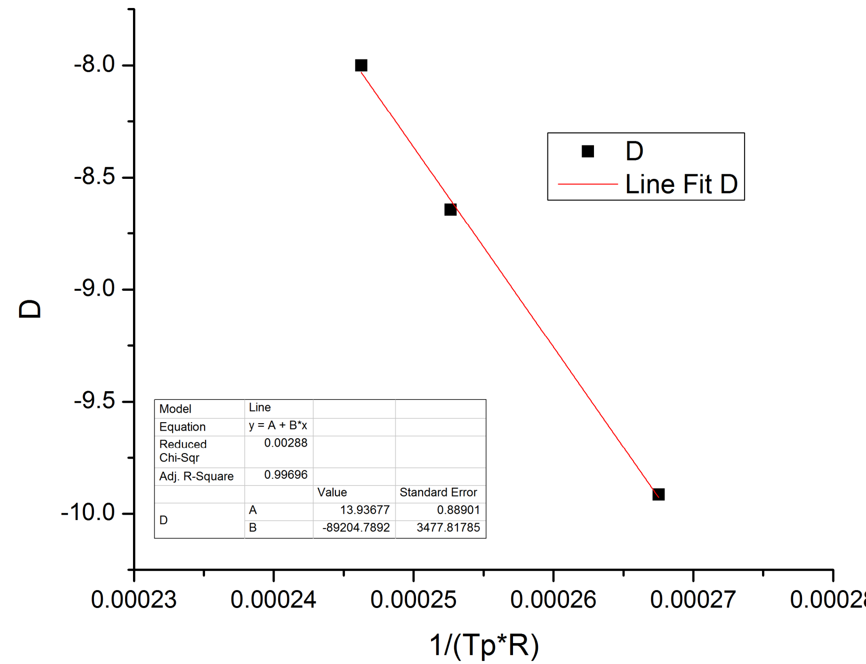
RVR7

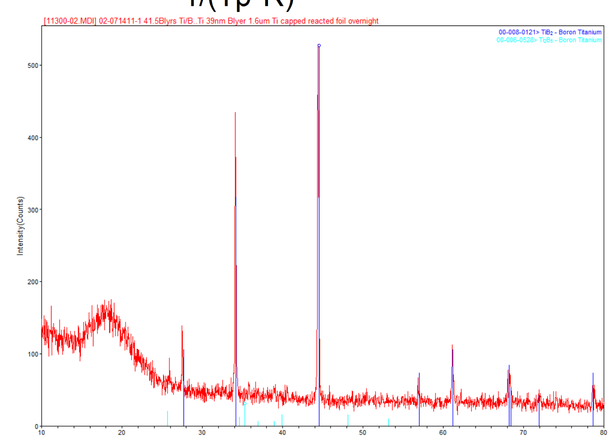
Differential Scanning Calorimetry of Ti/2B Reactive Foils





Ti/2B foil, 50 nm bilayer single reaction to completion

XRD traces indicate complete conversion to TiB_2 phase



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RVR7 Comparison of spinning and non spinning Co/Al
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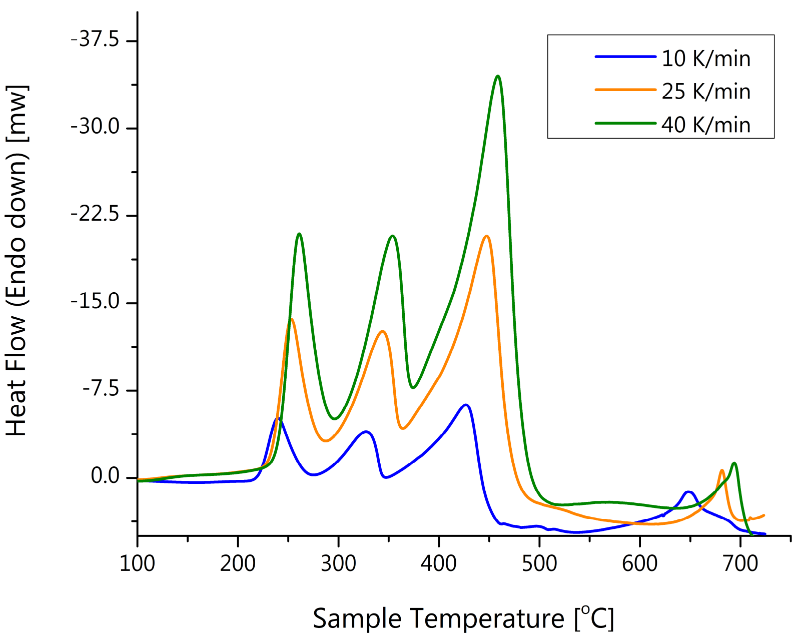
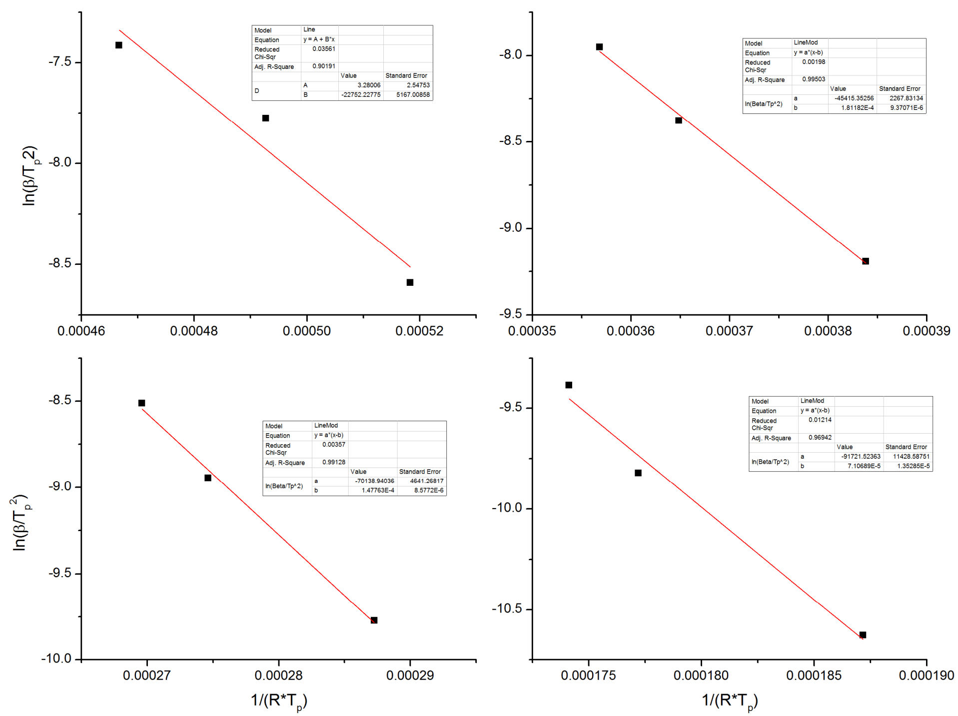
RVR6

Differential Scanning Calorimetry of Reactive Foils





Co/Al foil 66.4 nm bilayer

-4 distinct exotherms

-Exhibits spin in SHS mode

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RVR6 Figures for heat release for each material

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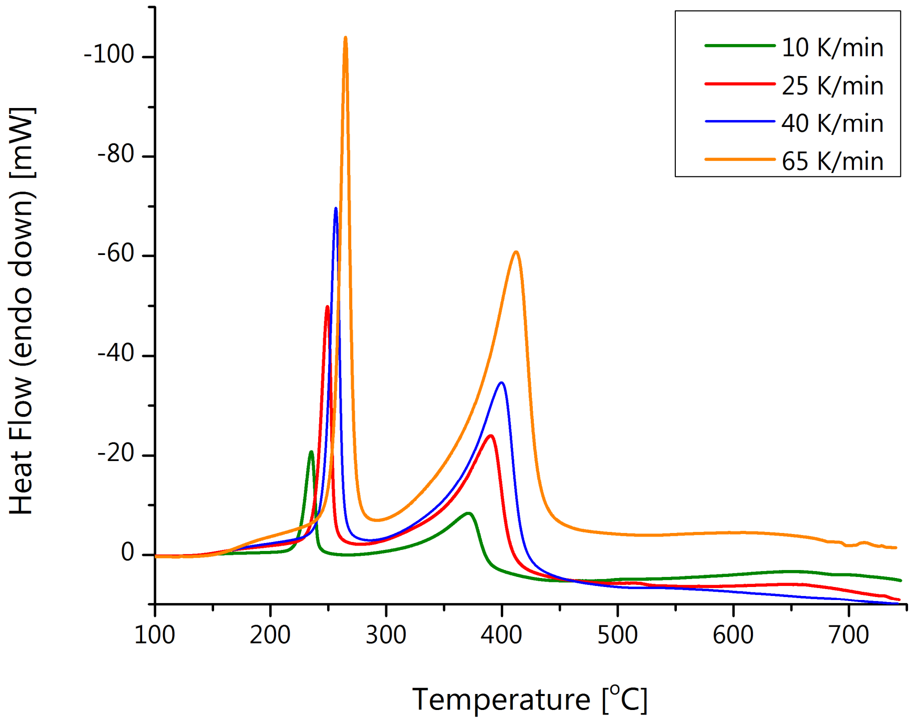
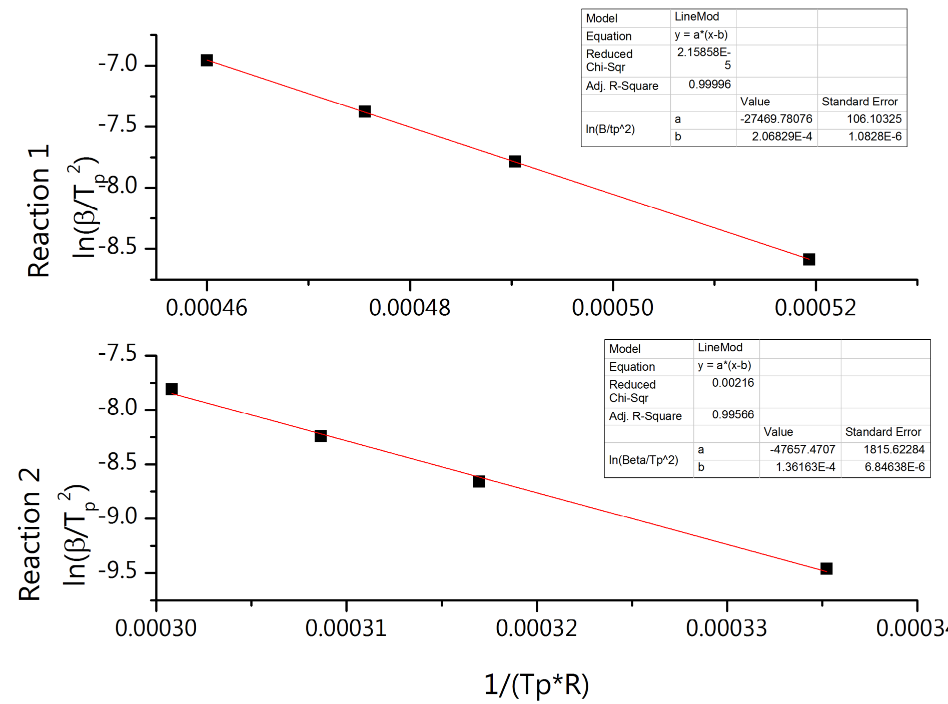
RVR9

Differential Scanning Calorimetry of Reactive Foils





Co/Al foil 21 nm bilayer

- Only exhibits 2 exotherms

- Exotherms at same onset temperature as in 66.4 nm design

- Does not exhibit spin in SHS mode

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RVR9 Figures for heat release for each material
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Kinetics Results

- Initial reactions have corresponding apparent activation energies despite bilayer thicknesses
  - Interfaces the same despite thicker bilayers
- Much higher apparent Ea for complete conversion to Co/Al at thicker bilayers
  - Likely due to formation of successive diffusion couples at Co/Al interface during reaction

Material	E_{a1} [J/mol]	E_{a2} [J/mol]	E_{a3} [J/mol]	E_{a4} [J/mol]
Co/Al 21 nm bilayer	27470±106	47658±1816	N/A	N/A
Co/Al 66.4 nm bilayer	22752±5167	45415±2268	70139±4641	91722±11429

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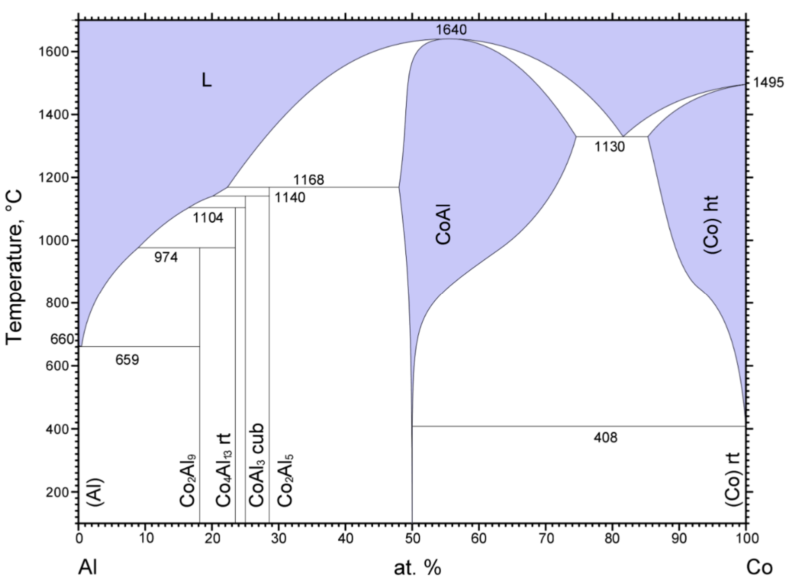
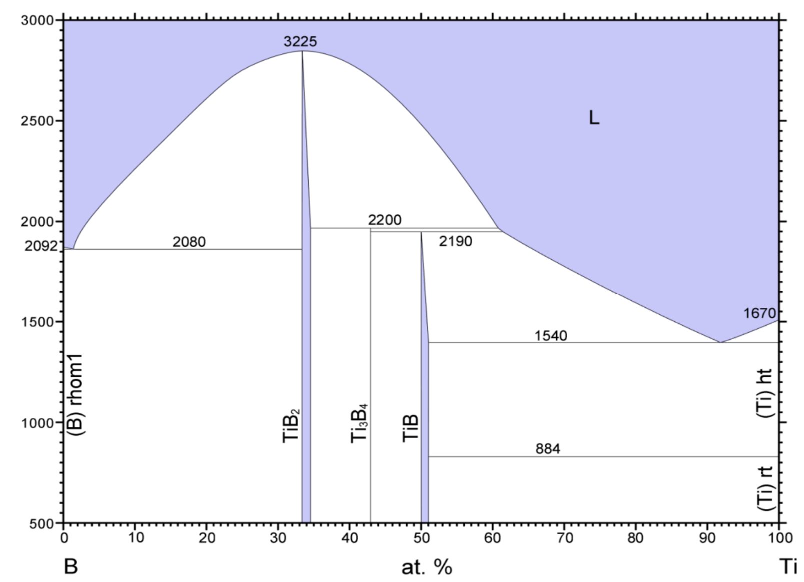
Kinetics Results

- Additional reaction steps in Co Al designs that exhibit spin – as seen on phase diagram
- Large bilayer forms diffusion coupes at boundaries causing all intermediate phases to form
- Thin bilayers allow fast mass transfer and more direct conversion to final phase
- Increase heat release rate and drives faster reaction propagation

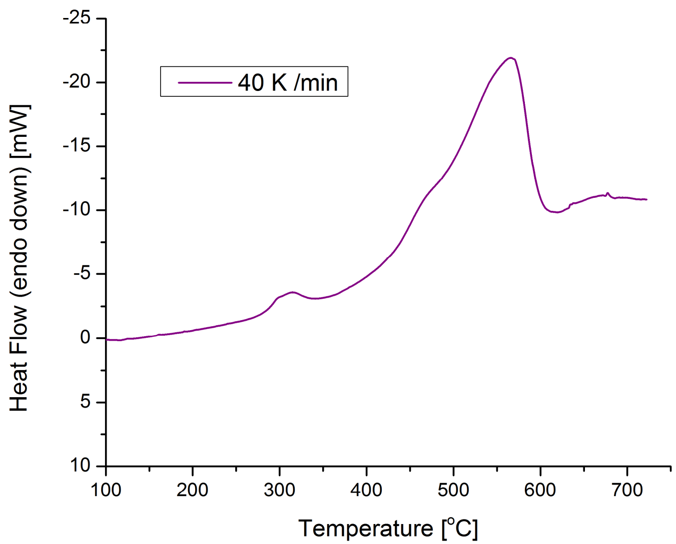


Ti/B phase diagram exhibits fewer intermediat e phases than Co/Al



RVR10

Differential Scanning Calorimetry of Ni/Ti Reactive Foils



Ni/Ti foil, 30 nm bilayer

- Ni/Ti exhibits a single dominating reaction step for completion
- Likely reason for its ability to propagate with such low exothermicity

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RVR10 Comparison of spinning and non spinning Co/Al
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Conclusions

- Spin not solely based on exothermicity
- Also dependent on kinetics and reaction pathway
  - Kinetics are design dependent
  - Defines energy input to initiate reaction
  - Also defines *rate of heat release* to continue reaction propagation
  - Favorable kinetics increase likelihood of propagation and SHS stability
    - Ni/Ti's ability to propagate despite weak exothermicity
    - Unfavorable reaction progress in thick bilayer Al/Co

Additional efforts

- Determine phases formed at each reaction step
  - Co/Al (2 bilayer designs), Ti/2B, Ni/Ti
- Thermal imaging to compare size and magnitude of preheat zone

Crowd Participation

- Any Questions?

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