Conference materials
UDC 621.793.182
DOI: https://doi.org/10.18721/JPM.163.246

Effect of Al-CuO multilayer thermite structures thickness on combustion behavior

M.E. Shiryaev[1]✉, R.M. Ryazanov[2], A.V. Sysa[2], E.A. Lebedev[1,2]

[1] National Research University of Electronic Technology (MIET), Zelenograd, Moscow, Russia;

[2] Scientific-Manufacturing Complex "Technological Centre", Zelenograd, Moscow, Russia

✉ shiryaevme@bk.ru

Abstract. This study investigates multilayer structures of Al-CuO formed on the surfaces of sitall substrates using magnetron sputtering. The influence of changing the total thickness of the multilayer structure on the character and speed of the combustion front, characteristics of the reaction products, and gas emission levels is analyzed. Increasing the total thickness of the multilayer structure leads to changes in the speed and behavior of the combustion wave, as well as variations in the quantity, size, and form of the combustion products. The analysis includes theoretical data obtained using the Thermo software and experimental investigations. Gas emission values at different reaction temperatures have been evaluated.

Keywords: multilayer structure, Al-CuO, thermite, combustion

Funding: The study was supported by the Russian Science Foundation grant 22-29-01177, https://rscf.ru/project/22-29-01177/.

Citation: Shiryaev M.E., Ryazanov R.M., Sysa A.V., Lebedev E.A., Effect of Al-CuO multilayer thermite structures thickness on combustion behavior, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 16 (3.2) (2023) 267–272. DOI: https://doi.org/10.18721/JPM.163.246

Материалы конференции
УДК 621.793.182
DOI: https://doi.org/10.18721/JPM.163.246

Влияние толщины многослойных термитных структур Al-CuO на характеристики горения

М.Е. Ширяев[1]✉, Р.М. Рязанов[2], А.В. Сыса[2], Е.А. Лебедев[1,2]

[1] Национальный исследовательский университет «МИЭТ», Зеленоград, Москва, Россия;

[2] «Научно-производственный комплекс «Технологический центр», Зеленоград, Москва, Россия

✉ shiryaevme@bk.ru

Аннотация. В данном исследовании изучаются многослойные структуры Al-CuO, формируемые на поверхности ситалловых подложек методом магнетронного распыления. Анализируется влияние изменения общей толщины многослойной структуры на характер и скорость фронта горения, характеристики продуктов реакции, а также величины газовыделения. При увеличении общей толщины многослойной структуры наблюдается изменение скорости волнового горения и его характера, а также варьируются количество продуктов горения, их размер и форма. Приведен анализ теоретических данных, полученных с помощью программы Thermo и экспериментальных исследований. Оценены значения газовыделения при различных температурах реакции.

Ключевые слова: многослойные структуры, Al-CuO, термиты, горение

Финансирование: Исследование выполнено при поддержке гранта Российского научного фонда No. 22-29-01177, https://rscf.ru/project/22-29-01177/.

Ссылка при цитировании: Ширяев М.Е., Рязанов Р.М., Сыса А.В., Лебедев Е.А. Влияние толщины многослойных термитных структур Al-CuO на характеристики горения // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2023. Т. 16. № 3.2. С. 267–272. DOI: https://doi.org/10.18721/JPM.163.246

Introduction

Currently, extensive research is being conducted on various nanostructured thermite materials. Reactive compositions utilizing thin-film multilayer reactive structures as local heat sources offer efficient solutions for a range of technological challenges in microelectronics. In such structures, self-propagating exothermic reactions can occur after local initiation. The temperatures reached in these reactions can reach several thousand degrees Celsius, making thermite materials valuable for various applications, including welding, reactive bonding, and the aerospace industry.

Most thermite materials used in electronics consist of powder mixtures and multilayer thin films of aluminum combined with various oxidizers (MoO_3, CuO, WO_3, Fe_2O_3, I_3O_4, and Bi_2O_3) [1, 2]. Typically, these materials are fabricated by mixing nanoscale particles of pure metal (fuel) with nanoscale particles of metal oxide (acting as the oxidizer) in the form of powders [3, 4], granules [5], or thin films [5, 6].

Al-CuO thermite systems are widely used due to their high specific heat of combustion (approximately 4.1 kJ/g) and adiabatic reaction temperature (2843 K). Multilayer Al-CuO materials can be formed using magnetron sputtering. Control over the characteristics of combustion reaction products, including composition and morphology, is crucial in many application areas. Furthermore, the ability to control the propagation rate of the combustion front in wave thermite reactions, as well as gas emission, can be crucial for specific applications.

In this study, we investigated the influence of the thickness of the Al-CuO multilayer film on gas emission, combustion front propagation velocity, and characteristics of the reaction products.

Materials and Methods

The multilayer Al-CuO thermite structure was fabricated using magnetron sputtering on the URM-71 system. The aluminum and copper oxide layers were deposited sequentially by sputtering the respective targets in an argon environment at a pressure of 3×10^{-3} Torr. The copper oxide and aluminum targets were operated under a constant current mode at powers of 250 W and 500 W, respectively.

The geometric characteristics and combustion products of the multilayer structures were examined using scanning electron microscopy (SEM).

In this study, samples with different total structure thicknesses were considered, where the bilayer thickness (the combined thickness of one aluminum layer and one copper oxide layer) was 100 nm. The multilayer structure consisted of 20, 30, and 40 layers, resulting in total thicknesses of 2 μm, 3 μm, and 4 μm, respectively.

The combustion front propagation speed was measured using high-speed video recording. The combustion of the structures was recorded using a high-speed camera at a frame rate of 10,000 fps. Wave-like combustion was initiated using an electrical spark at the lower part of the sample. Based on the recorded combustion videos, the distance traveled by the combustion front, the time taken to cover that distance, and the velocity were calculated.

The Thermo program was used to obtain gas emission values at different reaction temperatures. The mole amounts of the components were calculated based on a total volume of 1 cm^3. The Thermo program, developed at the Institute of Structural Macrokinetics of the Russian Academy of Sciences, is designed to calculate thermodynamic equilibrium in complex multicomponent heterophase systems. The calculation results include the composition of equilibrium products (both condensed and gaseous) and adiabatic temperature [7].

Results and Discussion

Fig. 1 shows a storyboard of the combustion process of multilayer thermite structures with a total thickness of 2, 3 and 4 μm. The calculated values of the propagation velocity of the combustion front were 6.7, 10.2 and 7.8 m/s, respectively. The maximum speed of the front was achieved by combustion structures thickness of 3 μm.

The study of the combustion process revealed differences in the propagation speed of the wave combustion front depending on the structure thickness. For the sample with a thickness of 2 μm, as shown in Fig. 1,*a*, the reaction products were practically not expelled, and there was no gas emission. In the case of the sample with a thickness of 3 μm, Fig. 1,*b*, gas emission and a significant amount of various-sized reaction products expelled from the surface were observed. Meanwhile, the combustion of the structure with a thickness of 4 μm, Fig. 1,*c*, was characterized by intense gas emission and a large quantity of expelled reaction products, which were larger in size compared to those expelled from the 3 μm thick structure.

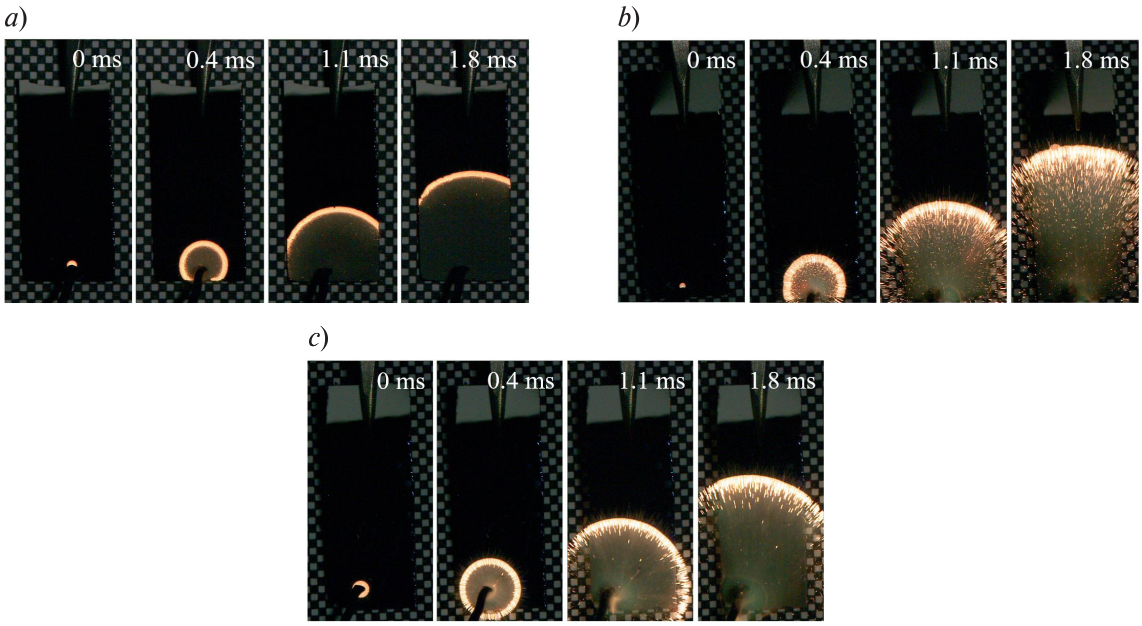

Fig. 1. Storyboard of the combustion process of multilayer Al-CuO thermite structures with a total thickness of 2 μm (*a*), 3 μm (b) and 4 μm (*c*)

Investigation of combustion products in multilayer thermite structures (MTS) using scanning electron microscopy yielded the results presented in Fig. 2.

For MTS with a total thickness of 2 and 3 μm, Fig. 2, *a*, *b*, respectively, the combustion products appeared as elongated droplet-shaped objects with spherical inclusions. However, for MTS with a total thickness of 3 μm, the reaction products exhibited significantly larger sizes, reaching several tens of μms. In addition to the larger particles, smaller particles were uniformly distributed on the substrate surface. In contrast, for MTS with a total thickness of 4 μm, Fig. 2,*c*, large objects with inclusions were practically absent. Instead, smaller droplet-shaped objects formed on the substrate surface, exhibiting similar structures to the reaction products in the other two cases, and they were covered with short and thin fibers.

To justify the differences in the form and quantity of reaction products, modeling was performed using the Thermo program. Theoretical values of gas emission were obtained within the specified temperature range of the reaction, ranging from 2250 K to 2850 K. The obtained data is depicted in Fig. 3.

In the analysis of multilayer thermite structure combustion, it's crucial to consider heat dissipation through the substrate, as some energy will disperse, leading to a decrease in reaction temperature. The substrate's influence on heat dissipation depends on the total thickness of the multilayer structure - the thinner the structure, the stronger the substrate's influence. This study experimentally found that multilayer thermite structures with a total thickness of less than 1.5 μm cannot sustain self-propagating combustion.

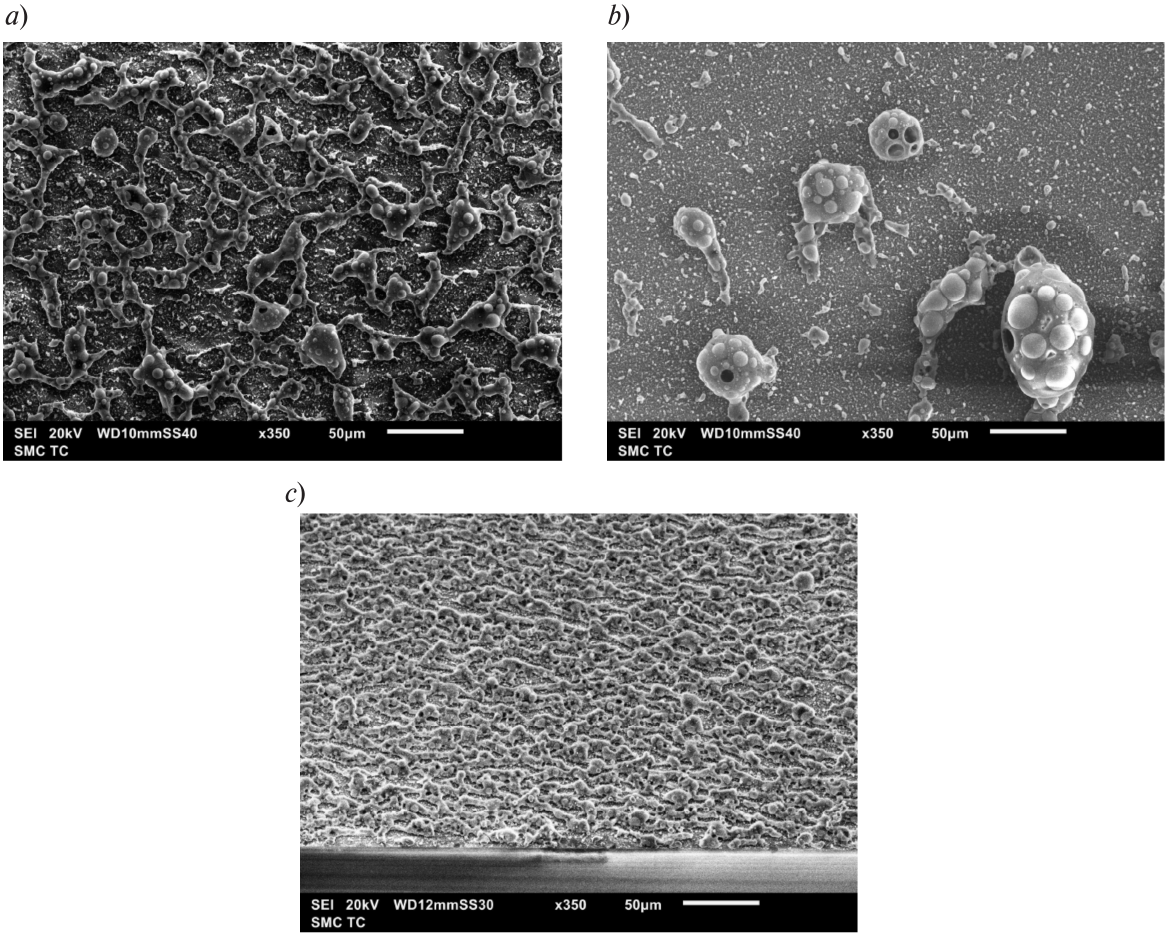

Fig. 2. Results of SEM studies of reaction products on the surface of multilayer Al-CuO thermite structures with thicknesses of 2 μm (*a*), 3 μm (*b*), 4 μm (*c*)

The comparison of experimental results obtained through SEM and high-speed video recording with theoretical gas emission calculations (presented in Fig. 3) allows us to estimate the change in reaction temperature depending on the total thickness of the multilayer thermite structure. The upper temperature limit is constrained to 2666 K (the adiabatic reaction temperature of Al + CuO, corresponding to the chosen component ratio, obtained in the Thermo program). This temperature value (marked in Fig. 3 as T_{adiab}) is below the melting points of aluminum (2743 K, Al T_{boil}), copper (2835 K, Cu T_{boil}), and aluminum oxide (3250 K), meaning these components do not contribute to gas emission.

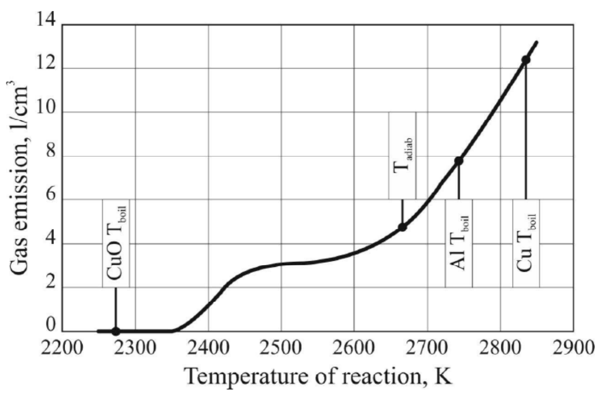

Fig. 3. Relationship between gas emission and the temperature of the reaction

The combustion of a 2 μm thick multilayer thermite structure visually does not accompany gas emission (Fig. 1,*a*), and SEM images clearly show that most reaction products remain on the substrate surface. The reaction products are hardened droplets of aluminum oxide with copper inclusions. Thus, for a multilayer thermite structure with a total thickness of 1.5 μm, the reaction temperature lies in the range from 2345 K (melting temperature of aluminum oxide) to 2400 K, when gas emission is still insignificant.

The combustion processes of multilayer thermite structures with a total thickness of 3 and 4 μm visually differ little from each other according to high-speed video recording results (Fig. 1,*b* and 1,*c*). However, compared to 2 μm thick multilayer thermite structures, significant gas emission is observed. SEM images clearly show that after the combustion of thicker multilayer structures, significantly fewer reaction products remain on the substrate surface, which may be associated with more intense gas emission.

Analyzing the character of the graph presented in Fig. 3, it can be assumed that the front temperature values for multilayer structures with a total thickness of 3 and 4 μm should be on different sides of the plateau observed in the range from 2450 to 2550 K, where significant changes in gas emission intensity do not occur. Thus, the reaction temperature for a 3 μm thick multilayer structure may lie in the range from 2400 to 2450 K, and for a 4 μm thickness from 2550 to 2666 K. It can also be assumed that further increase in the total thickness of the multilayer structure (more than 4 μm) will not lead to a significant change in the combustion character, as the adiabatic reaction temperature cannot be exceeded.

Conclusion

During the study, the influence of the thickness of the multilayer Al-CuO thermite structures on the characteristics of combustion reaction products was investigated. The results showed that the thickness of the multilayer structure had a significant impact on both the reaction characteristics and its products. High-speed video analysis revealed that the combustion front velocity increased with increasing thickness, while SEM analysis demonstrated changes in the morphology and composition of the reaction products. An analysis of theoretical calculated characteristics and obtained experimental data was conducted, revealing a relationship between them. This study provides an estimation of reaction temperature values for multilayer thermite structures of various thicknesses, contributing to a better understanding of their combustion behavior.

REFERENCES

1. **Nellums R., Terry B., Tappan B., Son S., Groven L.G.**, The effect of silicon powder characteristics on the combustion of silicon/teflon/viton nanoenergetics // Propellants, Explosives, Pyrotechnics. 39 (3) (2014) 337–347.

2. **Yarrington C., Son S., Foley T., Obrey S., Pacheco A.**, Nano aluminum energetics: The effect of synthesis method on morphology and combustion performance // Propellants, Explosives, Pyrotechnics. 36 (6) (2012) 551–557.

3. **Guidotti R., Odinek A., Reinhardt F.**, Characterization of Fe/$KClO_4$ Heat Powders and Pellets // Journal of Energetic Materials. 24 (4) (2006) 271–305.

4. **Petrantoni M., Rossi C., Salvagnac L., Conedera V., Esteve A.**, Multilayered Al/CuO thermite formation by reactive magnetron sputtering: Nano versus micro // Journal of Applied Physics. 108 (8) (2010) 84–32.

5. **Fritz J., Grzyb A., Knio O., Grapes M., Weihs T.**, Characterizing solid-state ignition of runaway chemical reactions in Ni-Al nanoscale multilayers under uniform heating // Journal of Applied Physics. 118 (13) (2015) 101–135.

6. **Grapes M., Weihs T.**, Exploring the reaction mechanism in self-propagating Al/Ni multilayers by adding inert material // Combustion and Flame. 172 (2016) 105–115.

7. Institute of Structural Macrokinetics of the Russian Academy of Sciences. “Thermo”. URL: https://ism.ac.ru/thermo/. Accessed Mar. 20, 2023.

THE AUTHORS

SHIRYAEV MAKSIM E.
shiryaevme@bk.ru
ORCID: 0000-0002-0028-4061

RYAZANOV ROMAN M.
r.m.ryazanov@gmail.com
ORCID: 0000-0002-2464-8712

SYSA ARTEM V.
sysa.artem@yandex.ru
ORCID: 0000-0002-5010-7071

LEBEDEV EGOR A.
dr.beefheart@gmail.com
ORCID: 0000-0002-5085-5408

Received 21.07.2023. Approved after reviewing 17.08.2023. Accepted 17.08.2023.