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

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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 ºC 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 Zr/Ni 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 10^{10} photons s^{-1} mm^{-1}, on free-standing (~20-40 μm thick) Al/Ni and Zr/Ni reactive foils with predicted overall compositions of Al_3Ni_2 and near Zr_2Ni (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 30º to 50º 2θ 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 Zr/Ni reactive foils, for example, form orthorhombic ZrNi first, with tetragonal Zr_2Ni 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_3Ni_2 and cubic AlNi, form simultaneously during the first 100 μs.

Discussion

From phase diagrams and predicted compositions, the expected stable phases are Al_3Ni_2+AlNi and Zr_2Ni+ZrNi. When Zr/Ni and Al/Ni multilayers are heated at a rate of 20-40 ºC min^{-1}, metastable amorphous ZrNi and Al_9Ni_2 and Al_3Ni 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 ºC and 1600 ºC 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 Zr_2Ni 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 Zr_2Ni phase appears restricted in the initial 100 μs.

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References

[1] J.A. Floro, J. Vac. Sci. Technol. **4**, 631 (1986).
[2] C.E. Wichersham, J.E. Poole, J. Vac. Sci. Technol. **6**, 1699 (1988).
[3] L.A. Clevenger, C.V. Thompson, K.N. Tu, J. Appl. Phys. **67**, 2894 (1990).
[4] T.P. Weihs, Self-Propagating Reactions in Multilayer Materials, Chapter F7, IOP Publishing Ltd (1997).
[5] A.J. Gavens, D. VanHeerden, A.B. Mann, M.E. Reiss, T.P. Weihs, J. Appl. Phys. **87**, 1255 (2000).
[6] B.M. Clemens, W.L. Johnson, R.B. Schwarz, J. Non-Cryst. Solids **61&62**, 817 (1984).
[7] K.J. Blobaum, D. VanHeerden, A.J. Gavens, T.P. Weihs, Acta Mater. **51**, 3871 (2003).
[8] Y.-T. Cheng, W.L. Johnson, M.-A. Nicolet, Appl. Phys. Lett. **47**, 800 (1985).

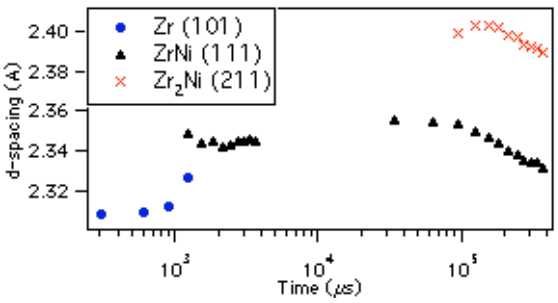

Fig. 1.

FIGURE 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 Zr_2Ni forms 120 ms later.