Preparation of AlB_{12} powder by self-propagating high-temperature synthesis (SHS)

Chao Wang[1]*, Xiaoming Cao[2], Mengge Dong[3], Lu Zhang[4], Jianxing Liu[3], Xiaozhou Cao[3], and Xiangxin Xue[3]*

[1]Department of Mechanical Engineering, the University of Texas at Dallas, Richardson, TX, 75080, USA
[2]Institute of Metal Research, Chinese Academy of Science, Shenyang, Liaoning, 110016, China
[3]School of Metallurgy, Northeastern University, Shenyang, Liaoning, 110819, China
[4]School of Energy and Environment, Anhui University of Technology, Ma'anshan, 243002, China

Chao Wang, Xiaoming Cao, and Mengge Dong made an equal contribution.

*Corresponding author. E-mail: wang.chao@utdallas.edu, xuexx@mail.neu.edu.cn

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Abstract. Self-propagating high-temperature synthesis (SHS) process is used to prepare AlB_{12}. The phase analysis results of preparing AlB_{12} with Al and B_2O_3 as raw materials show that under air and argon conditions, the self-propagating and acid-washed self-propagating powders all have α-Al_2O_3 impurities when Mg, Al and B_2O_3 are used as raw materials. The phase analysis results of the preparation of AlB_{12} show that under argon conditions, the self-propagating and acid-washed, self-propagating powder has un-removable $MgAl_2O_4$ impurities, and the root cause of the low purity of AlB_{12} prepared by the self-propagating method is the presence of un-removable impurities.

Introduction

Most of the borides are crystals with high hardness and melting point [1-4]. Stable chemical properties and a wide range of applications make it widely used in composite materials, semiconductors, and in various areas of national defense, such as radiation protection [5-8]. Among them, AlB_{12} has a special electronic structure and bonding characteristics [9, 10]. It can effectively adjust the conductivity of semiconductor materials, and thus is extensively employed in conductors and semiconductor materials. In addition to the above characteristics, the content of boron in AlB_{12} is extremely high, reaching 82.8%, which is very promising as neutron shielding material [11-13].

Ceramic powders are usually synthesized by traditional sintering methods [14-16]. However, the use of this method to synthesize ceramic powder takes a long time, consumes a great deal of energy and pollution [17]. Self-propagating high-temperature synthesis (SHS) is a unique technique for synthesizing materials by self-heating and self-conduction of high chemical reaction heat between reactants. This technology was first discovered by Merzhanov et al., in their research on the combustion of solid propellants in rockets and was announced in 1967. Compared with the conventional sintering method, the advantages of the SHS method can be summarized as follows: (1) It is time saving and makes full use of energy [18]. (2) It requires only simple equipment and processes [19]. (3) The high product purity and product conversion rate are close to 100% [20]. (4) It can not only produce ceramic powder, but if the proper amount of pressure is applied at the same time, high-density combustion products can also be produced [21]. (5) High output [22].

In previous studies, AlB_{12} powder was fabricated by through the use of the SHS method [23]. The calculation results of preparing AlB_{12} with Mg, Al_2O_3 and B_2O_3 as raw materials show that the adiabatic temperature of the system is 2789.5K, which satisfies the self-propagating reaction conditions. Further, the phase analysis results show that there is no matter in air or argon, the self-propagating powder and the acid-washed self-propagating powder all have $Mg_{0.4}Al_{2.4}O_4$ impurities, and the purity of the prepared AlB_{12} is not high.

Although AlB_{12} is produced, $Mg_{0.4}Al_{2.4}O_4$ has not been removed and still exists. In this paper, Al, B_2O_3 and Mg, Al, B_2O_3 were used as raw materials to conduct experimental studies on self-propagating synthesis of AlB_{12}.

Experimental procedure

The starting materials used in this research were Al powder (purity＞99% Al, average particle size 50 μm), B_2O_3 powder (purity＞99%, average particle size 96 μm), and Mg powder(purity＞98%, average particle size 100 μm).

The steps used in the self-propagating process to synthesize AlB_{12} ceramic powder are as follows: (1) Weigh a certain amount (in proportion to the reaction equation) of the original material powder, place it in the ball milling tank, and mix the ball mill for 2 hours. (2) Intercept the resistance wire and connect it to the two poles of the self-propagating device and place the material in the atmosphere with one end close to the resistance wire. (3) Start the ignition device and slowly increase the current. When the pointer fluctuates sharply, reduce the current and keep the current increasing steadily. Finally, the resistance wire will reach a molten state when the material is induced to burn, and the current is turned off. (4) The reaction product is pulverized and sieved with 160 meshes, and samples under the sieve are sampled for detection and analysis.

This article focuses on the study of two reaction systems, system 1: Al and B_2O_3, and system 2: Mg, Al, and B_2O_3. Two experimental atmospheres are used in both systems (Table 1).

Table 1 The experimental scheme of tests

Serial	Reactant	Atmosphere	Pickling condition
S1	$Al+B_2O_3$	Air	Before pickling
S2	$Al+B_2O_3$	Air	After pickling
S3	$Al+B_2O_3$	Ar	Before pickling
S4	$Al+B_2O_3$	Ar	After pickling
S5	$Mg+Al+B_2O_3$	Ar	Before pickling
S6	$Mg+Al+B_2O_3$	Ar	After pickling

In the Al-B_2O_3 system, the following chemical reactions mainly occur:

$$13Al+6B_2O_3 \rightarrow AlB_{12}+6Al_2O_3 \quad (1)$$

In the Mg-Al-B_2O_3 system, the following chemical reactions mainly occur:

$$3Mg+B_2O_3 \rightarrow 2B+3MgO \quad (2)$$

$$Al+12B \rightarrow AlB_{12} \quad (3)$$

$$18Mg+Al+6B_2O_3 \rightarrow AlB_{12}+18MgO \quad (4)$$

After the combustion synthesis, the extraneous components were leached out from the synthesized powder with 60° C in diluted HCl. The phase analysis of the synthesized powder was carried out using an X-ray diffractometer（XRD, X’Pert Pro MRD）with a Philips diffractometer using Cu Ka. The microstructure of powders was investigated using a scanning electron microscope (SEM, S-3400N).

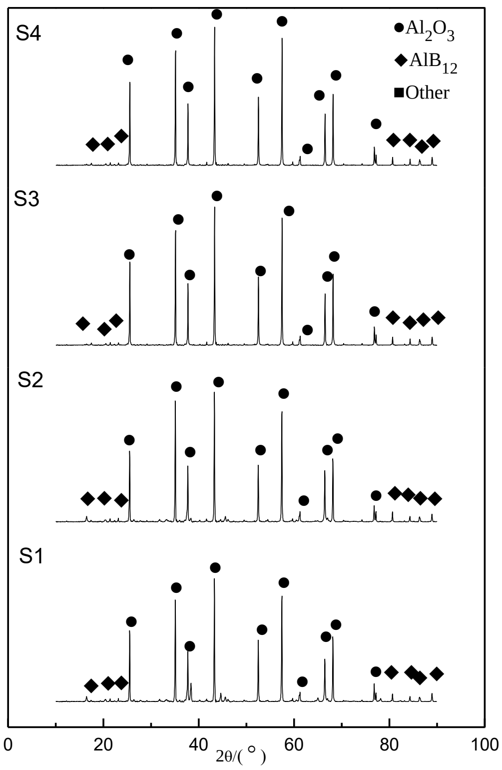

Fig. 1 XRD patterns of fabrication AlB_{12} from Al and B_2O_3

Results and discussion

Figure 1 is the X-ray diffraction pattern of Al and B_2O_3 prepared under both air conditions (before and after pickling) and argon conditions (before and after pickling) respectively. It can be seen from the figure that in either air or argon conditions, irremovable Al_2O is found in the bottom3. Analysis of its crystal structure revealed that the α-Al_2O_3 is corundum, an extremely stable substance that is difficult to remove through physical and chemical reactions. Therefore, AlB_{12} prepared from Al and B_2O_3, contains a large amount of inseparable corundum, which contributes to the failure of the self-propagating preparation of AlB_{12} using Al and B_2O_3 as raw materials.

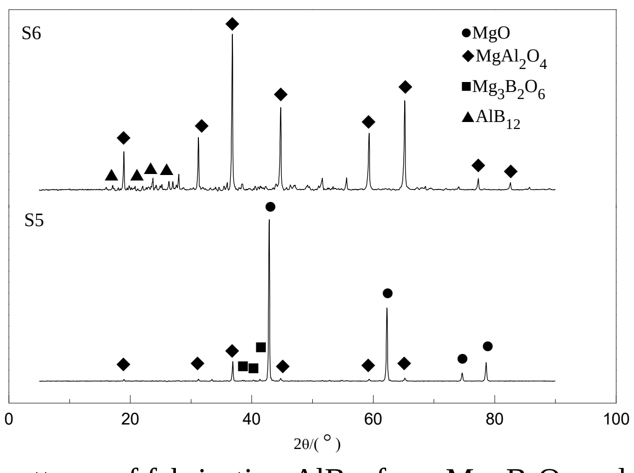

Fig. 2 XRD patterns of fabrication AlB_{12} from Mg, B_2O_3 and Al_2O_3

Figure 2 is the X-ray diffraction pattern of powder prepared with Mg, Al and B_2O_3 under argon conditions (before pickling). From the results of phase analysis, the main components of the coarse powder before pickling are MgO, $Mg_3B_2O_6$, $MgAl_2O_4$, and AlB_{12}, while in the powder after pickling, when MgO and $Mg_3B_2O_6$ are removed, the main impurity is $MgAl_2O_4$. This shows that the purity of AlB_{12} is not high when prepared by self-propagating, self-propagation when the raw materials used are Mg, Al and B_2O_3.

Figure 3 shows the microscopic morphology of powder prepared through use of the self-propagating method under argon conditions with Al and B_2O_3 as raw materials after pickling. From an analysis of the energy spectrum results, the A particles – with obvious layering phenomenon on the left are Al_2O_3 particles – while the B particles – with more obvious granular shape on the right – are AlB_{12}. This situation shows that despite the pickling treatment, Al_2O_3 is still untreated. It also shows that the unremovable by-product Al_2O_3 uses Al and B_2O_3 as raw materials and is the biggest obstacle to self-propagating preparation of AlB_{12}.

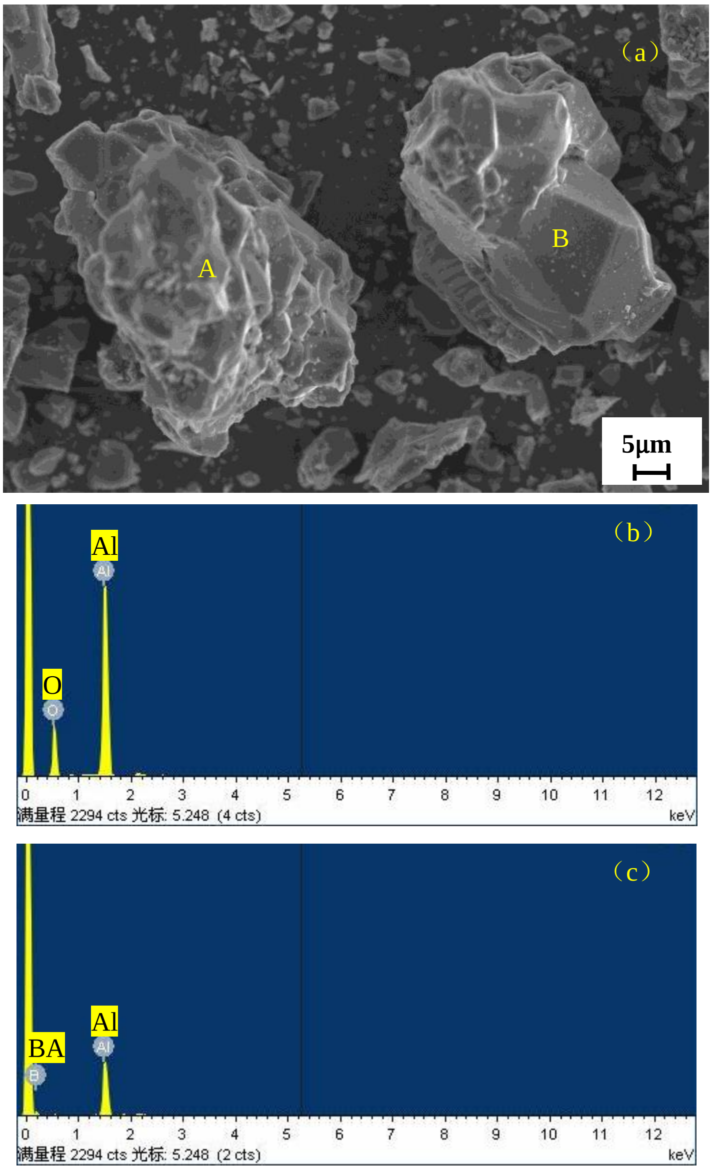

Fig. 3 (a): Microstructure photographs of powder by SHS; (b): EDS analysis of A area; (c): EDS analysis of B area

Table 2 shows the elemental analysis results after pickling of Al and B_2O_3 as raw materials and self-propagating preparation of AlB_{12} with Mg, Al, and B_2O_3 as raw materials. From the results in the table, the acid wash product prepared with the use of Al and B_2O_3 as raw materials has the highest content of O element, followed by B and Al. Observing the test samples, there are still insoluble substances, so the test results are also relatively incomplete. This shows that under argon conditions, using Al and B_2O_3 as raw materials to prepare AlB_{12}, and using the self-propagating method, the purity of AlB_{12} in the prepared product is low, and the B content is insufficient.

Table 2 The results of elementary analysis (mass fraction, %)

Serial	B	Mg	Al	O
S4	12.6	—	2.21	85.19
S6	58.5	9.62	3.43	28.45

Summary

The phase analysis results of preparing AlB_{12} using Al and B_2O_3 as raw materials shows that there are α-Al_2O_3 impurities in the self-propagating powder regardless of either the air condition or the argon condition, and it cannot be removed. Consequently, the purity of the prepared AlB_{12} is not high. The phase analysis results of preparing AlB_{12} using Mg, Al and B_2O_3 as raw materials indicates that the self-propagating and acid-washed self-propagating powder has unremovable $MgAl_2O_4$ impurities under argon conditions, and the purity of the prepared AlB_{12} is not high, causing self-propagation. The fundamental reason for the low purity of AlB_{12} prepared by this method is the existence of impurities that cannot be removed.

Notes

The authors declare that they have no competing financial interest.

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