

Additive Exploration of Al/CuO Aluminium Thermite Systems

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Abstract: Aluminium thermite research has made significant progress in recent years, especially in the Al/CuO system to show its scientific value and application potential. Starting from the history of aluminothermite development, types of aluminothermite production methods, and exploration based on Al/CuO system. The development history of aluminothermics is briefly described. Advantages and disadvantages of various aluminothermic agent preparation methods, existing new methods. Overview of titanium-containing polymers, silicone-containing polymers, fluorine-containing polymers, and other additives based on the Al/CuO system to enhance the performance of aluminothermic agents.

Keywords: Aluminium thermite, Al/CuO system, additives, energy release, combustion performance, safety.

1. Introduction

The historical evolution and technological innovation of aluminothermics constitute a complex and fascinating subject area whose roots can be traced back to the end of the 19th century, with the pioneering discovery of the principle of aluminothermics by the German chemist Hans Goldsmith, a process that successfully extracted high-purity metals without the need for a carbon medium by using the chemical interaction between aluminium and specific metal oxides, laying the cornerstone of the advances in metallurgy and pyrotechnics. In 1895, Goldsmith improved and patented this technique, establishing the famous 'Goldsmith Method' [1]. Subsequently, in 1899, the first industrial application of aluminium thermal welding technology in Essen, Germany, marking the aluminium thermite technology in the field of practical engineering took a substantial step forward [2].

Into the 20th century, especially during the two world wars, aluminium thermite gained widespread attention in military applications due to its unique high-temperature reaction properties and rapid energy release capability, becoming a key component in the manufacture of incendiary bombs, armour-breaking bombs and rocket propellants [3-6]. In the Cold War era, the application of aluminothermic agents was further expanded to the aerospace field, becoming one of the important fuels for solid rocket motors [7], while at the same time, the focus of the research shifted to formulation optimisation, reaction control and safety enhancement, opening up new horizons for the application of aluminothermic agents.

In the 21st century, with the rapid development of nanotechnology, the research on aluminothermic agents has entered a new era. Scholars are committed to exploring the potential of aluminothermic agents at the nanoscale, aiming to enable the materials to exhibit superior performance in micro-propulsion, precision detonation technologies, and catalytic applications by increasing energy density, accelerating reaction rates, and reducing sensitivity [8-10].

The history of aluminothermics is also the history of their preparation techniques, and the evolution of the technology covers from basic physical mixing to highly sophisticated

nanosynthetic methods. Physical mixing methods [11-14], despite their ease of operation, are limited in their application by uneven mixing and inefficient reactions. In contrast, co-precipitation [15-18] provides a more homogeneous product by simultaneous precipitation of metal and aluminium ions in solution via a chemical pathway, but requires tight control of reaction conditions to ensure product stability. Partially reduced iron oxide nanoparticles [19-20], on the other hand, use chemical reduction to modulate the oxide morphology and improve the efficiency of the reaction, although high-temperature treatment may affect the stability of aluminium nanoparticles. The solvothermal method [21-23] prepared core-shell structured oxides, especially through the ethylene glycol/water system, enables fine regulation of the reaction conditions and provides a new path for the structural design of aluminium nanothermite. The electrostatic spinning technique [24-25] and sol-gel method [26-30], on the other hand, facilitated the homogeneous dispersion of the materials and optimisation of their properties through their respective unique physical and chemical mechanisms. The template method of preparation further promotes the structural design of energy-containing materials by virtue of its precise regulation of porosity and morphology. In particular, the supersonic airflow collision method, innovatively proposed by the Institute of High Speed Aerodynamics of the China Aerodynamic Research and Development Centre (CARD), cleverly exploits aerodynamic principles to induce reactant particles to undergo high-intensity frictional collisions in supersonic environments, transforming kinetic energy into molecular internal energy, and realising highly activated and rapid low-heat solid-phase synthesis of the reactants [31]. The method aims to enhance the yield and explore micron-sized aluminotherms as a viable alternative to nanoscale materials, showing potential to become a mainstream preparation technique in the future.

Compared with other aluminothermic agents, the Al/CuO aluminothermic system exhibits remarkable properties in terms of energy release, safety control, and reaction efficiency [32-34], which are mainly attributed to the following: firstly, the system has a high energy density, and shows excellent combustion performance through the combination of

aluminium powders and copper oxide (CuO) in different ratios [35-36]; secondly, the system design is highly controllable, which greatly enhances the combustion performance [35-36]. Secondly, the system design is highly controllable, which greatly enhances the practical potential of the material [37]; furthermore, the interfacial effect at the nanoscale significantly enhances the reactivity of aluminium and CuO, accelerating the energy release process, which is attributed to the close contact between the two and the enhancement of the kinetics [38]; furthermore, the material skeleton can be designed through the preparation technique, which can also enhance the conductive and thermally conductive properties of the material, and facilitate the aluminium-thermal reaction process [39-40]. Finally, the aluminothermic agent exhibits an ultra-low electrostatic ignition threshold (only 10.11 mJ) [41], which is much lower than the standard of conventional materials, and greatly enhances the safety in the use scenario, which provides a favourable condition for the risk control in the practical application.

2. Research Exploration Based on Al/CuO System

2.1. Effect of titanium-containing polymer additives

Titanium is a chemically active metal, and the basic

research and performance optimisation of aluminothermic agents have been the key to drive their technological progress. The instability and reaction efficiency problems of traditional aluminothermic agents under some specific conditions have prompted scientists to search for new methods to improve their performance [42]. Jian Cheng et al [43] effectively solved the problem of accidental activation of nano-aluminium thermite by introducing Ti3C2 MXene to the Al/CuO system and innovatively using the microwave radiation ignition method. It was shown that Ti3C2 MXene was not only sensitive to microwave radiation, but also could significantly regulate the heat release characteristics of Al/CuO/MXene composites, especially reaching the maximum heat release value of 2138.5 J-g⁻¹ at 7.5 wt% content, and at the same time, optimising the combustion performance and improving the system safety and Adaptability. By introducing Ti3C2 MXene into the Al/CuO system, the team of scientists not only improved the microwave responsiveness and achieved a safer ignition method, but also significantly enhanced the heat release efficiency and reaction control (e.g., Fig. 1). This finding provides an innovative strategy to solve the problem of accidental activation of nano-alumina thermite, and also reveals the important role of additives in regulating the reaction properties of alumina thermite. Innovation in structural design is another important way to enhance the performance of aluminothermic agents.

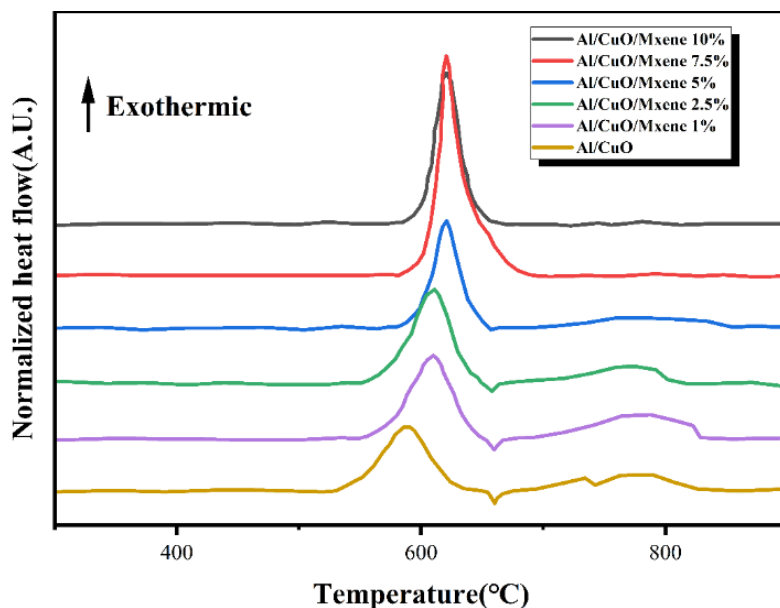


Figure 1. Effect of different Ti3C2 MXene contents on exothermic reactions [43].

Yongjin Chen [44] and his team successfully synthesised Al/Ti/CuO micro- and nano-composites with a unique 'lychee-like' core-shell structure, which originated from the uniform distribution of fine Ti and CuO nano-particles embedded in the surface or sub-surface of the larger Al particles under the action of high-intensity mechanical force. The rough surface with granular protrusions resembles a lychee, hence the name 'lychee-like' structure (see Fig. 2). X-ray diffraction (XRD) analyses confirmed that no new substances were generated during the ball milling process, and the purity of the original components was maintained. Differential scanning calorimetry (DSC) results showed that the introduction of Ti led to an increase in the onset reaction temperature of the Al/Ti/CuO composite aluminothermite and transformed the original double exothermic peak into a three-

stage exothermic process, with the first stage of exotherm occurring prior to the melting of Al at a peak temperature of 595.3°C, followed by a heat absorption effect triggered by the melting of Al, and the final stage of liquid-solid reaction between the melted Al and the Ti with nano. The final stage is the liquid-solid reaction between molten Al and Ti with nano-CuO, with a heat release peak at 916.5 °C. The 'lyotropic' Al/Ti/CuO composites prepared by high-energy ball milling not only exhibit a unique core-shell structure, but also optimise the heat release process through fine structural modulation, which improves the safety and adaptability of the materials. Such studies highlight the possibility of improving reaction kinetics and energy release efficiency through material design.

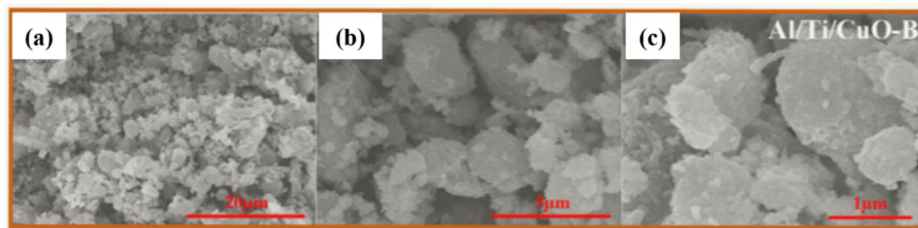


Figure 2. SEM images of Al/Ti/CuO-B with different magnifications [44].

Emilian Tichtchenko et al [45] revealed the particle size dependence of the combustion properties of Al/CuO aluminium thermite through theoretical modelling studies, highlighting the importance of the nanoscale for improving the combustion rate. While Vidushi Singh et al [46] revealed the effect of metal layer stacking order on the reactivity through the construction of Al-TiB₂/CuO multilayer structure, where the introduction of TiB₂ significantly enhanced the combustion rate and energy efficiency, while pointing out the possibility of optimising the metal layer arrangement to improve the reactivity. The study not only improved the combustion rate, but also revealed the significant effect of metal layer arrangement on the reactivity efficiency, which provided a theoretical basis for the design of efficient and safe aluminium thermite formulations. While the study by Y K Chang et al [47] revealed the modulating effect of Ti nanoparticles on the ignition and combustion behaviour of Al/CuO aluminothermics, suggesting that an optimal balance of reactivity and combustion efficiency can be achieved with a Ti content of 30 mol%.

2.2. Effect of silicone-containing polymer additives

The work of Lixiao Shen and her team [48] was devoted to

improving the energy efficiency stability of aluminothermic agents for the Al/CuO system, and aluminium-silicon alloy aluminothermic agents were prepared by adding silicon to aluminothermic agents by gas atomisation. It is shown that the introduction of silicon not only significantly enhances the combustion stability of the Al-Si/CuO system, but also improves the exothermic capacity by 110.2% and 93.1%, respectively, and accelerates the ignition rate by 569.06 ms and 540.48 ms, respectively, compared with the pure aluminium thermite through the optimization of the silicon content (20% versus 12%). Experimental observations reveal that the silicon The experimental observation reveals that the addition of silicon leads to the nonlinear change of the combustion process, which affects the morphology of the solid particles and the generation of the product phases, as well as the temperature of the adiabatic flame, and makes the combustion mechanism more complicated. In addition, the activation energy of the exothermic phases was analysed, and it was noted that the simultaneous existence of multiple reaction paths, including solid-solid and liquid-solid reactions, increased the complexity of the activation energy calculation (see Figure 3).

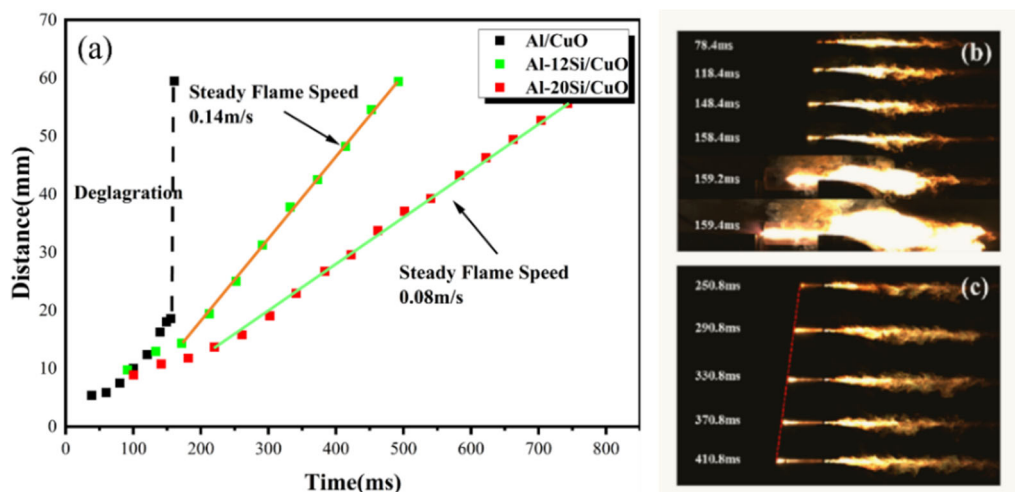


Figure 3. Flame velocity and combustion photography for different thermite: a) Flame front position obtained by fitting with time and flame velocity b) Non-stationary flame propagation of Al/CuO c) Representative single replicates of stabilised flame propagation of Al- 20si /CuO [48].

Zhou Jianzhong et al [49] investigated the effect of Si-Bi₂O₃ mixtures on the aluminothermic agent of Al/CuO system. It was found that when the mass ratio of the components in the aluminothermite was Al/CuO/Si/ Bi₂O₃ = 14.7/65.3/4/16 (wt %), it was able to significantly reduce the excitation temperature (ignition threshold) of the aluminothermite to only 613 °C. This suggests that the incorporation of Si and Bi₂O₃ at specific ratios can most effectively catalyse the

aluminothermic reaction and reduce the temperature required for ignition, while maintaining the high exothermic capacity of the aluminotherm. A study by Zou Xuan and colleagues [50] revealed that PTFE/Al/Si/CuO composites, compared to PTFE/Al/CuO, exhibit superior energy release capacity in terms of energy release capability. performance. This study further delved into the effect of different mass ratios of PTFE/Si on the energy release characteristics. Specifically,

the PTFE/Al/Si/CuO composites prepared at a PTFE/Si mass ratio of 20% exhibited the lowest ignition onset height of 48.27 cm. It was found that the PTFE/Al/Si/CuO composites reached the highest flame temperature of 2315.85 °C, also at a PTFE/Si ratio of 20%. This finding not only emphasises the easy ignition properties of this particular ratio material, but also highlights the extremely high thermal effects it is capable of generating during combustion, suggesting a more significant thermal damage potential.

2.3. Effects of fluoropolymer additives

In a study by Zhang Yue et al ^[51], an innovative energy-containing material combination was proposed, i.e., the addition of Al/PTFE to Al/CuO to form a mixed-ratio energy-containing material. This new material combination has a lower reaction excitation rate compared to single Al/CuO, which reduces the impact reaction threshold of aluminothermic agents. The study reveals that the addition of Al/PTFE can effectively reduce the ignition threshold of the whole energy-containing material system and simultaneously enhance the energy release efficiency of Al/CuO. Through the optimised ratio of the materials, the ignition capability of the diesel fuel tank can be significantly improved, which solves the problem that the traditional single kinetic energy intruder is difficult to provide sufficient ignition energy. The work of Wenchao Zhang et al ^[52] focuses on optimising the performance of the Al/MOX/PVDF system, and through the addition of different levels of graphene oxide, it was found that the Al/CuO/PVDF nanocomposites exhibited energy release characteristics beyond those of conventional firearms, reaching a high energy level of 3829 J-g⁻¹, highlighting the superiority of the Al/CuO combination in terms of energy performance. Hu Xiang et al ^[53] by electrophoretic deposition technique. Successfully prepared PVDF/Al/CuO organic/inorganic hybrid energy-containing thin films, through the systematic study of its components, structure, morphology and energy-containing properties found that when the addition of PVDF is 2 wt%, the combustion performance of the prepared PVDF/Al/CuO composite film is significantly improved, and the heat release energy reaches 3924 J/g, which is much higher than the heat release energy of the separate Al/CuO system. CuO system's heat release energy. This study improves the heat release efficiency of the energy-containing films and reveals the mechanism behind the improved performance: the oxidative properties of PVDF enable it to react with aluminium and release heat; meanwhile, the fluorine generated from the thermal decomposition of PVDF can erode away the

passivation film (Al₂O₃) on the surface of aluminium, which turns aluminium into a reactive state, thus greatly enhancing the exothermic effect of the Al/CuO system.

On the other hand, the work of Kunyu Xiong et al ^[54] explored the effect of fluoropolymers such as polytetrafluoroethylene PTFE and polyvinylidene difluoride PVDF on the performance of aluminium thermite for Al/MxOy system. They prepared a series of Al/MxOy blends containing different fluoropolymer concentrations and evaluated their thermal properties of reaction by bomb calorimetry. It was shown that the peak temperature of the reaction between Al and CuO could be significantly reduced to about 480 °C under specific conditions, e.g., at a PVDF content of 37 wt%, which is about 250 °C lower than the original temperature, revealing the significant effect of high content of PVDF in reducing the initial exothermic temperature. Further optimisation revealed that the optimum exothermic performance of the system was achieved at a fluorine content of ~6 wt%, which could reach ~2481 J/g. Their study on the introduction of fluoropolymers further demonstrated the potential of additives in modulating the reaction properties of aluminothermic agents. By optimising the content of PVDF, the onset temperature of the reaction was significantly reduced and the level of energy release was enhanced, demonstrating the effectiveness of fluoropolymers as a modulating factor and opening up a new direction for the optimisation of the properties of aluminothermic agents.

In addition, Yanjun Yin et al ^[55] applied electrophoretic deposition technique to prepare PTFE/Al/CuO film, which significantly enhanced the combustion efficiency, the introduction of PTFE not only increased the local hot spots, but also accelerated the flame propagation speed, and the energy release of the composite film increased significantly with the increase of the PTFE content up to 8784 J/g, which was attributed to the decomposition of the PTFE-generated gases interacting with the aluminium nanoparticles to promote the aluminothermal reaction at low temperatures (e.g., Fig. 4). The electrophoretic deposition method provided a more homogeneous structure and better reactivity compared to the physical mixing method. The study by Yanjun Yin's team not only enhanced the combustion efficiency, but also significantly enhanced the reactivity and mechanical properties of the material through the introduction of PTFE, demonstrating the positive impact of material processing technology on the performance of aluminothermic agents. Safety and environmental adaptability are elements that cannot be ignored in aluminium thermite research.

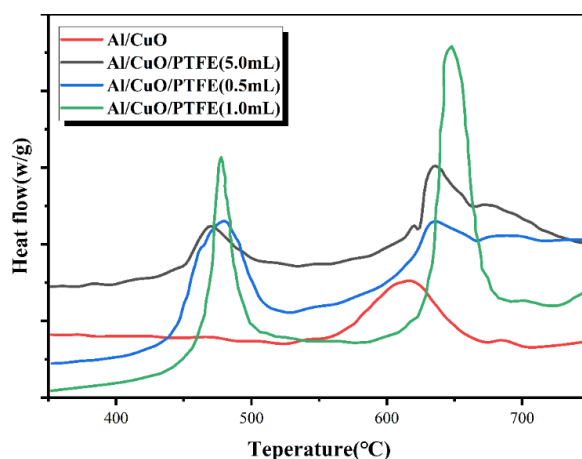


Figure 4. Effect of different PTFE contents on the exothermic reaction ^[55].

2.4. Impact of other factors

In terms of novel structural design, Keer Ouyang et al [56] investigated an Al/AIH₃/CuO hybrid system by using ultrasonic dispersion and magnetic stirring techniques in order to enhance the microthrust and combustion efficiency of aluminothermic agents. In the experiments, the DSC curves demonstrated three main peaks containing two exothermic and one absorptive process of Al melting with the change of AIH₃ addition. The introduction of AIH₃ induced the first aluminothermic reaction to advance, especially in the AIH₃-80 sample, where the peak temperature was advanced by 16.7°C. Meanwhile, the heat uptake of the first pyrolysis

reaction decreased with the increase of the percentage of AIH₃ substitution, while the second pyrolysis reaction heat uptake increased significantly (e.g., Fig. 5). The study indicated that the optimal replacement rate of AIH₃ is 20%, beyond which the combustion rate will be slowed down or even unable to maintain stable combustion, suggesting that the moderate addition of AIH₃ can optimise the onset of pyrolysis reaction and the distribution of exothermic heat in the system. Through the study of Al/AIH₃/CuO system, the precise control of the reaction onset timing and exothermic amount was achieved by changing the composition of the aluminothermic agent, revealing the flexibility and strategy of the additive in regulating the exothermic process.

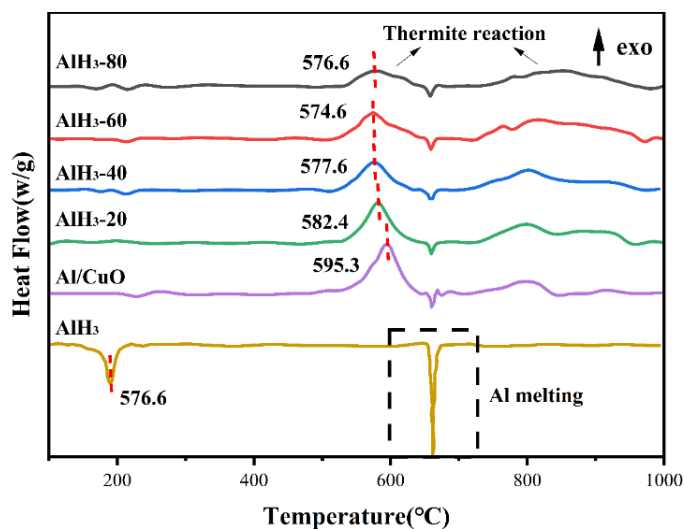


Figure 5. Effect of different AIH₃ additions on the exothermic reactions [56].

Yao Shu et al [57] evaluated the oxidation reactivity, ignition delay, burning rate, agglomeration properties and condensed combustion products of aluminium water propellants with different CuO loadings by laser ignition test and thermogravimetric-differential scanning calorimetry. The introduction of binary MIC reduces the initial temperature of Al and increases its oxidation activity compared to conventional aluminium water propellants. Both combustion rate and heating rate were linearly related to CuO loading, and increased fivefold and sixfold, respectively, when CuO loading reached 5 wt.%. The combustion efficiency of the modified propellant increased by 5-12%. As a result of the addition of MICs, the average size of the coalesced combustion products decreased from more than 400 μm to about 200 μm , indicating that agglomeration was weakened. The ignition delay time was slightly reduced, and the combustion intensity first increased and then decreased with increasing CuO loading. Their work can help guide the development of advanced aluminium water propellants for various propulsion and energy applications. Yuxiang Li et al [58] successfully enhanced the gas generation capability of energy-containing composites by combining Al/CuO with TACP. The incorporation of TACP not only lowered the decomposition temperature, but also significantly enhanced the bursting performance, suggesting that the comprehensive performance of aluminothermic composites can be effectively tuned through the introduction of suitable additives and enhance the comprehensive performance of aluminothermic agents. Rui Cui et al [59] explored in depth the in situ growth mechanism of CuO through the core-shell structured

Al@CuO energy-containing microspheres prepared by the alcohol-thermal method, emphasizing the importance of optimizing the preparation conditions to reduce the homogeneous nucleation of CuO and to improve the performance of the material. Chong Chen et al [60] demonstrated the potential of TFA@Al in the surface modification of porous aluminium in increase the reaction temperature and heat of explosion, which provides a new material option in the field of energy materials. Haolun Wang et al [61] developed a low-current-sensitive, high-thermal-conductivity flexible energy-containing thin film by blowing and spinning technology, which achieves the excellent performance of not generating hotspots and maintaining structural integrity at high voltages, and proves the potential of its application in the field of transient electronics and other fields, especially in the field of transient electronics which needs the flexibility and high energy release in microelectromechanical systems.

Another study led by Yuxuan Zhou [62] focused on the combustion performance of aluminium nanothermite in low-pressure environments, and prepared highly loaded Al/CuO aluminium nanothermite thin films using 3D direct writing technique. It was confirmed that the combustion rate was independent of air pressure changes and the ignition temperature was maintained constant when the film thickness exceeded 182 μm , while the film exhibited excellent hydrophobicity and combustion performance at the air-water interface. The film combustion process was divided into three clearly defined stages, each releasing 7.714 J/g, 78.03 J/g, and 548.60 J/g of heat, respectively, accompanied by an obvious

mass reduction effect (shown in Fig. 6). Nevertheless, there was a risk of flame extinction when the combustion rate decreased to the 300-500 mm/s interval. The double-edged effect of air pressure on the combustion rate is reflected in the fact that, on the one hand, the pressure reduction inhibits the chemical reaction rate, especially processes that depend on

gas-phase reactions; on the other hand, it promotes the mass and heat exchange between the burning surface and the surroundings. These findings provide an important theoretical basis and practical guidance for understanding and regulating the combustion behaviour of aluminium nanothermite under extreme conditions.

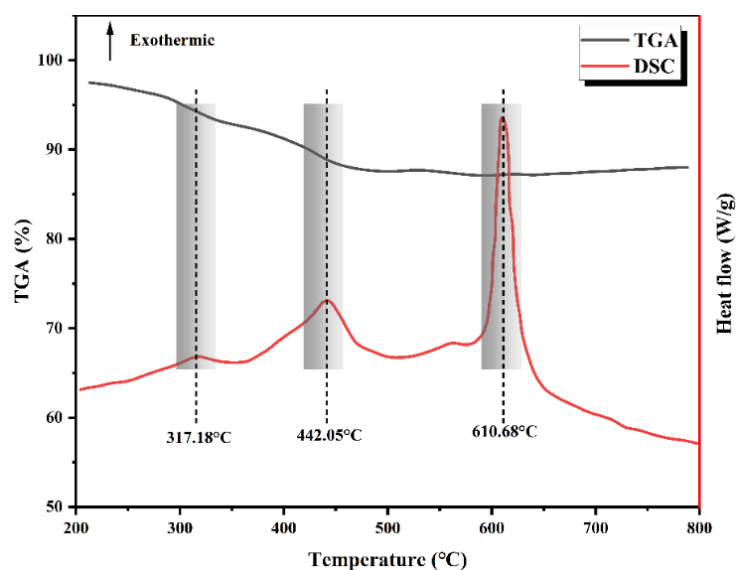


Figure 6. DSC curves of Al/CuO thin films [62].

From a comprehensive point of view, from basic theory to application practice, the research on aluminium thermite is gradually moving towards a more efficient, safer and more flexible direction. Through continuous exploration of new additives, optimization of the material structure, in-depth understanding of the reaction mechanism, the performance of aluminium thermite has been significantly improved, and its application areas are also expanding, from the traditional welding, military use, expanding to energy conversion, microelectronics technology and other broader areas. In the future, as materials science, nanotechnology and cross-disciplinary research continue to deepen, aluminium thermite technology is expected to achieve even greater technological breakthroughs, providing innovative solutions to address the challenges of energy conversion efficiency, materials synthesis, and energy demand in special environments.

3. Conclusions and Prospects

Aluminium thermite, as an efficient energy-releasing material, exhibits significant scientific value and application potential in the Al/CuO system. In recent years, the research mainly focuses on the following aspects:

1. Modification strategies and performance optimisation: The reaction properties and energy release efficiency of aluminothermic agents have been significantly improved by introducing additives, adjusting stoichiometric ratios and optimising structural design. For example, the addition of two-dimensional materials not only solved the problem of accidental ignition, but also achieved precise ignition in a specific way to improve heat release efficiency and safety. Adding graphene oxide to the Al/CuO system or using electrophoretic deposition technology to prepare composite films significantly improves the energy characteristics by adjusting the ratio of components.

2. New materials and composite systems: innovatively designed composite materials, such as the Al-TiB₂/CuO

ternary system, improve the combustion rate and reduce the combustion energy by optimising the order of the metal layers. In addition, the composite energy-containing material enhances the gas generating capacity and bursting performance, as well as constructs energy-containing microspheres with core-shell structure and surface-modified porous aluminium, which further improves the reaction efficiency.

3. Theoretical and Mechanistic Explorations: The effects of particle size and stoichiometric ratio on combustion characteristics were thoroughly investigated using theoretical models, and the important role of nanoscale on reaction kinetics was revealed. The development of low-current-sensitive, highly thermally conductive flexible energy-containing films enriches the theoretical understanding of the application field of aluminothermic agents.

4. Safety and application outlook: While pursuing high performance, safety has become the key to research. The combustion characteristics of aluminothermics are regulated by additives to balance the reactivity and safety. Future research will focus on the exploration of new additives, deepening the understanding of the reaction mechanism, expanding the application areas, strengthening the research on environmental friendliness and enhancing the safety assessment, so as to ensure the healthy development of aluminium thermite technology.

Looking ahead, the research on aluminothermics will tend to the following directions: first, continue to explore new additives and composite material design, with a view to further improving the energy release efficiency and reaction control precision without sacrificing safety. The second is to deepen the understanding of the reaction mechanism of aluminium thermite, and through a combination of theoretical simulation and experimental verification, precisely regulate the reaction kinetics and optimize the microstructure of the material. Third, to expand the application fields, especially in new energy, micro-nano-electronics technology, aerospace

and national defence and security and other high-tech fields, to develop more innovative and practical applications of aluminothermics. Fourth, to strengthen environmentally friendly research, explore degradable, low-pollution aluminium thermite formulations, in line with the requirements of sustainable development. Fifth, strengthen the safety assessment to ensure the stability and reliability of new aluminium thermite in various application scenarios, and promote the healthy and rapid development of aluminium thermite technology.

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