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Full densification of molybdenum powders and multilayer materials obtained by Spark Plasma Sintering

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Abstract. Molybdenum powders with two different particle sizes were sintered using the Spark Plasma Sintering (SPS) process. A near to 1 density are obtained in a few minutes. A Molybdenum/Aluminum Nitride multilayer was obtained while sintering in one step, using the SPS process.

Introduction

The conventional package of power module contains different type of materials such as semiconductors dies, metallized ceramic substrate, interconnection provided by wire bonding.... Etc [1]. Here we focus on a specific part of the package, namely the metallized ceramic substrate. The most substrate used in conventional package is the Direct Bonded Copper (DBC). The DBC is usually formed by a ceramic layer (Aluminum oxide Al_2O_3) between two copper layers.

In regard of power module's operation, the whole package is subjected to a thermal cycling (ranging from $-55\text{ }^\circ\text{C}$ to $200\text{ }^\circ\text{C}$). By repeating this thermal variation, it generates a mechanical stress at the interface between two different materials with vastly different Coefficients of Thermal Expansion (CTE). Moreover, the DBC leads to a high differential stress due to the materials CTE mismatch ($17 \times 10^{-6}/^\circ\text{C}$ for Cu and $7.2 \times 10^{-6}/^\circ\text{C}$ for Al_2O_3), that may cause failure at high temperature [2].

Silicon Carbide (SiC) is one of the promising candidates as a semiconductor device that can operate at much higher temperature ($>200\text{ }^\circ\text{C}$). It has a CTE equal to $3.7 \times 10^{-6}/^\circ\text{C}$. In order to avoid the problem described above, a new structure of metallized ceramic substrate needs to be realized using materials that has adequate CTE and very close to that of SiC. Molybdenum (Mo) and Aluminum nitride (AlN) were found to be the promising candidates for the new metallized ceramic substrate, with a CTE of $4.8 \times 10^{-6}/^\circ\text{C}$ and $4.1 \times 10^{-6}/^\circ\text{C}$ respectively.

The work on Mo/AlN based substrate was recently reported using the Spark Plasma Sintering (SPS) process to sinter Mo powder alone or with a disk of AlN [3,4]. In this paper, we will present the sintering of two Mo powders with two different particle sizes, and then we will present some experiment on the Mo/AlN/Mo multilayer co-sintering in one step.

Experimental Procedure

Commercial powders (Goodfellow, Cambridge, UK) were used for SPS consolidation:

- Two molybdenum powders with a purity of 99.99+% with different particle sizes:
 - The first with a range of 55-355 μm (cf. Fig. 1 a)
 - The second with an average particle size of 2 μm ((cf. Fig. 1 b)
- Aluminum nitride powder with a purity of 99.5+% with an average particle size of 1.4 μm (cf. Fig. 1 c).

The experiments were carried out in a SPS FCT systeme GmbH, HP D 25 model.

Our study has followed two-step: (i) the first was about consolidating both molybdenum powders separately, (ii) and the second consist of co-sintering Mo/AlN/Mo powders in one step. For both steps, powders were consolidated using a cylindrical graphite die, with an inner-diameter of 20 mm, under a vacuum (10^{-2} Torr), without any additive. Several sintering parameters were fixed such as the holding pressure of 77 MPa, a holding time of 5 min, a heating rate of 700 °C/min.

Precise density measurements of the consolidated bulk molybdenum samples were made using the Archimedes' principle according to ASTM Standard B328-94. The relative density was calculated based on the theoretical density of Mo (10.22 g/cm^3).

At the end of each experiment, the samples were cross-sectioned, mounted in a hot mounting resin and then prepared metallographically (mechanical grinding and polishing down to $0.06 \mu\text{m}$ silica slurry). The samples were examined using a scanning electron microscopy (SEM). Energy dispersive X-Ray spectroscopy (EDX) was used to identify the interface materials reaction.

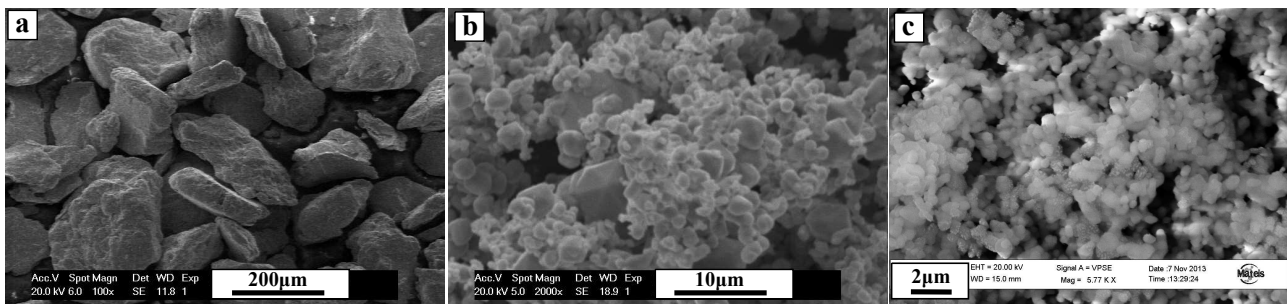


Fig. 1 SEM image of the commercial powders: molybdenum powders (a) with particle size range of 55-355 μm (b) with average particle size of 2 μm ; (c) aluminum nitride powder with average particle size of 1.4 μm

Mo powders sintering

As we mentioned before, the first step was to study the sintering of two molybdenum powders with two different particle sizes. Fig. 2 shows the relative density in function of sintering temperature, ranging from 1200 °C to 1950 °C. For both molybdenum powders, we see that the density increases as the sintering temperature increases:

- 84.4% to 99.5% for Mo powders with particle size of 55-355 μm
- 92.7% to 100% for Mo powders with average particle size of 2 μm

We can see that decreasing the particle size enhances the consolidation's kinetic and so, it results in increasing the relative density for the molybdenum powder with an average particle size of 2 μm . This is in good agreement with Herring's scaling Law [5].

Mo/AlN/Mo co-sintering multilayer

The second step was the co-sintering of molybdenum and Aluminum nitride powders in a single step. Mo powder with an average particle size of 2 μm has been chosen for different reason: (i) first sintering two powders with a close particle size and (ii) second, a satisfying relative density was obtained for Mo powder even at low temperature.

The sintering of AlN powders has been reported, whether with or without additives, using SPS technic [6,7]. In this study, Mo/AlN/Mo powders were sintered in one step, without any additives for both powders. Three different temperatures have been tested, with a large holding of 20 min in order to enhance the density at low temperature, and a holding pressure of 77 MPa.

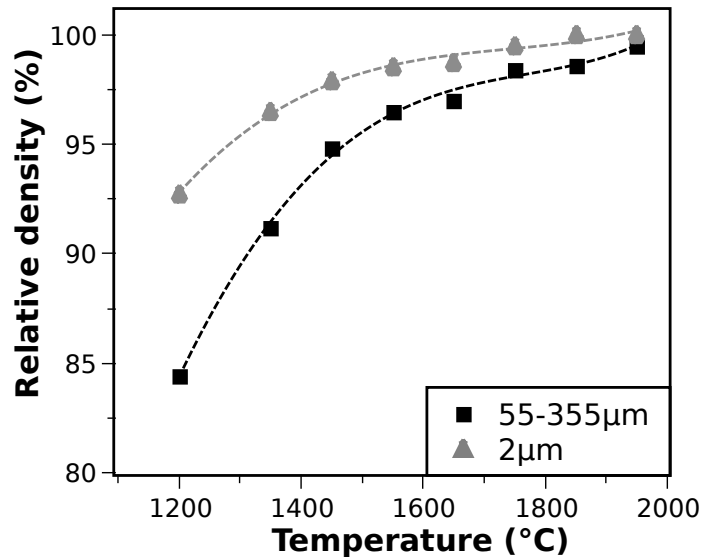


Fig. 2 Graph of the relative density in function of sintering temperature for the two molybdenum powders

Fig. 3 shows the cross-sectional SEM photos of AlN (upper photos) and Mo/AlN interfaces (lower photos) for the three sintering temperature. At 1450 °C, Mo/AlN's bulk presents pores at its microstructure and a chemical reaction were produced at the interface between Mo and AlN as we can see in Fig. 3.a. While increasing the temperature, it improves the assembly by decreasing the pores and makes a full-densified microstructure, although the presence of the chemical reaction at the interfaces, as we can see in Fig. 3.b & c. A good bond was obtained for all samples sintered at different temperature, even this unknown layer of ~20 µm-thick at Mo/AlN interface.

In order to reveal the chemical composition of the unknown layer, an EDX line's characterization was performed for all the samples, beginning from the Mo and end up at the AlN. The x-marks level were found to contain molybdenum with a count lesser than the pure one, without any of {Al, N or O} elements. Another evidence for this unknown layer is that it appears to be in a different contrast in regard of pure Mo, while taking a back-scattered SEM photos (cf. Fig. 3.c).

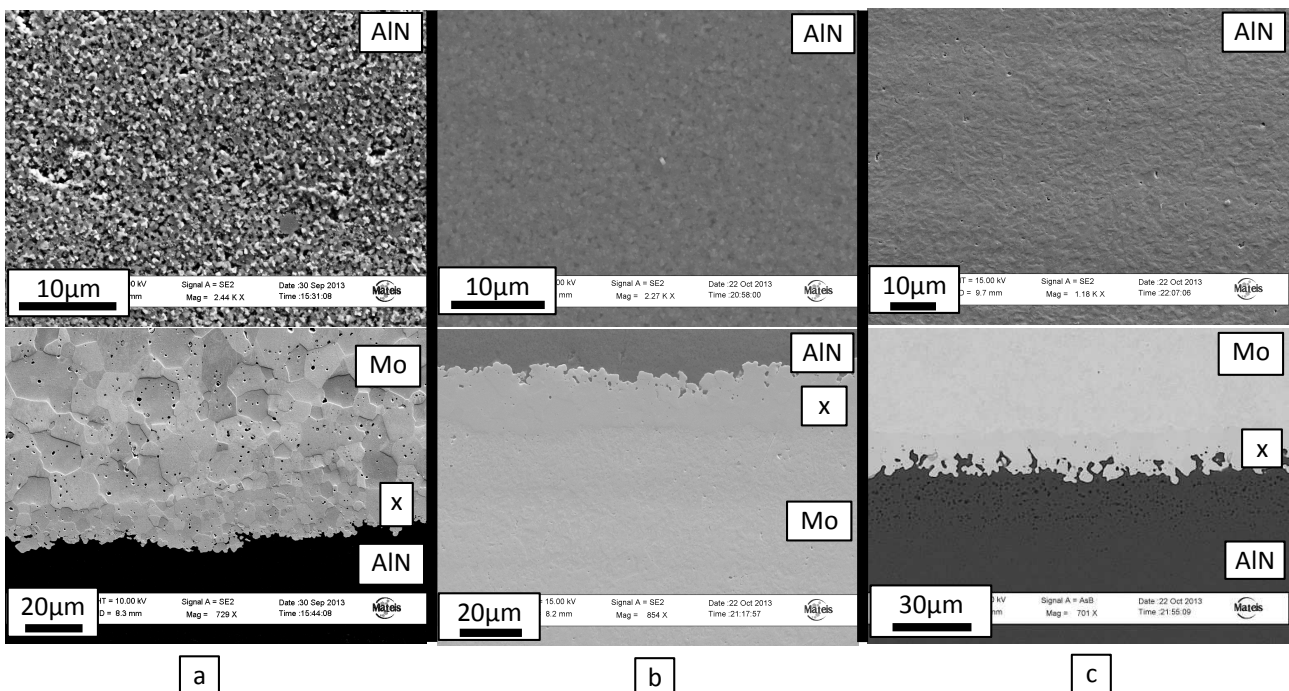


Fig. 3 Cross-sectional SEM photos of AlN (upper photos) and Mo/AlN interfaces (lower photos) for a co-sintering at (a) 1450 °C, (b) 1550 °C and (c) 1650 °C

Discussion

SPS allows for molybdenum sintering at “low” temperature (much lower than the melting point of molybdenum, at 2623 °C), with a high density (up to 100%). Decreasing the initial particle size of molybdenum enhance the relative density and allows to get a satisfied density at lower temperature.

Using the same equipment, a bond between molybdenum and aluminum nitride was successfully achieved, although the unknown layer at the interface of Mo/AlN. A more precise characterization for this layer must be conducted in order to get a better insight of the phenomena that have taken place. Moreover, a bending test will now be carried out to ensure that the bonding strength is good enough for high-temperature applications. At last but not least an electrical characterization (dielectric strength) is required as well as a thermal cycling to study the reliability of the substrate.

Summary

Two molybdenum powders with two different particle sizes were sintered to bulk using a Spark Plasma Sintering process. This process allows achieving full densification in a few minutes. Moreover, a molybdenum/Aluminum nitride multilayers was obtained while sintering in one step using the SPS, and results a good bond between the materials, although the presence of a chemical reaction at the interface.

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