

## **EFFECT OF TiB<sub>2</sub> ADDITION ON ALUMINUM ALLOY SINTERED BY SPS**

\*Y. Komiya, K. Negishi, and H. Izui

*Nihon University*

*Funabashi, Chiba, Japan*

*(\*Corresponding author: komiya.y@aero.cst.nihon-u.ac.jp)*

### **ABSTRACT**

Aluminum has the disadvantage of low strength. In order to obtain the high strength composites, crystals were refined in the composites. Grain refinement was approached at two process points, generating new grains and grain growth. It is important for refinement how to control the grain growth. Spark plasma sintering (SPS) technique and TiB<sub>2</sub> powder addition prevent grain growth. To investigate effects of strain and additives to the alloy, aluminum powder was sintered at 70 MPa and 80 MPa, changing the content of TiB<sub>2</sub> additive powder by SPS method. As the results, the crystal grain size was finer in the composite sintered at higher sintering pressure and with larger amount of additive, though over 1.0mass% TiB<sub>2</sub> addition dissuades grain refining. These grain refinements improve hardness and tensile strength of sintered aluminum alloys.

### **KEYWORDS**

Aluminum matrix composite, TiB<sub>2</sub>, Grain refinement, Spark plasma sintering, Hardness, Tensile strength

## INTRODUCTION

Aluminum matrix composites have low density, high specific strength, and low cost. Their applications are growing continuously in the field of automotive and aerospace industries. Recent studies to improve their good mechanical and tribological properties use different ceramic materials such as  $\text{Al}_2\text{O}_3$  (Das, Munroe, & Bandyopadhyay, 1996; Ashwatha & Anthony Xavier, 2014),  $\text{SiC}$  (Hassani, Bagherpour, & Qods, 2014),  $\text{B}_4\text{C}$  (Ehsan, Masoud, Touradj, Amir Hossein, & Ali, 2015),  $\text{TiC}$  (Ehsan, Ali, Masoud, Kamyar, & Touradj, 2017), and  $\text{TiB}_2$  (Fei & Zhong-Hua, 2013). On the other hand, grain refinement is also an important techniques for improving the properties of aluminum alloys. Finer grains improve the mechanical properties of the alloy. For cast and wrought alloys, addition of grain refiners in molten alloys accelerates generation of nucleus and suppresses grain growth. Al-Ti-B alloy is generally added as grain refiners in the molten aluminum alloys (Easton & Stjohn, 2001). Especially,  $\text{TiB}_2$  has very high melting point, low density, and excellent hardness. Therefore,  $\text{TiB}_2$  was selected as additions into aluminum alloys. Liquids reaction leads to high cost, oxidation, discoloration of molten alloy, and clustering of additive powder as compared with solid reaction such as sintering. However, sintering of aluminum alloy powder with ceramic powders is difficult for oxide layer surface. Recently, we take attention to spark plasma sintering (SPS) as advanced sintering process. SPS process can easily sinter a high quality specimen at a lower sintering temperature and in a shorter time than conventional sintering processes. By SPS process, we have sintered titanium powders that are as difficult to be sintered by any other processes as aluminum powders, and improved mechanical properties by adding hard ceramic powders  $\text{TiC}$  and  $\text{TiB}$  (Kamegawa, Izui, Komiya, Kobayashi, & Arimoto, 2015; Sampei, Izui, Komiya, Shigimura, & Suzuki, 2015). Furthermore, it is important to consider not only pure aluminum but alloys for solid-solution or precipitation strengthening. Actually, pure Al does not have enough strength compared with the alloys. Al-Cu alloy and Al-Mg alloy demonstrate adequate strength for industrial and commercial sectors. Al-Mg alloy retains good formability, and has high resistance to corrosion. It is therefore used for many applications. We must consider alloying processes. The most cost effective process is blended elemental technology where alloying elements are added to base powder as elemental or master alloy powder (Smugeresky & Dawson, 1981; Yasue, Radjai, Miwa, & Sakaguchi, 2003; Bolzoni, Esteban, Ruiz-Navas, & Gordo, 2012).

In order to improve the strength of sintered aluminum alloy, in this study, sintered aluminum alloy matrix composites with  $\text{TiB}_2$  powder were fabricated by SPS using blended element powder. In addition, the effect of the amount of  $\text{TiB}_2$  powder and sintering pressure on microstructure and mechanical properties was investigated.

## EXPERIMENTAL

As matrix of the composites, we used the commercial 99.9% pure aluminum powder (particle size 106–180  $\mu\text{m}$  from Kojundo Chemical Laboratory Co., Ltd., Japan) and 99.98% pure magnesium powder (average size 60  $\mu\text{m}$  from SFM SA, Swiss) in this study. An additional powder is  $\text{TiB}_2$  (average size 1.81  $\mu\text{m}$  from Japan New Metals Co., Ltd., Japan). The corresponding morphologies are shown in Figure 1.

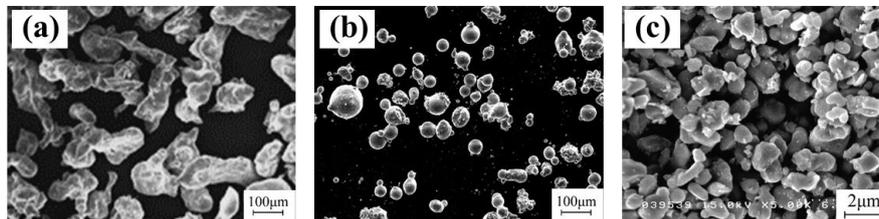


Figure 1. SEM micrographs of (a) pure-Al powder, (b) pure-Mg powder, and (c)  $\text{TiB}_2$  powder

The mixtures of powders were with a planetary ball-mill (P-6, Fritsch Japan Co., Ltd., Japan) at a rotation speed of 200 rpm for 10 min in air. Pre-compacted pressure was 20 MPa for 15 s before setting in the SPS chamber. We sintered these powders in a graphite die (Toyo Tanso Co., Ltd., Japan) in vacuum using an SPS system (Dr. Sinter, SPS-3.20IV, Sumitomo Coal Mining Co., Ltd., Japan). The heating rate was 50 K/min. The condition of sintering was at 773 K under a pressure of 70 MPa or 80 MPa for 10 min, respectively. It was necessary to carry out homogenization at 673 K for 6 h in air furnace for Al-Mg alloy based composites to diffuse Al and Mg.

Phase compositions of the composites were analyzed using an X-ray diffractometer system (Rint 2000, Rigaku Corporation, Japan) with Cu-K $\alpha$  radiation at 50 kV and 100 mA. The relative densities of the sintered specimen were measured by the Archimedeian method. Vickers microhardness tests of these specimens were carried out using a microhardness tester (HVM-2(T), Shimadzu Corporation, Japan). The microstructures of sintered samples were observed using optical microscope (OM, DSX510, Olympus Corporation, Japan) and scanning electron microscope (SEM) with energy dispersive X-ray analysis (EDX, SUPERSCAN SSX-550, Shimadzu Corporation, Japan) by standard metallographic methods. Grain size was measured using ImageJ software. Tensile tests of the composites were carried out using an INSTRON machine (MODEL1125). The crosshead-head speed was 0.5 mm/min, and the tests were conducted at room temperature.

## RESULTS AND DISCUSSION

For investigating an existence of the reaction during sintering and homogenization process, we carried out XRD analysis about sintered Al-2.5mass%Mg alloy with homogenization and Al-2.5mass%Mg-1.0mass%TiB<sub>2</sub> composite with homogenization. As the result in Figure 2, XRD patterns show only Al phase peaks and TiB<sub>2</sub> phase peaks. It has been decided that the powders were sintered without significant interfacial reactions as any other composites sintered in this study, even with addition of Mg and TiB<sub>2</sub> powder after homogenization at 673 K. However, it is necessary for details of the new compounds formation to observe the microstructures.

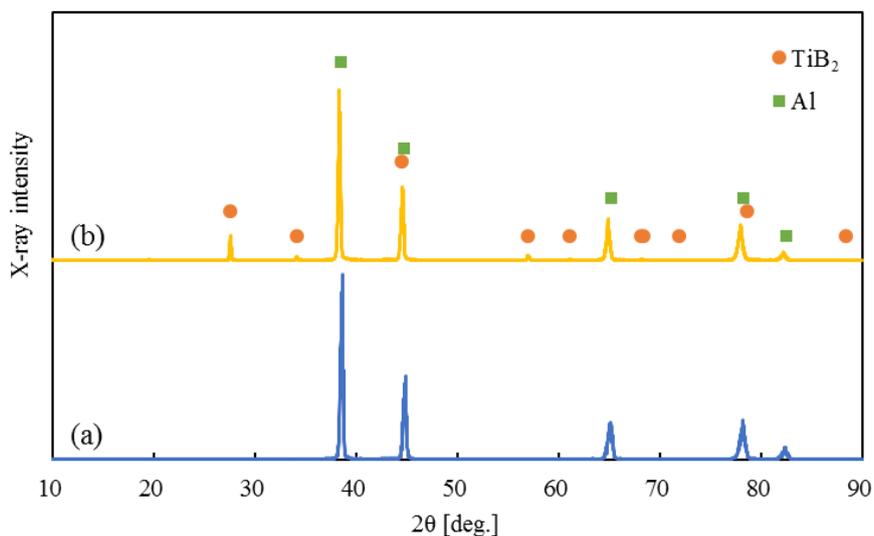


Figure 2. XRD patterns of (a) Al-2.5mass%Mg alloy with homogenization and (b) Al-2.5mass%Mg-1.0mass%TiB<sub>2</sub> composite with homogenization

As shown in Figure 3, relative densities of the sintered composites were measured. From this result, even if the additive amount was increased, the relative density of all the composites was not less

than 99% in this study. Therefore, it has been decided to obtain highly dense composites by SPS. This value slightly decreases as additive amount increase. Melting point of  $TiB_2$  is much higher than sintering temperature 773 K. Therefore, clustering of  $TiB_2$  powders occurs among aluminum powders and cavities increase in the composites. Al-Mg alloy matrix composites with homogenization have a lower density after diffusion of Mg in Al. The lattice constant of Al increases with the increase of Mg concentration (Uesugi & Higashi, 2013). It has been decided that Mg solid-solution in Al indicates lower density, and Mg solution decreases with the increase of  $TiB_2$  additive amount. Mg solution behavior can also be seen in Figure 4, which shows EDS Mg mappings of Al-Mg- $TiB_2$  composites. Actually, the distribution of Mg spreads for homogenization, but shrinks for  $TiB_2$  addition.

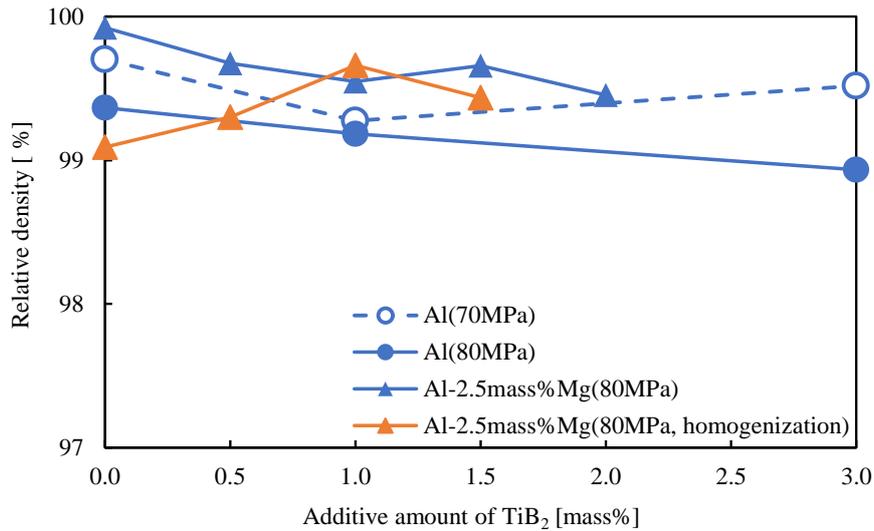


Figure 3. Relative density of composites as a function of  $TiB_2$  additive amount

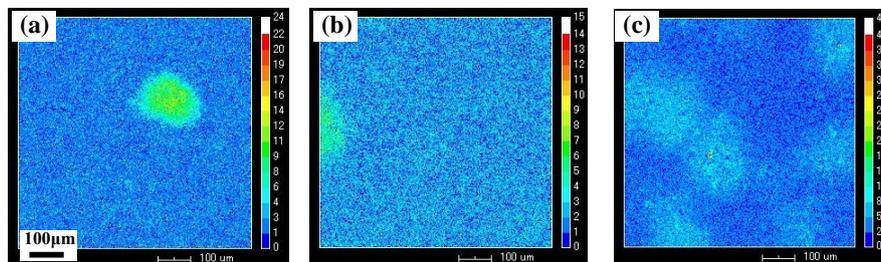


Figure 4. EDS Mg mapping of the (a) Al-2.5mass%Mg alloy without homogenization, (b) Al-2.5mass%Mg alloy with homogenization, and (c) Al-2.5mass%Mg-1.0mass% $TiB_2$  composite with homogenization

Using OM images such as in Figure 5, average grain size of the sintered composites was measured. The microstructures in Figure 5 show grain boundaries etched by hydrofluoric acid in the sintered powder. As the result of average grain size measurements in Figure 6, grains were made smaller by higher sintering compression, 2.5mass% Mg addition, or slightly  $TiB_2$  addition. Increase of strain energy by the high pressure or different kind powder addition causes grains fine during sintering process. However, addition of over 1.0 mass%  $TiB_2$  dissuades grain refining and accelerates grain growth. Furthermore, homogenization at 673 K is also one factor of the grain growth, as shown in Figure 6.

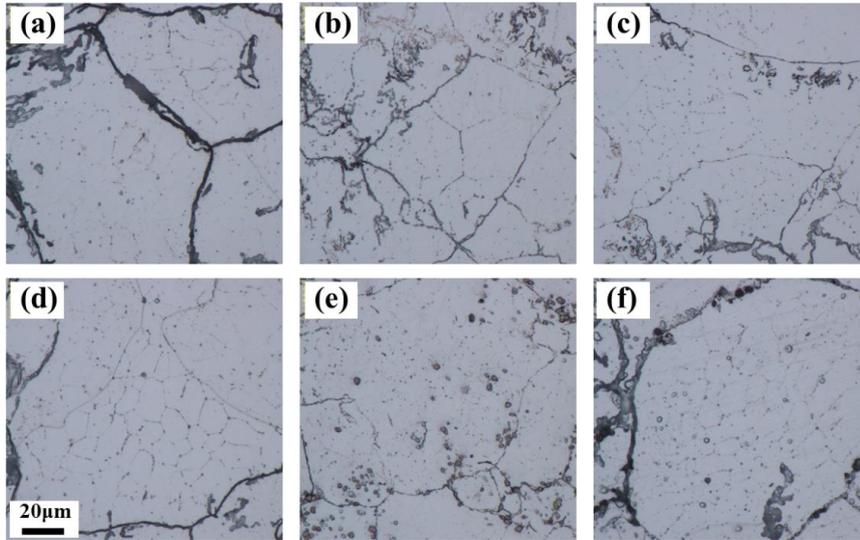


Figure 5. OM images of (a) pure-Al, (b) Al-2.5mass%Mg alloy without homogenization, (c) Al-2.5mass%Mg alloy with homogenization, (d) Al-1.0mass%TiB<sub>2</sub> composite, (e) Al-2.5mass%Mg-1.0mass%TiB<sub>2</sub> composite without homogenization, and (f) Al-2.5mass%Mg-1.0mass%TiB<sub>2</sub> composite with homogenization

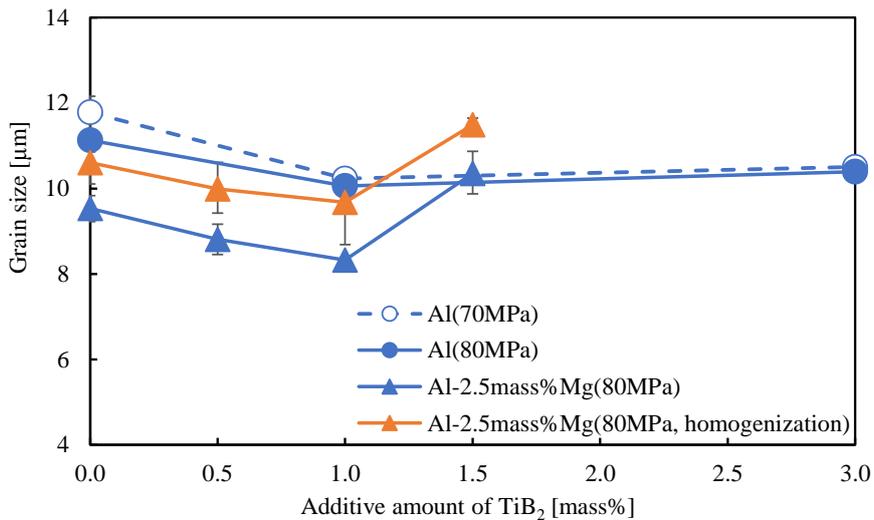


Figure 6. Grain size in the composites as a function of TiB<sub>2</sub> additive amount

The variation in Vickers microhardness of the sintered composites as a function of TiB<sub>2</sub> additive amount is shown in Figure 7. Addition of TiB<sub>2</sub> causes no significant change for all of the composites. The effect of sintering pressure cannot be shown in microhardness. On the other hand, the Vickers microhardness increases by addition of Mg and solid solution strengthening after homogenization. From variation of Vickers microhardness of homogenized composites, Mg solute strengthening decreases with increasing additive amount of TiB<sub>2</sub>. These around 1.0 mass% TiB<sub>2</sub> additions are too little to dispersion-strengthening.

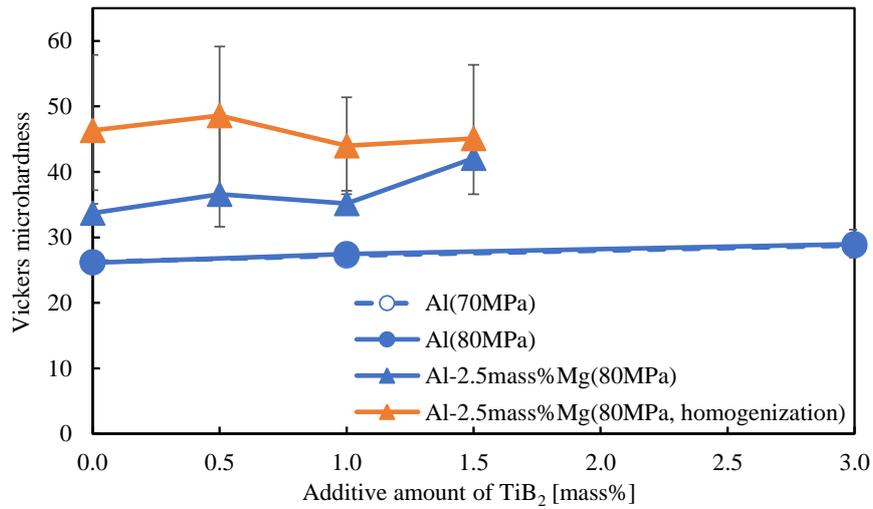


Figure 7. Vickers microhardness of composites as a function of TiB<sub>2</sub> additive amount

The effect of the additive amount of TiB<sub>2</sub> in tensile strength of the sintered composites is shown in Figure 8. The effect of sintering pressure on tensile strength of pure-Al was not significant. On the other hand, Al-Mg alloy based composites has a maximum value at 1.0 mass% TiB<sub>2</sub>, with and without homogenization. In comparison with result of grain size in Figure 6, this can be primarily attributed to Hall-Petch law, the strength of the alloy increases with grain refinement. The homogeneous TiB<sub>2</sub> prevents grain growth during sintering or homogenization. For composites with high additive reinforcement, clustering of reinforcements is inevitable. Therefore, it is also necessary to strengthen matrix by solute-strength or grain refinement dispersing small ceramic powders.

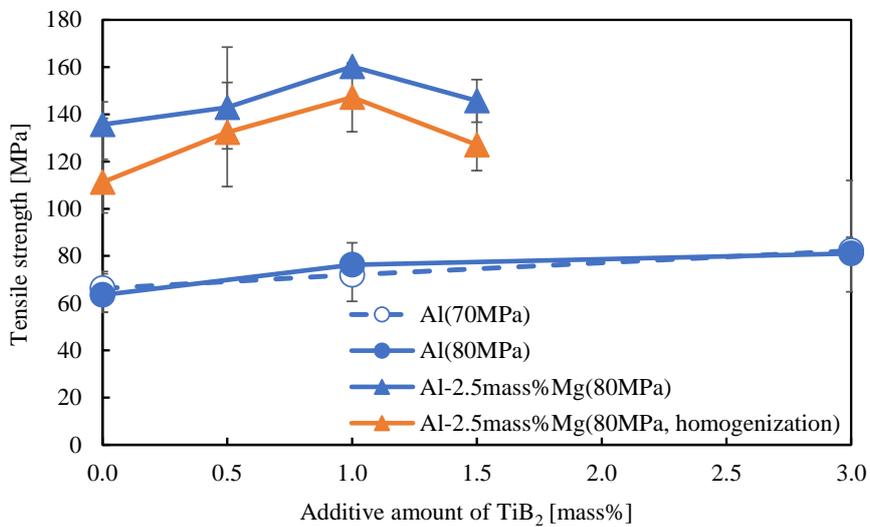


Figure 8. Tensile strength of composites as a function of TiB<sub>2</sub> additive amount

## CONCLUSIONS

The sintered aluminum alloy matrix composites with TiB<sub>2</sub> powder by SPS using blended element powder were fabricated, and the effect of the amount of TiB<sub>2</sub> powder and sintering pressure on microstructure and mechanical properties were investigated.

1. Highly dense Al-2.5mass%Mg-TiB<sub>2</sub> composites can be obtained by SPS.
2. Average grain sizes in the composite were made smaller by higher sintering compression, 2.5mass% Mg addition, or slightly TiB<sub>2</sub> addition. However, addition of over 1.0 mass% TiB<sub>2</sub> dissuades grain refining and accelerates grain growth.
3. Al-Mg alloy based composites has a maximum tensile strength at 1.0 mass% TiB<sub>2</sub>, with and without homogenization. This result can primarily be attributed to the strength of the alloy increases with grain refinement.

## REFERENCES

- Ashwath, P., & Anthony Xavier, M. (2014). The effect of ball milling & reinforcement percentage on sintered samples of aluminium alloy metal matrix composites. *Procedia Engineering*, 97, 1027–1032.
- Bolzoni, L., Esteban, P. G., Ruiz-Navas, E. M., & Gordo, E. (2012). Mechanical behaviour of pressed and sintered titanium alloys obtained from prealloyed and blended elemental powders. *Journal of the Mechanical Behavior of Biomedical Materials*, 14, 29–38.
- Das, T., Munroe, P., & Bandyopadhyay, S. (1996). The effect of Al<sub>2</sub>O<sub>3</sub> particulates on the precipitation behaviour of 6061 aluminium-matrix composites. *Journal of Materials Science*, 31, 5351–5361.
- Easton, M. A., & Stjohn, D. H. (2001). A model of grain refinement incorporating alloy constitution and potency of heterogeneous nucleant particles. *Acta Mater*, 49, 1867–1878.
- Ehsan, G., Masoud, A., Touradj, E., Amir hossein, P., & Ali, R. (2015). Investigation on microstructural and mechanical properties of B<sub>4</sub>C–aluminum matrix composites prepared by microwave sintering. *Journal of Materials Research and Technology*, 4, 411–415.
- Ehsan, G., Ali, R., Masoud, A., Kamyar, S., & Touradj, E. (2017). Evaluation of microstructure and mechanical properties of Al-TiC metal matrix composites prepared by conventional, microwave and spark plasma sintering methods. *Materials*, 10, 1255.
- Fei, X., & Zhong-hu, X. (2013). Characterizing an in situ TiB<sub>2</sub> particulates reinforced aluminium-based composite and its heat treatment. *Physics Procedia*, 50, 13–18.
- Hassani, A., Bagherpour, E., & Qods, F. (2014). Influence of pores on workability of porous Al/SiC composites fabricated through powder metallurgy + mechanical alloying. *Journal of Alloys and Compounds*, 591, 132–142.
- Kamegawa, S., Izui, H., Komiya, Y., Kobayashi, K., & Arimoto, R. (2015). Effect of matrix and reinforcement powder types on tensile and wear properties of TiB/Ti and TiC/Ti composites prepared by SPS. *TMS2015 Annual Meeting Supplemental Proceedings*, 203–210.
- Sampei, Y., Izui, H., Komiya, Y., Shigimura, H., & Suzuki, C. (2015). Mechanical properties of TiB-reinforced Ti-6Al-4V using matrix powders with different particle sizes and morphologies. *TMS2015 Annual Meeting Supplemental Proceedings*, 219–226.

- Smugeresky, J. E., & Dawson, D. B. (1981). New titanium alloys for blended elemental powder processing. *Powder Technology*, 30, 87–94.
- Uesugi, T. & Higashi, K. (2013). First-principles studies on lattice constants and local lattice distortions in solid solution aluminum alloys. *Computational Materials Science*, 67, 1–10.
- Yasue, K., Radjai, A., Miwa, K., & Sakaguchi, Y. (2003). Semisolid forming of Al-10mass%Mg alloy by blending of elemental powders. *Journal of Materials Science*, 38, 3591–3595.