

TEM Analysis of Structural Transformation in Al/Ni Nanomaterials under High Energy Ion Irradiation

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The paper addresses the effect of irradiation by accelerated ion beam on the structural transformation of Al/Ni multilayer nanomaterials studied by Transmission Electron Microscopy (TEM). The Al/Ni multi-layered nanomaterials are promising nanostructured energetic composite materials [1-2] that exhibit tunable ignition properties to a variety of external excitation methods including friction, shock waves, electrical sparks, and local heating. Because the ignition of such materials depends on their atomic structure and composition, irradiation may provide a novel approach for modification of the reactivity of the nanostructured energetic composite materials. Here we study the structural transformation in Al/Ni layers under irradiation.

Magnetron sputtering and electron beam evaporation have been used to fabricate free-standing reactive multilayer nanostructured foils [3]. High energy carbon and aluminum ion beams with different charge states and intensities were used to irradiate the samples. The samples were analyzed by TEM using both high resolution TEM (HRTEM) and High Angle Annual Dark Field (HAADF) scanning TEM (STEM) modes at FEI Titan 80-300 electron microscope. The microscope was operated at 300 keV and equipped with an Oxford Inca EDX detector. A TEM cross-sectional sample that included the NiO-Ni interface was prepared from the top surface by Focus Ion Beam (FIB) using FEI Helios SEM/FIB dual beam equipment.

It has been demonstrated that a significant enhancement of reactivity of Al/Ni materials after relatively short-term (40 min) high energy (20 MeV) irradiation by $^{12}\text{C}^{4+}$ ions (below the ignition threshold), is associated with structural transformations that lead to a decrease in the thermal self-ignition temperature and ignition delay time. Indeed x-ray diffraction (Fig. 1) indicates that defect formation in the samples under irradiation leads to a decrease of the diffraction peak intensities of Al and Ni in irradiated materials as compared to the original foils. At the same time, the full width at half maximum (FWHM) of the Al and Ni peaks (not shown) exhibits different trends with irradiation time indicating that longer irradiation can facilitate the growth of Al crystallites while decreasing Ni crystallite size. This observation agrees well with TEM/STEM analysis (Figs. 1 and 2) that evidences the intermixing of Al and Ni at layer interfaces. Both high resolution TEM and electron diffraction indicate that formation of amorphous materials at the interfaces with small (2-3 nm) crystals of the Al_3Ni intermetallic phase occur in the amorphous regions. It can be seen that the nuclei of Al_3Ni crystals are distributed in the Al-rich phase close to every other Al/Ni interface for the 40 min irradiated foil (Fig. 2). It is interesting that these nuclei line-up perpendicular to the direction of the incident beam. Such structures confirm that the beam induces solid-state diffusion of Ni into the Al layer, where nucleation of Al_3Ni phase takes place.

Thus, the enhancement of multilayer energetic nanomaterial reactivity is shown to be associated with

radiation-induced structural transformations including defect formation and intermixing of metals at the interfaces that lead to presence of amorphous layers at the interfaces with Al_3Ni intermetallic nuclei.

References:

- [1] Zhou, X.; et al. *ACS Appl. Mater. Interfaces* **2014**, *6*, 3058–3074
 [2] Dreizin, E. L. *Prog. Energy Combust. Sci.* **2009**, *35*, 141–167.
 [3] Weihs, T.P., In *Metallic Films for Electronic, Magnetic, Optical and Thermal Applications: Structure, Processing and Properties*, K. Barmak and K. R. Coffey, Eds.; Woodhead Publishing, Swaston, UK, **2014**, Chapter 6, 160-243

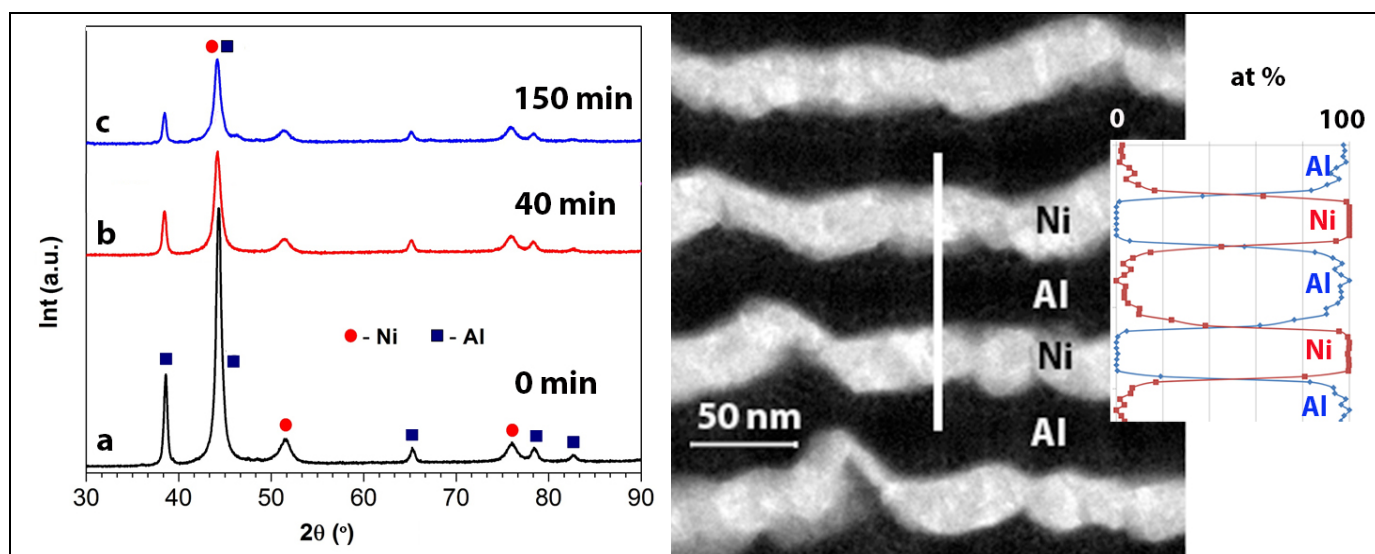


Figure 1 XRD data (left) of irradiated Ni/Al foils as compared to the original one and HAADF STEM image (right) of the Ni/Al foil irradiated for 40 minutes. Inserted is Al and Ni distribution along the vertical line showing intermixing of Al and Ni at the interfaces.

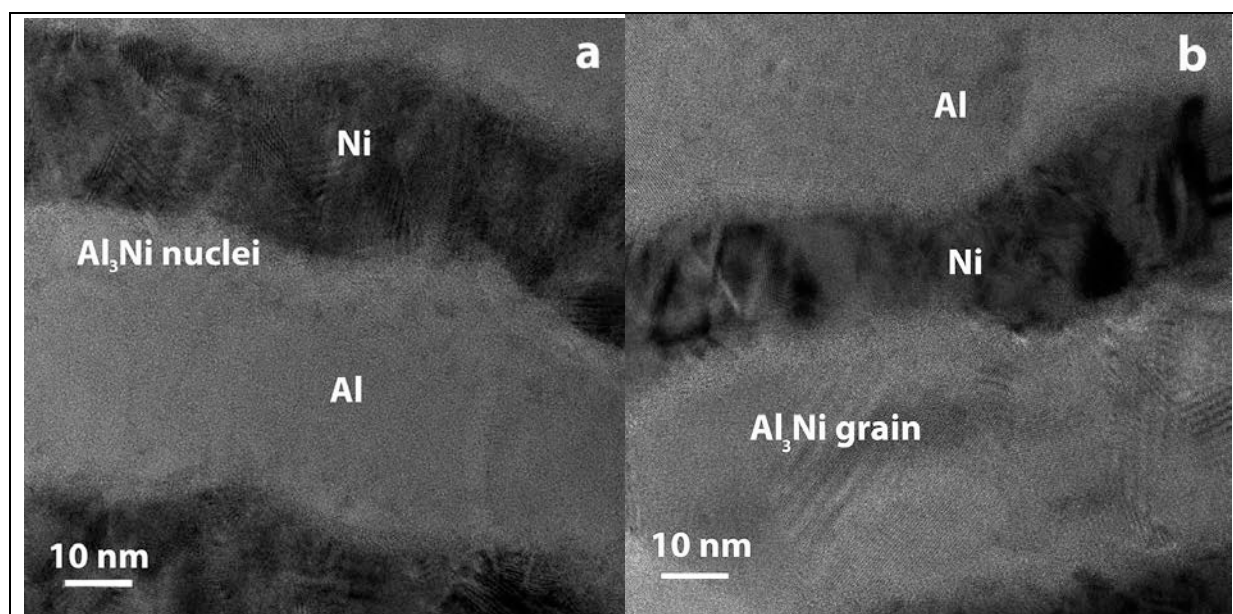


Figure 2 TEM images of Al-Ni foils irradiated for 40 (a) and 150 (b) minutes showing the Al and Ni intermixing and the formation of Al_3Ni nuclei (a) and small Al_3Ni grains (b).