Technical Report

Effect of Particle Size on the Quality Characteristics of Pure Titanium Fabricated Using Metal Additive Manufacturing

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ABSTRACT

In recent years, the use of additive manufacturing (AM) technology in the fields of aviation, medical devices, and so forth has been investigated. Electron beams and lasers are used for metal AM as heat sources. Metal powder can be re-used for both processes, although the particle size of the powder differs for each process. However, there is concern over the change in the characteristics of the re-used powder. The re-used powder particle distribution may vary from the original distribution. In this study, we investigated the effects of different particle sizes on pure titanium fabricated by AM using a laser beam as the heat source. Correlations of the laser strategy and quality characteristics of products fabricated using four types of powder particle size distribution were experimentally verified.

KEY WORDS

additive manufacturing, titanium, selective laser melting, metal powder, particle size

1 Introduction

Powder Metallurgy (PM) is a metal-forming process by sintering in which a high temperature below the melting point is applied to powders to bond the particles to one another under a certain atmosphere in order to exert consolidated characteristics such as those of metals or ceramics. PM technology plays an important role as a manufacturing method for preparing advanced materials and products in industrial fields. In PM technology, new forming processes that can realize both the easy formation of complicated shapes and the achievement of high density have been much expected for the last ten years. One such newly developed process is known as additive manufacturing (AM)¹⁻³⁾.

In AM processes with metal, a laser or an electron beam acting as a heat source scans a thinly spread metal powder according to electrical signals from a computer-aided design model and melts powders locally. Subsequently, more powder is spread on the formed layer and melted as before. This process is repeated layer by layer. These processes are called SLM (Selective Laser Melting) or EBM (Electron Beam Melting). One of the features of SLM is its ability to produce complicated shapes and structures that cannot be attained by machining or plastic forming. The other is that SLM is conducted at a temperature higher than the melting point of the metal powder material, whereas sintering is

performed at a temperature lower than the melting point. SLM is utilized industrially for the rapid production of prototypes and the development of small-lot parts of molds and machine parts⁴⁾. In the field of medicine, taking advantage of the features described above, tailor-made implants (artificial joints, artificial osteosynthesis materials, dental prosthesis, artificial bone, etc.) have been researched and developed for application to clinical operations⁵⁾.

Conversely, Ti metal and its alloys are widely used not only as industrial products, but also as medical biomaterials due to their high biocompatibility. Because they are hard to machine or work plastically, AM has been suggested as a way for preparing implants with complicated shapes and structures that fit to bone defects⁶⁾.

In research on SLM or EBM in recent years, there have been many reports about the effects of parameters in the AM process on the mechanical properties of the products⁷⁻⁸⁾ and melting behavior such as temperature distribution and molten pool sizes⁹⁾. Almost all of them were conducted using a certain distribution of powder particle size. In the AM process, the particles' sizes and shapes are important factors that dominate powder flow and layered powder density. From the metallurgical viewpoint, because the powders are melted and rapidly cooled and because the powders around the melt zone are influenced by transferred heat, a change in microstructure may occur^{10,11)}. Then, the mechanical properties of the products can also be varied through heat cycles. Furthermore, in the AM process, powders not irradiated are re-usable when they are heated.

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When this procedure is repeated, the powder particle distribution may vary from the original distribution of particles and may result in different mechanical properties of the final product. Therefore, the supplied characteristics of the powder should be essentially managed to obtain controlled and stable mechanical properties of the product.

In the present study, to establish a manufacturing method that will ensure stable product quality, the effects of powder particle size distribution on the mechanical properties and microstructure of commercially pure titanium parts produced by SLM processes are evaluated under different conditions of supplied power and scanning speed.

2 Material and methods

Commercially pure titanium gas atomized powder ($\leq 45\text{ }\mu\text{m}$, grade 2) was used. Its chemical composition is shown in Table 1. The powder was classified using two types of sieves into four types of powder particle distributions as shown in Table 2. Powder D is a powder of 50: 50 wt% powder mixed in the particle size range of both ends of powder A. Each powder was supplied into the SLM equipment (EOSINT M270 system (laser spot size: 0.1 mm, Germany EOS Inc.)) to form a prismatic specimen ($L7 \times D6 \times H5\text{ mm}$) and a tensile test specimen, as shown in Fig. 1. The specimens were formed in an argon atmosphere, and the residual oxygen concentration was set to 0.1%. The laser parameter is shown in Table 3, and the layer thickness was set to 30 μm ; the scanning pitch was held constant (0.12 mm). The direction of scanning was changed by rotation of 67° between consecutive layers. Using the prismatic specimens, the effects of laser power and scanning speed on the density of product and its microstructure

Table 1 Chemical composition (weight%) of Ti powder.

Fe	H	N	C	O
0.047	0.004	0.012	0.007	0.137

Table 2 Four types of Ti powder characteristics.

	Powder A	Powder B	Powder C	Powder D
Type	Standard	Fine	Coarse	Mix
Particle size (D50)	23.3	19.6	34.3	22.0
Tapping density (g/cm ³)	2.79	2.77	2.63	2.85

Fig. 1 Shaped test specimens: (a) prismatic specimens; (b) tensile specimens. (unit: mm)

were evaluated. The macrostructures and the microstructures of the prism specimens after wet etching were observed using an optical microscope. Tensile and hardness tests were conducted using the tensile test specimen, which was surface polished after SLM to remove surface irregularities. Density was determined by the Archimedes method. Based on the Japanese Industrial Standard (JIS), the tensile test speed was set to 10 mm/min using a JIS14 tensile specimen to determine the strength and elongation. Hardness was measured using a Vickers testing machine at a load of 10 kg.

In this study, the total energy of the laser needed to melt one volume unit of powder, i.e., the energy density ($E: \text{J/mm}^3$), was used as the index. The effects of laser strategy on the product's characteristics such as strength, elongation, hardness, relative density, and microstructure were investigated. Here the energy density, $E (\text{J/mm}^3)$, was defined as follows using the laser power $P (\text{W})$, the scanning speed $V (\text{mm/s})$, the scanning pitch $S (\text{mm})$, and the layered powder thickness $T (\text{mm})$.

Energy density $E = P/(V \cdot S \cdot T) (\text{J/mm}^3)$

3 Results

3.1 Relative density and the microstructures of the prism specimens

The relative density increased with the increase of the energy density (true density of pure titanium metal: 4.51 g/cm³), as shown in Fig. 2. However, in the case where the energy density was 125 J/mm³ ($P = 90\text{ W}$ and $V = 200\text{ mm/s}$), the relative density was lower than that of the material produced at the lower energy density of 111 J/mm³ ($P = 120\text{ W}$, $V = 300\text{ mm/s}$). Fig. 3 shows the macro structures of prismatic specimens using Powder C (a) and Powder B (b) in the

Table 3 Laser parameters

Laser Parameter	Prismatic specimens	Tensile specimens
Laser power (W)	90, 120, 150	90, 120, 150
Scanning speed (mm/s)	100, 200, 300	100, 200, 300
Scanning pitch (mm)	0.12	0.12
Layer thickness (mm)	0.03	0.03

Fig. 2 Relationship between the energy density and relative density of prismatic specimens. (Relative densities: the average value)

Fig. 3 Macrostructure of prismatic specimens, (a) relative density: 96.3%, (b) relative density: 99.1%.

section perpendicular to the laminated direction. Although pores were often observed in the cross-section of the specimen with a relative density of 96.3% ($E = 125 \text{ J/mm}^3$), no pores were observed in the specimen with a relative density of 99.1% ($E = 333 \text{ J/mm}^3$). This difference is attributed to the fact that the supplied energy was insufficient to melt the powders at lower energy density. Furthermore, as shown in Fig. 2, the difference in the relative densities of the Powders A, B, C, and D is large when the energy density is small, which suggests that the relative density depends on the powder-size distribution. This relative density difference among the powder types becomes smallest when the supplied energy density is 208 J/mm^3 .

Fig. 4 shows the microstructures of the prismatic specimens produced at different laser powers of 90, 120, and 150 W and at a scan speed of 200 mm/s using Powder A. Conversely, Fig. 5 shows the microstructures of the prismatic specimens at $P = 150 \text{ W}$ and $V = 200 \text{ mm/s}$ using Powders A, B, C, and D. The grain size of the specimen produced using a powder with small-sized particles (Powder B) is relatively small compared with that of the specimen produced with a powder of larger-size particles (Powder C). Powder D, of which the average particle size is almost the same as that of Powder A, showed a mixed microstructure containing large and small grain sizes, because it was a mixture of large and small particles.

3.2 Mechanical properties of the tensile specimens

Fig. 6 shows the tensile strengths and elongations of the tensile specimens produced by SLM at a laser power of 150 W and a scan speed of 200 mm/s. In Fig. 6 (a), the tensile strength is almost the

Fig. 4 Microstructure of the prismatic specimens with Powder A: (a) $P = 90 \text{ W}$; (b) $P = 120 \text{ W}$; (c) $P = 150 \text{ W}$ at $V = 200 \text{ mm/s}$.

Fig. 5 Microstructure of the prismatic specimens for various types of powders ($P = 150 \text{ W}$; $V = 200 \text{ mm/s}$): (a) Powder A; (b) Powder B; (c) Powder C; (d) Powder D.

same in the range of relative density above 99.6% for each particle size of Powders A, B, C, and D. Moreover, there exist different magnitudes of strength among these powders. The tensile strength is highest for Powder B and lowest for Powder C. Conversely, in Fig. 6 (b), the elongations are almost the same for all specimens produced by SLM using powders of different particle sizes. It was also confirmed that the surface hardness varied with the powder particle size as well as the tensile strength.

In Fig. 7, the oxygen contents of the tensile specimens formed by SLM under laser powers of 150 and 120 W at $V = 200 \text{ mm/s}$ using the four types of powder are shown. It is clear from Fig. 7 that the larger the powder particle size, the smaller the oxygen content.

Fig. 6 Tensile strength and elongation versus relative density of SLM parts for various types of powders: ($P = 150 \text{ W}$, $V = 200 \text{ mm/s}$). ((a) Tensile strength (b) elongation)

Fig. 7 Oxygen content of specimens formed by SLM using various types of powders ($P = 150$ W and 120 W; $V = 200$ mm/s).

Fig. 8 Elongation of specimens formed by SLM at various energy densities using various types of powders.

Fig. 9 Relationship between tensile strength and elongation as SLM for various types of powders ($P = 150$ W, $V = 200$ mm/s).

This tendency is consistent with the observation that particles of smaller size have larger oxidized surface areas per unit volume, causing higher oxygen content in the molten titanium metal. Fig. 8 shows the elongation of tensile specimens formed using the four types of powders at the given energy densities. As energy density increased, the elongations of the four types of powders converged until they showed almost the same elongation at an energy density of 208 J/mm^3 .

Fig. 9 shows the relationship between tensile strength and elongation for the tensile specimens formed using powders of

different particle size at an energy density of 208 J/mm^3 ($P = 150$ W, $V = 200$ mm/s). As shown in Fig. 9, the plotted values of tensile strength can be divided into three groups (Groups H, M, and L). Powder B ($D_{50} = 19.6 \text{ μm}$) in Group H led to the highest tensile strength, Powders D ($D_{50} = 23.3 \text{ μm}$) and A ($D_{50} = 22.0 \text{ μm}$) in Group M led to middle tensile strengths, and Powder C ($D_{50} = 34.3 \text{ μm}$) in Group L had the lowest tensile strength. This result indicates that tensile strength is dependent on the powder particle size in SLM processing.

4 Discussion

The mechanical properties and microstructure of the SLM parts are governed by the laser power, scanning speed, scanning pitch, layer thickness, and powder material. Many experimental results^{12,13)} for SLM using titanium or Ti alloy powder have been reported, and the mechanical properties are comparable to those of the forged product. In this study, based on the assumption that the metal powders are re-used in SLM processes, the effects of differences in the particle size distribution on the mechanical properties of the SLM parts were examined in detail.

In general, higher energy densities in the SLM process delivered higher relative densities of product, and this behavior was also observed in this experiment (Fig. 2). However, it was found that the highest relative density was obtained in the case of Powder B and the lowest relative density was obtained in the case of Powder C for different energy densities. This means that it is possible to manufacture a product of higher relative density at lower energies when small-sized particles are used in an SLM process. For manufacturing with SLM, it is important that when the powder layer is melted locally by a laser, the laser beam should melt at least one or more of the previous layers simultaneously to make a melt pool. When a higher energy density is supplied to the powder, the depth of the melt pool increases; therefore, interlayer defects can be easily decreased.

In the field of powder metallurgy, the higher the relative density, the higher is the tensile strength. In powder-forging and MIM, engineers devote themselves to raising the relative density. In AM technology, the strength of a product is also increased according to its increased relative density. Note that smaller powder particles result in a higher tensile strength in SLM processes. As is evident from Figs. 6 and 8, the tensile strength is affected by the powder particle size and increases in the following order: Powder B > Powder D $\geq$ Powder A > Powder C. In addition, elongation is in the range from 22% to 27% and slightly depends on powder particle size. Oxygen content is considered to be one of the causes for differences in the tensile strength. In other words, as the surface area including oxide film per unit volume of fine powder increases, the amount of oxygen that is incorporated at the time of melting (Fig. 7) also increases. When the oxygen content of tensile specimens formed using the large Powder C was 0.114 wt%, which

is smaller than the amount of oxygen (0.137 wt%) in the starting raw material powder (Powder A), the tensile strength also became low. It is presumed that the amount of oxygen in each powder (Powders B, C, and D) is changed by sieving before the SLM process.

According to the material specifications of pure titanium, the strengths of grades 1 to 4 depend on the oxygen and iron content. When handling powder in SLM processing, the change in iron content is almost negligible. However, preventing oxygen uptake due to oxidation of the powder surface is extremely difficult. The starting raw material powder is of grade 2, of which the amount of oxygen is ≤ 0.20 wt% in the JIS standard. Therefore, the four types of powder can be used as grade 2 pure titanium powders. However, Powder B, which contained many smaller particles, showed a higher strength compared with those of the other three types of powders. This suggests that oxygen content management is essentially important for maintaining the quality of a product.

According to the Japanese Industrial Standard (JIS), the tensile strength and elongation of pure titanium for Grade 2 and Grade 3 powders are ≥ 345 MPa and $\geq 20\%$ and ≥ 450 MPa and $\geq 18\%$, respectively. As can be seen from the results shown in Fig. 6, the strength and elongation of the SLM parts using for Grade 2 powder is 540~600 MPa and 22%~27%, respectively. SLM parts correspond to grade 3 or grade 4 materials.

Although the influence of oxygen content for increasing strength is described above, another reason due to which SLM can achieve high-strength products should be noted. In general, titanium powder is melted layer-by-layer to form a melt pool by supplying sufficient energy, and the pool is then rapidly cooled through the surrounding powders and solid. This leads to an acicular structure of cooled titanium metal. However, as shown in Figs. 4 and 5, the structure after SLM processing is a re-crystallized structure, and grain size caused slight differences in the four types of powder used. When the acicular structure is exposed repeatedly to high temperatures below the melting point for a short time from other melting zones during SLM processing, it may be changed into a re-crystallized microstructure¹⁴⁾.

Moreover, the specimens formed by SLM using the four types of powders showed different microstructures, as shown in Fig. 5. This may also have been caused by slight differences in their thermal histories. There exists a requirement for further investigation of microstructural changes during SLM processing.

5 Conclusion

In this study of the SLM process, the effects of titanium powder particle size on the mechanical properties and microstructures of SLM parts were investigated. The tensile strengths of the SLM

parts varied according to the powder particle sizes, although there was little change in elongation. Especially, the tensile strength is the highest when Powder B containing many smaller sized particles, nevertheless supplied energy density is same. One of reasons for this was the increase in the oxygen content during the sieving of powders and the processing of SLM, which is derived from oxidized film on the surface of titanium particles. Another is that crystal grains become fine due to repeated heat cycles after solidification caused by heat conduction from other melting zones. The metallurgical phase-change mechanism from rapidly cooled structure to re-crystallized fine structure must be investigated in detail.

Finally, the obtained results in this study, which showed that the powder particle size may govern the mechanical and metallurgical properties of SLM parts, suggest viewpoints of reusing powder in SLM processing. Therefore, to obtain stable product quality, it is necessary to establish a method for determining a suitable particle size distribution in SLM processing.

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