**In-situ** X-ray Diffraction of Phase Transformations in Nanostuctured Reactive Multilayer Foils.

J.C. Trenkle, 1 L. Koerner, 2 M.W. Tate, 2 S. Gruner, 2,3 T.P. Weihns, 1 T.C. Hufnagel 1


**Introduction**

Reactive multilayer foils are composed of nanoscale layers of materials that can sustain a self-propagating exothermic reaction when a thermal pulse is applied [1-5]. For a complete discussion of reaction mechanisms, see ref. [4]. The reaction zone is ~100 µm wide and propagates at ~1-10 m s⁻¹, reaching temperatures in excess of 1500 °C in less than 100 µs [4,5]. These reaction characteristics provide a unique opportunity to study kinetically limited phase transformations.

We have used a pixel array detector (PAD) with microsecond temporal resolution to study these phase transformations *in-situ* in Al/Ni and ZrNi reactive multilayer foils at the Cornell High Energy Synchrotron Source (CHESS).

**Methods and Materials**

Scattering was done in transmission at CHESS D1 beam-line, which has a flux of 10¹⁶ photons s⁻¹ mm⁻¹, on free-standing (~20-40 µm thick) Al/Ni and ZrNi reactive foils with predicted overall compositions of Al,Ni₃ and near Zr,Ni (a line compound), respectively. We used a beam energy of 8.2 keV and a spot size of ±200 µm, which is similar to the width of the reaction zone.

We initiated the reaction in the foils using a resistively-heated filament. Data collection by the PAD was triggered through an optics system, which detected light emitted from the propagating reaction front as it approached the x-ray beam. The PAD, which is 15 x 15 mm with 150 x 150 µm pixels, was centered on diffracted beams, capturing partial diffraction rings in a 30° to 50° 20 range.

Detector integration times ranged from 50 µs to 30 ms. The PAD stores eight progressive frames per experiment in each pixel. We introduced initial time delays in the data collection in order to acquire diffraction data out to ~500 ms after the reaction front had passed the x-ray beam.

**Results**

In both multilayer systems, the initial reaction occurs within the first 100 µs and forms one or more of the final phases, without any metastable intermediate phases being observed. The ZrNi reactive foils, for example, form orthorhombic ZrNi first, with tetragonal Zr₂Ni being first detected some 120 ms later (during cooling) and taking an additional 100 ms to grow to completion (Fig. 1). In the Al/Ni reactive foils, all final phases, trigonal Al,Ni₃ and cubic AlNi, form simultaneously during the first 100 µs.

**Discussion**

From phase diagrams and predicted compositions, the expected stable phases are Al,Ni₃, AlNi and Zr₂Ni. When ZrNi and Al/Ni multilayers are heated at a rate of 20-40 °C min⁻¹, metastable amorphous ZrNi and Al,Ni₃ and AlNi form, respectively, prior to any final phase formation because of kinetic and thermodynamic barriers [6,7]. In a self-propagating reaction, however, the reaction temperatures (1000 °C and 1600 °C in Zr,Ni and Al/Ni foils, respectively) significantly reduce these barriers, allowing for diffusion and nucleation of final phases in <50 µs.

Kinetic limitations are still realized in the delayed development of the ZrNi phase, which is possibly the result of the high ratio of layer thicknesses (3:1 Zr:Ni) and limited Zr diffusion observed in Zr/Ni couples [8]. With a 50 nm bilayer period (thickness of a Zr and Ni layer), complete mixing of the layers and growth of the ZrNi phase appears restricted in the initial 100 µs.

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**References**


**Fig. 1:** Plot of measured d-spacing vs. time. The first two data points are before the reaction front has entered the x-ray beam. The Zr to ZrNi transformation happens within 100 µs while Zr₂Ni forms 120 ms later.