Journal of Alloys and Compounds 689 (2016) 218–224

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Effect of carbon on the nitridation behavior of aluminum powder

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ARTICLE INFO

Article history:
Received 24 February 2016
Received in revised form
14 June 2016
Accepted 11 July 2016
Available online 14 July 2016

Keywords:
Nitridation
Aluminum
Powder
Carbon
EELS
TEM

ABSTRACT

The effect of carbon on Al powder nitridation was investigated from an atomic-scale chemical and crystallographic viewpoint. Carbon delays the initiation of nitridation and melting of Al powder, possibly because of its insulation effect. Therefore, carbon provides a pathway through which a constant supply of nitrogen gas is provided before molten Al powder is fully consolidated. Carbon also alters the nitridation mechanism by acting as a reaction agent of aluminum oxides and providing the path for nitrogen gas for constant nitridation, thereby stimulating the formation of the AlN with a typical triplet band structure. As a result, when carbon covers the entire surface of the powder, about 85% nitridation can be achieved.

1. Introduction

Aluminum nitride has high thermal conductivity (320 W/m K, ten times higher than that of Al_2O_3), high electrical insulation (9×10^{13} Ω cm), low thermal expansion coefficient (4×10^{-6}/°C, close to that of silicon), and excellent mechanical and chemical stability [1–12]. As such, aluminum nitride is widely used as a thermal barrier material for semiconductors and compound semiconductor substrates and as a reinforcing phase in composite materials. The application scope of this material has recently been broadened to heat-dissipating materials in light-emitting devices (LEDs) owing to its high thermal conductivity and low thermal expansion coefficient [1–12].

Two methods, the (i) carbothermal reaction and nitridation of alumina with a reaction agent (carbon) and (ii) direct nitridation of Al powder in a nitrogen atmosphere, have been extensively used for the commercial fabrication of aluminum nitride (AlN) powder [13–18]. The typical precursors of carbothermal reaction are aluminum oxide (Al_2O_3) and a source of carbon serving as a reaction agent. However, this process is limited by the surface areas of the precursors and requires an excess amount of carbon (~15%–30% additional carbon) to complete the nitridation. Hence, it is costly, energy intensive, and emits a significant amount of carbon-based byproducts (CO and CO_2) leading to a severe environmental impact. Preparing a homogeneous mixture of precursors is also essential in accomplishing the successful synthesis of AlN [15–18]. In comparison, the direct nitridation method uses cheaper starting materials and a lower manufacturing temperature as well as involves a simpler process than that associated with the carbothermal reaction. The direct nitridation reaction of aluminum powder proceeds in accordance with Equation (1) and is thermodynamically possible at both room and elevated temperatures [19–22].

$$Al\ (l) + 1/2N_2\ (g) = AlN\ (s) \quad (1)$$

This reaction is highly exothermic and generates a considerable amount of heat. As a result, unreacted aluminum (melted by this reaction heat) consolidates, which then impedes further nitridation by blocking the diffusion pathways that supply nitrogen gas. To prevent this blockage, commercial direct nitridation methods employ prolonged heating at elevated temperatures, of 1000 °C–2000 °C, leading to the complete nitridation of the consolidated aluminum. To increase the overall yield, the resulting aluminum nitride must also be subjected to repeated nitridation and

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http://dx.doi.org/10.1016/j.jallcom.2016.07.109

pulverization. Otherwise, further processing steps, such as the addition of aluminum tri-fluoride (AlF_3) or AlN, are required to facilitate the reaction completion [13]. These processing steps may, however, have negative side effects. For instance, during the pulverization process, the fraction of impurities (e.g., oxygen) may increase and have a negative impact on the thermal conductivity. These additional steps may also increase the overall cost of manufacturing.

Recently, the addition of AlN powder or other additives (e.g., carbon) have been reported to prevent the coalescence of unreacted aluminum, thereby greatly enhancing the degree of nitridation at lower temperatures during the process [3–6,23,24]. However, the effects of these additives on the nitridation behavior have yet to be clearly examined. As such, in the current study, we used relatively cheap lamp carbon (LC) as an additive to suppress the coalescence of unreacted aluminum. The new facile process proposed in this study facilitates a higher degree of nitridation at a relatively low temperature, which has yet to be achieved by the aforementioned processes, although it may generate reaction

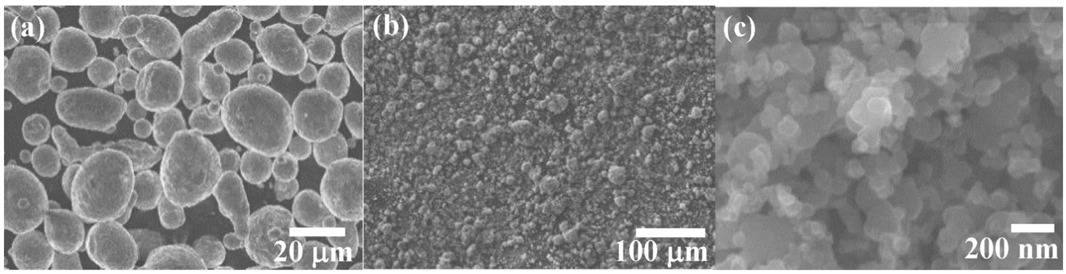

Fig. 1. SEM images of **(a)** aluminum and **(b)** Al and **(c)** LC powder at **(b)** low and **(c)** high magnification.

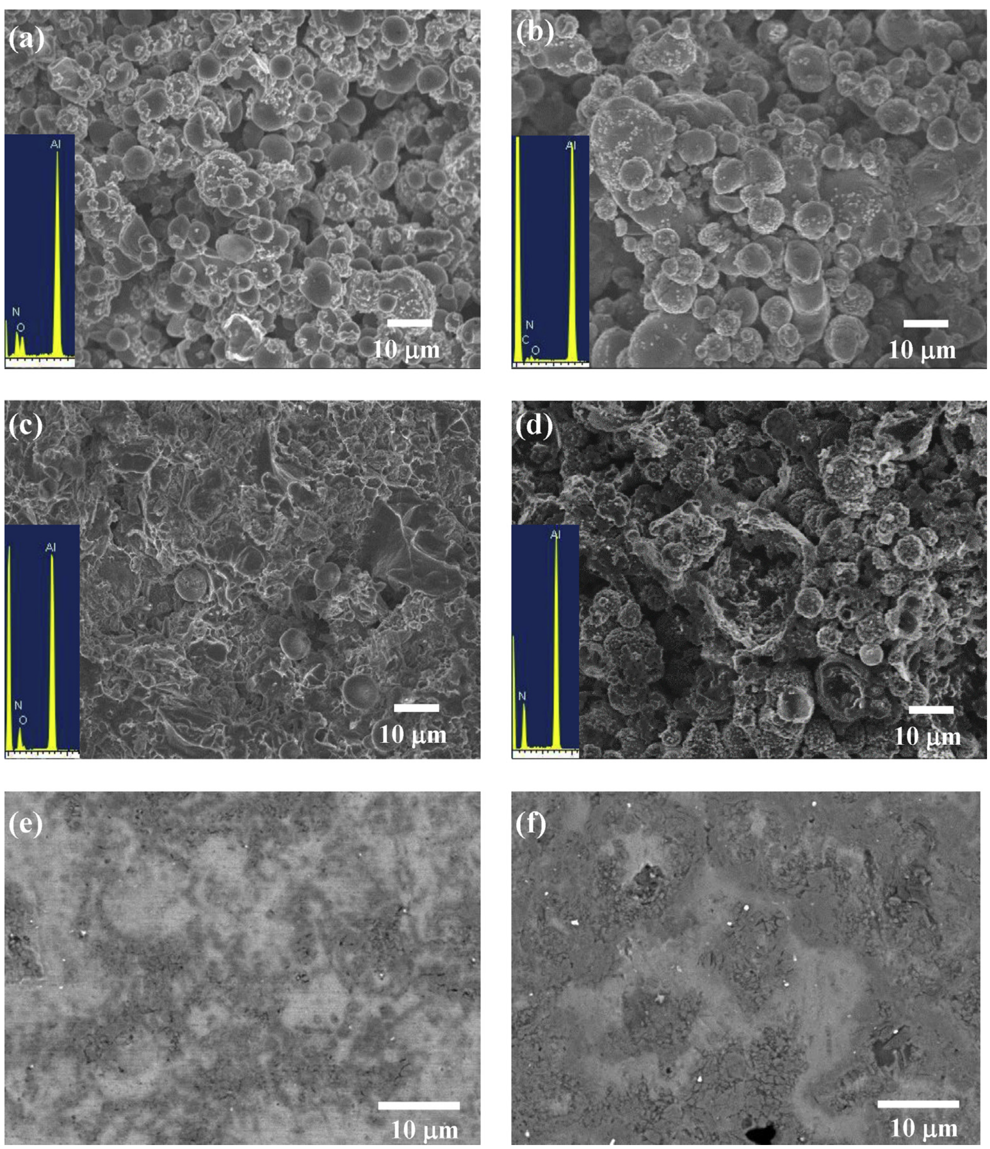

Fig. 2. SEM images of LC-free and Al-3 wt% powder beds after heat treatment. (**a**), (**c**), and (**e**) show LC-free powder beds after heating for 20, 30, and 60 min, respectively; **(b)**, **(d)**, and **(f)** show Al-3wt% LC powder beds after heating for 20, 30, and 60 min, respectively; insets show EDS spectra obtained from the corresponding area.