



Influence of the high energy ball milling on structure and reactivity of the Ni + Al powder mixture



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ABSTRACT

Investigation of the micro- and atomic structures for the planetary ball-milled Ni + Al mixtures has revealed existence of intermediate nano-crystalline and amorphous phases, which are not detectable by XRD analysis, but can be observed by means of HRTEM. Annealing of the milled mixtures at moderate temperature, 205–280 °C, transforms the nano-phases into crystal state and makes them XRD-detectable. These nano-scale structures may serve as nucleus for the intermetallic phases produced via reaction between Ni and Al and, therefore, decrease the activation energy of this reaction and diminish the temperature of the reaction initiation. Apparently, the active nano-phases are formed due to sliding of Al and Ni layers during the ball milling. The simple model is suggested, which allows non-contradictory explanations of the nucleus formation and their influence on the reactivity of the mixture.

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1. Introduction

Exothermic reactions in powder mixtures are widely used for combustion synthesis of materials, also named self-propagating high temperature synthesis (SHS) or solid flame [1,2]. This method utilizes the heat generated by exothermal heterogeneous reactions for making the process of chemical synthesis self-sustained. For example, reaction between the Ni and Al powders, resulting in formation of intermetallic compounds:



($x = 0.33; 1.0; 1.5; 1.67; 3.0; \dots$, depending on the desired intermetallic phase), can occur in self-sustained and self-propagating modes. The reactive mixture must be heated locally up to reaction initiation temperature T_i (600–700 °C for Ni + Al), then the mixture self-heats up to 1640 °C or less (depending on x), and chemical transformation completes. High-energy ball milling (HEBM) of such mixtures may significantly increase their reactivity, which means decreasing of T_i and, in some cases, increasing of the reaction wave self-propagating velocity [3–13]. The results are summarized in the

recent review [14]. Increasing of reactivity cannot be explained by trivial grinding (reducing of the particles sizes), since in many cases average particle size increases after the mechanical treatment. The experimental data on the reaction wave propagation velocity are at first view contradictory. On the one hand, activated 3Ni + Al mixtures with loose density [3,5] and pressed samples of stoichiometric Ni + Al mixture with relatively high initial densities (70–80%) [12,13] demonstrate significant (several times) increase of the combustion velocity. On the other hand, pressed samples of stoichiometric Ni + Al mixture with porosity 40–50% exhibits lower combustion velocity as compared to not activated ones [15]. Temperature of self-ignition is determined by relationship between rate of reactive heat generation inside the sample, and rate of heat dissipation into environment (heat loss). All data, published up to now, show that self-ignition and onset reaction temperatures decrease due to mechanical activation [6–20]. The energy of the minimum mechanical impact required for reaction initiation also decreases [12,13]. It was shown that HEBM diminishes the reaction apparent activation energy [10].

It is reasonable to suggest that mechanical treatment causes some transformations in the reactive mixture, which can be named *activating factors*, and these activating factors determine increased reactivity of the mixture. Several hypotheses, suggested in the literature to explain the influence of HEBM on the reactivity of heterogeneous powder mixtures, were analyzed in [8,11,14]. The most

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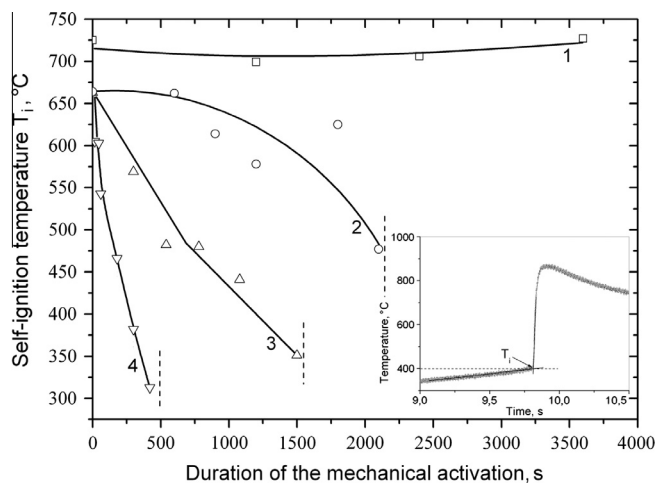


Fig. 1. Dependence of self-igniting temperature of Ni + Al mixture on duration of activation at various acceleration values: 1–12G (air), 2–20G (Ar), 3–40G (Ar), 4–60G (Ar). Heating rate 25 °C/s.

important ones are: (i) decreasing apparent activation energy by the magnitude of the energy stored into reactants during MA; (ii) increasing atomic mobility due to high concentration of the grain boundaries and defects created by plastic deformation; (iii) formation of non-equilibrium meta-stable phases; (iv) increase of the contact (interfacial) surface area between reactants; (v) formation of freshly formed reactive surfaces; and (vi) nano-scale mixing of the reagents. Through high resolution electron microscopy [11], it was shown that short term HEBM not only increases the contact surface area between reagents by thinning and folding the materials, but more importantly, creates regions near the Al/Ni interface where nanometric Ni is embedded in an Al matrix, without oxide barriers. The appearance and development of the nanometrically mixed region is attributed to the significant change in apparent activation energy.

In the present work, we report results of the experimental study on the composition, crystal and atomic structures of these nanometrically mixed boundary regions, which provides deeper insight on the possible mechanism of reaction activation by short term high energy ball milling.

2. Materials and methods

Powder mixtures were prepared using nickel powder PNE-1 (Ni content 99.5 mass.%, particles size <71 μm, loose density 3.4 g/cm³) and aluminum powder ASD-4 (Al content 99.7 mass.%, particles size 30–50 μm). Equimolar Ni + Al blends were ball-milled in the high-energy planetary mill AGO-2 with different centrifugal accelerations of the milling balls (approx. 20G, 40G, and 60G, where $G = 9.8 \text{ m/s}^2$ is a free fall acceleration at normal gravity). The reaction mixture (10 g) and steel milling balls (200 g, $\varnothing 7\text{--}9 \text{ mm}$) were put into the cylindrical steel containers (jars). The jars were evacuated and filled with Ar at 1 bar. For the comparison, some Ni + Al mixtures were ball-milled using Fritsch Pulverisette 4 at lower acceleration, 12G, but with larger balls ($\varnothing 15 \text{ mm}$), in air. These blends were prepared with Aesar Ni and Al powders, 99.9% purity, –325 mesh (average diameter 5–7 μm). Some mixtures were annealed in the vacuum furnace at moderate temperature, from 120 °C to 300 °C for several hours. For the annealing, the powders were spread as thin layer on the copper plate, to avoid self-heating up and self-ignition.

The activated mixtures are composed of lamellar Ni/Al particles, as described in the previous works [4,5,11,12,20]. In order to measure the T_i , small disk-shaped pellets with diameter 3 mm and thickness 0.5–1.0 mm, were cold-pressed from these mixtures. Then the pellets were placed in a boron nitride ceramic crucible and heated by carbon strip heater in Ar (1 bar) atmosphere at constant rate 25 °C/s (for comparison between the different mechanical activation modes) or 50 °C/s (for comparison between the different annealing regimes). When self-sustained exothermic reaction was initiated, the slope of temperature curve sharply increased, as shown in Fig. 1 insert. The point of drastic increase of the temperature rise rate (maximum of the second time derivative) was determined as temperature of reaction initiation (or temperature of self-ignition), T_i . The value of T_i depends on

heating rate, and cannot be considered as intrinsic attribute of the reaction mixture. However, when the heating rate is the same for some set of testing samples, the T_i provide important direct characteristic for comparative reactivity of the mixtures.

The X-ray diffraction analysis of the mixtures was made using ARL XTRA powder diffractometer (Thermo Fisher Scientific, Switzerland). Micro- and nano-structure, local chemical composition, and local atomic structure were examined with High Resolution TEM TITAN (FEI, USA). The samples for the TEM investigations were prepared using Focused Ion Beam Nanolab 6000Helios (FEI, USA). Thin rectangular plates $20 \times 20 \times 2 \text{ μm}$ were cut out of the ball-milled lamellar particles by ion beam and thinned down to 70 nm by ion etching for further HR TEM investigation.

3. Experimental results

The dependences of T_i on regimes of mechanical activation for Ni + Al mixture are presented at Fig. 1. The results show that reactivity of the mixture strongly depends on the acceleration of the planetary ball mills. At lower acceleration (12G) we did not detect any increase of reactivity (i.e., decrease of T_i) even after 1 h of activation. For the acceleration values equal or higher than 20G, significant decrease of T_i was observed; this effect became stronger when the acceleration increased. Temperature of self-ignition gradually decreases with increasing of HEBM time until some critical duration time of activation is exceeded, when the mixture suddenly becomes not ignitable (dashed vertical lines). The powder XDR analysis has revealed that all not-ignitable mixtures consisted mostly of the NiAl B2 phase. Therefore, at critical time of mechanical treatment the reaction (1) occurs inside the jar. For the annealing experiments and structure investigation, we have selected the powders ball-milled at maximum acceleration (60G) for 5 min, which is close to the critical time. It can be expected that the content of the activating factor achieves the highest level in these samples.

Influence of the annealing regime on T_i is shown in Table 1. Annealing does not affect reactivity of the powder mixture prepared in the porcelain mortar without ball-milling. However, the ball-milled mixtures undergo significant influence of the annealing. The gradual increase of T_i with increasing annealing temperature, clearly shows that our hypothetical activating factor deactivates at relatively low temperatures.

The XRD spectra presented at Fig. 2 shows the atomic structure transformations due to ball milling and annealing. Not-milled mixture gives relatively sharp and narrow peaks of Ni and Al (Fig. 2a). After the ball milling, these peaks become broader and less intensive (Fig. 2b), however, no additional peaks appear at the diffraction pattern. No peaks of intermetallic phases are determined after annealing at 120 °C (Fig. 2). The annealing at 205 °C results in appearance of many minor peaks attributed to the orthorhombic phase NiAl₃ (PDF card 02-0416), which become more pronounced after annealing at 280 °C (Fig. 2d and e). Also, some traces of the NiAl B2 phase can be mentioned at Fig. 2e. Annealing at 300 °C leads to formation of NiAl as a major phase (Fig. 2f). Finally, it should be noted that non-milled mixture do not generate any new crystalline phases even after annealing at highest temperature (Fig. 2g). Thus, crystalline intermetallic NiAl₃ and, probably, traces of NiAl appear in the ball-milled samples during annealing in the temperature range 120–280 °C. Formation of these phases corre-

Table 1
Dependence of T_i on the regimes of mechanical activation and annealing.

Annealing regime	Temperature of self-ignition T_i , °C (heating rate 50 °C/s)	
	Non-activated mixture	Activated (5 min, 60G)
No annealing	740	440
120 °C; 6.5 h	715	425
205 °C; 24 h	730	490
280 °C; 6 h	720	550
300 °C; 6 h	735	No ignition

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