Effects of plasma rotating electrode process parameters on the particle size distribution and microstructure of Ti-6Al-4?V alloy powder

doi: 10.1016/j.powtec.2020.08.027

Effects of plasma rotating electrode process parameters on the particle size distribution and microstructure of Ti-6Al-4V alloy powder

Yujie Cui[a*], Yufan Zhao[a*], Haruko Numata[b], Huakang Bian[a], Kimio Wako[c], Kenta Yamanaka[a], Kenta Aoyagi[a], Akihiko Chiba[a]

[a] *Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

[b] *Department of Materials Processing, Tohoku University, Sendai 980-8577, Japan*

[c] *JAMPT Corporation, Sendai 985-0874, Japan*

* *Corresponding author. E-mail address: cuiyujie@imr.tohoku.ac.jp* (Yujie Cui*, Tel: +81-022-215-2630*)；*zhyf90215@imr.tohoku.ac.jp*（Yufan Zhao, *Tel: +81-022-215-2452*）

Abstract

The starting powder quality significantly influences the process window optimization and properties of parts fabricated by additive manufacturing. In this study, we investigate the influence of plasma rotating electrode process (PREP) parameters on the particle size distribution and microstructure of Ti-6Al-4V alloy powder. The martensite size in the powder decreased with increasing rotating electrode speed owing to the

higher cooling rate. Numerical simulations using computational thermal fluid dynamics were found to be feasible for quantitative evaluation of the temperature variation, cooling rate, and powder size during PREP, and proves to be a new method to study the mechanism of powder formation. In addition, to reduce the experimental cost, a statistical model combining principal component analysis and the Monte Carlo methods was proposed to evaluate the relationships between PREP parameters and average powder diameter based on the limited collected experimental data. The proposed statistical model can also be applied in research fields where multivariable problems exist.

Keywords: Powder fabrication; Particle size; Martensite; Cooling rate; Statistic analysis

1. Introduction

The powder bed fusion technology for additive manufacturing (AM) has attracted significant attention [1–3]. Considerable research has been conducted on the production of metal powder [4–7], whose quality has significant influence on the process window optimization and properties of AM-built parts [8]. The packing density on the build plate and the density of the AM-built parts are affected by particle size distribution [9].

The decreased particle size significantly changed the powder layer quality, thus affecting the subsequent melting process [10]. Thin layer deposition, surface quality, and mechanical properties of AM-built parts can be improved using smaller particles [11]. In addition, the higher sphericity and less amount of satellite powder contribute to the increased flowability and packing capability of metal powder, which in turn significantly affects the AM process. For instance, an ideal spherical powder without satellite powder does not result in powder interlocking due to minimized surface tension. Moreover, since the powder microstructure is closely related to the compatibility and thermophysical properties of powders at elevated temperature [12], the original microstructure of the powder has significant influence on the process parameters and the properties of the AM-built parts, especially for sintering-type AM when the powder is only partially melted. Gas-atomized (GA) powder has been extensively used in powder bed fusion technology of AM. During the gas atomization process, molten metal flows out from a nozzle, thereby impacting the inert gas jet and atomizing into particles. Some fine particles are attached to the partially molten particles during the atomization process, resulting in the formation of satellite particles. When the high-speed gas atomization jet impacts the molten metal, the gas is entrapped after solidification, contributing to the formation of pores within the solidified particles

and subsequently becoming a significant source of pore formation within the AM-built sample. The gas pores and satellite powder present in the starting GA powder result in the formation of defects, which greatly affects the mechanical properties of the AM-built parts. For instance, Shui et al. [13] reported that the pore defects can act as crack initiation sites, which deteriorated the fatigue strength of AM-built Ti-6Al-4V (Ti64) alloy. Masuo et al. [14] demonstrated that the size and number of pores in AM-built parts can be reduced by hot isostatic pressing (HIP). However, the grain growth after HIP deteriorated the mechanical properties of the AM-built parts. In addition, Tammas-Williams et al. [15] reported that pores in the as-produced Ti64 AM component reappeared in the same locations after HIP and β-annealing heat treatment. Ahmed et al. [16] studied the influences of powder porosity on powder recycling during AM. The pore size increased by 54% after repeated 10 builds, which limited the application of the recycled powder. Therefore, porosity in the AM-built materials limits the implementation of powder bed fusion technologies of AM.

In contrast, the plasma rotating electrode process (PREP) is a centrifugal atomization method, which produces powders with higher sphericity, fewer satellites, and fewer pores in comparison with GA powders [17, 18]. Although GA remains the leading

powder manufacturing technology for AM, PREP is emerging as an important powder manufacturing technology applied to AM raw materials. The PREP parameters have a close relationship with the average powder diameter. The quantitative relationships between rotation speed during supreme-speed PREP and the average powder diameter were evaluated [19]. The rotational speed of the alloy electrode mainly determined the particle size distribution. A larger percentage of fine EP741NP/Inconel718 alloy powder was obtained by increasing the rotational speed of the alloy electrode [20]. The average PREP powder size also decreased with increasing diameter of the alloy electrode [21]. Tang, et al. [22] proposed that the formation of Ti64 alloy satellite powder was related to the cooling rate during PREP. Less amounts of satellite powders were obtained at higher rotational speed, but the cooling rates were not evaluated at the different rotational speeds.

Although the relationships between PREP parameters and average powder diameter have been preliminarily studied, the effects of these parameters on the powder formation and microstructure have not yet been elucidated.

To study the influence of PREP parameters on powder formation, the cooling rate under

different PREP parameters should be studied; however, it is challenging to evaluate the cooling rate during PREP using the available experimental methods. Multiphysics simulations can be performed for analogous analysis of the cooling rate as a function of process conditions. The numerical simulations involving computational thermal fluid dynamics (CtFD) provide insights into the heat transfer and fluid flow mechanisms. In addition, because conducting PREP experiments is expensive, and several PREP parameters must be considered simultaneously, it is difficult to empirically investigate the relationships between the average powder diameter and each PREP parameter. Therefore, we propose to apply a statistical model to clarify these relationships. Principal component analysis (PCA) is an effective method to filter out the experimental "noise," which does not obey the "objective law". The Monte Carlo (MC) method can be applied to generate substantial pseudo-data by randomly sampling the known probability density functions of whole input quantities. Herein, the PCA and MC methods would be applied to clarify the intrinsic relationships between the average powder diameter and PREP parameters.

We used this combination of experiments, CtFD simulations, and statistical modeling to investigate the effects of PREP parameters on the powder diameter and microstructure

of Ti64 alloy. The powder microstructure was characterized by scanning electron microscopy (SEM), X-ray diffractometry (XRD), electron backscatter diffraction (EBSD), and transmission electron microscopy (TEM). The CtFD simulations were applied to characterize the cooling rate during PREP, and a statistical model was established to clarify the relationships between PREP parameters and average powder diameter. This statistical model can also be applied in several other process technologies, where several factors must be considered simultaneously.

2. Experimental methods

The Ti64 alloy electrodes for PREP were obtained from Aichi Steel Corporation. Figure 1 shows the schematic of the PREP atomizer used (Fuji Electronic Industrial Co., Ltd). The plasma arc was applied to melt the rapidly rotating Ti64 alloy rod. The molten metal was ejected by centrifugal forces from the Ti64 alloy rod, where it solidifies into spherical particles within the chamber. The PREP experiments were conducted in inert argon atmosphere. The Ti64 alloy powder was produced under argon gas flow plasma discharge of 6–9 kVA between cathode and anode in the PREP equipment. The cathode was a copper-tip, whereas the anode was a Ti64 alloy rod with a diameter of 15–25 mm. We chose an arc electric current of 80 A during PREP. The electrode rotating speed

ranged from 7000 to 11 700 rotations per minute (rpm). The chemical compositions of the Ti64 alloy electrode and the powder fabricated at different rotational speeds were tested by inductively coupled plasma (ICP) atomic emission spectroscopy. A laser diffraction particle size analyzer (Beckman Coulter LS230) was used to evaluate the particle size distribution. When particles floating in the dispersion medium are irradiated with light from a diode laser and a tungsten/halogen lamp, a scattering pattern corresponding to the particle size is detected. The particle size parameter a can be expressed as $a = \pi \cdot x/\lambda$, where x is the particle size and λ is the wavelength of light. Fraunhofer law was applied for measuring relatively large particles ($10<a<10^4$), while Mie law was applied for measuring relatively small particles ($a<10$). The PSD results obtained by this laser diffraction method are close to the actual dimensions of particles in case of round particles, while a deviation will be generated when fibrous particles were present [23]. Consequently, the shape of the powder should be considered. The roundness of the powder was calculated as follows [24]:

$$Roundness = \frac{4S}{\pi (F_{max})^2} \quad (1)$$

where S denotes the area of the particles, and F_{max} denotes the maximum Feret diameter, which is defined as the distance between the parallel lines of the two projections of the particle projection profile. The surface morphology of the powder was observed by

SEM (Hitachi, Tokyo, Japan). XRD (Malvern PANalytical, Netherlands) was conducted to characterize the phase composition of the powder. X-ray computed tomography (X-CT) (Comscantecno Co., Ltd, Yokohama, Japan) with a resolution of 10 μm was conducted to verify the porosity of the powder. To observe the cross-section of the powder, the samples were hot-mounted, ground, and polished using an automatic lapping machine with an oxide polishing suspension. The cross-sectional samples were observed by EBSD using a data acquisition software (TSL Solutions, Japan), with a step size of 0.04 μm. Focused ion beam milling (FEI QUANTA 200 3D) was applied to prepare samples for TEM observation. A double-Cs-corrected G2 60-300 Titan 3 TEM was applied to conduct high-angle annular dark-field scanning TEM (HAADF-STEM) observations at an operating voltage of 300 kV. STEM energy-dispersive X-ray (EDX) mapping was used to identify the elemental distribution of the Ti64 alloy. In addition, PCA and MC methods were applied to build a statistical model using R studio software developed by R Core Team [25].

3. Numerical simulation

To analyze the cooling rate during PREP under various process conditions, CtFD simulations were carried out. A three-dimensional transient model of the melt pool was

established by applying a commercial CtFD software FLOW-3D® [26]. The fluid-free surface was modeled using the volume-of-fluid method [27], wherein index F represents the volume fraction of the fluid occupying each grid in the computational domain:

$$\frac{\partial F}{\partial t} + \nabla \cdot \left(F\vec{V}\right) = 0 \tag{2}$$

where $0 \leq F \leq 1$ and $\vec{V}$ is the fluid velocity.

Since the qualitative simulation results are valid and applicable to the present research, some simplifications have been made to the modeling. Fig. 2 shows a circular fluid source placed on the end face of the rotating electrode instead of the melting process by a heat source. Moreover, the size of the electrode and fluid source and the corresponding fluid supply rate were scaled down to reduce the burden of calculation. Diameters of 2 mm and 1 mm were set for the electrode and fluid source, respectively. The scaled down fluid supply rate (0.025 g/s) was determined by the melting rate, which was calculated from the mass of the powder produced and the corresponding processing time. In the practical PREP process, the heating of the plasma arc can result in the instantaneous temperature of the alloy exceeding its vaporization temperature. Thus, the initial temperatures of the fluid source and electrode were set to 3000 K (slightly lower than the boiling point of Ti64) and 1923 K (liquidus of Ti64),

respectively. Concerning the molten alloy, the thermal conductivity, specific heat, viscosity, and density were evaluated as a function of temperature; surface-effect terms of fluid contained capillary and Marangoni forces. The capillary force, $\gamma c\vec{n}$, is dependent on the curvature of the melt surface c, and is caused by the surface tension; γ. $\gamma c\vec{n}$ acts in the normal direction, $\vec{n}$, to the melt surface, thereby limiting the melt volume and minimizing the surface tension potential. The Marangoni force is produced by the temperature-dependent surface tension:

$$\gamma(T) = \gamma_{\mathrm{L}} + \frac{d\gamma}{dT}(T - T_{\mathrm{L}}) \tag{3}$$

where γ_{L} is the surface tension (J/m^2) at the referenced temperature (liquidus) and $d\gamma/dT$ is the temperature coefficient of surface tension (J/m^2·K). The Marangoni force that acts along the tangent of the melt surface is expressed as follows:

$$\frac{d\gamma}{dx} = \frac{d\gamma}{dT}\nabla T \tag{4}$$

where ∇T denotes the temperature gradient (K/m) of the melt surface. In addition to thermal conduction, the heat radiation (j^*) at temperature (*T)* was also activated and expressed by the Stefan–Boltzmann law:

$$j^* = \epsilon\sigma(T^4 - T_0^4) \tag{5}$$

where ϵ is the material emissivity, σ denotes the Stefan–Boltzmann constant (W/m^2·K^4), and T_0 denotes the ambient temperature (K). Within the computational

domain, the electrode was surrounded by a void region with a pressure of 0.1 MPa. Thermal conduction between fluid and void (atmosphere) was simulated by setting a heat transfer coefficient (h_{FV}) for fluid to void.

The thermophysical properties of Ti64 alloy and some of the coefficients applied in the simulation are shown in Table 1. Some of the properties were taken from the software database, while the others were obtained from the research by Boivineau et al. [28]. Regarding the computational cycle, the input data contained an automatic time-step selection configuration, where the time-step size was adjusted according to the stability conditions. These conditions meet the rule that no quantity should diffuse more than approximately one mesh cell during a single time-step. An available simulation with a processing cycle time of 0.06 s took approximately 49 h to finish by employing an Intel® Xeon® CPU E5-2683 v4 (2.10 GHz)×2 with 96 GB of RAM. Based on the simulation results, the cooling rates (dT⁄dt) were calculated by differentiating the spatial-temporal thermal field of the fluid.

$$\frac{dT}{dt} = \frac{T_t - T_{t-1}}{\Delta t} \tag{6}$$

Table 1. Thermophysical properties of Ti64 alloy and the coefficients used in the simulation.

Name	Symbol and unit	Value
Density	ρ $(\mathrm{g/cm^3})$	4.42-3.75
Viscosity	μ $(\mathrm{mPa \cdot s})$	3.5-2.06
Thermal conductivity	κ $(\mathrm{W/m \cdot K})$	7-34.6
Specific heat	C_P $(\mathrm{J/kg \cdot K})$	546-831
Heat transfer coefficient for fluid to void	h_FV $(\mathrm{W/m^2 \cdot K})$	1000
Emissivity	ε	0.40
Surface tension at T_L	γ_L $(\mathrm{J/m^2})$	1.53
Temperature coefficient of surface tension	$\frac{d\gamma}{dT}(\mathrm{J/m^2 \cdot K})$	-0. 00028
Stefan–Boltzmann constant	σ $(\mathrm{W/m^2 \cdot K^4})$	5.67e-08
Environment pressure	P_0 (Pa)	0.01
Environment temperature	T_0 (K)	298

4. Results

Table 2 shows the chemical compositions of the Ti64 electrode and alloy powders fabricated at rotational speeds of 7000, 9000, and 11 700 rpm and tested by ICP. The contents of Al and V in the powder decreased with slower speeds compared to the original Ti64 alloy electrode. For example, the contents of Al and V decreased from 6.29% and 4.36% in the electrode to 5.82% and 4.00% in the powder fabricated at the rotational speed of 11 700 rpm, respectively. The abovementioned result is mainly attributed to the vaporization of Al and V during PREP. Moreover, such vaporization

was more significant in the powder fabricated at lower rotational speeds, owing to the longer contact time between molten metal and plasma.

Table 2. The chemical compositions of the Ti64 alloy powder fabricated at various rotational speeds (mass, %). (Rotating electrode diameter: 20 mm).

Samples	Al	V	H	O	N	Fe	C
Ti64 electrode	6.2900	4.3600	0.0031	0.1800	0.0040	0.2020	0.0050
Powder (7000 rpm)	5.5900	3.8850	0.0020	0.1690	0.0072	0.2100	0.0077
Powder (9000 rpm)	5.7350	3.9800	0.0020	0.1675	0.0021	0.2050	0.0067
Powder (11 700 rpm)	5.8200	4.0000	0.0020	0.1835	0.0089	0.2150	0.0078

Near-spherical Ti64 alloy powder without satellites was obtained at different rotation speeds and diameters of the electrode, as shown in Fig. 3. The calculated powder roundness using the present conditions by equation 1 was higher than 0.95. Fig. 4 shows the particle size distribution of Ti64 alloy powders produced using various PREP parameters. The peak position of particle size distribution decreased with an increase in the rotational speed from 7000 to 11 700 rpm or increased electrode diameter from 15 to 25 mm. Moreover, the average powder diameter also decreased with an increasing rotational speed or diameter of the Ti64 alloy electrode, as shown in Fig. 5. Consequently, increasing the rotational speed or enlarging the electrode diameter is effective in producing smaller-sized Ti64 alloy powder. The cross-sectional CT images

in Fig. 6 show that no pores above the CT resolution of 10 μm were observed in the Ti64 alloy powder at different rotational speeds.

Fig. 7 depicts the inverse pole figures (IPF) of the Ti64 alloy powder fabricated at 7000, 9000, and 11 700 rpm. The microstructure mainly consisted of martensite with an acicular/lath morphology. The average martensite-crystal width decreased from 2.03 μm at 7000 rpm to 1.05 μm at 11 700 rpm, which was mainly ascribed to the increase in the cooling rate. During the PREP cooling process, the initial β phase transforms to martensite that obeys the Burgers orientation relationship, i.e., $\{\bar{1}10\}_\beta//(0001)_\alpha$, $\langle 111\rangle_\beta//\langle 11\bar{2}0\rangle_\alpha$ [29]. Fig. 8 shows the misorientation angle distribution in Ti64 alloy powder fabricated at the rotational speed of 11 700 rpm. Four peaks are observed around 10°, 60°, 63°, and 90°. The corresponding rotation axes around the peaks are shown in the IPF curves (Fig. 8). This is consistent with misorientation distribution existing in the 12 possible martensite variants transformed from prior β phase [30]. The rotation axis is [0 0 0 1] (type □) and [1 1 $\bar{2}$ 0] (type □) around the peak of 10.53° and 60°, respectively. The rotation axis is [$\overline{1.377}$ $\bar{1}$ 2.377 0.359] (type □) around a peak of 60.83° and [$\overline{10}$ 5 5 $\bar{3}$] (type □) around a peak of 63.26°. The peak around 90° is related to the rotation axis of [1 $\overline{2.38}$ 1.38 0] (type □).

TEM analyses were conducted to further verify the martensite structure. Fig. 9 shows that a high density of dislocations was generated in the acicular/lath microstructure, which is characteristic of martensite transformed from the prior β phase [31]. Moreover, some twins were observed in the Ti64 alloy powder fabricated at 11 700 rpm, as indicated by the white arrows in Fig. 9(b). In contrast, twins were not observed in the Ti64 alloy powder fabricated at 7000 rpm, as shown in Fig. 9(a). Twin formation was closely related to the cooling rate within the temperature range of martensite transformation (823–1123 K) [32]. Twins were not observed in the martensite at a cooling rate lower than 10^4 K/s. Herein, the twinning was promoted at higher rotational speeds, owing to the higher cooling rate. The martensite size in the Ti64 alloy powder fabricated at 11 700 rpm was smaller than that formed at 7000 rpm, as shown in Fig. 9. Fig. 10 shows the XRD patterns of Ti64 alloy powder fabricated at different rotation speeds. Only the peaks of martensite were detected. To further verify whether all the prior β phase transformed to martensite during PREP, STEM-EDX experiments were conducted. The EDX maps in Fig. 11 of Ti64 alloy powder show that no aggregation of V occurred in the microstructure, indicating that no β phase remained, as V generally aggregates into the β phase.

5. Discussion

Ti64 alloy powder without pores and satellites was obtained by PREP. The average powder size was decreased by increasing the rotational speed or diameter of the electrode. Relatively small-sized martensite was verified at high rotational speeds owing to correspondingly high cooling rates. The dependence of the cooling rate variation and atomization mechanism on the PREP parameters are discussed in the following sections.

5.1 Cooling rate during PREP

The cooling rate when the particles hit the wall is significantly higher than that during their flight in the chamber. However, most of obtained PREP powder is essentially of perfect spherical shape without deformation as shown in Fig. 3; this proves that the droplet solidified before it hit the chamber wall. Therefore, we mainly consider the cooling rate during the flight of the droplet. Fig. 12 shows the simulation snapshots with the contours of the cooling rate during the PREP process at different rotational speeds. Most particles experience a high cooling rate that reached a level of 1×10^3–1×10^4 K/s. Although there might be some inaccuracy because these are qualitative simulation results, the range of cooling rates conformed to those in actual processing. Because of the hot fluid supply and strong convection, heat exchange occurred within

the melt and inside the droplet. The convection can heat specific parts of a droplet; therefore, a negative cooling rate was also observed (blue regions in Fig. 12). The cooling rate during PREP was higher than the critical cooling rate for martensite formation (410 K/s) [32], which is consistent with the powder microstructure with martensite, as shown in Figs. 7 and 9. The variations in maximum cooling rate with processing time derived from simulations at different rotation speeds are shown in Fig. 13. The beginning of the fluid source supply (0 s) was the starting point of the simulation. There was a transition period between the initial and steady state of the fluid centrifugal motion (approximately 0.02 s). From 0.02 to 0.06 s, the maximum cooling rate increased as the rotational speed was increased from 7000 to 11 700 rpm. Greater heat transfer from fluid to void (atmosphere) occurred at higher rotational speeds. The heat transfer rate $\dot{Q}$ is expressed as follows:

$$\dot{Q} = h_{\mathrm{FV}} \cdot A(T_{\mathrm{F}} - T_0) \tag{7}$$

where A is the specific surface area of the fluid droplets; h_{FV} denotes the coefficient of heat transfer; and T_{F} and T_0 are the temperatures of the fluid and environment, respectively. The cooling rate of droplets are affected by (i) the convective heat transfer between the droplets and the filling gas in the tank and (ii) the heat radiation from the droplets surface. The linear velocity of the electrode edge increases, and the initial

velocity when the droplets fly out increases, which increase the heat convection between the droplets and the filling gas, thus increasing the cooling rate. Secondly, under high rotational speeds, the droplet size decreases; however, the melting rate does not change significantly under the same arc current. The specific surface area A of the liquid flying out of the electrode increases, which, in turn, increases the heat radiation from the droplets to the environment (heat loss rate), thereby increasing the cooling rate. Subsequently, the accelerated cooling rate promoted martensite nucleation with increased rotational speed, resulting in the formation of smaller-sized martensite, as shown in Figs. 7 and 9.

In addition, agglomerates of droplets were observed in the periphery of the electrode. This phenomenon occurred more frequently at lower rotational speeds, as shown in the magnified images at the right upper positions in Fig. 12, contributing to the larger particle size.

Our results indicated the applicability of the CtFD simulations to predict the temperature variation, cooling rate, and powder size during PREP. Thus, it is feasible to predict and analyze the influence of more factors and provide

suggestions for the refinement of powder particles. Furthermore, the CtFD simulation involves the flow field and the free surface of the fluid; therefore, for the high-speed centrifugal granulation process during PREP, the granulation behavior and disintegration mode, which are difficult to observe through experimental means, may be studied. In particular, CtFD simulation is potentially ideal to study the mechanism of powder formation so as to develop novel methods to control the particle size of the powder.

5.2 Atomization mechanism during PREP

Three disintegration models were reported for PREP [33, 34], namely, direct drop formation (DDF), ligament disintegration (LD), and film disintegration (FD). Primary and secondary particles are formed in the case of DDF, resulting in two dependent peaks in the particle size distribution. The model transitions from DDF to LD with an increased melting rate and a higher electrode rotating speed. One main peak exists in the particle size distribution in the case of LD. The FD model occurs under extremely high electrode rotational speeds, where the liquid metal flows rapidly from the molten surface of the electrode, spreading out in the form of a film and breaks up into agglomerates owing to surface tension, and forms small droplets. The classification of

the disintegration model is determined by the Hinze–Milborn number (Hi) [20]:

$$Hi = \frac{\mu^{0.17} Q \rho^{0.71} \omega^{0.6}}{\gamma^{0.88} D^{0.68}} \tag{8}$$

where μ denotes the viscosity of liquid metal, Q denotes the melting rate (Q was calculated from the mass of the powder produced and the corresponding processing time), ρ is the density of liquid metal, ω denotes the rotational speed, γ represents the surface tension of liquid metal, and D is the electrode diameter. The disintegration model is DDF when Hi is smaller than 0.07, changes to LD when Hi is between 0.07 and 1.33 and is FD if Hi is larger than 1.33. In this study, PREP conditions were calculated using equation 8 with Hi values ranging from 0.05 to 0.12. This indicates that the disintegration modality in the present study is a combination of DDF and LD. Further, the CtFD simulated scenarios shown in Fig. 12 also represent the transition between DDF and LD, which indicates the equivalence and reliability of the simulation. The average DDF powder diameter, d_{DDF}, can be calculated as follows [20]:

$$d_{\mathrm{DDF}} = \frac{1}{\omega}\sqrt{\frac{12\gamma}{\rho D}} \tag{9}$$

and the average LD powder diameter, d_{LD}, can be obtained as [35]

$$d_{\mathrm{LD}} = 2.0\frac{1}{\omega}\sqrt{\frac{\gamma}{\rho D}} \tag{10}$$

where ω denotes the rotational speed, γ represents the surface tension of liquid Ti64

alloy, ρ is the density of liquid Ti64 alloy, and D stands for the electrode diameter. Fig. 14 shows the calculated average powder size using equations 9 and 10 based on the utilized PREP parameters. We found that the experimental results were between calculated DDF values and LD values. It is challenging to predict the average diameter with high accuracy using DDF or LD model. Thus, new models are required to better describe the relationship between the average powder diameter and PREP parameters.

5.3 Statistical model

Several PREP parameters affect the average powder size simultaneously, and it would be costly to investigate the relationships between all the variables and average powder size. In this study, we applied a combination of PCA and MC methods to establish a statistical model based on the obtained experimental data.

Fig. 15 shows the schematic of the statistical model by combining the PCA and MC methods. The experimental data was first obtained at selected PREP conditions, and experimental data in the original space was converted to its feature space and combined with its eigen subspace. The low eigenvalues related to the experimental errors can be separated and ignored. The high eigenvalues remained in the PCA model. Subsequently,

MC method was applied to randomly generate 20 000 pseudo-data points basing on the data processed by the PCA model. Fig. 16 shows the variation of average powder size with the rotating electrode diameter, illustrating the significantly expanded results compared to the initial experimental data shown in Fig. 5. The diameters of the rotating electrode range from 8.5 to 35.0 mm in the statistical model, whereas there were only three electrode diameters (15, 20, and 25 mm) used in the experimental results. The black points indicate all the generated 20 000 data points; yellow, green, and red points indicate the generated data at the rotational speed of 7000, 9000, and 11 700 rpm, respectively.

The results indicate that the average powder size decreased linearly with increasing diameter of the rotating electrode, ranging from 15 to 30 mm. In addition, the average powder size at some conditions can be predicted by this statistical model. For instance, Fig. 17 shows the predicted average powder diameter at the rotational speed of 10 000 rpm while the PREP experiment was not conducted yet in this condition. To verify the validation of prediction based on the obtained data using the statistical model, the PREP experiment was conducted at 10 000 rpm after the prediction. The experimental data at 10 000 rpm are shown in Fig. 17 (the pink points). Only small deviations exist between

experimental data and predicted data. The statistical model was established based on the obtained experimental results within a specific range in this study. More experimental data are required to investigate the relationships between the average powder size and PREP parameters at more extensive ranges. The PREP equipment will be revised to obtain more experimental data under the conditions of larger electrode diameter and higher rotational speeds in our future studies.

In addition to the effects of the diameter and rotational speed of the alloy electrode, those of the plasma power and cooling gas on the average powder size during PREP will be investigated in future research. The statistical model combining PCA and MC models can exhibit greater advantages when more PREP parameters are considered simultaneously. It can thus become an efficient method to clarify the relationship between the average powder size and any one of many PREP parameters, based on limited experimental data collected thus far. This statistical model can also be helpful in building a database for industrial production of PREP powder under various processing conditions. The average powder diameter can be predicted using the statistical model, and the production of differently sized powder according to the requirements can be feasible.

6. Conclusions

The effects of PREP parameters on average powder size and powder microstructure of Ti64 alloy were investigated via experiments, CtFD simulations, and statistical modeling. The results are summarized as follows:

(1) Increasing either the rotational speed or diameter of the electrode can effectively decrease the average powder size of Ti64 alloy.

(2) CtFD simulations can be applied to characterize the temperature variation and cooling rate during PREP. The cooling rate was enhanced by increasing the rotational speed of the electrode.

(3) The microstructure of Ti64 alloy PREP powder consisted entirely of martensite, with relatively small martensite crystals forming at high rotational speeds; this is attributed to the high cooling rates at high speeds.

(4) The statistical model combining the PCA and MC models can be applied to investigate the relationships between PREP parameters and average powder size. The validity of the statistical model for predicting average powder size within the evaluated range was also demonstrated.

To summarize, powder fabrication by PREP can ensure powder quality in terms of

high sphericity and low porosity. However, owing to the power limitation of the present equipment, it is difficult to attain extremely high rotational speeds. Further manipulation of the PREP parameters to produce a finer powder and free control of the powder size distribution are the subsequent efforts to be pursued. Enhancing the motor power of the PREP equipment and the diameter of rotating electrode will be carried out after modifying the current equipment. The production of improved PREP powder appropriate for AM at reduced production costs through the development of a PREP atomizer with higher efficiency will continue to be our goal.

Figures

Fig. 1. Schematic of PREP atomizer.

Fig. 2. The PREP model established by Flow-3D. In the computational domain, a circular fluid source placed on the end face of the rotating electrode was used instead of melting by a heat source. The initial temperature of the fluid source and electrode were set to 3000 and 1923 K, respectively.

Fig. 3. SEM images of powder fabricated at different rotational speeds and diameters of the Ti64 alloy electrode.

Fig. 4. (a, b, c) Particle size distributions and (d) corresponding peak positions of Ti64 alloy powder fabricated at various PREP parameters. Electrode diameter: (a) 15, (b) 20, and (c) 25 mm. Rotational speeds: 7000, 9000, and 11 700 rpm.

Fig. 5. Variation of average powder diameter with rotational speed and diameter of the Ti64 alloy electrode.

Fig. 6. Cross-sectional CT images of Ti64 alloy powder fabricated at the rotational speeds of (a) 7000, (b) 9000, and (c) 11 700 rpm (rotating electrode diameter: 20 mm).

Fig. 7. (a, b, c) IPF maps and (d, e, f) boundary maps of Ti64 alloy powder fabricated at the rotational speeds of (a, d) 7000, (b, e) 9000, and (c, f) 11 700 rpm (rotating electrode diameter: 20 mm).

Fig. 8. Misorientation angle distribution in Ti64 alloy powder fabricated at the rotational speed of 11 700 rpm (rotating electrode diameter: 20 mm).

Fig. 9. Bright-field images of Ti64 alloy powder fabricated at the rotational speeds of (a) 7000 and (b) 11 700 rpm (rotating electrode diameter: 20 mm).

Fig. 10. XRD patterns of Ti64 alloy powder fabricated at various rotational speeds (rotating electrode diameter: 20 mm).

Fig. 11. (a, e) Dark-field (DF)-HAADF images and (b-d, f-h) STEM-EDX maps of Ti64 alloy powder fabricated at the rotational speed of 11 700 rpm (rotating electrode

diameter: 20 mm).

Fig. 12. Simulation snapshots with contour of the cooling rate during PREP process at rotational speeds of (a) 7000 rpm, (b) 9000 rpm, and (c) 11 700 rpm.

Fig. 13. Variation of maximum cooling rate derived from simulations with processing time at different rotational speeds.

Fig. 14. Experimental results and the calculated results based on DDF and LD models at different rotational speeds (rotating electrode diameter: 20 mm).

Fig. 15. Schematic of the statistical model using the combination of the PCA and MC methods.

Fig. 16. Generated data based on the experimental data in Fig. 5 by using the combination of the PCA and MC methods. Black points indicate all generated data; yellow, green, and red points indicate the generated data at the rotational speeds of 7000, 9000, and 11 700 rpm, respectively.

Fig. 17. Experimental verification of predicted data based on the statistical model. Black points indicate all generated data; purple points indicate the generated data at the rotational speed of 10 000 rpm; and pink points indicate the supplemental experimental data at the rotational speed of 10 000 rpm.

Table 1. Thermophysical properties of the Ti64 alloy and coefficients used in the

simulation.

Table 2. Chemical compositions of Ti64 alloy powder fabricated at various rotational speeds (mass%) (rotating electrode diameter: 20 mm).

Acknowledgement

The authors are grateful for the financial support from the Cross-ministerial Strategic Innovation Promotion Program (SIP). Yujie Cui acknowledges the financial support from the Japan Society for the Promotion of Science (Grant no. 20K14619).

CRediT authorship contribution statement

Yujie Cui: Investigation, Methodology, Writing - original draft, Visualization. **Yufan Zhao**: Investigation, Methodology, Writing - original draft, Visualization. **Haruko Numata**: Investigation. **Huakang Bian**: Writing - review & editing. **Kimio Wako**: Investigation. **Kenta Yamanaka**: Writing - review & editing. **Kenta Aoyagi**: Writing - review & editing. **Akihiko Chiba**: Funding acquisition, Supervision, Project administration, Writing - review & editing.

References

[1] W. Ding, G. Chen, M. Qin, Y. He, X. Qu, Low-cost Ti powders for additive manufacturing treated by fluidized bed, Powder Technol. 350 (2019) 117–122. https://doi.org/10.1016/j.powtec.2019.03.042

[2] A.A. Kadyrov, V.Y. Frolov, B.A. Yushin, Numerical Simulation of a Plasma Jet with Several Electrodes Used in the Technology of Creating Powders for 3d Printers, 2019 IEEE Conference of Russian Young Researchers in Electrical and Electronic Engineering (EIConRus), Saint Petersburg and Moscow, Russia (2019) 808-811. https://doi: 10.1109/EIConRus.2019.8657308.

[3] G. Zeng, Z. Han, S. Liang, P. Zhang, X. Chen, P. Zhang, The applications and progress of manufacturing of metal parts by 3D printing technology, Materials China 33 (2014) 376-382. https://doi.org/10.7502/j.issn.1674-3962.2014.06.09

[4] G. Chen, Q. Zhou, S.Y. Zhao, J.O. Yin, P. Tan, Z.F. Li, Y. Ge, J. Wang, H.P. Tang, A pore morphological study of gas-atomized Ti-6Al-4V powders by scanning electron microscopy and synchrotron X-ray computed tomography, Powder Technol. 330 (2018) 425–430.

[5] J.M. Park, T. Na, H. Park, S. Yang, J. Kang, T.W. Lee, Preparation and characterization of spherical niobium silicide-based powder particles by electrode

induction gas atomization, Mater. Lett. 243 (2019) 5–8. https://doi.org/10.1016/j.matlet.2019.01.155

[6] T.W. Na, K.B. Park, S.Y. Lee, S.M. Yang, J.W. Kang, T.W. Lee, J. M. Park, K. Park, H.K. Park, 2020. Preparation of spherical TaNbHfZrTi high-entropy alloy powders by a hydrogenation-dehydrogenation reaction and thermal plasma treatment, J. Alloys Compd. 817, 152757. https://doi.org/10.1016/j.jallcom.2019.152757

[7] A.A. Kadyrov, B.A. Yushin, V.Y. Frolov, Investigation of the Properties of a Two-Jet Arc Plasma Torch to Obtain a Metal Powder, 2020 IEEE Conference of Russian Young Researchers in Electrical and Electronic Engineering (EIConRus), St. Petersburg and Moscow, Russia (2020) 1024-1026. https://doi: 10.1109/EIConRus49466.2020.9039081.

[8] W.S.W. Harun, N.S. Manam, M.S.I.N. Kamariah, S. Sharif, A.H. Zulkifly, I. Ahmad, H.Miura, A review of powdered additive manufacturing techniques for Ti-6Al-4V biomedical applications, Powder Technol. 331 (2018) 74-97. https://doi.org/10.1016/j.powtec.2018.03.010

[9] Q.B. Nguyen, M.L.S. Nai, Z. Zhu, C.N. Sun, J. Wei, W. Zhou, Characteristics of inconel powders for powder-bed additive manufacturing, Engineering, 3 (2017)

695-700. https://doi.org/10.1016/J.ENG.2017.05.012

[10] C. Meier, R. Weissbach, J. Weinberg, W.A. Wall, A.J. Hart, Critical influences of particle size and adhesion on the powder layer uniformity in metal additive manufacturing. J. Mater. Process. Technol. 266 (2019) 484-501. https://doi.org/10.1016/j.jmatprotec.2018.10.037

[11] A.B. Spierings, N. Herres, G. Levy, Influence of the particle size distribution on surface quality and mechanical properties in AM steel parts, Rapid Prototyp. J. 17 (2011) 195-202. https://doi.org/10.1108/13552541111124770

[12] N. Minoru, M. Yasuhiro, K. Tomoaki, S. Jun, C. Akira, K. Ryohei, Microstructure dependence of compactibility of rapidly solidified Ti-rich TiAl alloy powders produced by plasma rotating electrode process, Mater. Trans. 38 (1997) 334‑343. https://doi.org/10.2320/matertrans1989.38.334

[13] X. Shui, K. Yamanaka, M. Mori, Y. Nagata, A. Chiba, Effects of post-processing on cyclic fatigue response of a titanium alloy additively manufactured by electron beam melting, Mater. Sci. Eng. A 680 (2017) 239–248. https://doi.org/10.1016/j.msea.2016.10.059

[14] H. Masuo, Y. Tanaka, S. Morokoshi, H. Yagura, T. Uchida, Y. Yamamoto, Y. Murakami, Influence of defects, surface roughness and HIP on the fatigue strength

of Ti-6Al-4V manufactured by additive manufacturing, Int. J. Fatigue 117 (2018) 163–179. https://doi.org/10.1016/j.ijfatigue.2018.07.020

[15] S. Tammas-williams, P.J. Withers, I. Todd, P.B. Prangnell, Porosity regrowth during heat treatment of hot isostatically pressed additively manufactured titanium components. Scr. Mater. 122 (2016) 72–76. https://doi.org/10.1016/j.scriptamat.2016.05.002

[16] F. Ahmed, U. Ali, D. Sarker, E. Marzbanrad, K. Choi, Y. Mahmoodkhani, E. Toyserkani, 2020. Study of powder recycling and its effect on printed parts during laser powder-bed fusion of 17-4 PH stainless steel. J. Mater. Process. Technol. 278, 116522. https://doi.org/10.1016/j.jmatprotec.2019.116522

[17] G. Chen, S.Y. Zhao, P. Tan, J. Wang, C.S. Xiang, H.P. Tang, A comparative study of Ti-6Al-4V powders for additive manufacturing by gas atomization, plasma rotating electrode process and plasma atomization. Powder Technol. 333 (2018) 38–46. https://doi.org/10.1016/j.powtec.2018.04.013

[18] G. Zeng, X. Mao, Y. Zhao, C. Huang, S. Liang, Z. Han, F. Yan, P. Zhang, Microstructures and Mechanical Properties of Hot Isostatic Pressed Parts of Ti-6Al-4VELI Alloy Spherical Powders with Three Different Diameter Distributions, Rare Metal Materials and Engineering 46 (2017) 3197–3203.

https://doi.org/10.1016/s1875-5372(18)30028-6.

[19] Y. Liu, X. Zhao, Y. Lai, Q. Wang, L. Lei, S. Liang, A brief introduction to the selective laser melting of Ti6Al4V powders by supreme-speed plasma rotating electrode process, Progress in Natural Science: Materials International 30 (2020) 94-99. https://doi.org/10.1016/j.pnsc.2019.12.004

[20] Y. Liu, S. Liang, Z. Han, J. Song, and Q. Wang, A novel model of calculating particle sizes in plasma rotating electrode process for superalloys, Powder Technol. 336 (2018) 406–414. https://doi.org/10.1016/j.powtec.2018.06.002

[21] M. Zdujić, D. Uskoković, Production of atomized metal and alloy powders by the rotating electrode process, Sov. Powder Metall. Met. Ceram. 29 (1990) 673‑683. https://doi.org/10.1007/BF00795571

[22] J. Tang, Y. Nie, Q. Lei, Y. Li, Characteristics and atomization behavior of Ti-6Al-4V powder produced by plasma rotating electrode process. Adv. Powder Technol. 30 (2019) 2330–2337. https://doi.org/10.1016/j.apt.2019.07.015

[23] A. Califice, F. Michel, G. Dislaire, E. Pirard, Influence of particle shape on size distribution measurements by 3D and 2D image analyses and laser diffraction, Powder Technol. 237 (2013) 67‑75. https://doi.org/10.1016/j.powtec.2013.01.003

[24] M.N. Pons, H. Vivier, K. Belaroui, B. Bernard-Michel, F. Cordier, D. Oulhana,

J.A. Dodds, Particle morphology: from visualisation to measurement. Powder Technol. 103 (1999) 44-57. https://doi.org/10.1016/S0032-5910(99)00023-6

[25] R Core Team, 2016. R: A Language and Environment for Statistical Computing. https://www.r-project.org/

[26] FLOW-3D® Version 11.2 [Computer software]. (2017). Santa Fe, NM: Flow Science, Inc. https://www.flow3d.com.

[27] C.W. Hirt, B.D. Nichols, Volume of fluid (VOF) method for the dynamics of free boundaries. Journal of Computational Physics 39 (1981) 201–225. https://doi.org/10.1016/0021-9991(81)90145-5

[28] M. Boivineau, C. Cagran, D. Doytier, V. Eyraud, M.H. Nadal, B. Wilthan, G. Pottlacher, Thermophysical properties of solid and liquid Ti-6Al-4V (TA6V) alloy. International Journal of Thermophysics 27 (2006) 507–529. https://doi.org/10.1007/PL00021868

[29] R. Banerjee, A. Genç, P.C. Collins, H.L. Fraser, Comparison of microstructural evolution in laser-deposited and arc-melted In-Situ Ti-TiB composites. Metall. Mater. Trans. A 35 (2004) 2143–2152. https://doi.org/10.1007/s11661-004-0162-0

[30] H. Beladi, Q. Chao, G.S. Rohrer, Variant selection and intervariant crystallographic planes distribution in martensite in a Ti-6Al-4V alloy. Acta Mater.

80 (2014) 478–489. https://doi.org/10.1016/j.actamat.2014.06.064

[31] M. Matsumoto, H. Yoneda, K. Sato, S. Kurosu, E. Maire, D. Fabregue, T.J. Konno, A. Chiba, Room-temperature ductility of Ti–6Al–4V alloy with α' martensite microstructure. Mater. Sci. Eng. A 528 (2011) 1512–1520. https://doi.org/10.1016/j.msea.2010.10.070

[32] S. Cao, B. Zhang, Y. Yang, Q. Jia, L. Li, S. Xin, X. Wu, Q. Hu, C. Lim, On the role of cooling rate and temperature in forming twinned α' martensite in Ti-6Al-4V. Journal of Alloys and Compounds 813 (2020) 152247. https://doi.org/10.1016/j.jallcom.2019.152247

[33] B. Champagne, R. Angers, REP ATOMIZATION MECHANISMS. Powder Metall. Int. 16 (1984) 125–128.

[34] K. Isonishi, M. Tokizane, Production of Ti-alloy powder by plasma rotating electrode process. Tetsu-to Hagane. 12 (1990) 2108–2115. https://doi.org/ 10.4144/rpsj1986.37.215

[35] T. Kamiya, A. Kayano, Disintegration of viscous fluid in the ligament state purged from a rotating disk, J. Chem. Eng. Japan 4 (1971) 364‑369. https://doi.org/10.1252/jcej.4.364