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Microwave Induced Reduction/Oxidation Reaction by SHS Technique

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마이크로파를 이용한 SHS 방법에 의한 분말의 산화-환원반응

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Abstract

A reduction/oxidation reaction between Al metal powder and SiO_2 powder was performed by Self-propagating High-temperature Synthesis(SHS) reaction induced by microwave energy to produce a composite of Al_2O_3 and Si powders by using a 2.45 GHz kitchen model microwave oven. A Microwave Hybrid Heating(MHH) method was applied by using SiC powders as a suscepting material to raise the temperature of the disk samples and the heat increase rate of over 100°C/min were obtained before the reaction. The reaction started around 850°C and the heat increase rate jumped to over 200°C/min after the reaction took place.

요 약

가정용 2.45 GHz 마이크로파 오븐을 사용하여 Al 금속분말과 SiO_2 분말간에 SHS 방법에 의하여 산화/환원 반응을 통한 Al_2O_3 분말과 Si분말간의 복합체를 얻을 수 있었다. 분말간의 반응을 일으키기 위한 온도까지 승온시키기 위하여는 SiC 분말을 susceptor로 이용한 마이크로파 복합가열(Microwave Hybrid Heating)방법을 사용하여 분당 100°C의 승온 속도로 가열하였으며 반응은 850°C 근처에서 일어났으며 가열 속도는 반응이 시작 되면서는 분당 200°C 이상의 온도상승이 일어나면서 원하는 반응을 얻을 수 있었다.

1. Introduction

Microwave processing of materials has attracted much interest in recent years in ceramics, polymers, and composites.[1-3] It has many advantages over conventional processing methods such as selective heating, short processing time by rapid heating, cost savings, processing not possible with conventional means. In addition, self-propagating high-temperature synthesis (SHS) is another promising technology since it has various advantages such as low energy requirement, higher purity of the product, and a short processing time and requires low capital investment for process equipments. Many materials have been produced by SHS using various energy sources to ignite the reaction such as a metal coil heating filament, a conventional oven, and laser beam.[4,5] Recently microwave energy was used to ignite the reaction between ceramics and metal powders. Microwave energy heats the sample from the center of the body and ignites the reaction in the interior of the sample, and a combustion wavefront propagates radially outward.[6]

The power absorbed by the sample is given by

$$P_{ab} = \sigma E^2 = 2\pi f \varepsilon_0 \varepsilon_r' \tan\delta E^2 \quad (1)$$

where σ is the conductivity, f is the frequency, ε_0 is the permeability of vacuum, ε_r' is the relative dielectric constant, $\tan\delta$ is the loss factor, E is the electric field in the sample, The skin depth(δ) at which the incident power is reduced to 1/e of its value at the surface is simplified as

$$\delta = 2\rho\ 10^{-2}\ (\rho\ \lambda)^{1/2} \quad (2)$$

where λ is the incident wavelength and the ρ is the resistivity of the metal in ohm · meter. The skin depth(δ) of flat metal surface is not deep at 2.45 GHz, usually less than 3 μm, while the penetration depth of a mixture of a metallic powder and a ceramic powder can be deeper since the particle size is small. Hence more metallic powders inside the sample can be radiated by microwave. Each particle is affected by surface current down to the skin depth(δ) and can act as multiple ignition sites to heat up the sample. The heat is propagated to the rest of the bulk by heat transfer.[3,7]

In this experiment, a composite of Al_2O_3/Si was made by a reduction/oxidation reaction between aluminum(Al) metal powder and SiO_2 powder by using a simple 2.45 GHz kitchen model microwave oven. In order to make the SHS reaction between aluminum powder and SiO_2 powder, the sample was heated with SiC powder as a suscepting material by using a microwave hybrid heating (MHH) method. Most oxide ceramic materials do not absorb microwave energy well at low temperature, but SiC absorbs the microwave energy and heats up very fast. The sample itself absorbs the microwave energy and converts it to heat effectively when the effective dielectric loss factor becomes high as the temperature of the sample becomes high by the radiating heat from the susceptor.

2. Experimental Procedure

A simple kitchen model microwave oven (LG Electronics: Model MR-M274, 750W) was used in this experiment. Aluminum metal powder (High Purity Chemicals, Co., 99.9%) and SiO_2 powder (Shinyo Pure Chemicals Industries Co., Ltd) were mixed in a 4:3 molar ratio as given in Eq. (3),

$$4Al+3SiO_2 \rightarrow 2Al_2O_3+3Si+\triangle H(\text{exothermic}) \quad (3)$$

All powders were mixed with mortar and pestle and sieved through a 75 μm mesh screen to remove agglomerates. One gram of this mixed powder was pressed in a mold by applying 1 ton pressure and the diameter of the sample was 13 mm. This sample was put in the insulation box where alumina powder was layered at the bottom. The sample was covered with the alumina powder and 10 g of SiC powder was covered on top of the alumina layer. Again alumina powder was layered on top of the SiC powder. After the top of the insulation box was covered, the K-type thermocouple was inserted through the hole at the center of the top cover (Fig. 1). The insulation box with the sample was put in the microwave oven and power was turned on and temperature was monitored with the aluminum shielded K-type thermocouple. After temperature reached 800℃, the thermocouple was taken out and power of the microwave oven was on another 5 minutes. The reacted specimen were cut by half and X-ray diffraction analysis was performed and the SEM micrographs were taken to see the microstructures.

Extra mole of Al(x=2) metal powder were added to increase the strength of the reacted specimen by infiltrating the porous SHS samples with excess liquid aluminum under the same procedure as follows,

$$(4+x)Al+3SiO_2 \rightarrow 2Al_2O_3+3Si+xAl+\triangle H\ (\text{exothermic}) \quad (4)$$

3. Results and Discussion

Full power of the microwave oven was used

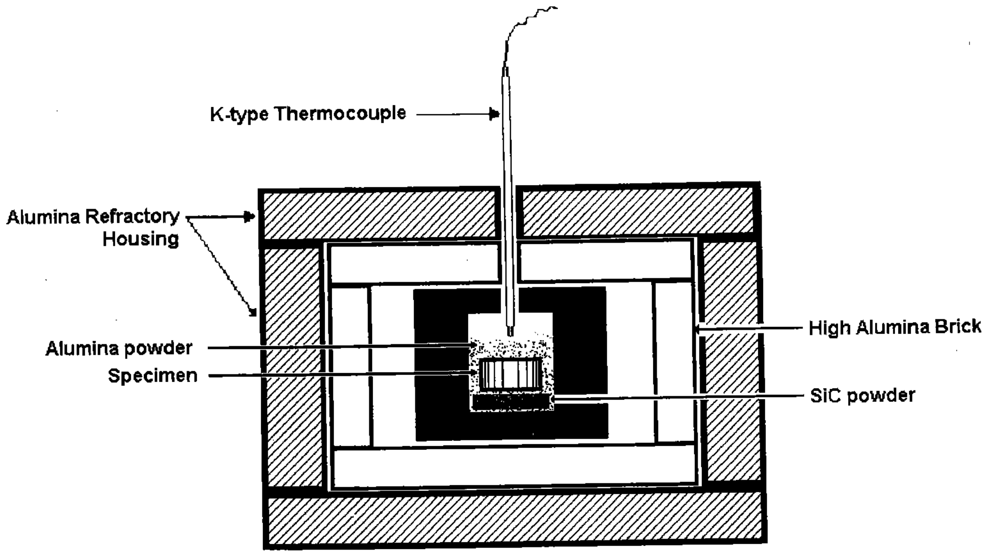

Fig. 1. Schematic Diagram of the insulation box.

in this experiment, and the temperature increase rate was over 100℃/min up to the reaction temperature which was around 850℃ and it jumped to over 200℃/min after the reaction started. The starting temperature of the reaction varied a little around 850℃ and the thermocouple was taken out of the mold after the reaction had started since the K-type thermocouple could not measure the temperature over 1,200℃. After the reduction/oxidation reaction had started, the reaction was continued by SHS reaction and a Si/Al_2O_3 composite was produced from SiO_2/Al powders after the reaction had completed.

The formation of Al_2O_3 and Si was conformed by the XRD analysis of the reacted samples and the XRD patterns were shown in Fig. 3 which were not shown in the XRD patterns of the starting powder. The color of the reacted samples turned black from light gray color of the starting powders and the size of the sample had shrunk as shown in Fig. 2. The reacted samples showed cracks on the surface of the sidewall and were crushed by finger pressure. Hence extra 2 molar ratio of aluminum powder was added in the starting mixture to strengthen the Al_2O_3/Si composite.

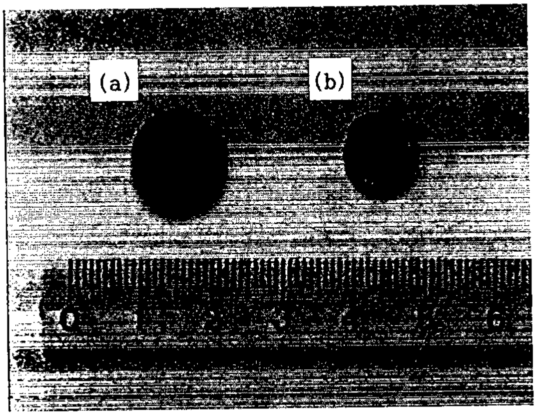

Fig. 2. The size of the samples (x=0). a) before reaction and b) after reaction.

SHS reaction also took place in these samples, but part of the samples did not react under the same experimental conditions. It was assumed that uneven mixing of the additional aluminum metal powders might have affected the reaction under the radiated microwave energy. The reacted samples with x=2 were much stronger than the samples with x=0. The SEM micrograph of the sample with x=2 showed that whiskers were formed inside of the sample during the SHS reaction in Fig. 4. These whiskers are believed to be formed from the aluminum metal and alumina vaporized

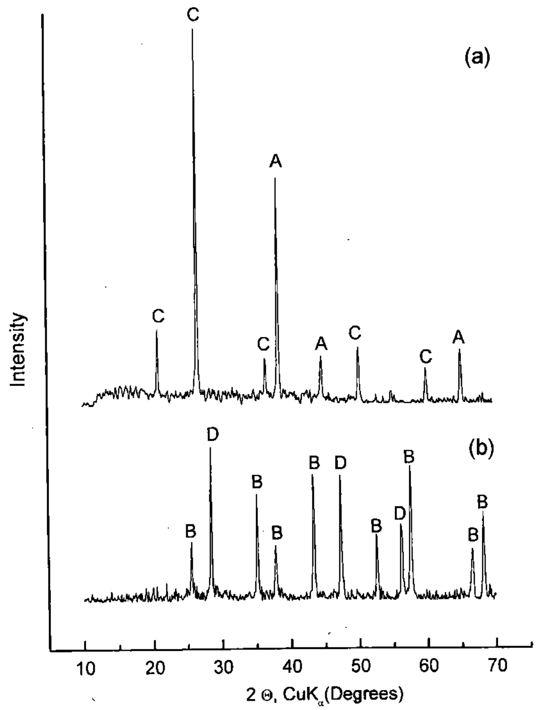

Fig. 3. X-ray diffraction patterns of mixed powders. a) before reaction (A: Al, C: SiO_2) and b) after reaction (B: Al_2O_3, D: Si).

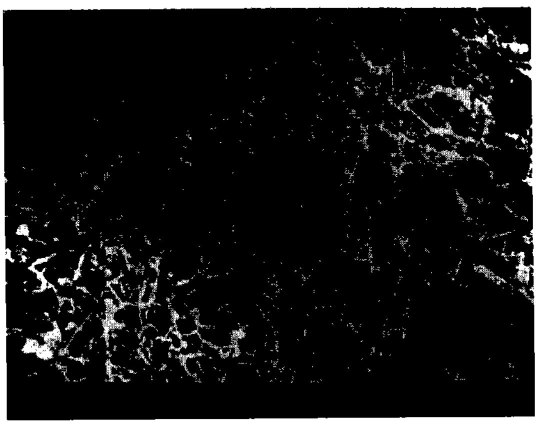

Fig. 4. Microstructure of the reacted sample with *x*=2.

due to the high temperature combustion synthesis process. Whiskers might help to work as a reinforcing source in this ceramic-metal composite.

4. Conclusions

The reduction/oxidation reaction by SHS using a 2.45 GHz microwave energy of a kitchen microwave oven was performed with the mixture of 4:3 molar ratio of aluminum metal powder and SiO_2 powder. A ceramic-metal composite of Al_2O_3/Si was made by applying the MHH method using SiC powders as a suscepting material. Samples with extra 2 molar ratio of aluminum metal powder reacted under the same experimental conditions. The liquid aluminum metal infiltrated the pores of the Al_2O_3/Si composite and increased the strength of the sample, and whiskers were formed in this reaction. Further research is needed to find out the role of the extra aluminum metals and whiskers in this Si/Al_2O_3 composite as a reinforcing source.

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References

1) Clark, D. E., Folz, D. C. and Schulz, R. L., "Recent Developments in the Microwave Processing of Ceramics", *MRS Bull.*, 41-46, Nov. (1993).
2) Lewis, David A., LaMaire, S. J. and Viehbeck, Alfred, "Microwave Processing of Polyyimide Thin Films", Microwaves: Theory and Application in Materials Processing III, *Ceram. Trans.*, **59**, 433-439 (1995).
3) Willert-Porada, M., Fisher, B. and Gerdes, T., "Application of microwave heating to combustion synthesis and sintering of Al_2O_3-TiC ceramics", *Ceram. Trans.*, **36**, 365-375 (1993).
4) Munir, Z. A., "Synthesis of high temperature materials by self-propagation combustion", *Amer. Ceram. Soc. Bull.*, **67**(2), 342-24 (1988).
5) Sheppard, L. M., *Amer. Ceram. Soc. Bull.*, **68**(6), 1165-1167 (1989).
6) Ahmad, I., Dalton, R. and Clark, D., *J. of Microwave Power and Electromagnetic Energy*, **26**(3), 128-138 (1991).
7) Bescher, Eric P., Sarkar, Urmimala and Mackenzie, John D., "Microwave Processing of Aluminum-Silicon Carbide Cermets", *MRS Sym. Proceedings*, **269**, 371-378 (1992).