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Investigation of the aluminum nitride formation during the aluminum nanopowder combustion in air

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Abstract

The phase formation sequences, intermediate and final products of aluminum nanopowder combustion are studied. The experiments were performed in the Budker Institute of Nuclear Physics, Siberian Branch of the Russian Academy of Sciences, Novosibirsk, at the "Precision Diffractometry II" station (SR beamline No. 6 of the VEPP-3 electron storage ring). The main combustion product was found to be aluminum nitride. In the combustion of aluminum nanopowder aluminum γ-oxide is the first to form, and aluminum nitride arises next. The formation of aluminum nitride probably occurs by successive replacement of oxygen by nitrogen from the aluminum oxide. The use of synchrotron radiation with high photon flux made it possible to determine with moderate time resolution the sequence of stages of formation of crystalline products during combustion of the aluminum nanopowder.

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Keywords: nanopowder; aluminum; combustion; synchrotron radiation; combustion products; aluminium nitride; phase formation sequence.

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1. Introduction

According to existing conceptions [Korshunov et al. (2010)], during Al nanopowder (NP) combustion the formation of an independent crystal phase of aluminum nitride assumes interaction of aluminum only with nitrogen, in a certain volume in the presence of oxygen within a short period of time. Stabilization of nitride can be connected with kinetic suppresion of oxygen diffusion through a nitride layer caused by thermodynamically allowed process of aluminum nitride oxidation by air oxygen. Earlier thermodynamic calculations showed that aluminum nitride can appear as an intermediate product during powdery aluminum combustion, but it has to be oxidized by air oxygen and does not remain in the final products. Reaction of nitride formation in air with high content of aluminum nitride in the final products becomes possible only due to aluminum nanopowder (NP Al). Thus, aluminum nitride synthesis by means of aluminum nanopowder combustion in air seems to be an alternative way of AlN production [Zakorzhevskii et al. (2002) and Wilmanski et al. (2014)].

The aim of this work was to elucidate phase formation sequence during aluminum nanopowder combustion in the air using time resolved X-ray diffraction of synchrotron radiation.

2. Experimental procedure

NP of aluminum was obtained by electrical explosion of aluminum wires, aerogel density is 0.08 g/cm^3. During passivation, additives air is formed "argon coats" for oxygen, nitrogen and water. Wherein the bulk density of NP increases and reaches 0.15 - 0.20 g/cm^3. If not passivated, aluminum NP being stored forms and sintered agglomerates. The bulk density and the degree of agglomeration significantly affect the combustion rate and composition of the reaction products: the content of aluminum nitride reduces, and the unburned aluminum fraction increases. Compaction of NP aluminum (0.5 - 1.0 kg/cm^2) reduces flammability and sintering.

Aluminum NP produced by an electric explosion of conductors in argon was studied. The NP was produced using an UDP-4G installation developed at the Tomsk Polytechnic University which provides an aluminum NP yield of 50 g/h [Gromov and Teipel (2014)]. The diameter distribution of the resulting powder particles is close to a lognormal one with a maximum of 120 nm and an asymmetry to large particle size (Fig. 1). The content of metallic impurities did not exceed 0.2%. The NP was passivated by small addition of air, after which it remained stable when heated in air up to 400° C.

Under condition of thermal explosion (2200-2400°C), aluminum nitride is formed in the gas phase in the second stage of combustion and is stabilized due to the endothermic process, followed by crystallization into a solid phase (exothermic process) [Il'in et al. (2011)]. The growth of crystals during combustion of aluminum NP in air occurs toward the heat flow, which is consistent with the classical concepts of crystal growth [Laudise (1970)]. The time of combustion of aluminum NP and subsequent crystallization of the product was ~ 1 min. The combustion products of the aluminum NP obtained in the mode of thermal explosion were analyzed by Auger electron spectroscopy (ESO-5UM). It was found that the surface consisted of aluminum (42%), nitrogen (28%), and oxygen (30%), which corresponds to the oxidized surface of aluminum nitride.

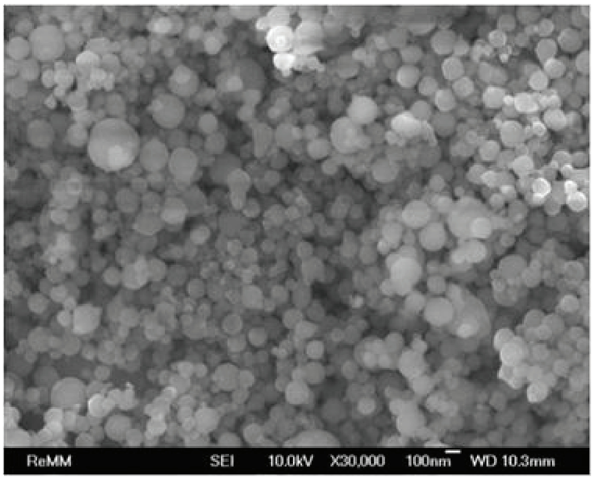

Fig. 1. Microphotography of aluminum nanopowder.

The samples for time resolved XRD analysis were prepared by compaction of the aluminum NP in a steel press mold under a pressure of 7.5 MPa. The diameter of the obtained cylinders was 10 mm, the height was 7 mm, and the weight was 0.4 g. The pressing did not destroy the oxide-hydroxide layers on the surface of nanoparticles (additional oxidation and sintering did not occur), which was monitored by measuring the electrical resistance: the samples after compression did not conduct electric current, which indicated that the durable protective oxide-hydroxide layer on the particles was not damaged. Thus, the pressed samples were nonconductive.

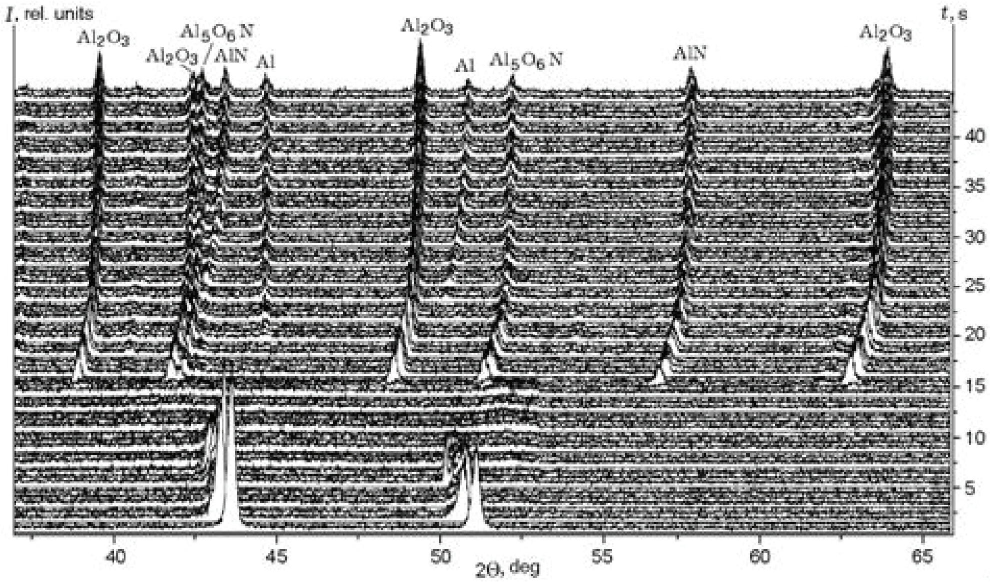

Fig. 2. Time scan of diffractograms of the burning compacted aluminum nanopowder (surface).

The experiments were performed in the Budker Institute of Nuclear Physics, Siberian Branch of the Russian Academy of Sciences, Novosibirsk, at the “Precision Diffractometry II” station (SR beamline No. 6 of the VEPP-3 electron storage ring) [Il'in et al. (2013) and Levichev E.B. (2016)] with OD-3M position sensitive detector [Aulchenko et al. (2009)].

3. Results and discussion

The combustion of the sample in the form of aluminum NP conical shape proceeds in two stages, if the mass of the sample exceeds the critical value. In the first step the temperature does not exceed 600 – 800° C: the process occurs in the form of thermal waves spread over the surface of the powder. It was established, at this stage occurs combustion of hydrogen which was accumulated in aluminum nanoparticles and warming the sample from surface into the bulk. The hydrogen content of the additives to passivate aluminum NP air reaches 1.5 wt. %. After ten seconds spontaneously in one or more dots bright glow appears covering the whole sample surface (second stage). At this stage, the combustion process is accelerated, the temperature raises up to 2200 - 2400° C and the binding of atmospheric nitrogen occurs to form aluminum nitride crystal phase.

In experiments on combustion of compacted aluminum NP *in situ* sample was placed on the diffractometer, and synchrotron radiation illuminated the sample surface. The XRD frames were recorded in the angle range of 35 – 65 degrees (fig. 2) every second after initiation of burning. In the first step reflections of aluminum (38.5 and 45.0 deg) were detected, which, when melting the aluminum, were not observed on XRD patterns.

According to the *in situ* XRD phase analysis of the burning sample obtained on synchrotron radiation (fig. 2), the formation of the crystalline phases goes through the following stages.

1. After initiation of the combustion and heating of the aluminum NP, the intensity of the diffraction maxima of metallic aluminum decreased: the diffraction pattern shows the first stage of the two-stage combustion process, which involved melting of aluminum inside the powder nanoparticles at a temperature 660° C.

2. During the reaction of nitride formation, which corresponds to the second stage of combustion, the XRD patterns did not show reflections of metallic aluminum and the temperature of the sample abruptly increased.

3. About 15 s after the initiation of combustion, the crystalline phases of aluminum oxide and oxynitride (Al_5O_6N) began to form.

4. The formation of the crystalline phase of aluminum nitride and metallic aluminum was observed after about 22 s from the start of combustion.

Formation of all crystalline phases during combustion occurs substantially simultaneously. After cooling, the sample is disaggregated in an agate mortar and examined by conventional X-ray diffraction analysis. It was found that the main crystalline phase in aluminum NP combustion product is aluminum nitride: its content was 80%. Electron microscopy revealed combustion product particles to have typical dimensions ranging from 0.3 to 1.5 mm.

4. Summary

1. In the products of complete combustion of pressed samples of aluminum nanopowder, the main phase is aluminum nitride, and the content of the remaining crystalline phases does not exceed 27%.

2. The composition of the final products of combustion of freely poured state aluminum nanopowder consists of 86 % aluminum nitride crystalline phase, and in the packed - average (surface and bulk) 80%. In both cases, no restriction in the flow of air, and stirring was conducted.

3. The formation of aluminum nitride probably occurs by successive replacement of oxygen by nitrogen from the aluminum oxide: NP $Al + O_2 + N_2 \rightarrow Al_5O_6N$ and $Al_5O_6N + N_2 \rightarrow AlN$.

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