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ScienceDirect

Energy Procedia 66 (2015) 109 – 112

Energy Procedia

The 12th International Conference on Combustion & Energy Utilisation – 12ICCEU

Ignition and combustion characterization of nano-Al-AP and nano-Al-CuO-AP micro-sized composites produced by electrospray technique

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Abstract

Metal powders such as aluminum nanoparticles (Al NPs) have been found huge potential as reactive additives to highly increase energy density in various energetic systems such as propellants, explosives and pyrotechnics. However, it is suffering issues of agglomeration and post-combustion aggregates, which largely reduce the energy utilization efficiency and the energy release rate. One option to eliminate this disadvantage is to coat the nanoparticles with gas generator which can produce gas to prevent the sintering. This work use electrospray technique to assemble Al NPs and Al-CuO NPs into microparticles, with coating of gas generator-ammonium perchlorate (AP) to produce gas to prevent possible sintering, thus obtaining a highly reactive Al-based composites. The Al/CuO NPs composites are ignited in a confined cell to measure its combustion pressure history. The peak pressure and the pressurization rate of Al/CuO/AP is more than 3X higher and faster, compared to the physically mixed Al/CuO nanothermite. This work provides an ideal approach to prepare Al NPs based energetic materials such as solid propellant or solid fuel air explosives.

Peer-review under responsibility of the Engineering Department, Lancaster University

Keywords: aluminum, nanothermite, electrospray, ammonium perchlorate, microparticles

1. Introduction

Metal powders such as aluminum powder have been commonly used in propellants, explosives and pyrotechnics owing to high energy density. [1, 2] However, conventional micro-sized aluminum powders are suffering long ignition delay and post-combustion agglomeration issues. Recent years, nano-sized aluminum powders were employed as additives instead of micron ones, showing much shorter ignition delay time and faster energy release rate. [3] Whereas, the aluminum nanoparticles (Al NPs) have some severe disadvantages which might impede the further development of Al NPs-based energetic materials.

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Peer-review under responsibility of the Engineering Department, Lancaster University

doi:10.1016/j.egypro.2015.02.063

Owing to high viscosity of Al NPs, the mixing process of the Fuel (such as aluminum), the Oxidizer (such as ammonium perchlorate) and the polymers (such as HTPB) are difficult. It limits the mass loading of Al NPs in the energetic system thus lower the potential maximum energy density. [4] It will be one ideal option if the assembly of Fuel (Al NPs) and Oxidizer (AP, CuO, etc.) can be completed in nano scale within one step. In the previous study, the Al NPs, Al-CuO nanothermites were assembled into microspheres using an electrospray technique, showing much higher reactivity than physically mixed cases. [5, 6] In the present paper, we incorporate the commonly used oxidizer-ammonium perchlorate and commonly used fuel-Al NPs (also Al-CuO nanothermite) into micron particles, achieving close assembly to obtain desirable energetic systems.

2. Experimental Section

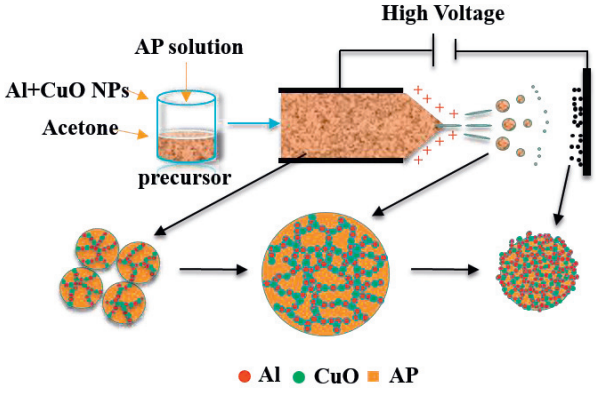
Materials:

Al NPs (~50 nm) and CuO NPs (~40 nm) were purchased from Aladdin-Reagent, were used as receives. Nitrocellulose (NC) comes from the collodion (4%-8% NC in ether and ethanol), which was purchased from Sigma-Aldrich. The needle used in this paper was in diameter of 0.4 mm. The syringe pump (TYD01-02) was purchased from Leifu Company in Heibei, China. The high voltage supply (0-50 kv) was purchased from Dongwen High voltage suppler in Tianjin, China.

Precursor Preparation:

Al/AP: In a typical experiment, 50.0 mg AP was dissolved in 0.5 ml deionized water. At the same time, 50.0 mg Al NPs was dispersed in nitrocellulose (NC) solution (10 mg NC in 1 ml acetone), then ultrasonicate it for 60 min. Add the AP solution drop by drop into the above Al NPs suspension while vigorous stirring. At last, stir the final suspension for 24 hr.

Al/CuO/AP: In a typical experiment, 52 mg AP was dissolved in the mixture of 1.2 ml deionized water and acetone. 261 mg CuO NPs was dispersed in 3 ml ethanol, and ultrasonicate it for 60 min. Then add 87 mg Al NPs into the suspension and ultrasonicate it for another 60 min. Add the AP solution drop by drop into the above Al/CuO suspension while vigorous stirring. At last, stir the final suspension for 24 hr.



Electrospray Process:

Take Al/CuO/AP micro-sized composites for example. At first, the precursor was loaded in a syringe with a metal needle. The syringe was pumped by a syringe pump with a speed of 2.5 ml/h. There is a receiving board (aluminum foil, 24 cm*24 cm) at 10 cm away from the metal needle tip. A voltage of 19 kv was added between the board and the needle.

Figure 1. Schematic showing of electrospray process of Al/CuO/AP micro-sized particle

Pressure, Pressurization Rate and Burn Time Measurement:

25.0 mg Al/CuO/AP composites was placed in a confined cell with a constant volume of 13 ml. A heated Ni coil was employed as igniter on the top of the composites. A pressure sensor and an optical sensor were installed in the wall of the cell, which will obtain the pressure history and optical history simultaneously once the sample was ignited. The pressurization rate was caculated by measuring the slope of pressure-time history and the burning time was obtained by measuring the half peak width of the optical emission history.

3. Results and Discussion

3.1 Al-AP composites

The mixing of Al NPs with oxidizer and polymers is always a key issue when prepare solid propellant. Better mixing means much shorter mass diffusion distance and larger interfacial area between the fuel and oxidizer, thus achieving higher reactivity. [5] In this paper, we dissolved the oxidizer-AP in the precursor beforehand, achieving close contact between the fuel and oxidizer in nanoscale by coating the AP on the surface of Al NPs in liquid phase. As Figure 2c and 2d shows, the Al and AP were mixed well in the composites. As Figure 2a-2d shows, by employing different content (10%, 30%) of polymer-nitrocellulose (NC) in the composites, the porosity of the particles can be adjusted. As our previous study confirmed, once ignited, the gas generator-NC as well as the oxidizer, AP, another good gas generator, will release gas to prevent the possible sintering among Al NPs. Moreover, NC is expected to form a protective layer on AP crystals to prevent any absorption of moisture.

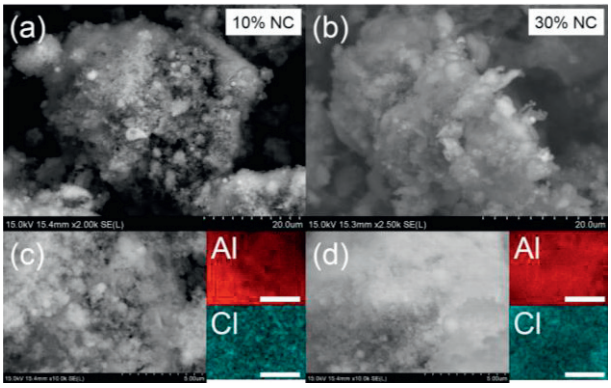


Figure 2. Scanning Electron Microscope (SEM) images (a, b) and Energy Dispersive X-ray Detector (EDX) results (c, d) of Al/AP composites with 10 wt. % NC (a, c) and 30 wt. % NC (b, d). Note: the scale bars in Fig. 2c and Fig. 2d is 5 μm

3.2 Al-CuO-AP composites

As the previous study shows, a proper amount (5 wt. % of whole mass) of gas generator-nitrocellulose in Al-NPs based thermite (Al/CuO, Al/Fe_2O_3, Al/Bi_2O_3) will produce gas to prevent possible sintering among Al NPs, thus obtaining much higher reactivity than physically mixed thermites. [6] In this study, we incorporate another gas generator-AP, into Al/CuO nanothermite. As Figure 3 shows, Al/CuO/AP composites were successfully produced by electrospray without using any binder. As Figure 3a shows, the produced microparticles are several micron with narrow size distribution. As Figure 3b-3e show, with the increase of AP content (from 1% to 13%) in Al/CuO, the mixing condition of Al/CuO/AP is becoming better. However, with further increase of AP to 15%, AP formed micro-sized crystals which were separated from Al/CuO particles.

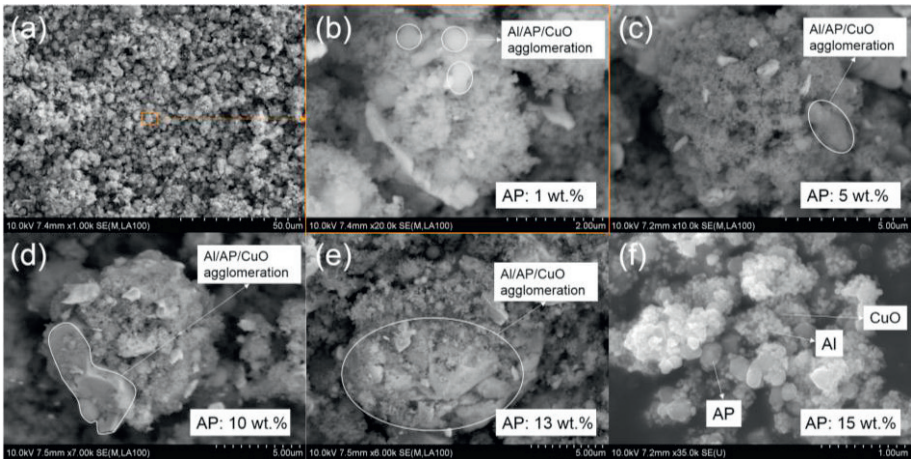
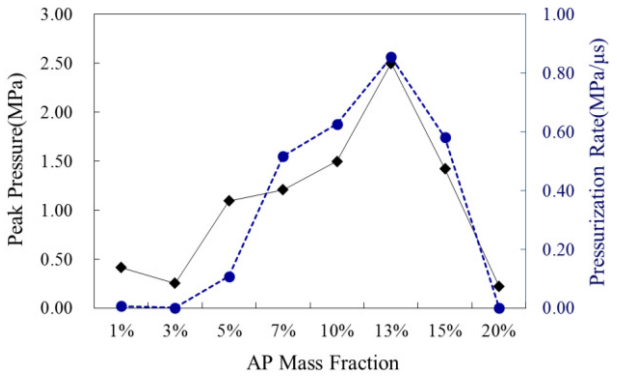


Figure 3. The low (a) and high (b) magnification SEM images of Al/AP/CuO micro-sized particles (1 wt. % AP content). The high magnification SEM images of Al/AP/CuO micro-sized particles with 5 wt. % AP (c), 10 wt. % AP (d), 13 wt. % AP (e) and 15 wt. % AP (f).

3.3 Ignition of Al/CuO/AP composites in confined cell



The produced Al/CuO/AP composites (25.0 mg) were ignited in a confined cell (13 ml). The pressure history and optical emission history were captured. Therefore, the peak pressure, pressurization rate and burning time can be obtained. As Figure 4 show, the composites with 13 wt. % AP has the highest peak pressure of ~2.5 MPa and also the highest pressurization rate of ~0.85 MPa/μs, which is almost 3X higher than the conventional Al/CuO nanothermites. On the contrast, the burning time of composites with 13 wt. % AP is 134 ms, which the shortest one is.

Figure 4. The peak pressure and pressurization rate changing with the AP content in Al/AP micro-sized particles. Note: all the sample were 25.0 mg.

Table 1. The burning time changing with the AP content in Al/AP micro-sized particles. Note: all the sample were 25.0 mg.

AP content (%)	1	3	5	7	10	13	15
Burn Time (ms)	198	220	142	141	136	134	137

4. Conclusion

Micro-sized Al/AP and Al/CuO/AP composites were formed by electrospray tehnique. Al NPs and AP were well mixed and the porosity of Al/AP composites can be adjusted by tuning the NC content. The Al/CuO/AP were successfully produced without using binder. Al/CuO/AP composites with 13 % AP have the closest assmbly condition thus have the best combustion performance, whose peak pressure and pressization rate is 3X higher than the conventional Al/CuO nanothermite.

Acknowledge

This work is supported by the National Science Foundation for Young Scientists of China (Grant No. 11102091) and Doctoral Scientific Fund Project of the Ministry of Education of China (Grant No. 20113219110010). Haiyang Wang is grateful for the finacial supporting from the Reseach Innovation Scheme for Graduate Student of Jiangsu Province, China (Grant No. CXZZ13_0188) and the China Scholarship Council.

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