

In situ X-ray Diffraction Studies of Rapidly Propagating Exothermic Reactions in Nanostructured Multilayer Foils

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Reactive multilayer foils are energetic materials comprised of alternating nano-scale layers of materials that can sustain a self-propagating exothermic reaction. The reaction characteristics, including the maximum temperature, heat released, and speed of propagation, can be precisely controlled by appropriate design of the multilayer architecture. However, the characteristic length and time scales make studying the reaction itself a significant challenge. For instance, the reaction zone is typically about $100\ \mu\text{m}$ wide and propagates at about $1\ \text{m s}^{-1}$, reaching temperatures in excess of $1500\ ^\circ\text{C}$ in less than $100\ \mu\text{s}$, followed by rapid cooling. One aspect that makes these reactions interesting from a scientific point of view is that the rapid heating and cooling may provide kinetic constraints which would alter the phase transformations relative to those observed in a slower reaction (*e.g.* by annealing) in the same multilayer system.

We have studied phase evolution during self-propagating reactions in Al/Ni reactive multilayers, *in situ* and in real time, by means of x-ray diffraction using a pixel array detector at the Cornell High Energy Synchrotron Source. The time resolution ($\sim 50\ \mu\text{s}$) and spatial resolution ($\sim 50\ \mu\text{m}$) of the measurements was sufficient to allow us to observe the phase transformation sequence in detail. In particular, we can track both the integrated peak area (which is proportional to the volume fraction) and the interplanar spacings for peaks from the various phases.

We observe that the initial reaction occurs within the first $100\ \mu\text{s}$, forming one of the final phases along with an amorphous (possibly liquid) phase. For example, Al/Ni multilayers with an overall composition of Al_3Ni_2 form the cubic intermetallic AlNi and an amorphous phase first, followed by hexagonal Al_3Ni_2 some 37 ms later (during cooling after the reaction front has passed). Formation of Al_3Ni_2 appears to occur by a peritectic transformation similar to that present in the equilibrium phase diagram. We also observe changes in lattice parameter that we attribute to the combined effects of interdiffusion and thermal expansion.