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Original Research Paper

The role of microstructure refinement in improving the thermal behavior of gas atomized Al-Eu alloy powder

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A B S T R A C T

Europium (Eu) was applied to the metal fuel for the first time in this study. A novel binary alloy powder consisting of Al-97 wt.% and Eu-3 wt.% was designed and prepared successfully by the close-coupled gas atomization. As a result of Eu addition and rapid solidification, a special micro-nano structure is found in the internal of the powder, which was formed by an interlaced distribution consisted of isolated Al phase and Al_4Eu phase. It was proved that this special microstructure promoted the oxidation of the Al-Eu alloy powder. The DTA results show that, the oxidation exothermic enthalpy of the Al-Eu alloy powder reached 8108.2 $\mu V \cdot s \cdot mg^{-1}$ at the temperature of 1065 °C, which is almost 5 times than that of the pure Al powder. The results of oxidation ratio calculation show that, the oxidation ratios of the Al-Eu alloy powder are both much higher than those of the pure Al powder. The present work indicates that Eu as a novel active additive can be applied to other Al-based alloy powders to improve their thermal behavior.

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1. Introduction

Al powder is one kind of very important metal fuel with high volume combustion enthalpy and large energy density, in the meanwhile, Al powder is abundant with low-cost and less pollution, therefore Al powder is widely used in solid propellant, explosives, pyrotechnics and other fields [1–3]. It was found that conventional Al powders have a few fatal flaws in practical applications, such as agglomeration, low burning rate and biphase flow loss. It is easy to form dense alumina protective film in the combustion process, causing ignition delay and high ignition temperature, preventing the further burning of elemental Al inside the particles [4–7]. Aiming at solving the problems existing in the practical application of conventional Al powders, intensive research efforts have been done on nano-Al powders [8–10]. Compared with conventional Al powders, nano-Al powders obtain many advantages, such as higher burning rate, greater specific impulse, lower ignition temperature and shorter ignition delay [11,12]. However, due to the extremely high activity of nano-Al powders, there are still many issues need to get settled in production, application and storage [13]. All of these issues cause the large-scale application of nano-Al powders to be very difficult. Therefore, the present work is focused on the study of Al-based alloy powders, additives such as Mg, Ti, Li and I are widely used in metal fuels, improving the thermal behavior of the pure Al powder [14–19].

There have been many patent reports on the applications and tests of rare earth (RE) metals or alloys in ammunition since 1960s [20–23]. RE metals have high chemical activity just after alkali metals and alkaline earth metals, RE metals also have low ignition temperatures, take Ce for example, it can be ignited when heated to the temperature of 165 °C. In addition, compared with other active metals, RE metals also have high combustion enthalpies. RE metals only oxidize slowly in the air, but they showed a good spontaneous combustion performance in practical application. Among the metals having spontaneous combustion property, the combustion enthalpies of RE metals are next only to Titanium (Ti) and Zirconium (Zr). But Ti and Zr are easy to form dense oxide protective films in the combustion process, and they are not conducive to the spread of combustion and easily lead to flame-out phenomenon, and the production safety of Zr is really poor, RE metals have no such shortcomings by comparison. Besides, Ti and Zr are expensive, they have no such rich resources as RE metals and the cost is about 3 times higher than RE metals [24,25].

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Therefore, as the active additive of Al-based alloy powders, RE metals have obvious advantages.

Since Europium (Eu) is the most active element in RE metals, as an active additive for Al-based alloy powders, it has the advantages of high activity and low ignition temperature, which could promote the combustion of Al-based alloy powders. In addition, Eu has variable oxidation states, which could adjust the degree of oxidation according to the content of oxygen in the environment by self, and also, its oxides have a catalytic effect in the combustion. The melting point of Eu is 822 °C, which is not much different from Al. This is conducive to the melting and the atomization of Al-based alloys. Moreover, Eu and Al can form various intermetallic compounds, such as Al_4Eu, Al_2Eu and AlEu, which could provide a broad window for the design of Al-based alloys [26,27]. In this study, Eu was used in the metal fuel for the first time, and a novel binary alloy powder consisting of Al-97 wt.% and Eu-3 wt.% was designed and prepared successfully by the close-coupled gas atomization (CCGA). Then the microstructure and the thermal behavior of the Al-Eu alloy powder were investigated through a series of experiments.

2. Experimental details

Due to the powder obtained by which is fine low-oxygen spherical and has rapid solidification microstructure, the CCGA, with its large-scale production possibility and low-cost, is one of the main methods for high-quality spherical metal and alloy powders production [28–30]. Composed of Al-97 wt.% and Eu-3 wt.%, a novel binary alloy powder was prepared by the CCGA equipment made by Phoenix Scientific Industries Ltd (PSI). The process of the CCGA can be divided into three stages: alloy melting, alloy disintegration and powder collection. And the alloy disintegration may also be divided into three stages: primary disintegration, secondary disintegration and solidification. The process parameters of the CCGA: the melting temperature is about 960 °C, the vacuum degree is below 10^{-2} Pa, the atomization gas is Ar and the atomization pressure is 3.5 MPa. The Al-Eu alloy powder was sieved with a 325 mesh sieve, and the powder with the size less than 46 μm was acquired. In the next experiments, the powder which has been screened out was used. Five different techniques were used to analyze the microstructure and the thermal behavior of the Al-Eu alloy powder. These are scanning electron microscopy (SEM), X-ray diffraction (XRD), energy dispersive X-ray spectrometer (EDS), thermogravimetric and differential thermal analysis (TG-DTA).

The morphology of the powder was examined by SEM (Nova NanoSEM 450). The phase composition of the powder was investigated by XRD (PANalytical B.V.) using Cu Kα radiation, and the patterns were collected between 10° and 90°. Further investigations are required to study the effect of Eu addition in the structure of the powder. Then the powder was embedded in epoxy resin, after mechanical grinding and polishing, the sample was obtained for the particle cross-section analysis. The morphology and distribution of elements of the particle cross-section were examined by SEM (Nova NanoSEM 450) coupled with an EDS.

In order to study the thermal behavior of the Al-Eu alloy powder in oxygen, TG-DTA (PerkinElmer Instruments Diamond) examination was carried out. As a contrast, the thermal behavior of the pure Al powder was also examined by TG-DTA. The pure Al powder was prepared by the CCGA and sieved with a 325 mesh sieve as well. The conditions of the TG-DTA examination: from room temperature to 1300 °C in oxygen atmosphere with gas flowing at 20 ml·min^{-1}, in which heating rate was 20 °C·min^{-1}, sample mass was about 1.0 mg each.

Titration experiments were carried out to calculate the oxidation ratio of the oxidized products at 1200 °C and compared with the results from TG analysis. The steps of titration experiment: configured 30 ml $Fe_2(SO_4)_3$ solution with consistency of 1 mol/L, added appropriate amount (about 0.15 g) of product, 50 ml H_2SO_4 with consistency of 10% and 70 ml saturated $NaHCO_3$. Closed the flask and left only one outlet connected with saturated $NaHCO_3$, then heated the flask until the end of the reaction. After the flask cooled, added 18 ml mixed acid ($V_{H_2SO_4} : V_{H_3PO_4} = 1:4$) and 35 ml water. At last, added four drops of $C_{12}H_{10}NO_3S$ indicator and titrated immediately with $K_2Cr_2O_7$ solution with consistency of 0.05 mol/L until the solution turned into purple from green. The elemental Al content can be calculated based on the weight of product and the consumption of $K_2Cr_2O_7$ solution.

The oxidation processes of the Al-Eu alloy powder and the pure Al powder were investigated through a series of experiments. The constituent developments of the Al-Eu alloy powder and the pure Al powder samples before and after the exothermic peak were examined by XRD (PANalytical B.V.) using Cu Kα radiation, and the patterns were collected between 10° and 90°. In order to compare the oxidation processes of the Al-Eu alloy powder and the pure Al powder, products at different stages of the oxidation process according to the TG-DTA results were sampled and analyzed. The powders before the exothermic peak were embedded in epoxy resin, after mechanical grinding and polishing, the samples were obtained for the particle cross-sections analysis. And the powders after the exothermic peak were acquired for the morphology analysis. SEM (Nova NanoSEM 450) examinations were carried out in these experiments.

3. Results and discussion

3.1. Microstructure analysis

A novel binary alloy powder with the composition Al-Eu (3 wt.%) was prepared by the CCGA. Fig. 1 shows the morphology of the Al-Eu alloy powder. It can be seen that all particles are fine and spherical in shape and the degree of agglomeration of the powder is also very low, as shown in Fig. 1(a). Apparent grain boundaries can be observed on the surface of the particles, and the surface of the particles is also very smooth, as shown in Fig. 1(b). All of these evidences indicate that high-quality alloy powder was prepared successfully by the CCGA.

In order to identify the phase constitution of the Al-Eu alloy powder, XRD examination was carried out. Fig. 2 shows the XRD pattern of the Al-Eu alloy powder. The results show that, only Al and Al_4Eu are identified in the Al-Eu alloy powder, which is consistent with the Al-Eu phase diagram [31]. The characteristic peaks of Eu are hard to find, suggesting that Eu has been totally alloyed with Al to form the intermetallic compound Al_4Eu, as shown in Fig. 2. It is worth mentioning that no any oxides is observed, indicating that the powder is low-oxygen combining with the results of the SEM examination (Fig. 1). Further investigations are required to study the effect of Eu addition in the microstructure of the Al-Eu alloy powder.

The SEM image and the EDS elemental spectrums of the particle cross-section of the powder sample are presented in Fig. 3. It can be clearly observed that there are many submicron-sized or even nano-sized grains and the corresponding grain boundaries in the powder in back scattering mode, indicating that the grains are refined due to Eu addition and rapid solidification [32]. The EDS results show that, the content of Eu distributed in the grain boundary (①) is as high as 20.07 wt.%, while no Eu is detected in the grain (②). According to the results of the EDS and the XRD (Fig. 2), it could infer that, during the process of the CCGA, Eu, after segregating at the grain boundaries, produced an intermediate compound Al_4Eu together with Al. The particle is micro-sized, but the refined

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