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Effects of processing and powder size on microstructure and reactivity in arrested reactive milled Al + Ni

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Abstract

Ball-milling Al-metal powders can result in self-sustaining high-temperature synthesis in intermetallic-forming systems. Here, Al and Ni powders with similar composition are used to investigate how microstructural differences affect the measured time to reaction (TTR) between powders of different sizes processed under milling conditions specified by statistically designed experiments. Linear statistical models predicting the TTR and the change in temperature (ΔT) are built from these experimental results. The time required to observe a self-sustained high-temperature synthesis of NiAl with different combinations of the powders and ball-milling conditions vary by almost an order of magnitude. Comparisons of powders milled to times corresponding to percentages of their averaged TTR show similar reaction initiation temperatures despite the difference in total milling time. Several distinct arrested reactions within the powder grains exhibit rapid solidification or incomplete diffusion of Ni into Al, forming porous Ni-rich layered structures. The partially reacted grains suggest that the composite laminate particles are not forming intermetallic on the grain scale, but on the localized scale between layers.
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1. Introduction

High energy ball-milling has been used extensively to intimately mix metal–metal or metal–metal-oxide powders. The powder is processed through repeated impacts of a "grinding" media usually made from elastically deforming spherical balls enclosed in a sealed container; this processing produces materials with highly refined microstructures, enhanced strength or metastable phases [1–16]. This technique has been shown to synthesize intermetallic compounds resulting from gradual and explosive formation at the grain level [3,4,6,9–13]. A self-sustained high-temperature synthesis (SHS) of intermetallics may release large amounts of heat during intermetallic phase formation on the order of that released by explosives [17]. The gasless reaction from reactive milled (RM) experiments forming nickel aluminides [3,4,6,8–10,12,15,16,18,19], titanium-based alloys [5] and combustion reactions in metal–metal-oxides [11,13,14] in a ball-mill are several examples of materials that exhibit SHS.

The microstructure within ball-milled powder grains is highly heterogeneous and is refined during processing until reaction occurs. The refinement process consists of the cold welding of powder grains within the contact areas where balls either impact with one another or with the walls of the vessel [5,8,21]. Prior to reaction, the particle microstructure may consist of many layers due to the flattening of the particles for materials of relatively high deformability. Many investigations have been reported on the kinetics of reaction in idealized multilayered intermetallic systems that may be applicable to the reactions observed

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in ball-milled powder particles [17–19,22,24,25]. It was shown in Ref. [17] that reaction propagation velocities in sputter-deposited Al/Ni and Al/Monel are affected by the layer thickness, temperature and intermixing at the material interface. Additionally, the heat of reaction was found to be lower for laminate thicknesses less than 50 nm due to the amount of interfacial mixing [17]. Atomistic simulations of Al/Ni nanolaminates under shock loading have shown that the introduction of voids either at the interface of the layers or within the Al enhanced the rate of intermetallic reaction [26]. By comparison, prerequisite conditions for reaction in Al/Ni powders observed in high-energy ball-milling are a suitable reduction in the layer thickness, and it has been suggested that solid-state diffusion times are decreased by several orders of magnitude as defect densities increase [8,27].

Recent investigations show that arrested reactive milling (ARM) techniques can be used to ball-mill constituent powders to a point where they are well blended but have not reacted; this constitutes an important class of candidate materials for heat sources in joining applications [17] or additions to explosive materials [20,28–31]. The layers within the particles may exhibit a very high defect density depending on the impact energy of the grinding media against the vessel walls and the deformability of the constituent powders, which changes with each subsequent impact.

In our recent work, it was shown that the reaction initiation threshold in mixed and ball-milled Ni + Al powders[1] under high-rate mechanical loading depends on the microstructure within the powder particles and the level of strain-hardening, which change as a function of processing time [18]. Comparisons between mixed and ball-milled powders showed a reduction in the mechanically induced reaction initiation for moderate milling times that is attributed, in part, to the high specific surface area between constituent materials. However, upon further milling and corresponding reduction in layer thickness within the powder grains, the reduced ductility resulted in a higher mechanically activated reaction initiation threshold. Thus, the optimum amount of ball-milling was found to depend on the grain microstructure and the level of strain hardening. This dependence is sensitive to the ball-milling process variables, which is addressed in this investigation.

Here we investigate high-energy ball-milling of nominally spherical Ni and Al powders in an equiatomic ratio and vary particle sizes and ball-milling conditions to compare the microstructural differences and reaction characteristics of ARM materials. The time required to observe SHS with different combinations of the powders and ball-milling conditions shown here vary by almost an order of magnitude.[2] Here the time to reaction (TTR) is used as a basis for comparison between the powders, which will be useful in both thermal and X-ray analysis. Three materials are selected to determine the average TTR for given particle sizes and milling conditions to compare the powders as a function of milling time needed for SHS. The temperature change on the outside of the vessel containing the powder is also measured and used to indicate the specific energy release during alloy formation. The resulting NiAl powder grains show signs of melting and void creation, as expected, and several partially reacted powder grains are shown and discussed.

2. Experimental methods

2.1. Ball-milling

For each ball-milling operation, the powders, stainless steel grinding media and process control agent (PCA) were loaded and unloaded into a stainless steel vial within a glovebox filled with Ar. Although Ar has been shown to increase the ambient temperature within the vial, it is used to prevent nitride or oxide formation (see Chapter 15 in Ref. [27]). Four stainless steel vials were used at random to reduce the dependence of milling on any contaminant deposits or difference in roughness on the inside surfaces. It is shown in Ref. [32] that contamination is an order of magnitude less when using stainless steel vials and grinding media: 0.35% Fe contamination was reported for 8 h of continuous milling [21]. Each ball-milling experiment is performed with a molar ratio of $Ni_{0.5}Al_{0.5}$ with three different Ni powders (Ni: 5–15 μm, −300 mesh, −150 + 200, 99.8% pure, Alfa Aesar) and Al powders (Al: H2, H30, H50, 99.7% pure, Valimet, Inc.). The powder was milled in a SPEX-8000 shaker mill (SPEX CertiPrep® Group) with varying amounts of stearic acid as a percentage of the total powder mass (95% reagent grade, Sigma Aldrich) [5]. A recent investigation shows that varying the milling intensity by grinding powders with media of different density affects the degree of refinement and subsequent reaction [14]. Here, changing the size of the milling media varies the milling intensity. Fresh grinding media is used for each experiment to aid in the reduction of Fe contamination due to surface deterioration [8,27].

In total, seven variables are considered: the diameter of the grinding media (d_b (mm)), the charge ratio (i.e. mass of grinding media to the mass of powder, r_{bp}), the total powder mass (m_p (g)), the controlled temperature (T_c (°C)), the size of the Ni and Al powders (d_{Ni} and d_{Al} (μm)), and the amount of PCA (m_{PCA}/m_p). Testing each variable at two different levels would require 128 experiments to quantify the individual or combined effects these changes have on the measured TTR and the temperature change of the vial. Design and analysis experiments principles were used to systematically reduce the number of experiments while maintaining as much information as possible for quantitative analysis [33]. These principles allow us to quantify our results while changing more than one variable at a time (i.e. a factorial design). The term "designed experiments" is

[1] The powders investigated in Ref. [18] are identical to those discussed herein.

[2] This is shown in Table 2 by comparing the time to reaction values from Experiments 10 and 17.