SAND2019-4495C

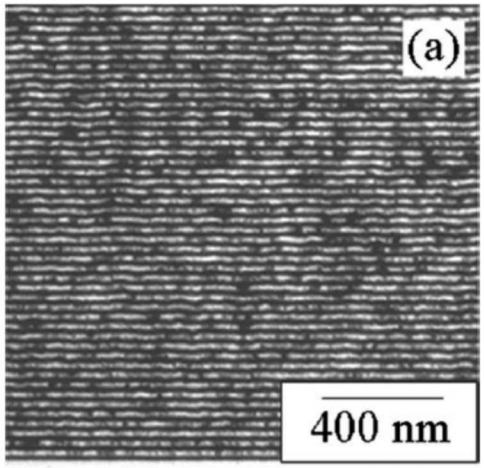
Analytical modelling of propagation velocity in electron transparent nanolaminates

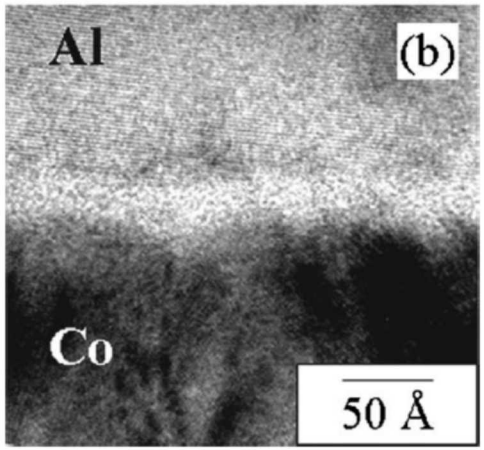
Michael J. Abere, Garth C. Egan, and David P. Adams

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Self-propagating reactions in Co/Al nanolaminates

Sandia National Laboratories



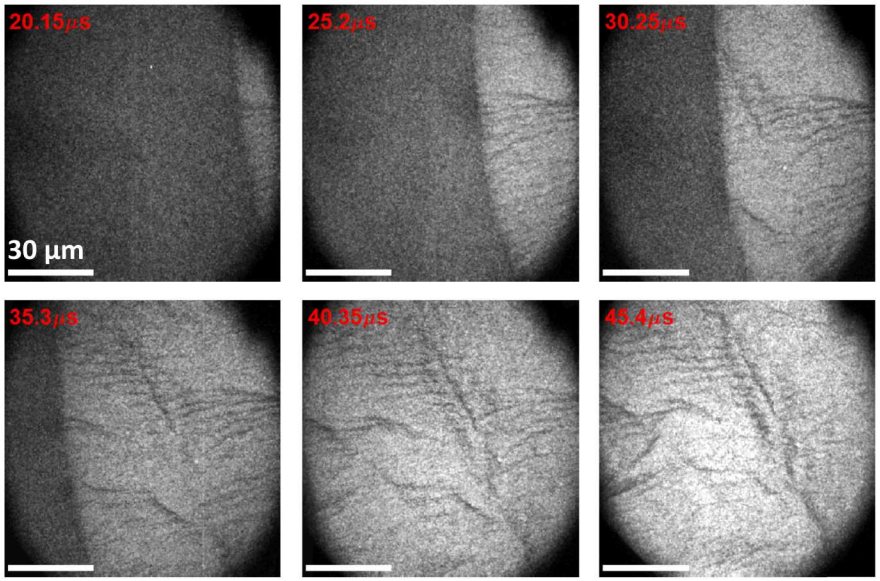


25 nm Bilayer, 6 m/s
15 μs per frame, played at 5 fps

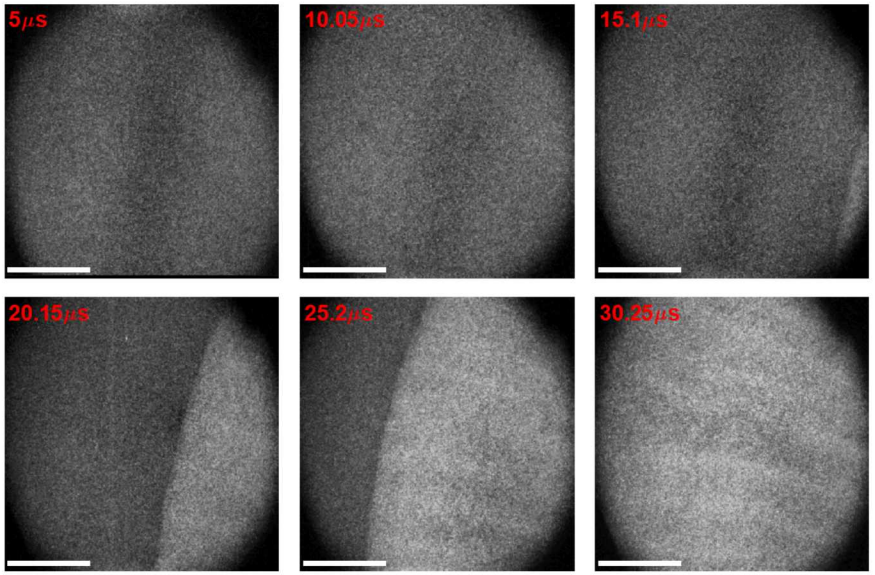
Dynamic TEM shows that foils still propagate at 150 nm total thickness

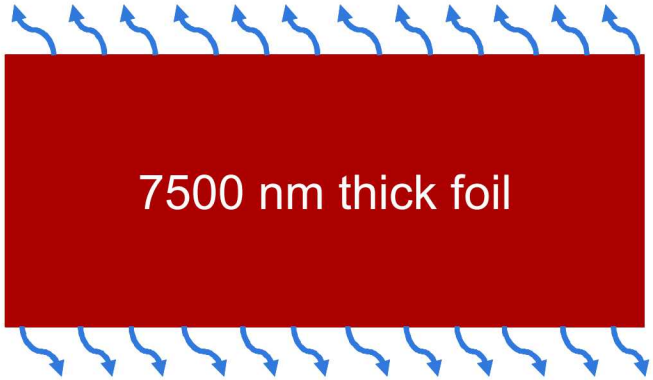
Sandia National Laboratories

6 Bilayers (25.2 nm thick)



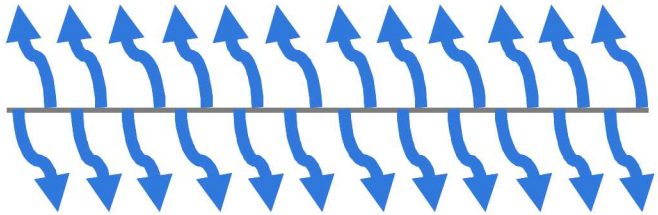
11 Bilayers (15 nm thick)



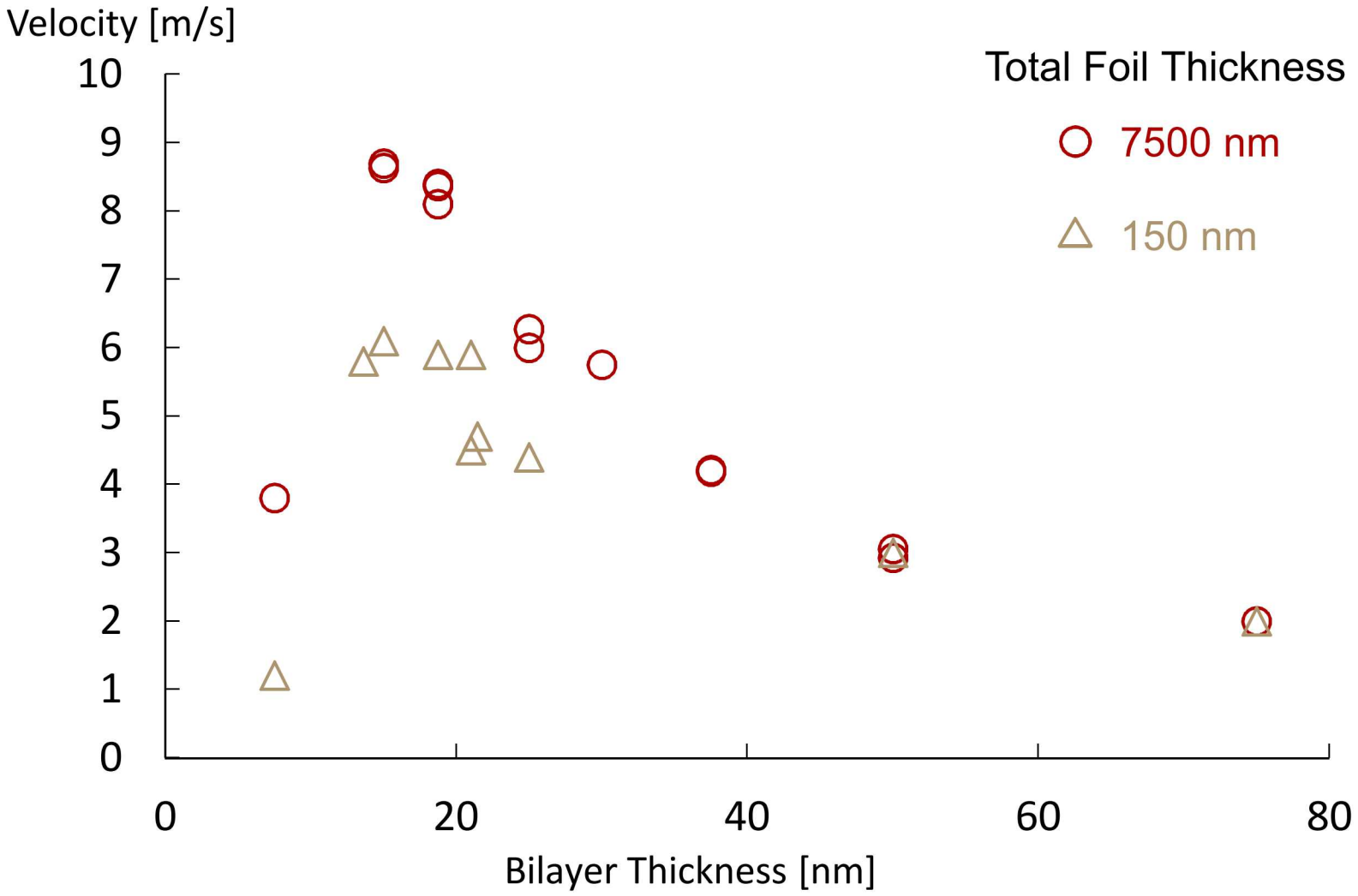


Vs.

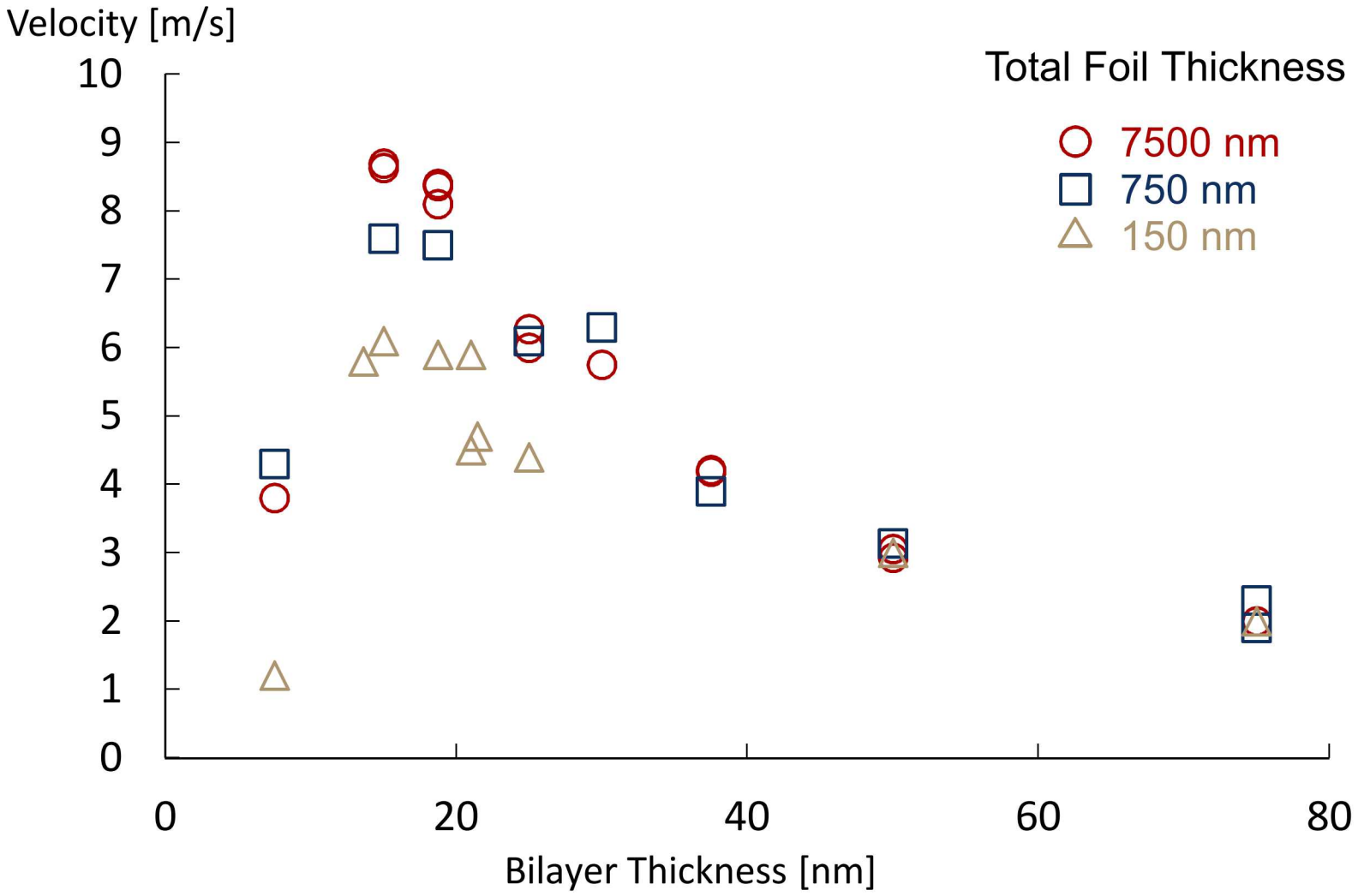
150 nm thick foil



Experimentally Measured Velocities



Experimentally Measured Velocities



The relationship between physical parameters and propagation velocity

From Mann, Gavens, Reiss, Van Heerden, Bao, and Weihs, J. Appl. Phys. 1997:

Fick's Second Law

$$\frac{dC}{dt} - \nabla \cdot D\nabla C = 0$$

Heat Equation

$$c_p \rho \frac{dT}{dt} - c_p \rho \lambda \nabla^2 T = \frac{dQ}{dt}$$

Solved for the case of a moving wave

$$v_x^2 = \left(\sum_{n=odd} \frac{k_n^2}{\alpha_n^2} \right)^{-1} \frac{4\lambda_x^2 R T_f^2 D}{E_A \lambda_y (T_{ad} - T_0)} exp\left(\frac{-E_A}{R T_f} \right)$$

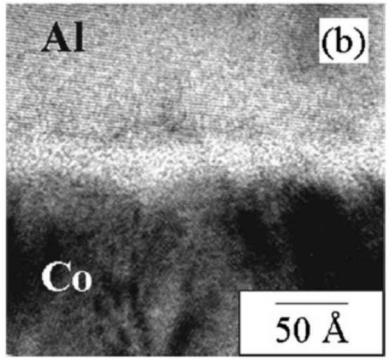
Values for model parameters are informed by experimental data

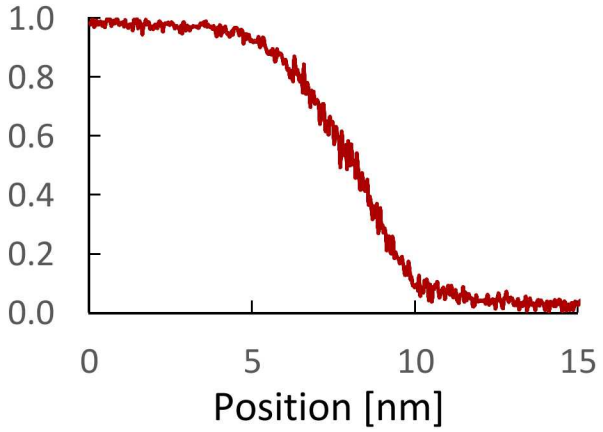
$$v_x^2 = \left(\sum_{n=odd} \frac{k_n^2}{\alpha_n^2}\right)^{-1} \frac{4\lambda_x^2 R T_f^2 D}{E_A \lambda_y (T_{ad} - T_0)} exp\left(\frac{-E_A}{R T_f}\right)$$

Series of eigenvalues, α_n, and Fourier coefficients, k_n, account for the chemical potential lost to imperfect interfaces as a function of initial concentration, $C_0(y)$, for quarter period δ.

$$\alpha_n = \frac{n\pi}{2\delta} \qquad k_n = \frac{1}{2\delta}\int_{-2\delta}^{2\delta} C_0(y)\, sin(\alpha_n y) dy$$

Concentration at each 1 Å wide mesh point comes from STEM-EDS to solve $C_0(y)$

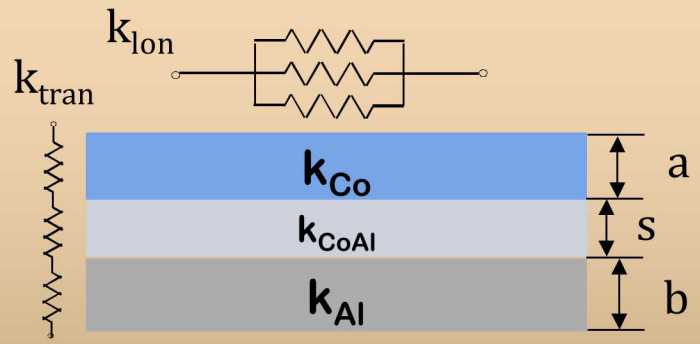




Thermal Conductivity in a three-parameter fit of velocity data

$$v_x^2 = \left(\sum_{n=odd} \frac{k_n^2}{\alpha_n^2}\right)^{-1} \frac{4\lambda_x^2 R T_f^2 D}{E_A \lambda_y (T_{ad} - T_0)} exp\left(\frac{-E_A}{RT_f}\right)$$

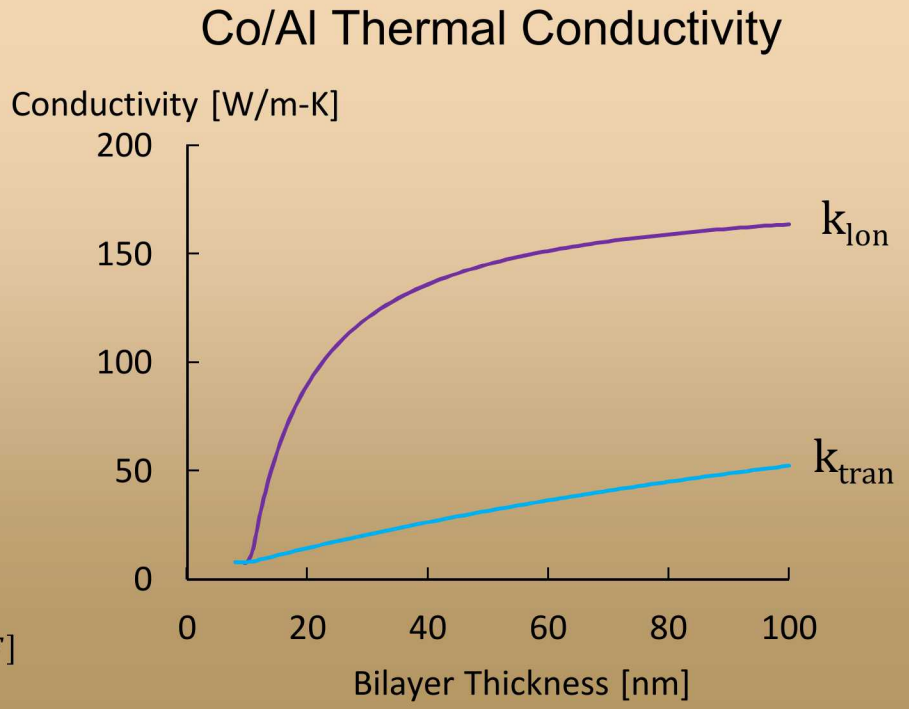
Heat of reaction from calorimetry and enthalpy-temperature diagrams allow for the calculation of adiabatic and flame temperatures



$$F = 1 - \frac{s}{a \cdot w} \qquad w = 1 + \frac{b}{a}$$

$$k_{tran}(t,T) = \left(\frac{F}{k_{Al}(T) \cdot w} + \frac{F(w-1)}{k_{Pt}(T) \cdot w} + \frac{1-F}{k_{AlPt}(T)}\right)^{-1}$$

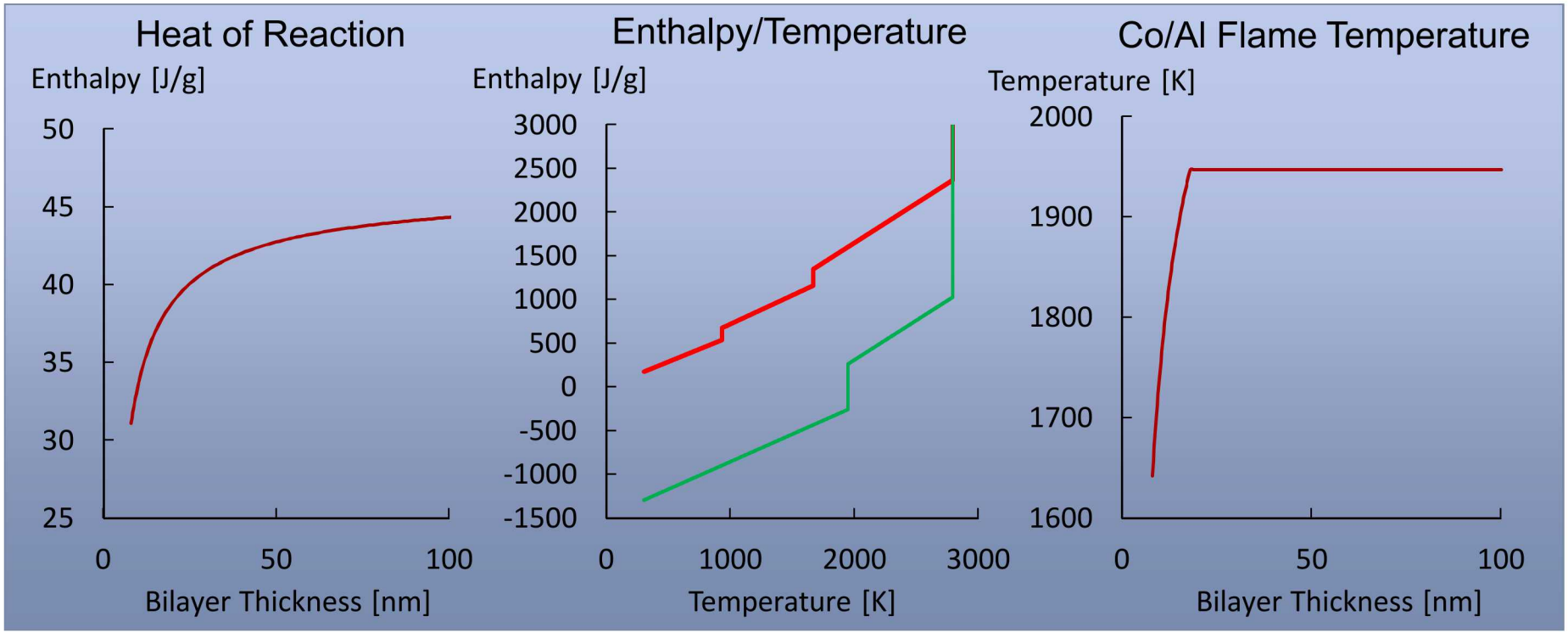
$$k_{lon}(t,T) = \frac{k_{Al}(T)F}{w} + \frac{k_{Pt}(T)F(w-1)}{w} + k_{AlPt}(T)[1-F]$$



Values for model parameters are informed by experimental data

$$v_x^2 = \left(\sum_{n=odd} \frac{k_n^2}{\alpha_n^2}\right)^{-1} \frac{4\lambda_x^2 R T_f^2 D}{E_A \lambda_y (T_{ad} - T_0)} exp\left(\frac{-E_A}{R T_f}\right)$$

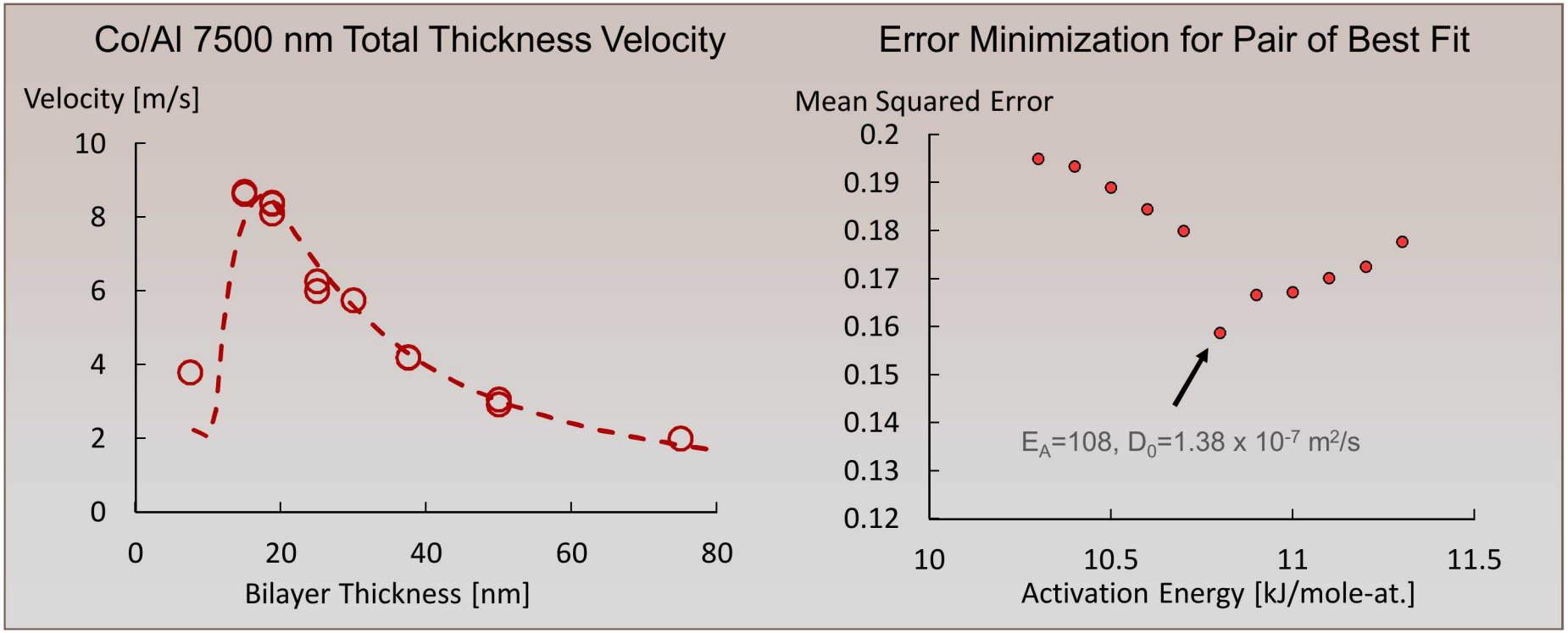
Heat of reaction from calorimetry and enthalpy-temperature diagrams allow for the calculation of adiabatic and flame temperatures



Pair of best fit for activation energy and diffusion coefficient

$$v_x^2 = \left(\sum_{n=odd} \frac{k_n^2}{\alpha_n^2}\right)^{-1} \frac{4\lambda_x^2 R T_f^2 D}{E_A \lambda_y (T_{ad} - T_0)} exp\left(\frac{-E_A}{R T_f}\right)$$

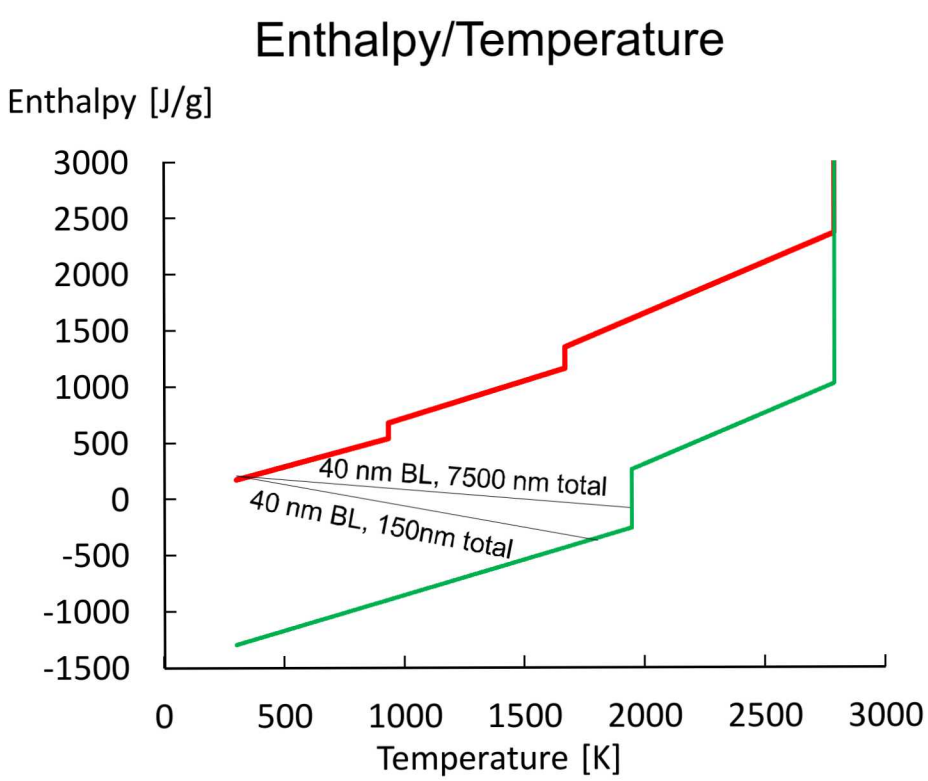
Heat of reaction from calorimetry and enthalpy-temperature diagrams allow for the calculation of adiabatic and flame temperatures

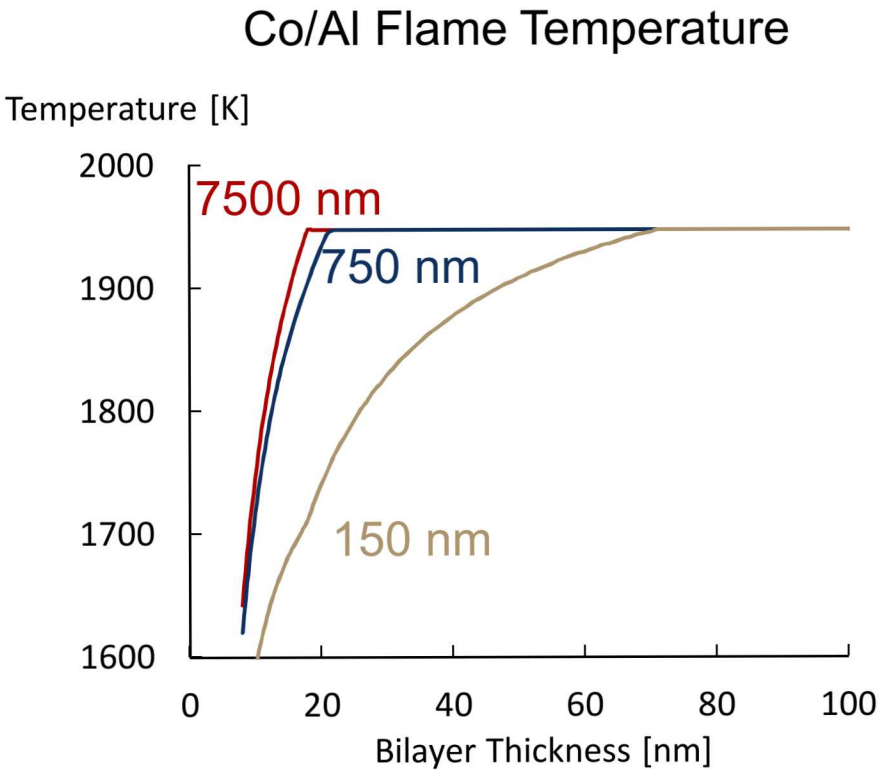


Accounting for Heat Loss in Model

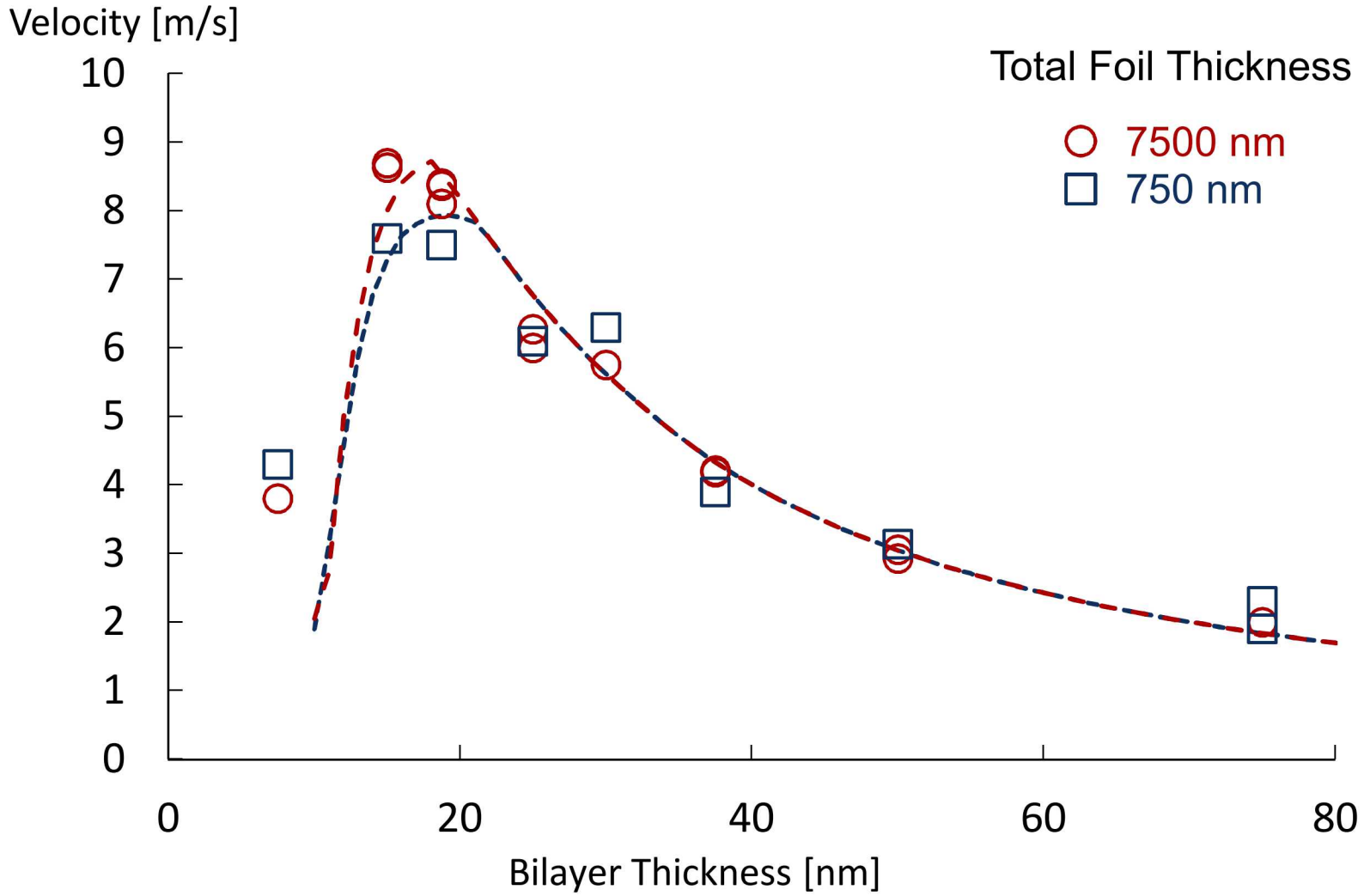
Flame temperature is determined based on the enthalpy/temperature diagram from the heat of reaction minus heat lost to radiation by:

$$Q_{rad}(T) = -\varepsilon \frac{2\sigma(T^4 - T_0^4)}{b_{Total\ Thickness}}$$

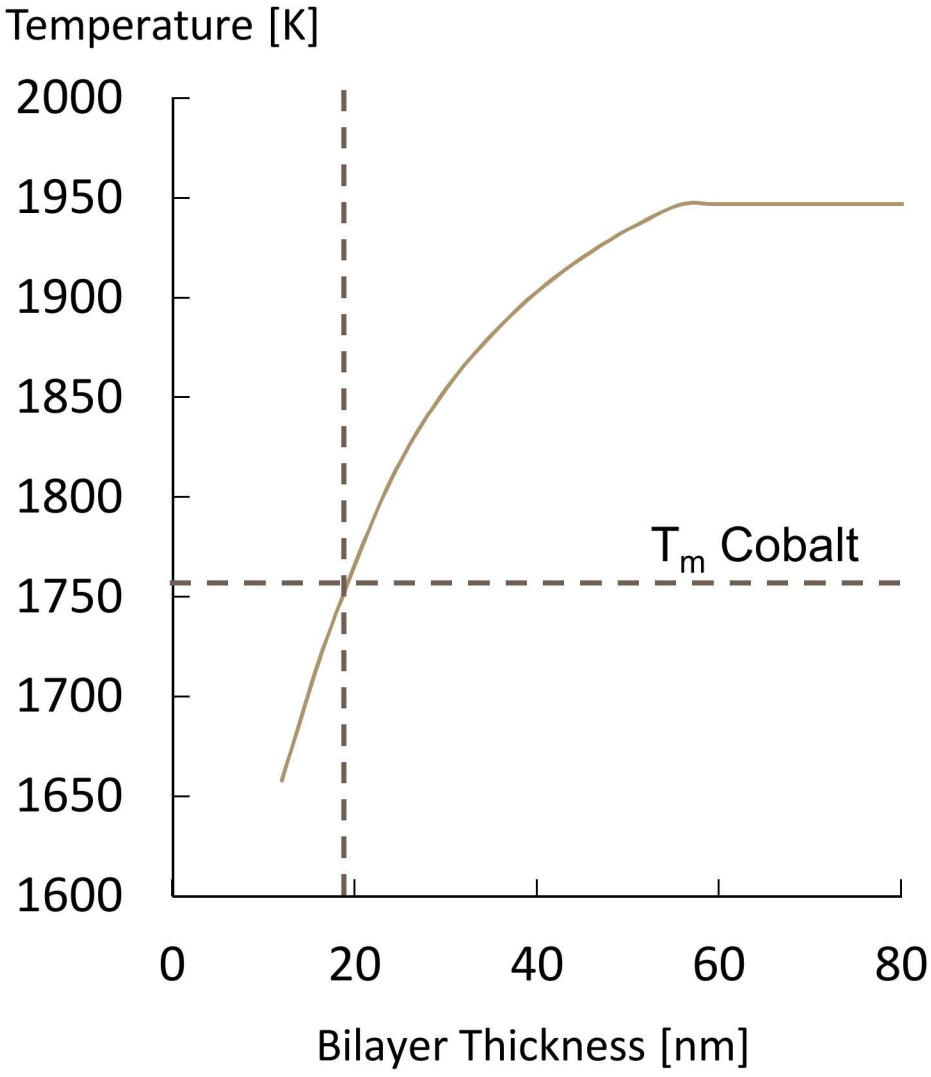


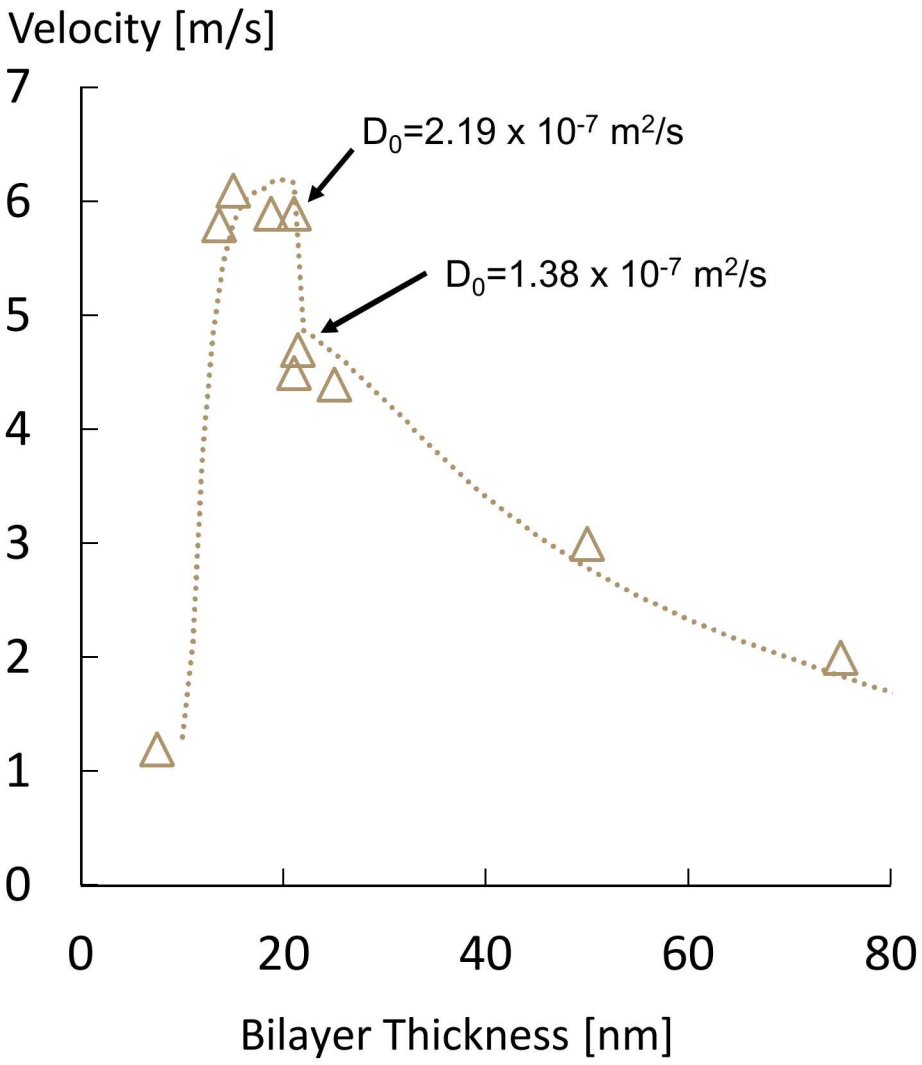


Radiation loss in 750 nm thick foil reduces peak velocity



Mass transport rate is dependent on whether both reactants melt

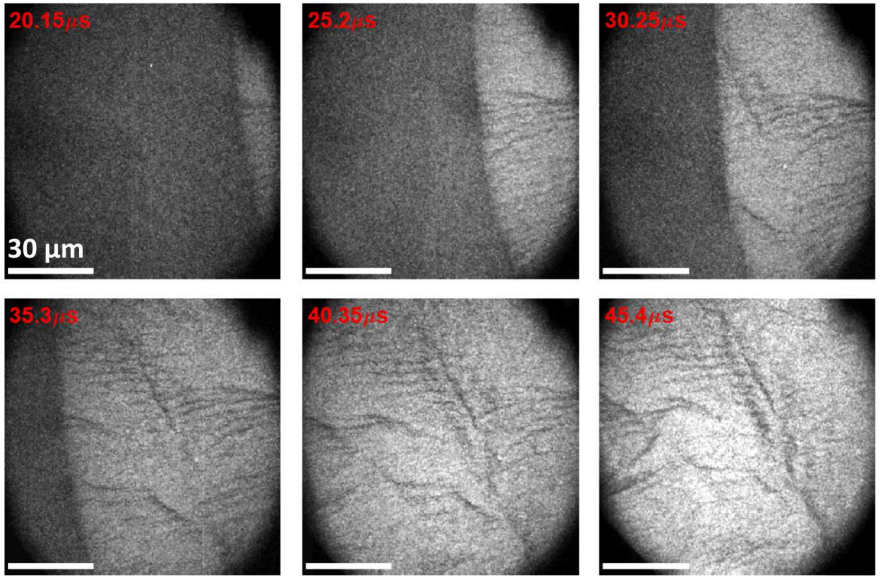




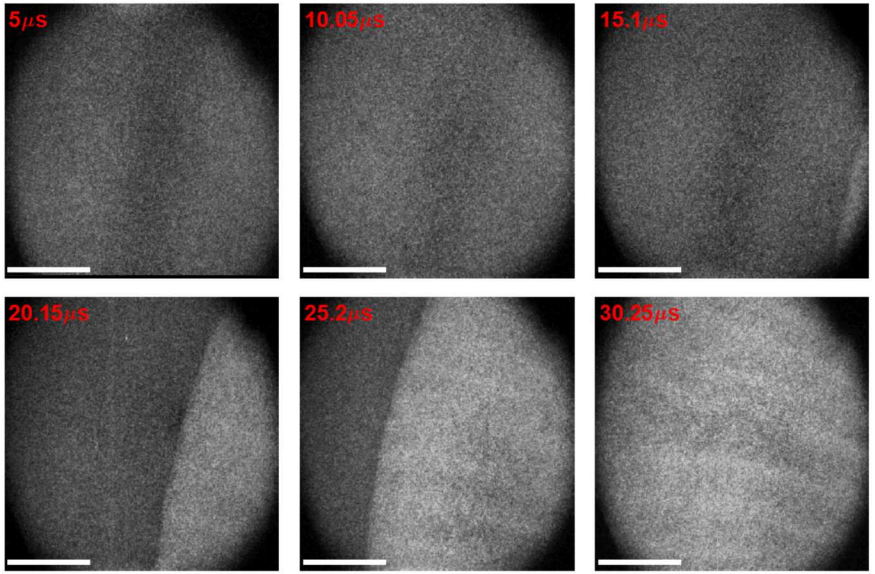
Reactant phase determines kinetic path to product formation

Sandia National Laboratories

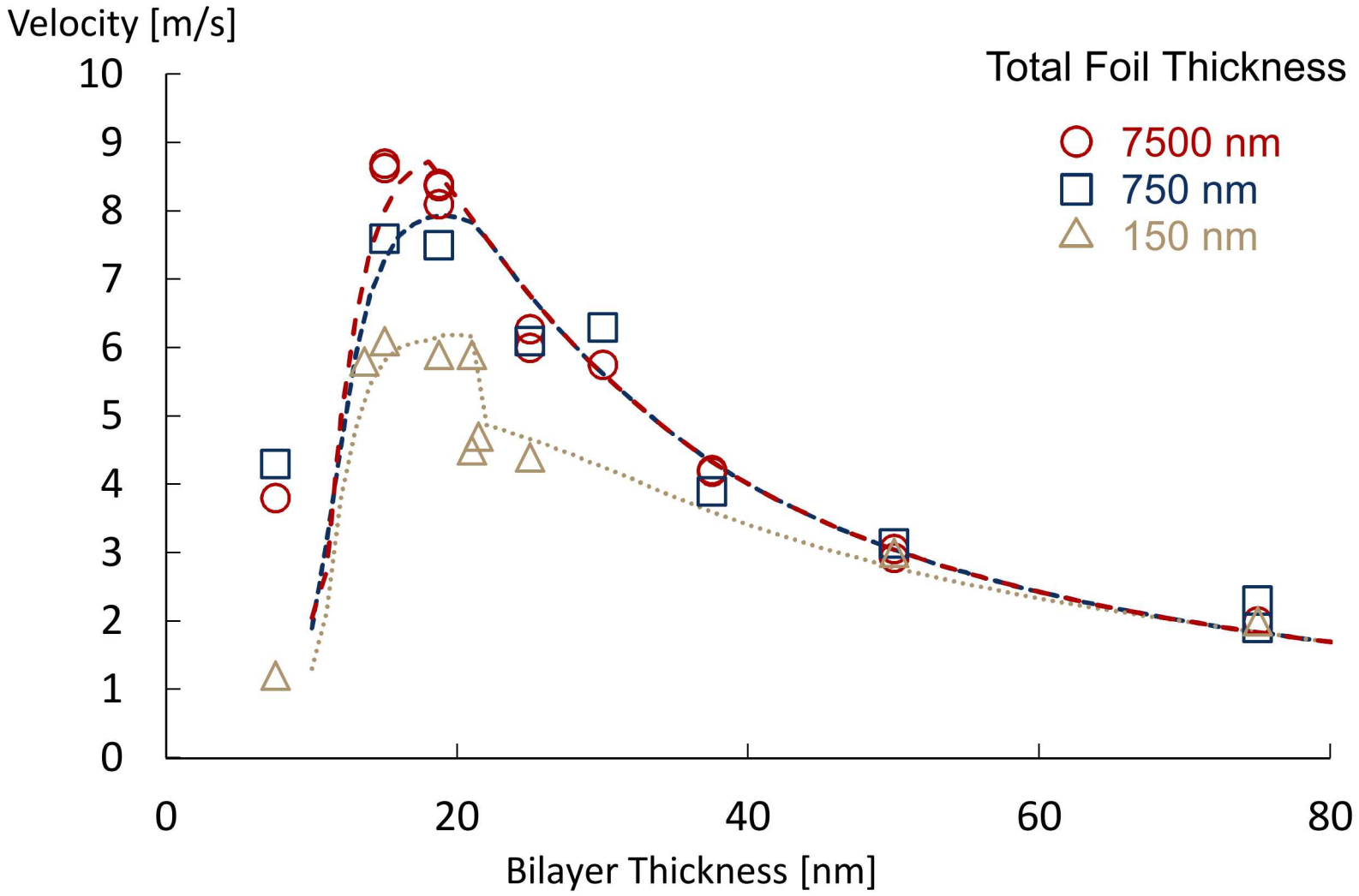
6 Bilayers (25.2 nm thick)



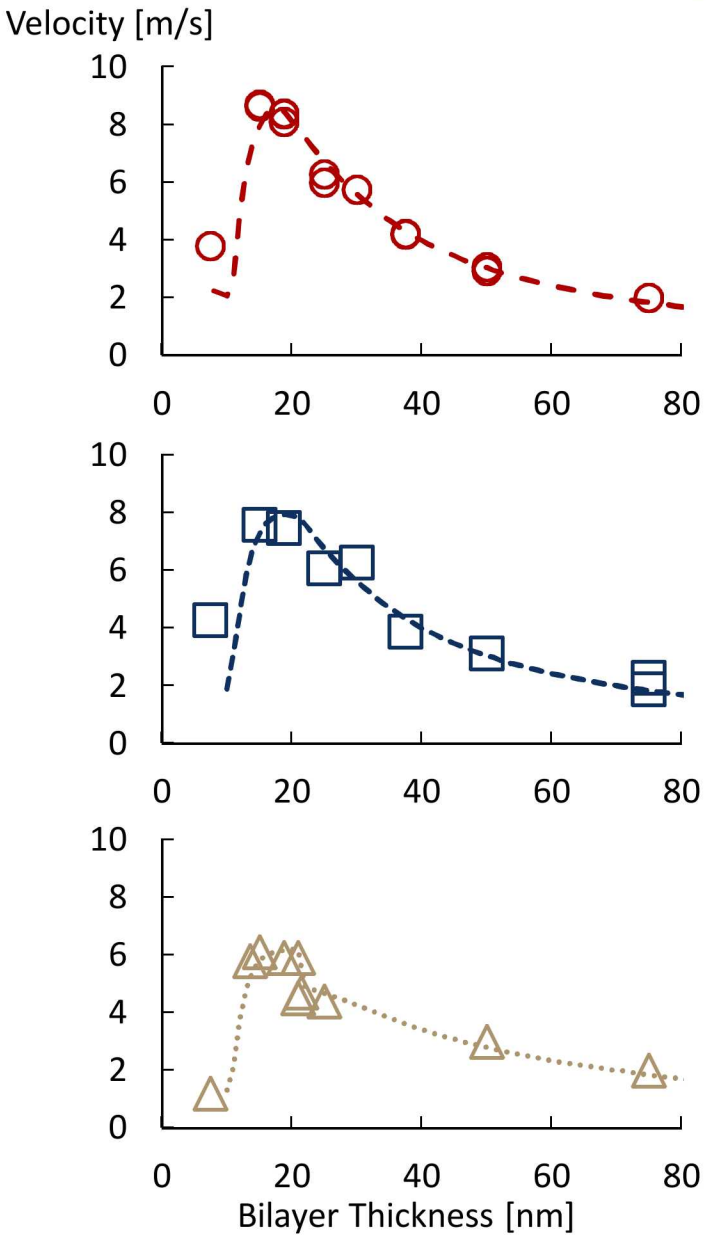
11 Bilayers (15 nm thick)



Model Fits to Measured Velocities



Summary



We have populated the Mann model based on experimental data in order to fit values for activation energy and diffusion coefficient for Co/Al

Energy loss from radiation can lower flame temperatures for thin foils and bilayer designs with reduced heat of reaction due to premixing

The propagation velocity of electron transparent Co/Al samples is dependent both upon radiation effects and phase path dependent reaction kinetics