*Transactions of the Materials Research Society of Japan* 29 [8] 3495-3498 (2004)

# Application of SPD/PM Process to Produce a TiC/$Ti_5Si_3$ Nano Grained Composite

N. Miyano, H. Iwasa*, K. Isonishi** and K. Ameyama***
Research Institute for Applied Mechanics, Kyushu Univ., Japan
Fax: +81-92-583-7758, e-mail: miyano@riam.kyushu-u.ac.jp
*Graduate School, Ritsumeikan Univ., Japan
**Faculty of Education, Shiga Univ., Hikone, Shiga, Japan
Fax: 81-77-537-7805, e-mail: isonishi@sue.shiga-u.ac.jp
***Faculty of Science and Engineering, Ritsumeikan Univ., Japan
Fax: 81-77-561-2756, e-mail: ameyama@se.ritsumei.ac.jp

SPD/PM (Severe Plastic Deformation / Powder Metallurgy) Process is one of the most effective means for producing non-equilibrium phase. Mechanical Alloying (MA) is one of such a SPD/PM process and it often realizes an amorphous phase formation as well as nano grained microstructure. We have applied the MA process to produce a TiC/$Ti_5Si_3$ nano composite for the micro-parts. Powders of elements Ti and SiC whose composition is Ti-20mass%SiC are blended for MA, and the MA powder whose average particle size is 20~30 μm, has an amorphous structure. The MA powder is filled into a micro-mold produced by LIGA process, and cast together by SPS (Spark Plasma Sintering) in order to fabricate new micro-parts as a transcription of the micro-pats by LIGA process. As a result, this process proved successful for making the micro-parts made by the TiC/ $Ti_5Si_3$ composite.
Key words: Mechanical alloying, Non-equilibrium, Superplasticity, LIGA, molding

## 1. INTRODUCTION

One of the authors proposed a non-equilibrium powder metallurgy (PM) process, which enables to fabricate a near net-shape product [1-3]. The non-equilibrium PM process combined with mechanical alloying (MA) [4] and sintering, such as hot pressing (HP) or spark plasma sintering (SPS), realizes to fabricate a ceramics parts at low temperature and pressure for the sintering. The concept of the process is shown in Fig. 1. Two kinds of powders, conventional forming and MA powders, were sintered at elevated temperatures to become ceramics composites. Because the MA powder has an amorphous structure[1-3], new phases and nano-size grain were nucleated during the sintering. As a result, the composites of the MA powder kept an equiaxial grain structure and indicated an extremely low stress deformation due to the grain boundary sliding and/or the stress relaxation by formation of new phase, under some specific condition of temperatures and strain rates. In another word, the deformation is progressed by a pseud-superplasticity.

In the present study, we apply the non-equilibrium PM (MA/SPS) process to fabricate TiC/$Ti_5Si_3$ ceramics composites for micro size parts (called micro-parts). The ceramic composites are suitable as a material for micro electro mechanical system (MEMS) parts in terms of the mechanical and physical properties. However, there are obstructions of processing for the ceramic micro-parts. Therefore, the non-equilibrium PM process is effective to fabricate a micro-parts made of the ceramic composites since it shows a superior formability.

## 2. EXPERIMENTAL PROCEDURE

The starting materials were commercially pure Ti (average particle size: 45μm) and SiC (9.4μm) powders.

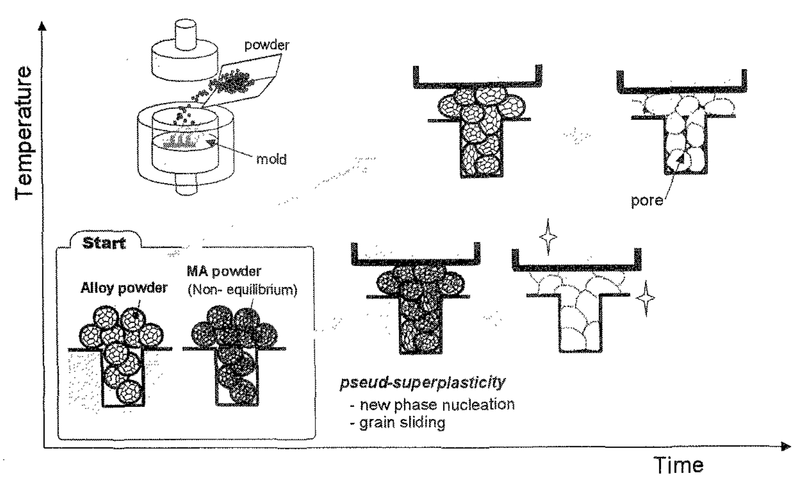


Fig. 1 Schematic illustration of the conventional PM process and the non- equilibrium PM process

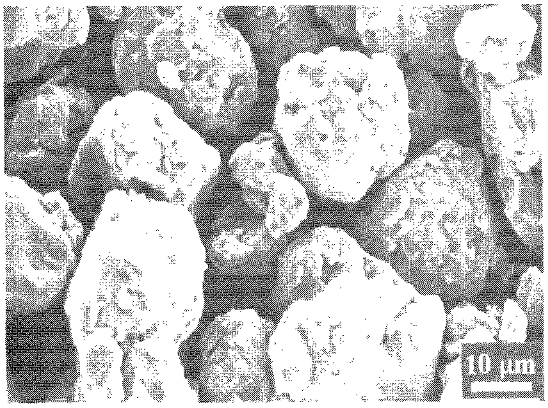


Fig. 2 SEM image of the (Ti+SiC) MA powder after 360 ks MA.

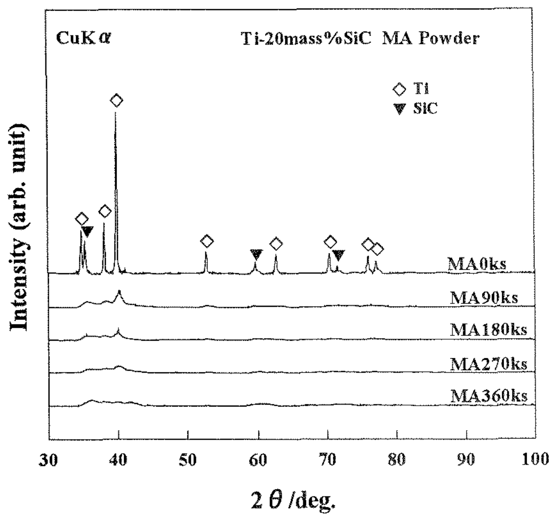


Fig. 3 XRD patterns of the MA powder milled for 0, 90, 180, 270 and 360 ks.

They were mixed to the composition of a Ti-20 mass% SiC, and mechanically alloyed by a planetary ball mill conducted at a rotating speed of 250 rpm for 360 ks with a SKD11 vial and SUS304 balls under an Ar gas atmosphere. The powder to ball weight ratio was 1 : 7.2. During the milling, 1.0 mass% of n-Heptane was added as a process control agent.

The MA powders were sintered by SPS and a vacuum hot press (VHP). The SPS was carried out at 773 K for 3.6 ks, under a pressure of 50 MPa and at a heating rate of 1.7 $Ks^{-1}$, followed by furnace cooling. The VHP was carried out at 753 K for 10.8 ks, under a pressure of 200 MPa and at a heating rate of 0.5 $Ks^{-1}$, followed by furnace cooling. The SPS and VHP compacts were cut to $\phi$3 mm x 4 mm column shape and were compression tested at temperature range between 773 K and 1073 K, at an initial strain rate of 4.2 x $10^{-4}$ $s^{-1}$. The MA powders and the compacts were examined by means of XRD, SEM and TEM.

## 3. RESULTS AND DISCUSSION

### 3.1 MA powder

Average particle size of the milled powders were approximately 20~30 μm, as shown in Fig. 2. The

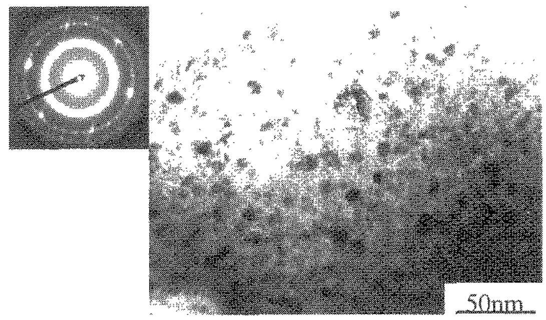


Fig. 4 TEM micrograph and a selected area diffraction pattern of the (Ti+SiC) MA powders after 360 ks milling.

Microstructural change during milling was examined by means of XRD analysis of the MA powders. Fig. 3 shows XRD patterns of the powder mechanically alloyed for 0 ~ 360 ks. Substantial broadening of the XRD peaks of the original species took place with the progress of milling in the powders. This indicates that the MA powders have an amorphous-like structure and/or an ultra fine grain structure. Fig. 4 shows a TEM micrograph and SADP (selected area diffraction pattern) of the Ti-20mass%SiC MA powder. The SADP indicates that the MA powder consisted of an amorphous phase and ultra fine grains of the average size of 10 nm.

Fig. 5 shows stress-strain curves of the specimens compression-tested at an initial strain rate of 4.1 x $10^{-4}$ at 773 K, 973 K and 1073 K. The constituent phases determined by XRD were also indicated below the figure. The flow stress at 1073 K was twice as high as that at 973 K. This is unusual as a high temperature deformation property. A TiC was the final phase at 973 K deformation, while $Ti_5Si_3$ phase appeared and the

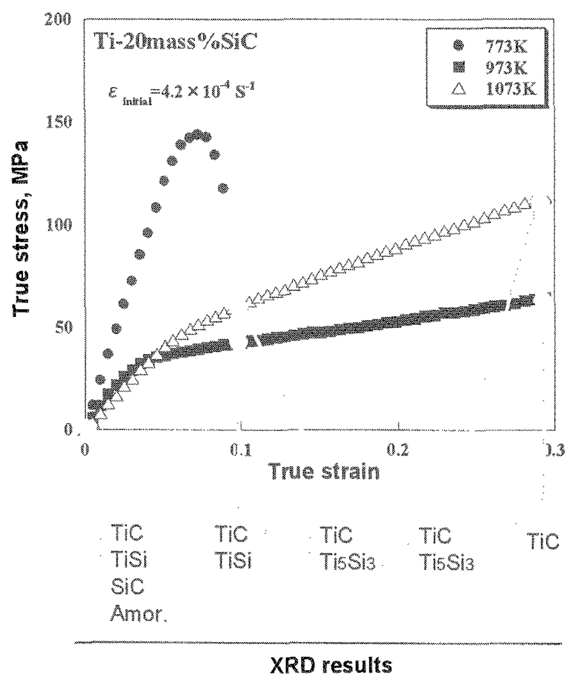


Fig. 5 Stress-strain curve for compression test at an initial strain rate of 4.2 x $10^{-4}$.

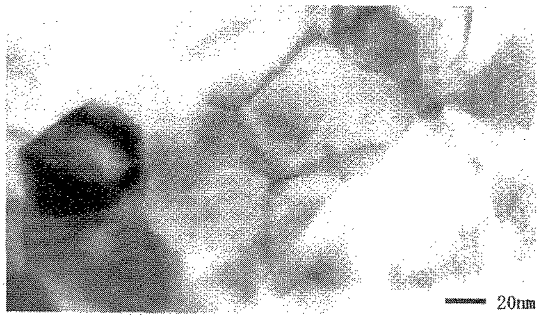


Fig. 6 TEM micrographs of the specimen compressed to 30 % at the initial strain rate of 4.2 x $10^{-4}$ $s^{-1}$ at 973 K

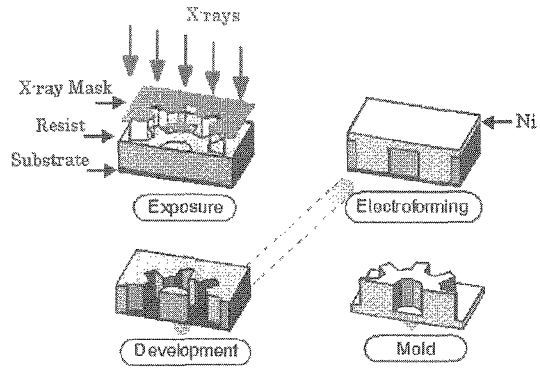


Fig. 6 A schematic drawing of the LIGA process

($TiC+Ti_5Si_3$) was the final structure at 1073 K deformation. The density of the compact after deformation at 973 K was superior to that at 1073 K; 83.2 % at 973 K and 81.1 % at 1073 K after compression test of e (true strain) = 0.3. The microstructure of the compact of 973K was examined by TEM, as shown in Fig. 6. From the figure, the specimens consisted of an equiaxial grain structure with an average grain size of 28 nm. Note that the compact shows no dislocation structure and have clear grain boundaries.

These results of the TEM observation and the XRD analysis strongly suggest that crystallization of an amorphous phase to a TiC phase and maintenance of a fine TiC grain structure decrease the deformation temperature as well as the deformation stress. The microstructure with no dislocation even after a large deformation was similar to that of a superplastically deformed material. From such a microstructural feature, it can be expressed as a *pseud-superplastic* deformation.

3.2 MICRO-MOLDING

The pseud-superplasticity enables to improve the formability of micro-parts. The MA powder was sintered by SPS with a Ni mold produced by a LIGA process. The LIGA process can fabricate micro-parts with a high aspect ratio because of the high directivity X-rays from the synchrotron radiation (SR) source [5,6]. The process is shown in Fig. 7. The details of the LIGA process are omitted from this paper (see ref. [6] and [7] for detail). The results of the sintering are

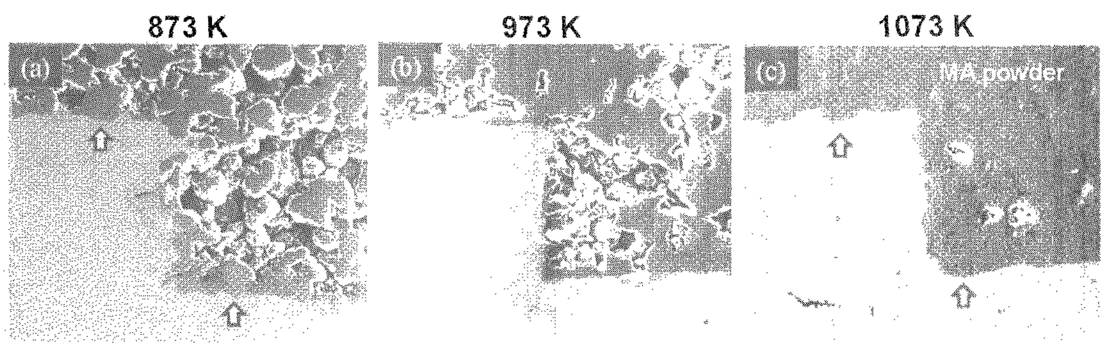


Fig. 8 SEM images of the cross section of the compacts sintered by SPS at various temperatures of the MA powder .

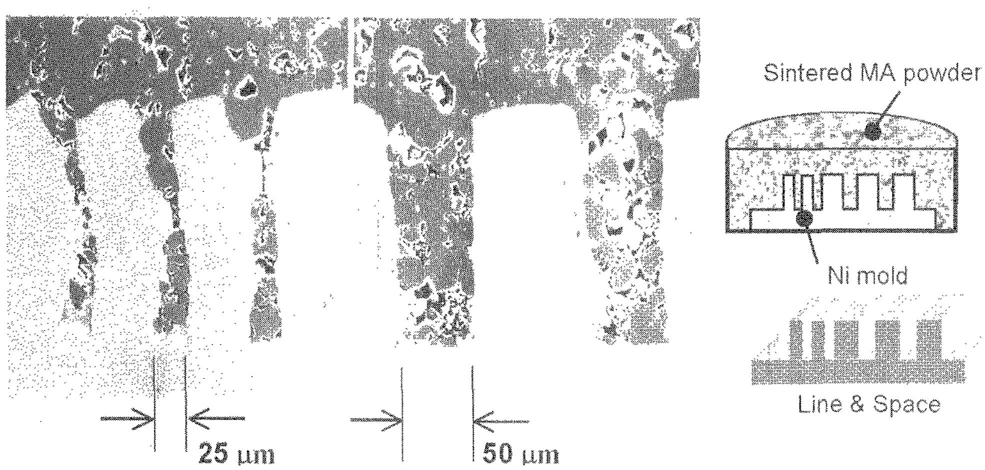


Fig. 9 SEM images of the cross section of the SPS compacts sintered at 1023K using line & space Ni mold.

shown in Figs. 8(a)-(c). These SEM images indicate a cross section of the each SPS compacts. The MA powder become the $TiC/Ti_5Si_3$ ceramic composites with rise of the sintered temperature. However, at 873 K in Fig. 8(a) and 1073 K in Fig. 8(c), the Ni molds were deformed wavy at interface between the Ni mold and the MA powder. This was caused by damage from the MA particle as indicated by arrows in the figures, because the MA particle was harder than the Ni mold. Nevertheless, there is no deformation of the Ni mold sintered at 973 K in Fig. 8(b). This was resulted from the pseudo-superplasticity, as the mentioned above. The phenomenon works as good effect for high densification and accuracy of making micro-parts. Fig. 9 demonstrates an appearance of the micro-parts produced by the SPS at 1053 K, using a line & space shape of Ni mold, whose slits are 25 μm and 50 μm in width, respectively. In both figures, the density of the compacts was not enough. The MA powder whose particle size was about 20~30 μm was filled in the slit of 50 μm in width, however, the slit of 20 μm was deformed. Optimization of the pseudo-superplasticity in the near future will enhance the usage of a $TiC/Ti_5Si_3$ ceramics composite for the micro-parts.

## 4. CONCLUSIONS

The non-equilibrium PM processing such as an MA/SPS process was examined in a Ti-20 mass% SiC materials systems. The results are as follows:

(1) The MA powder consisted of an amorphous and an ultra fine grain structure.

(2) Unusual high temperature deformation behavior was observed in Ti-20 mass% SiC compacts because of their metastable phase structure.

(3) Not only maintaining a fine grain structure but also avoiding a $Ti_5Si_3$ phase formation during hot deformation resulted in a pseudo-superplasticity, and thus the flow stress decreased. These results were attributed to the microstructural change during the deformation.

(4) The MA/SPS process was able to be applied to a Ni mold LIGA process. Optimization of the pseud-superplasticity realized to fabricate molds made of fine ceramics composites such a $TiC/Ti_5Si_3$.

## REFERENCES

[1] Y. Suehiro and K. Ameyama, Journal of Materials Processing Technology, 111, 118-121 (2001).

[2] K. Tagaya, Y. Suehiro and K. Ameyama, Proc. Fourth Pacific Rim Int. Conf. on Advanced Materials and Processing, JIM, 959-962 (2001).

[3] T. Namura, N. Nagumo, Y. Matsumoto, N. Sasaki and K. Ameyama, Mater. Sci. Forum 233, 251-260 (1997).

[4] J. S. Benjamin: Metal Trans, 1, 2943-2951 (1970).

[5] P. Bley and J. Mohr, FED Journal, Vol.5 Suppl. 1, 34-48 (1994).

[6] N. Miyano, K. Tagaya, K. Kawase, K. Ameyama and S. Sugiyama: Journal of JSMS, 52, 783-788 (2003).

[7] S. Sugiyama, Y. Zhang, H. Ueno, M. Hosaka, T. Fujimoto, R. Maeda R, T. Tanaka, MHS, 79-84 (1996).

(Received December 1, 2003; Accepted July 22, 2004)