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Ignition behavior and characteristics of microwave-combustion synthesized Al_2O_3–TiC powders

Duangduen Atong [a,*], David E. Clark [b]

[a] National Metal and Materials Technology Center, 114 Thailand Science Park, Paholyothin Road, Klong 1, Klong Luang, Pathumthani 12120, Thailand
[b] Virginia Polytechnic Institute and State University, Department of Material Science and Engineering, Blacksburg, VA 24601, USA

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Abstract

Al_2O_3–TiC powders were produced by self-propagating high temperature synthesis (SHS) using microwave heating (MH) and microwave hybrid heating (MHH). The central composite design (CCD) was used for conducting extensive experiments to study the effect of reaction parameters including amount of excess Al and Al_2O_3, particle size of C and Al_2O_3, and heating method on ignition behavior and characteristics of the resulting powders. The empirical models relating these important parameters and their interaction to the responses were then developed.

Keywords: Microwave; SHS; Combustion synthesis; Al_2O_3–TiC; CCD

1. Introduction

Al_2O_3–TiC composite has been widely used in industries as cutting tools and wear resistance coating because of its high hardness, chemical stability, good strength and toughness at elevated temperatures, and excellent wear resistance. Currently, it is manufactured primarily by pressureless sintering or hot pressing of TiC and Al_2O_3 powders [1]. The preparation of Al_2O_3–TiC composite powder by self-propagating high temperature synthesis (SHS) process has drawn much attention recently [2,3]. The principle of the SHS process is based on exothermic reactions between reactants which results in high-purity product. However, researches on the use of microwave energy to synthesize Al_2O_3–TiC composite are rare [4,5]. The union of these two technologies allow for enhanced process control and superior product quality. Previous work on producing Al_2O_3–TiC powders by SHS technique using microwave energy showed that the most significant factors affecting ignition behavior and characteristics of the ignited powders were amount of Al and Al_2O_3, particle size of C and Al_2O_3, and also heating method (microwave heating, MH and microwave hybrid heating, MHH). Hence, in this study, the central composite design (CCD) was employed to perform a comprehensive evaluation of the effects of those five important parameters in a system of TiO_2/C/Al/Al_2O_3.

2. Experimental

Al_2O_3–TiC powders were combustion synthesized using microwave energy from a mixture of TiO_2, C, Al, and Al_2O_3 reactants. The reaction involved was as follows

$$3TiO_2 + 3C + (4 + x)Al + yAl_2O_3 \rightarrow 3TiC + (2 + y)Al_2O_3 + xAl \quad (1)$$

The CCD was used for conducting extensive experiments to study the effect of reaction parameters including amount of excess Al and Al_2O_3, particle size of C and Al_2O_3, on ignition behavior and characteristics of the ignited powders. Two experimental set for MH and MHH were conducted in parallel to each other. Each reaction parameter was evaluated at five different levels (Table 1). The number of experimental runs required for four parameters at five levels was 30. The procedure for constructing 30 runs-CCD and their actual factor levels was given elsewhere [4]. The particle size of TiO_2 and Al used were 2–3 μm (99% rutile, Alfa Aesar) and 10 μm (99.7%, Alfa Aesar), respectively. The mixed reagent powders were pulverized in a porcelain mor-

* Corresponding author. Fax: +66-2-564-6447.
E-mail address: duangdua@mtec.or.th (D. Atong).

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Table 1
Factor and investigated levels of 30-runs CCD

Factor	Level				
C particle size (μm)	0.017 (99%)[a]	0.027 (>99%)[a]	0.037 (>99%)[a]	0.042 (99.9%)[b]	0.075 (>99%)[a]
Al_2O_3 particle size (μm)	0.49 (99.8%)[c]	<10 (99.7%)[d]	20–38 (99.9%)[b]	44–53 (99.9%)[b]	63–74 (>99%)[e]
Amount of excess Al (%)	0	10	20	30	40
Amount of excess Al_2O_3 (%)	0	10	20	30	40

[a] Cabot Corp.
[b] Alfa Aesar.
[c] Alcoa.
[d] Aldrich.
[e] Fisher.

tar, sieved, and then dried in an oven at 120 °C for 2 h prior to ignition. The samples were ignited under 800 W-microwave power operating at 100% duty cycle. The Alfrax-66–40% SiC was used as a susceptor for the MHH experiment. An inconel-shielded K-type thermocouple and a two color infrared pyrometer were used to determine temperatures. The particle size, density, and surface area of powders were analyzed. Analysis of variance (ANOVA) was used to provide a quantitative measure of confidence.

3. Results and discussions

Al_2O_3–TiC powders were successfully produced by SHS process using both MH and MHH. The JMP Start Statistics software was used to perform a multiple regression analysis for each of the responses measured. The final regression models for predicting all the investigated response values were summarized in Table 2. It was found that MH samples could be ignited within approximately 50 s while MHH ignited samples required longer times, 2.30–3 min. The MHH samples ignited at higher temperature than MH samples and resulted in lower combustion temperature. It could be seen from the model that as the particle size of carbon increased the ignition time decreased, but not significantly. More detail was shown on Fig. 1. The slowest ignition time occurred when the smallest carbon particle size was used. However, the sample with intermediate carbon size was ignited faster than one with the largest carbon size. This may be because it provided the best reactant particle arrangements and particle contact between TiO_2 and Al which is believed to be necessary for the initial reaction to TiO_2 and Al. In MH case, the sample with the largest carbon size was observed to have highest combustion temperature. The reason for this was thought to be due to a decrease surface area of the larger carbon size, decreasing heat loss. When using the MHH technique, the susceptor provided another heat source and helped reduced heat loss from sample, thus the variation of carbon size did not change combustion temperature significantly. It was also observed that as the alumina particle size increased the ignition time and temperature decreased (Fig. 2). This was because the rest of the reactants could act as a continuous phase and thus increased the particle contact between TiO_2 and Al, resulting in faster ignition. The sample with an intermediate alumina size was observed to have a lowest combustion temperature. With Al_2O_3 (20–38 μm) being close to Al size (10 μm), smaller TiO_2 particles (2–3 μm)

Table 2
Regression models for predicting ignition behavior and characteristic of microwave ignited Al_2O_3–TiC powders

	Prediction equations
(a) MH	
Ignition time (s)	$t_{ig} = 48.83 - 5.83X_1 + 13.17X_3 + 16.08X_4 - 7.5X_2X_4 + 6.88X_3X_4$
Ignition temperature (°C)	$T_{ig} = 145.67 + 30.08X_3 + 48.75X_4 + 35.77X_4^2$
Combustion time (s)	$t_c = 57.83 - 6X_1 + 14X_3 + 18.03X_4 - 7.63X_2X_4$
Combustion temperature (°C)	$T_c = 1929.17 + 23.20X_1 - 37.79X_3 - 61.38X_4 + 47.05X_1^2 + 59.05X_2^2 - 49.44X_1X_4 - 48.31X_2X_4 - 117.06X_3X_4$
Powder density (g/cm^3)	$\rho = 3.76 + 0.02X_1 - 0.17X_3 - 0.02X_4 - 0.02X_1^2 - 0.02X_2^2$
Particle size (μm)	$PS = 1.39 - 0.18X_3 - 0.12X_4 - 0.14X_1^2 - 0.12X_3^2 - 0.10X_4^2$
Specific surface area (m^2/g)	$Sp.SA = 1.16 - 0.17X_1 + 0.38X_3 + 0.19X_4 + 0.27X_1^2 + 0.17X_3^2 + 0.21X_2X_3$
Agglomeration size (μm)	$AS = 52.76 + 4.81X_1 + 6.70X_2 - 5.75X_4$
(b) MHH	
Ignition time (s)	$t_{ig} = 159.83 - 25.83X_1 - 20X_2 + 45.83X_3 + 48.33X_4$
Ignition temperature (°C)	$T_{ig} = 383.17 + 61.46X_3 + 78.13X_4 + 37.28X_4^2 - 46.94X_1X_4$
Combustion time (s)	$t_c = 163 - 26.61X_1 - 19.80X_2 + 45.63X_3 + 48.79X_4$
Combustion temperature (°C)	$T_c = 1726.17 - 55.83X_3 - 58.67X_4 + 66.96X_2^2 - 58.38X_1X_4 - 61.38X_2X_4 - 125.25X_3X_4$
Powder density (g/cm^3)	$\rho = 3.80 - 0.03X_1 - 0.17X_3 - 0.03X_4 - 0.02X_2^2$
Particle size (μm)	$PS = 1.51 - 0.11X_1 - 0.13X_3 - 0.14X_4 - 0.14X_1^2 - 0.20X_3^2 - 0.12X_4^2$
Specific surface area (m^2/g)	$Sp.SA = 1.05 + 0.22X_1 + 0.37X_3 + 0.29X_4 + 0.21X_1^2 + 0.28X_3^2 + 0.17X_4^2 + 0.22X_2X_3 + 0.19X_3X_4$
Agglomeration size (μm)	$AS = 54.32 + 5.24X_1 + 6.02X_2 - 6.74X_4 - 2.5X_3^2 - 6.64X_3X_4$

X_1, C particle size; X_2, Al_2O_3 particle size; X_3, wt.% excess Al; X_4, wt.% excess Al_2O_3.

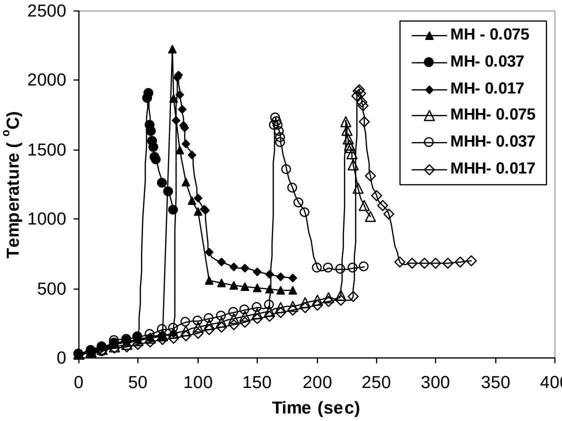

Fig. 1. Effect of carbon particle size (μm) and heating method on ignition behavior of composites.

would be distributed in the intersticies of packed larger particles of both Al_2O_3 and Al. Thus, only the Al in direct contacted with TiO_2 reacted and melting occurred locally. Results of the effects of diluent addition on ignition behavior of samples shown in various models (Table 2) were confirmed in more details in Figs. 3 and 4. The addition of Al or Al_2O_3 to the reactants increased ignition temperature, and decreased the combustion temperature. The ignition time increased as the amount of the diluents increased. This was due to the decreased heat generation rate. In addition, it was thought that the high microwave reflectivity of the Al partially shielded the sample and resulted in a shallow microwave penetration depth. The Al_2O_3 particles also may act as a barrier to reactant particle contact. It has low electrical conductivity and very low loss tangents thus allowing microwaves to pass through with very little absorption.

Nevertheless, it was found that both MH and MHH ignited powders showed no significant difference in the properties. The average values of powder densities, particle sizes, and specific surface areas were in the range of 3.7–3.8 g/cm^3, 1.4–1.5 μm, and 1–1.2 m^2/g, respectively. From the models, the effect of the addition of Al (X_3) on the final product density was much more significant as compared to the other effects. The density decreased with increasing amount of Al. It was clear that the agglomeration size of the ignited powders depended on the size of the reactant as much as the amount of excess Al_2O_3. The larger reactant particles produced coarser reacted powders while the addition of Al_2O_3 to the reactants decreased the agglomeration size. Some interactions between the parameters also have significant effects on the investigated responses. The interaction between the amount of excess Al_2O_3 (X_4) and other parameters (X_1, X_2, X_3) was found to have more effect on the responses than any other combinations of the interaction.

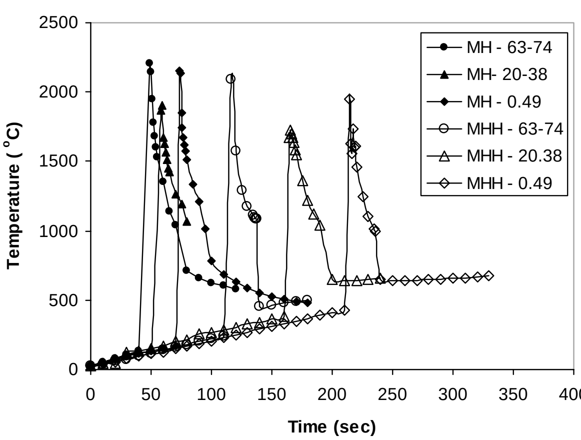

Fig. 2. Effect of alumina particle size (μm) and heating method on ignition behavior of composites.

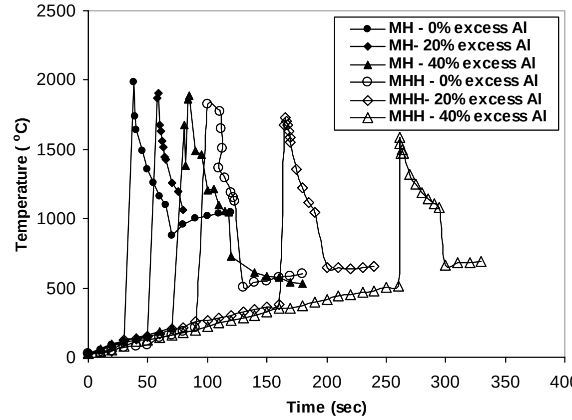

Fig. 3. Effect of excess amount of Al and heating method on ignition behavior of composites.

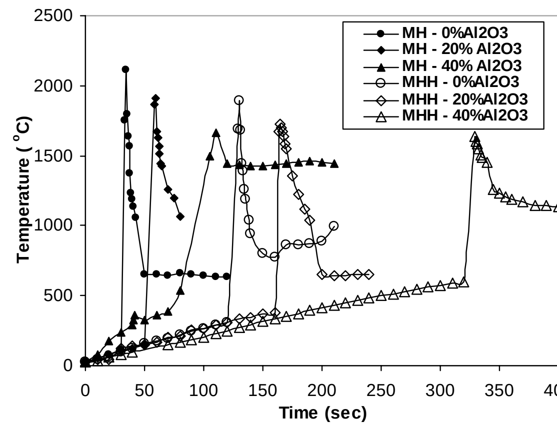

Fig. 4. Effect of amount of Al_2O_3 and heating method on ignition behavior of composites.

4. Summary

The results showed no significant difference in the characteristics of powders ignited by two heating method (MH and MHH), though a clear difference in ignition behavior was observed. Various empirical model relating parameters to the ignition behavior and characteristics of powders were developed. This provided the information required for adjusting parameters in order to obtain the desired products.

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References

[1] J.G. Baldoni, S.T. Buljan, Ceramics for machining, Am. Ceram. Soc. Bull. 76 (2) (1988) 381–387.

[2] C.R. Bowen, B. Derby, The formation of TiC/Al_2O_3 microstructures by SHS reaction, J. Mater. Sci. 31 (1996) 3791–3803.

[3] A.O. Kunrath, T.R. Strohaecker, J.J. Moore, Combustion synthesis of metal-matrix composites: Part III. The Al–TiC–Al_2O_3 system, Scripta Mater. 34 (2) (1996) 189–194.

[4] D. Atong, Microwave-induced combustion synthesis of Al_2O_3–TiC powder. Dissertation, University of Florida, 2000.

[5] D. Atong, D.E. Clark, Microwave-induced combustion synthesis of Al_2O_3–TiC powder, in: D.E. Clark, J.G.P. Binner, D.A. Lewis (Eds.), Microwave: Theory and Application in Materials Processing V, Ceram. Trans. Vol. III, American Ceramic Society, Westerville, OH, 2001, pp. 197–206.