Effects of solid lubricants on properties of Al-Si-SiC composite powder produced by gas atomization technique

Haris RUDIANTO[1,*], Sang Sun YANG[2], Yong Jin KIM[2], Ivo DLOUHY[3], and Wahyu CAESARENDRA[4]

[1] *Gunadarma University, Department of Mechanical Engineering, Jalan Margonda Raya No 100 Depok, West Java, 16424, Indonesia*
[2] *Korea Institute of Materials Science, Powder Metallurgy Research Group, 797 Changwondaero, Changwon, Gyeongsang Namdo, 642-831, Republic of Korea*
[3] *Brno University of Technology, Institute of Materials Science and Engineering, NETME CENTER, Technicka 2896/2, Brno, South Moravia, 61669, Czech Republic*
[4] *Diponegoro University, Department of Mechanical Engineering, Jalan Prof. Sudharto, SH, Tembalang, Semarang, Central Java, 50275, Indonesia*

[*]**Corresponding author e-mail:** harisrudianto@staff.gunadarma.ac.id

Received date:
18 December 2019
Revised date:
3 July 2020
Accepted date:
7 July 2020

Keywords:
Aluminum matrix composites
Powder metallurgy
Green properties
Sintering

Abstract

Al-Si-SiC composites powder was analyzed in this research. The powder was made by gas atomizing process. Powder characterization was done to observe its morphology, particle size distribution and thermal properties. Effects of lubricants were intensively investigated in this research with compositions and pressure variables. It is assumed that higher amount of solid lubricant put into the powder, the lower ejection force needed to take out compacted powder. Finding appropriate solid lubricant is necessary to improve green and sintering properties. EBS (ethylene bis stearamide), aluminum stearate, zinc stearate and paraffin wax were used for investigation. Sintering under ultra high purity nitrogen gas 99.9999% was carried out to produce high density material. SEM-EDS and XRD were carried to characterize this powder. SiC particles in this powder prohibit to achieve optimum sintering properties of Al-Si-SiC due to their resistance during compaction which leads to form pores and lower sintering density. And ethylene bis stearamide (EBS) seems to be suitable solid lubricant for this powder by giving lower ejection force, flowability and higher sintering density for 93.5%.

1. Introduction

Aluminum matrix composites have been investigated by researchers especially due to their lightweight properties for automotive or aerospace applications [1-4]. SiC particle reinforced aluminum matrix was investigated to optimize their wear and mechanical properties. Mechanical alloying is commonly used to produce Al-SiC composite involving cold welding, fracturing and rewelding of powder. Mechanical alloying is able to disperse SiC particles on aluminum matrix, but based on microscopic studies from this method, SiC particles will be located on grain boundary. Inability of SiC particles to wet on aluminum matrix is a result of high resistance and hardness of SiC particles compared to Aluminum matrix. As a result, there will be pore in between SiC particles and Aluminum matrix which leads to lower density and mechanical properties [1-7].

Presureless sintering of aluminum powder is considered as low cost processing compare to extrusion. Sintering of aluminum alloy powder was successfully investigated by G.B. Scaffer et al. and many other researchers. Nitrogen gas seems to be sufficient sintering atmosphere for aluminum powder because of possibility to support pore filling mechanism during sintering [8-9]. So, in this research, conventional sintering based process was carried out.

To produce compacted powder, solid lubricants are usually added into aluminum powder to improved their compactibility and sinterability. Solid lubricants are able to reduce the friction between metal powders and die walls during compaction and ejection, and to minimize tool wear. Possibility to form residual products which able to impede diffusion process during sintering, for this reason, lubricants should be removed prior to sintering process [10-12]. L. X. Liu et al and N. Showaiter et al. investigated effects of paraffin wax, lithium stearate and ethylene bistearamide (EBS) in aluminum alloys. According to their results, paraffin wax was found as a suitable lubricant for Al-Mg-Si-Cu alloy powder with acceptable green density and sintering density [8,11].

Al-Si-SiC composite powder contains ceramic material which has very high mechanical properties and melting temperature. In order to have high green density after compaction, this composite powder needs to be pressed at such high pressure. Compaction dies will wear easily if used at such high pressure. Lubrications play important role in this aspect to improve wearability of compaction dies. This research is very important because there are not many reports doing investigation in this matter. And higher

DOI: 10.14456/jmmm.2020.38

difference of melting temperature between aluminum and SiC leads to inability to wet each other at sintering temperature of aluminum around 550-600°C [13-15]. Atomization technique was done in this research in order to have better wettability between aluminum and SiC. High pressure flowing gas will be able to molten aluminum stick on SiC particles with no or less voids between them.

Ethylene bis stearamide (EBS), aluminum stearate, zinc stearate and paraffin wax were rarely found to be investigated in aluminum powder. In this research, investigation was on effect of EBS, paraffin wax, aluminum stearate and zinc stearate lubricants on sintering properties of Al-Si-SiC powder.

2. Experimental

Gas atomized Al-Si-SiC powder was prepared for this research. This powder has chemical composition of Al-9Si-0.5Mg-0.2Cu and 20% volume fraction of SiC (12 μm) and theoretical density of this powder is 2.72 $g \cdot cm^{-3}$. This powder was made by using 80 atm pressure and 7 0 0 °C melting temperature under ultra high purity nitrogen gas atmosphere at gas atomizing machine in Powder Technology Research Group, Korea Institute of Materials Science.

Aluminum stearate, zinc stearate, ethylene bis tearamide (EBS) and paraffin wax were prepared. The compositions of solid lubricant in powder vary from 0.5-1.5 wt%. Mixing process was done with using turbula mixer for 30 min. To observe morphology of this Al-Si-SiC powder, optical microscopy was carried out. Particle size analyzer with dry method also was carried out to investigate particle size distribution of this powder. For thermal properties characterization, thermogravimetry analysis and differential scanning calorimetry (DSC-TGA) were done simultaneously. The process was under ultra high purity nitrogen gas atmosphere with temperature range from 20-800°C.

In this work, the flowability of powders was measured using Carney flow meter which has 2.54 mm orifice. The procedures for the measurement of flow time were as follows: by blocking the bottom opening of the flow meter, a 50 g sample was poured into it. The powder was then released from the flow meter and the time taken for all the powder to flow out was recorded as the flow time.

Standard cylindrical specimens (8 mm diameter and 8 mm height) were compacted at room temperature and at different pressures using an instrumented floating die (D2) and a laboratory (100 T) hydraulic press. No die-wall lubrication was used in these experiments. The ejection behavior of the different mixes was evaluated by continuously recording the force during ejection using a load cell under the hydraulic cylinder and a position sensor fixed to the die. Typical ejection curves present the variation of the ejection shearing stress as a function of the displacement inside the die. The ejection shearing stress corresponds to the ejection force divided by the surface of the compact in contact with the die walls. The effects of the springs were subtracted from the curves before analysis.

Sintering was carried out under ultra high purity nitrogen gas at 555°C for 1 h. Before reaching sintering temperature, compacted powder was delubricated at 400°C for 30 min. Sintering density was done with using Archimedes method.

3. Results and discussion

This powder has irregular morphology as shown by Figure 1(a). This morphology is usually expected from powder made by gas atomizing process due to high pressure gas flows on molten metal. Molten aluminum will stick on SiC particles when high pressure gas flows. With high rapid cooling, SiC particles are able to infuse themselves on Aluminum grain which form intragranular SiC. Average of particle size after gas atomizing process is expected to be larger because of possibility of non homogenous air pressure and cooling when molten metal falls down. To be able to obtain certain size of particle, meshing was done. And the curve shows that average particle after 140 meshing is around 100 μm. Figure 1(a). shows that SiC particles are inside aluminum particles called intragranular SiC. Using gas atomizing to produce this powder with expectation to have better wettability between Al and SiC. Wetting behavior of powder is crucial to obtain high sintering density of materials, and this is related to pore filling mechanism during sintering. Due to high different of melting point between Al and SiC, it requires higher sintering temperature. Because separate form of Al and SiC, wetting angle of SiC particles on pure Al are at $\theta > 140°$ on 1000°C which is unacceptable for sintering of aluminum [9]. Gas atomizing technique seems to be suitable alternative to reduce wetting angle for SiC on Aluminum powder

For ejection force measurement, compaction pressure was at 620, 700 and 800 MPa. Figure 2 shows ejection force for Zn stearate, paraffin wax, Al stearate and ethylene bis stearamide (EBS) with 0.5, 1 and 1.5 wt%. For all solid lubricants, ejection force decreased with increasing content of solid lubricant and higher compaction pressure will give higher ejection force. This is expected, higher amount of lubricants will ease ejection process of compacted powder from the die, due to less friction with the die wall.

Lubrication is done to minimize friction between two interacting surfaces in relative motion. Friction occurs because a solid surface never microscopically smooth. Even the best machined surface has peaks and valleys called roughness. When two such surfaces come into contact, it is only the peaks on the surfaces that make actual contact. These contacts support the normal load and deform plastically and get cold welded. Depending upon the magnitude of the normal load more and more high spots or peaks come into contact and the 'real area' of contact increases in contrast to the 'apparent area', which is the geometrical area of the surfaces in contact. This phenomenon is called adhesion. Friction is believed to be caused by this adhesion. When two such surfaces have to be moved in relation to each other, some force will be required to sheer these contacts. This force is called frictional force [10-13]. Among other solid lubricants, aluminum stearate gave lowest ejection force and followed by EBS. This is related to low friction force between compacted materials with Al stearate lubricant with the die wall.

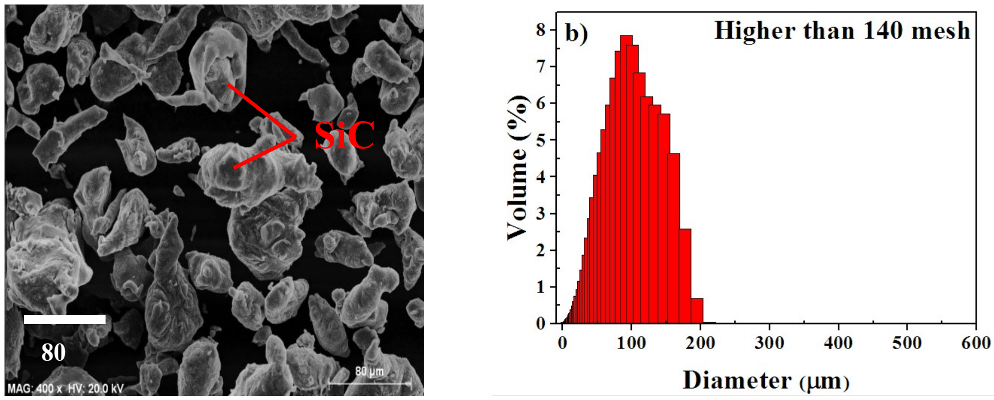

Figure 1. (a) Microstructure of Al-Si-SiC powder and (b) particle size distribution of Al-Si-SiC after meshed at 140 meshing number.

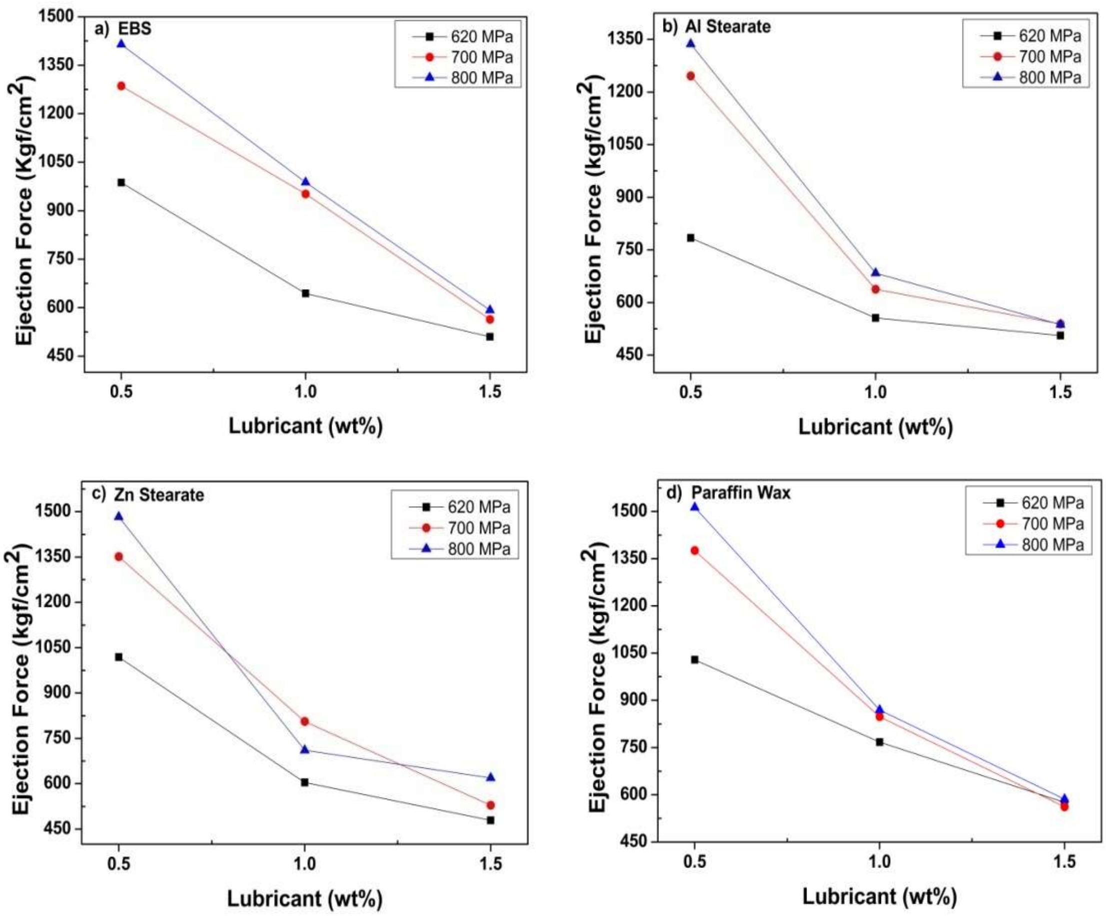

Figure 2. Effect of solid lubricants on ejection force starting with (a) EBS, (b) Aluminum Stearate, (c) Zinc Stearate and (d) Paraffin Wax

On Figure 3 shows that green density increased with increasing content of lubricant and compaction pressure. Al stearate gave better green density on 620 and 700 MPa compare to other lubricants, but at 800 MPa, all lubricants gave similar green density for up to 95% relative. This indicates that lubricants improved compactibility of powder. The effect of lubricants on interparticle behavior at high compaction pressure gives similar results with similar green density. But at lower compaction pressure, the effects of lubricants give different results. zinc stearate and paraffin wax give similar trend in green density, but aluminum stearate and EBS show higher green density. This indicates aluminum stearate and EBS give better effect on interparticle bonding during compaction process due to their adhesion behavior between particles.

The flowability characteristic of a powder is directly related to both the physical properties of the material itself, as well as the specific processing conditions in the handling system. The effect of lubricant type and concentration on powder flowability is an important aspect to consider. Inadequate flow may cause difficulties in filling the die and induce part to part dimensional changes. Different approaches might be considered to optimize the flowability of the powder. A flow agent might be added to the powder. Binder treatment or particle agglomeration is also alternative. In these cases, the flow agent or binder should have minimal negative impacts on the other powder characteristics and specimen properties. It is well known that problems may occur when a liquid binder is introduced into the powder mix. The most common problem is the reduction of powder flowability, or even worse, the complete loss of free flowing properties.

On Table 1, it shows flowing behavior of powder with Carney flow meter. Zinc stearate and paraffin wax seem not to give an effect of flowing behavior of Al-Si-SiC powder with showing random results between high and low content of lubricants. Aluminum Stearate seems to reduce flowability of powder with higher time needed to flow a 50 g of powder and it did not flow at 1.5wt% content. EBS seems to have reasonable results with higher content of lubricant improves flowability of the powder. It is assumed that this is because of reduced adhesion between the solid lubricant and aluminum particles.

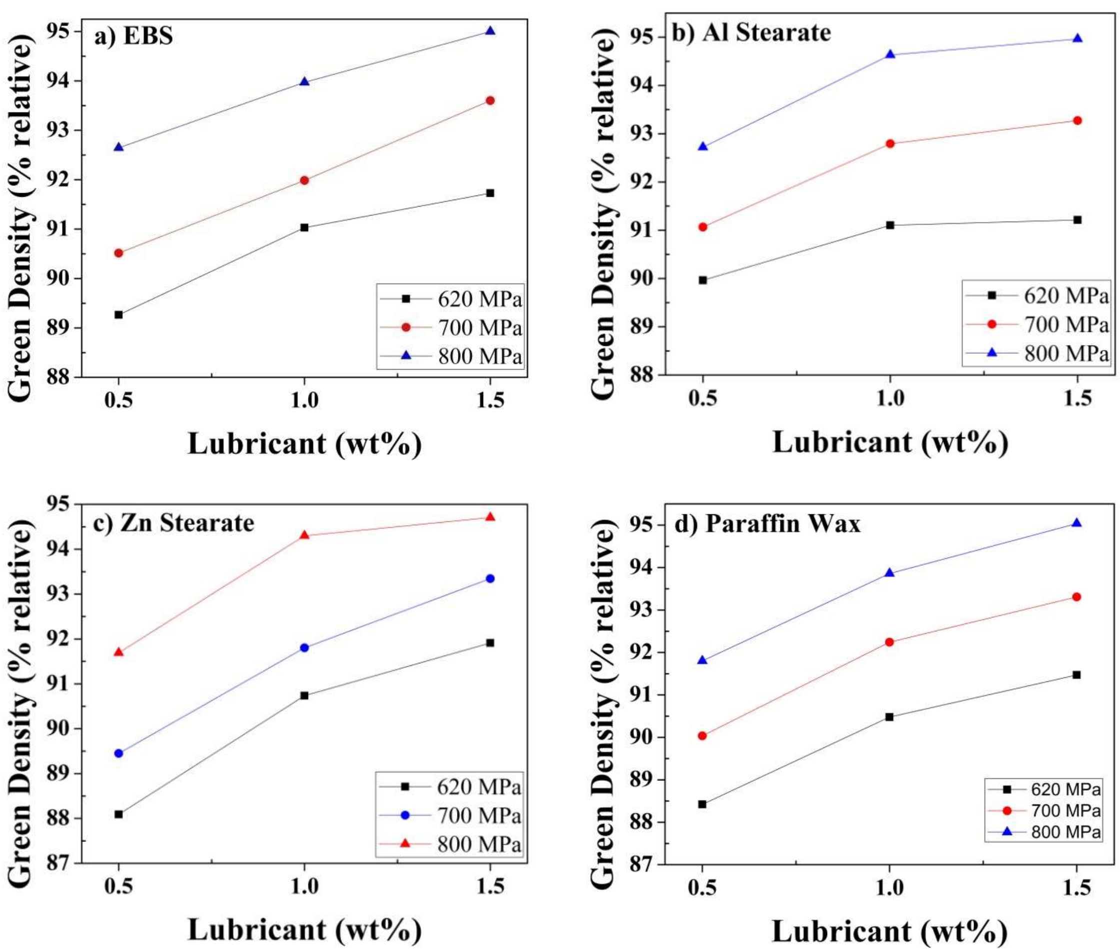

Figure 3. Effect of lubricants on green density starting with a) EBS, b) Aluminum Stearate, c) Zinc Stearate and d) Paraffin Wax.

Table 1. Effect of flowability of Al-Si-SiC powder with EBS lubricant.

No.	Sample	Content of Lubricant (wt%)	Fluidity (min:sec)
1	Al-Si-SiC+ EBS	1.5	2:10
2		1.0	2:11
3		0.5	2:15
4	Al-Si-SiC + Al stearate	1.5	Not Flowing
5		1.0	2:36
6		0.5	2:29
7	Al-Si-SiC + Zn stearate	1.5	2:09
8		1.0	2:05
9		0.5	2:09
10	Al-Si-SiC + paraffin wax	1.5	2:16
11		1.0	2:19
12		0.5	2:17

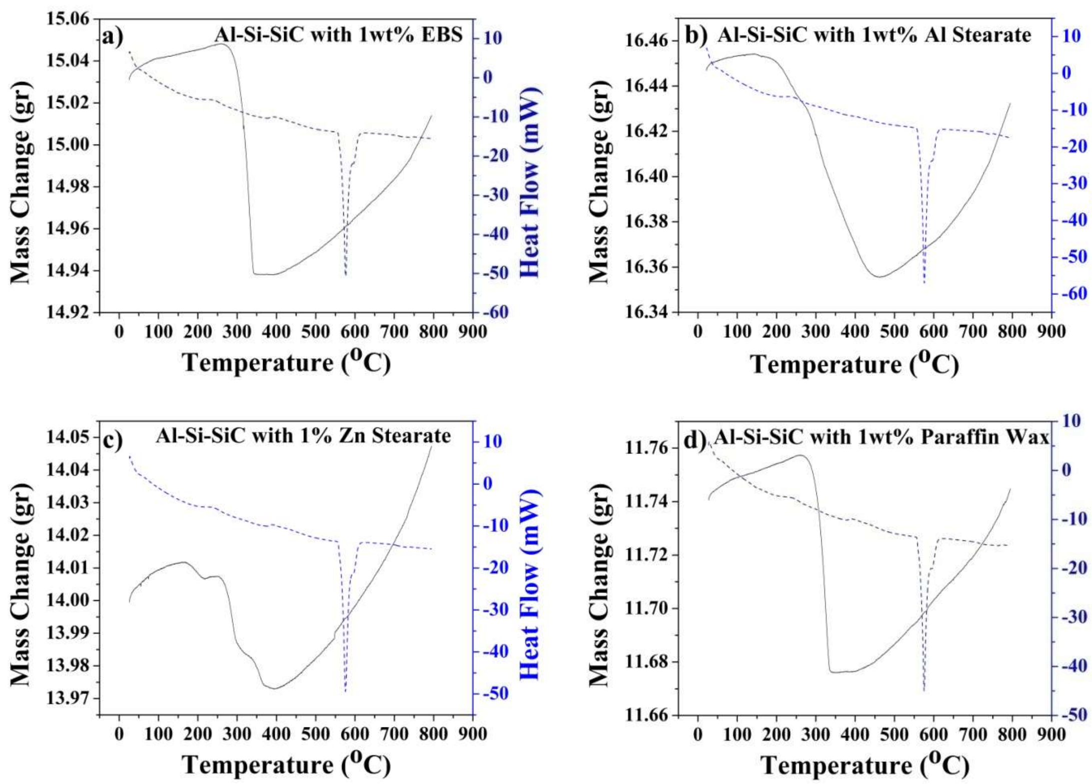

Figure 4. Thermogravimetry and differential scanning calorimetry curve to estimate delubrication and sintering temperature with different types of solid lubricants at 1 wt% of (a) EBS, (b) aluminum stearate, (c) zinc stearate, and (d) paraffin wax.

Figure 4 (a-d) show thermogravimetry (black line) and differential scanning calorimetry (blue line) curve of powder with different lubricant. From all thermogravimetry curves, it seems it has slightly different of delubricating temperature with each other. EBS and paraffin wax have similar thermogravimetry curve with delubrication starting at 300-400°C, and this is related to their melting point. Aluminum and zinc stearate have slow and higher delubrication process with more that 400°C delubrication temperature. Slow curve of thermogravimetry is assumed that removing lubricant prior to sintering will be longer and higher to be able to avoid resistant during delubrication. From all thermogravimetry curve after 400°C goes up, this indicates there is gaining mass in sample. This is assumed to be a reaction between magnesium-aluminum oxide to form spinel and nitrogen gas-aluminum to form AlN [16]. Differential scanning calorimetry of powders are similar, this means lubricants does not effect on thermal behavior of Al-Si-SiC powder.

According to DSC-TGA curve at Figure 4, endothermic peak of this powder is at 575ºC. This endothermic peak is assumed to be eutectic point of this powder. At 575°C, the powder starts to melt and form liquid phase. Liquid phase sintering seems to be appropriate technique as proved by researchers to obtain high density of aluminum powder by disrupting oxide layer on aluminum to ease densification. But high content of liquid phase will have opposite effect by formation particle coarseing which leads to lower sintering density called Ostwald ripening phenomena [5,12]. So, sintering was carried out at 555°C for 1 h and before reaching sintering temperature, compacted powders were delubricated at 400°C for 1 h under ultra high purity nitrogen gas.

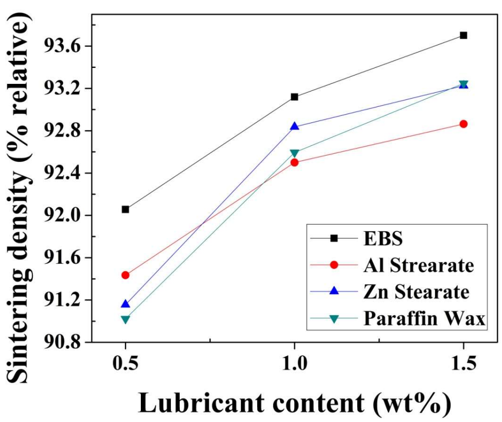

Figure 5. Sintering density of Al-Si-SiC with 400°C for 1 h delubricating temperature and sintered at 555°C for 1 h.

Figure 5 shows sintering density of compacted powders with different lubricants. From curve, sintering density increased with higher amount of lubricants. And EBS gave highest sintering density for about 93.5% relative. Some researchers reported that metallic stearates which are widely used in ferrous alloys, will leave decomposition products if the temperature below 420°C and are generally not used for P/M aluminum alloys. That is the reason why aluminum stearate and zinc stearate gave lower sintering density at this case [8,10]. EBS which is also commonly added on a commercial powder premix Alumix 231 for 1.5wt% shows good compactibility and sinterability [13].

In general, optimum sintering density especially for aluminum silicon based powder could reach up to more than 98% relative. Lower optimum sintering density in this composite powder possibly could be from the voids around SiC particles which are very hard to fill because of low wettability between Al and SiC. Optimization of chemical compositions are also the possibility, because certain amount of magnesium and tin could increase the sintering density of this composite powder [8,9,15].

On Figure 6, it shows microstructures of sintered Al-Si-SiC powder. SiC particles seem to homogenously dispersed throughout the surface and have good wettability with aluminum. And there are some sintering precipitates on and inside grain boundary. From sintering density curve, it indicates that this sintered material contains pores as it shown by SEM pictures. High content of oxide can be a reason for this condition. Oxide layer is preventing of aluminum particles to wet each other on lower sintering temperature due to high melting point of oxide. Depending on chemical compositions of aluminum alloy powder, sintering temperature of aluminum alloys is usually ranged between 500 to 600°C.

To investigate intermetallic and chemical distribution, alongside with SEM-EDS was as well carried out. There are several selected points (from 1 to 6) which indicate different chemical compositions as shown by Table 2. Based on chemical compositions, this powder has potential to form sintering precipitates. For chemical distribution, it shows aluminum, copper, magnesium and silicon carbide are homogenously dispersed. Magnesium and oxygen have higher concentrates on grain boundary and somewhere close to pore. This indicates that during sintering, magnesium starts to react to form spinel $MgAl_2O_4$ and decompose itself to grain boundary [17]. So, based on EDS point analysis data, it shows that no. 5 have high content of magnesium and oxygen. EDS point no. 3 seems to be α aluminum and point no. 2 and 4 with high content of iron seem to be AlFe precipitates. No. 6 is supposed to be SiC particle. AlCuMg, $CuAl_2$, Mg_2Si, AlFe and AlSiMg are possible sintering precipitates from this powder. XRD was carried out to find these precipitates as shown by Figure 8. α aluminum as matrix seems to have strongest intensity from this powder followed by silicon and silicon carbide. $CuAl_2$ and Mg_2Si are found and expected from sintering.

Table 2. EDS point analysis of sintered Al-Si-SiC powder.

Spectrum	Al (wt%)	Si (wt%)	Mg (wt%)	Cu (wt%)	C (wt%)	O (wt%)	Fe (wt%)
6	62.75	20.87	0.3	3.92	4.43	2.54	
2	62.75	14.54		0.98	3.08	1.01	18.65
3	92.84	0.75	0.28	3.12	2.84	4.37	
4	52.88	27.15	0.1	1.05	30.84		18.82
5	44.45	31.1	1.3	2.29	5.75	14.83	
6	0.28	68.19			31.53		

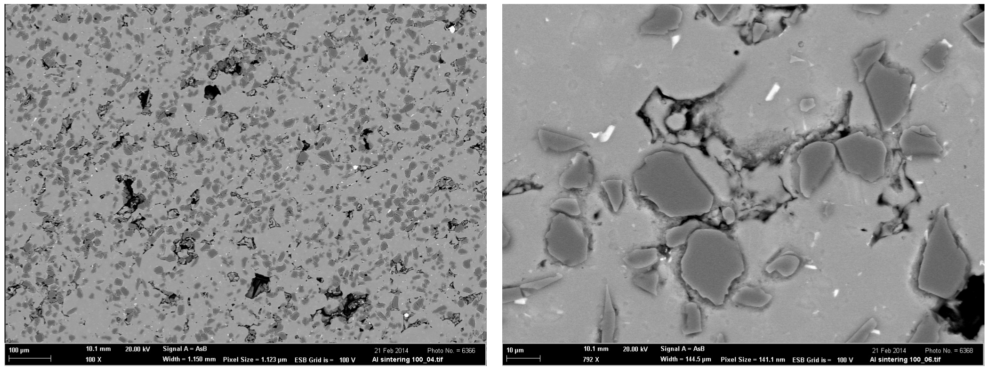

Figure 6. Microstructure of Al-Si-SiC by SEM showing there are some pores on surface with 1.5wt% EBS.

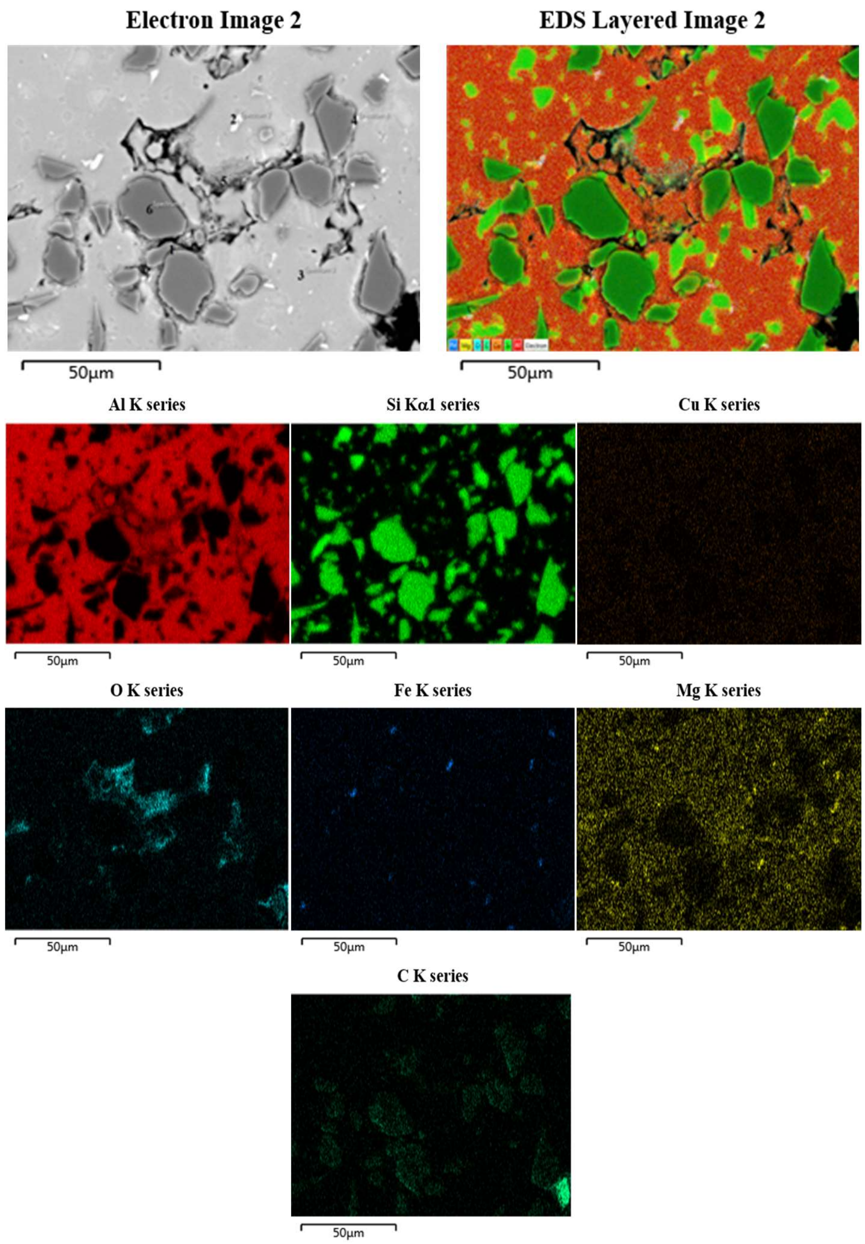

Figure 7. EDS analysis of sintered Al-Si-SiC powder.

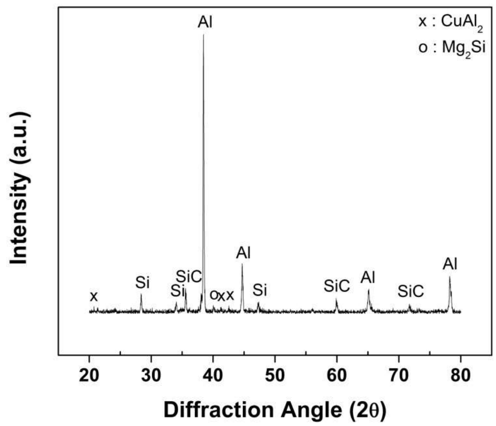

Figure 8. XRD analysis of sintered Al-Si-SiC powder.

4. Conclusions

Al-Si-SiC powder was successfully made by gas atomizing process. Intragranular SiC was found inside aluminum matrix. Optical microscopy showed better wettability between SiC particle and aluminum matrix. Ejection force increased with increasing compaction pressure and low lubricant content. Higher content of lubricant gave lower ejection force and this indicates there is adhesion reaction between lubricant and die wall. Green density increased with increasing content of lubricant and compaction pressure. Ethylene bistearamide (EBS) with composition of 1.5wt% was found as suitable lubricant for this powder with having good flowability, ejection force and green density.

Sintering has successfully homogenized this powder by showing sintering precipitates on grain boundary and inside grain. SEM-EDS and XRD characterized sintering precipitates and chemical compositions after sintering. α Al, Si, SiC $CuAl_2$ and Mg_2Si seem to be dominant precipitates from this powder. And optimum sintering density was achieved at 93.5% relative, by this number, there should be further research to improve sintering density.

5. Acknowledgements

This research is was supported by Fundamental R&D Program for Core Technology of Materials funded by the Ministry of Knowledge and Economics, Republic of Korea and Project NETME of Ministry of Education, Youth and Support of Czech Republic.

References

[1] N. Zhao, P. Nash, and X. Yang, "The Effect of Mechanical Alloying on SiC Distribution and the Properties of 6061 Aluminum Composite," *Journal of Materials Processing Technology*, vol. 170, issue 3, pp. 586-592, 2005.

[2] M. Bunma, P. Subarporn, R. Bobuangern, T. Patthannkitdamrong, T. Thuanwong, and T Patcharawit, "Process parameter-microstructure-mechanical property relations of SiCp-reinforced aluminum composites produced by powder-injection casting," *Journal of Metals, Materials and Mineral*, vol. 21, no. 2, 2011.

[3] S. Sivananthan, K. Ravi, and C. S. J. Samuel, "Effect of SiC particles reinforcement on mechanical properties of aluminium 6061 alloy processed using stir casting route," *Materials Today: Proceedings*, vol. 21, pp. 968-970, 2020.

[4] R. Ande, P. Gulati, D. K. Shukla, and H. Dinghra, "Microstructural and wear characteristics of friction stir processed Al-7075/SiC Reinforced Aluminium Composite," *Materials Today: Proceedings*, vol. 18, pp. 4092-4101, 2019.

[5] B. Rahmatian, K. Dehghani, and S. E. Mirsalehi, "Effect of adding SiC nanoparticles to nugget zone of thick AA5083 aluminium alloy joined by using double-sided friction stir welding," *Journal of Manufacturing Processes*, vol. 52, pp. 152-164, 2020.

[6] Ö. Yilmaz, B. Cem Turan, and M. Gübrüz, "Production and characterization of SiC reinforced aluminum alloy matrix composites from waste beverages cans," *Journal of Metals, Materials and Minerals*, vol 29, pp. 28-33, 2019.

[7] T. Ye, Y. Xu, and J. Ren, "Effects of SiC particle size on mechanical properties of SiC particle reinforced aluminum metal matrix composite," *Materials Science and Engineering A*, vol. 753, pp. 146-155, 2019.

[8] N. Showaiter and M. Youseffi, "Compaction, sintering and mechanical properties of elemental 6061 Al powder with and without sintering aids," *Materials and Design*, vol. 29, issue 4, pp. 752-762, 2008.

[9] G. B. Scaffer, S. J. Boner, S. H. Huo, and T. B. Sercombe, "The effect of the atmosphere and the role of pore filling on the sintering of aluminum," *Acta Materialia*, vol. 54, no.1, pp. 131-138, 2006

[10] R. M. German, "Powder metallurgy science," *Metal Powder Industries Federation*, 1994.

[11] Lian X. Liu, Graham B. Schaffer, and J. D. Litster, "Binder-treated segregation-free aluminum alloy powders," *International Journal of Powder Metallurgy*, vol. 41, no. 1, 2005.

[12] L. P. Lefebvre, Y. Thomas, and B. White, "Effects of lubricants and compacting pressure on the processability and properties of aluminum P/M parts," *Journal of Light Metals*, vol. 2, pp. 239-246, 2002.

[13] A. C. Ferro and B. Derby, "Wetting behavior in the Al-Si/SiC system interface reactions and solubility effects," *Acta Metallurgica et Materialia*, vol. 43, no. 8, pp. 3061-3073. 1995.

[14] D. W. Heard, I. W. Donaldson, and D. P. Bishop, "Metallurgical assessment of a hypereutectic aluminum–silicon P/M alloy," *Journal of Materials Processing Technology*, vol. 209, pp. 5902-5911, 2009.

[15] H. Rudianto, S. S. Yang, K. W. Nam, Y. J. Kim, " Mechanical properties of Al-14Si-2.5 Cu-0.5 Mg aluminum-silicon P/M alloy," *Reviews on Advanced Materials Science*, vol. 28, issue 2, pp.145-149, 2011.

[16] M. Yan, P. Yu, G. B Schaffer, and M. Qian, "Secondary phases and interfaces in a nitrogen-atmosphere sintered Al alloy: transmission electron microscopy evidence for the formation of AlN during liquid phase sintering," *Acta Materialia*, vol. 58, no. 17, pp. 5667-5674, 2010.

[17] T. Pieczoncka, T. Schubert, S. Baunack, and B. Kiebak, "Dimensional behavior of aluminum sintered in different atmospheres," *Materials Science and Engineering A*, vol. 478, no.1-2, pp. 251-256, 2008.