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Impact of Electron Beam Melting process recycling on defects and microstructure of Ti-6Al-4V powders

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Abstract

Metal powders are used as the feedstock material during the Electron Beam Melting processes. The process involves using an electron beam as the energy source to produce intricate parts with complex shapes in a layer-by-layer production system. The electron beam is directed by information from an STL file, and the process takes place in a pre-heated chamber that is maintained under vacuum. Once the production cycle is complete, the process yields the desired components along with a certain amount of residual powders that were not melted.
To improve process efficiency and reduce costs associated with powder atomization, it is feasible to reuse the excess powder for subsequent production cycles. Prior to starting a new cycle, the excess powder is initially sieved to ensure a more uniform powder batch, and subsequently, the sieved powder can be mixed with other virgin powder to decrease oxygen content.
This study examines the microstructure and defects present in a batch of virgin powders produced through plasma atomization and a batch of powders that were reused five times and mixed with Ti-6Al-4V grade 23 (ELI powders) at each cycle. The ELI powders are characterized by a low oxygen content.
The results of the analysis indicate a connection between the powder atomization process and the formation of porosities in virgin powders resulting from trapped gas, as well as surface irregularities, including the presence of satellites. With an increase in the number of reuses, there is a reduction in the number of satellites, potentially due to surface partial melting due to the pre-heating of the EBM chamber, leading to rougher surfaces on the recycled particles.
The microstructure of virgin powders is predominantly characterized by a fine acicular α’ phase, known as martensitic, formed due to the rapid cooling rate during the atomization process. Conversely, recycled powder tends to exhibit a coarser grain microstructure

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due to lower cooling rates. However, it is common to observe particles with a microstructure similar to that of newly manufactured powders, indicating that each individual particle has undergone a distinct thermal history.

Keywords: Additive Manufacturing; Ti-6Al-4V; Electron Beam Melting; powder recycling; internal porosity; microstructure; external defects.

1. Introduction

The ISO/ASTM52900 standard (ISO/ASTM, 2015) defines Powder Bed Fusion (PBF) as an Additive Manufacturing (AM) process in which specific regions of a powder bed are selectively fused by thermal energy. PBF techniques are regarded as the most effective approach to achieve high reproducibility and dimensional accuracy, and this is why these processes have been extensively studied in both the industrial and academic fields (Mostafaei et al., 2022). The most well-known processes under this category for producing metallic components are Electron Beam Melting (EBM) and Selective Laser Melting (SLM).

These two technologies are primarily distinguished by their ability to produce highly intricate shapes such as medical instruments and customized implants (Ishfaq et al., 2022), complex lattice structures (Bellini et al., 2021), aerospace components with high precision (Gardan & Schneider, 2015), and even custom-shaped jewelry (Cooper, 2016), through the addition of materials layer-by-layer in a single production cycle. EBM processes are primarily distinguished from SLM processes by the utilization of an electron beam instead of a laser as the heat source, and by taking place in a hermetically sealed chamber under vacuum to prevent the dissipation of the electron beam. Conversely, SLM processes occur in a controlled environment (Gordeev & Valentine, 2018). Furthermore, it is worth noting that the production chamber in EBM undergoes preheating at a specific temperature depending on the type of material used, such as approximately 1000°C for nickel-based alloys (Chandra et al., 2018) and approximately 400°C for pure copper (Guschlbauer et al., 2018). The preheating step represents a crucial aspect to take into account in EBM processes. The preheating process is crucial as it allows for the initial formation of sintering bridges between particles, preventing individual particles from becoming negatively charged and generating repulsive forces that would lead to the production chamber being filled with suspended powder, commonly referred to as the "smoke effect" (Milberg & Sigl, 2008).

With the exception of these minor differences, EBM and SLM are quite similar. Both methods start with an initial quantity of metallic powder that is selectively melted. At the end of the process, the desired product and some unmelted powders are obtained, which represents the production waste. Due to the high production costs associated with powders, which can also vary depending on the type of metal used (Hann, 2016), it is possible to reuse this waste powder in the subsequent production process (Bellini et al., 2022) (Foti et al., 2022). Currently, there are no established guidelines that govern recycling methodology, and thus, powder recycling is primarily based on user experience (Powell et al., 2020). Therefore, recycling procedures may vary, although typically the powder is initially sieved and, if required, may be combined with additional virgin powder of the same or different types before being introduced into the subsequent manufacturing cycle (Filipovic, 2016).

However, depending on the number of reuse cycles, recycled powders may not exhibit the same properties as virgin powders, primarily due to oxygen contamination and preheating, and can lead to worsened mechanical properties of components manufactured from these recycled powders. Hence, comprehending the alterations in powder properties is crucial to minimize the decline in the performance of the produced components. In fact, several studies have investigated the differences between unused and recycled powders, as well as the dissimilarities in the components produced from these powders. Typically, recycled powders exhibit inferior quality compared to virgin powders.

Specifically, according to research conducted by (Tang et al., 2015) and (Emminghaus et al., 2022), recycled powders in Ti-6Al-4V alloy generally exhibit elevated oxygen levels in comparison to virgin powders, whereas the concentrations of other elements such as V and Al are consistent. The increased oxygen content in recycled powders is primarily attributed to repeated circulation of powder in the process zone, leading to higher oxidation levels. Additionally, another reason is that when removed from the EBM machine, the powder is exposed to moisture and the surrounding atmosphere, which further contributes to oxygen absorption (Shanbhag & Vlasea, 2021).

Regarding the morphology, studies by (Carrion et al., 2019), (Emminghaus et al., 2021) and (Gatto et al., 2021) all suggest that recycled powders exhibit fewer satellites than their as-received counterparts, which is beneficial because it can significantly improve particle flowability. Moreover, several studies including (Carrion et al., 2019), (Yusuf et al., 2020) and (Nie et al., 2021), have reported that recycled powders demonstrate a narrower particle size distribution than unused powders when particle size is examined. Possible reasons for this change include particles sticking together during the manufacturing process to form larger clumps (Carrion et al., 2019), particles remaining suspended in the chamber and not being reusable (Seyda et al., 2012), and small particles sticking to larger ones, making them difficult to count (Sutton et al., 2016), although it should be noted that other research has shown that reused powders can sometimes have a broader size range than virgin powders (Gatto et al., 2021).

No differences were found in the microstructure of Ti-6Al-4V powders between virgin and reused powders, consisting mostly of acicular α’ martensite (Emminghaus et al., 2022) (Sun et al., 2018). Components produced from new or reused powders showed, as well, no dissimilarities in the microstructure, displaying slender acicular α' within the columnar prior-β grains oriented in the same direction as the building process (Carrion et al., 2019) (Strondl et al., 2015). In addition, recycled powders generally have a coarser microstructure compared to virgin ones due to slower cooling rates during manufacturing cycles (Opatová et al., 2020). However, the microstructure is also highly dependent on process parameters such as laser power and scanning speed (Emminghaus et al., 2021).

2. Experimental

In this study, plasma-atomized Ti-6Al-4V powders grade 5 and grade 23, both provided by Advanced Powders and Coatings, Inc. (AP&C), were utilized; more specifically, a batch of virgin powder grade 5 and another batch of powder that had been recycled 5 times. AP&C utilized their proprietary Advanced Plasma Atomization process (APA^{TM}) which is distinguished for its capability to generate highly spherical powders with accurate size distributions, minimal oxygen content, and low internal porosity (Capus, 2017).

The batch of recycled powder began with grade 5 powder during the first production cycle, and then, in each of the following cycles, grade 23 powder was added. Since it is not possible to attribute the recycled powder to specific regions within the building chamber, the percentage of powder reused can be regarded as a mixture of particles originating from both the pre-sintered and surrounding regions.
Table 1 below presents preliminary information regarding the chemical composition of both powders.

Table 1. Chemical composition of the Ti-6Al-4V powder particles

	% Al	% V	% C	% N	Fe %	% O	% Ti
Virgin	6.50	4.03	0.01	0.02	0.205	0.11	Remaining
Recycled 5 times	6.48	4.02	0.01	0.02	0.196	0.19	Remaining

ASTM F2924 (Ghods et al., 2020) specifies that the permissible limit for oxygen content in aerospace applications is 0.2%. It is noteworthy that the use of grade 23 Ti-6Al-4V powders enables the oxygen content to remain below this limit.

To investigate the morphology in virgin powders and after recycling, which include changes in shape, surface roughness, and imperfections, the FEI Quanta 650 Scanning Electron Microscope (SEM) was employed.

In order to analyze the porosity and voids within the particles, the powder particles were embedded into phenol-formaldehyde resin, creating cylindrical molds with a diameter of 30 mm that made the powder easier to handle. To polish the resin/powder samples, porous woven wool felt and 1 µm and 0.3 µm Alumina solutions were used exclusively, while grinding procedures were avoided due to their aggressive nature, which can increase the number of particles escaping from the resin during the process. The polished samples were then examined for any internal porosities using a Nikon Epithot inverted Metallurgical Microscope.

Starting from the optical microscope images of the powder cross-section, the ImageJ software was utilized to provide the trend of change in internal porosity. However, it should be noted that these values understimate the actual values as it is not known whether the diameter being considered is actually the maximum.

Finally, the analyzed particles were used to observe their microstructure and possible changes in the present phases. This was done by performing a chemical etching using a 0.1 Molar hydrofluoric acid (HF) solution, followed by rinsing under running water, and then observing them using the same optical microscope.

3. Results and Discussions

Based on the SEM images displayed in ***Fig. 1***, it was observed that both types of powders were mainly spherical in shape. However, a closer inspection revealed some notable differences between the two batches. Specifically, the virgin powder exhibited significantly higher concentration of fine particles, also known as satellites, compared to the recycled batch, where most of them were absent. The reason for the disappearance of the small particles was the pre-heating process that occurred inside the EBM chamber, during which the tiny particles fused onto the surface of the larger particles. The removal of satellites was found to enhance the flowability of the material (Seyda et al., 2012).

Furthermore, while the surface of the virgin powder appeared to be smooth and largely devoid of imperfections, the recycled powder exhibited some indications of degradation. For instance, a few broken particles were observed in the recycled powder, possibly due to the sieving procedures employed during the recycling process. In addition, the surface of the recycled powder appeared to be slightly rougher than that of the virgin powder, which could be attributed to the presence of the melted satellites.

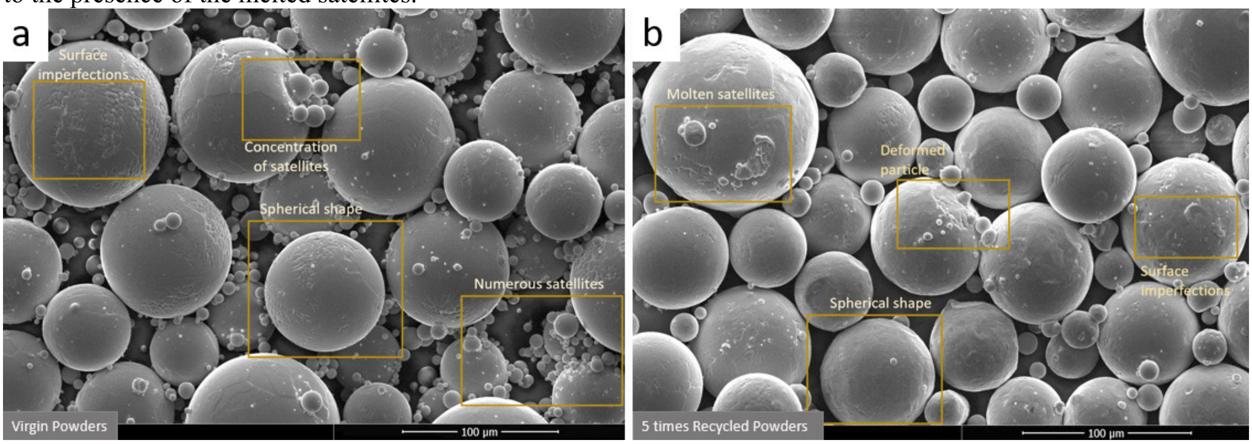

Fig. 1. SEM observation of virgin (a) and recycled (b) Ti-6Al-4V powder particles.

As shown in ***Fig. 2***, both types of powders exhibited internal defects in the form of pores caused by entrapped gases during the atomization process, which is a phenomenon also reported by other authors (Chen et al., 2018). Porosities in powders typically have a spherical morphology, which is a result of the high pressure inside the liquid droplets that acts on the gas bubbles. It should be noted that these pores can potentially affect the performance of the final parts, particularly in terms of their mechanical properties. Therefore, it is important to carefully examine and control the porosity levels of powder materials during the manufacturing process.

From these images, it is almost impossible to express opinions regarding the particle size, as they are embedded in resin, and the maximum observable diameter is unknown. The same applies to the pores, as they as well are spherical.

However, a qualitative analysis can be performed using the commercial software ImageJ with the purpose of showing the trend of how internal porosity varies as the number of reuses changes. With the use of this software, it was possible to observe that the virgin powder holds a defect percentage of 3.72%, while the recycled powder presents a defect percentage of 2.76%. It can be observed that internal defects in the recycled powder have decreased, and this is also consistent with findings from other authors (Bellini et al., 2022).

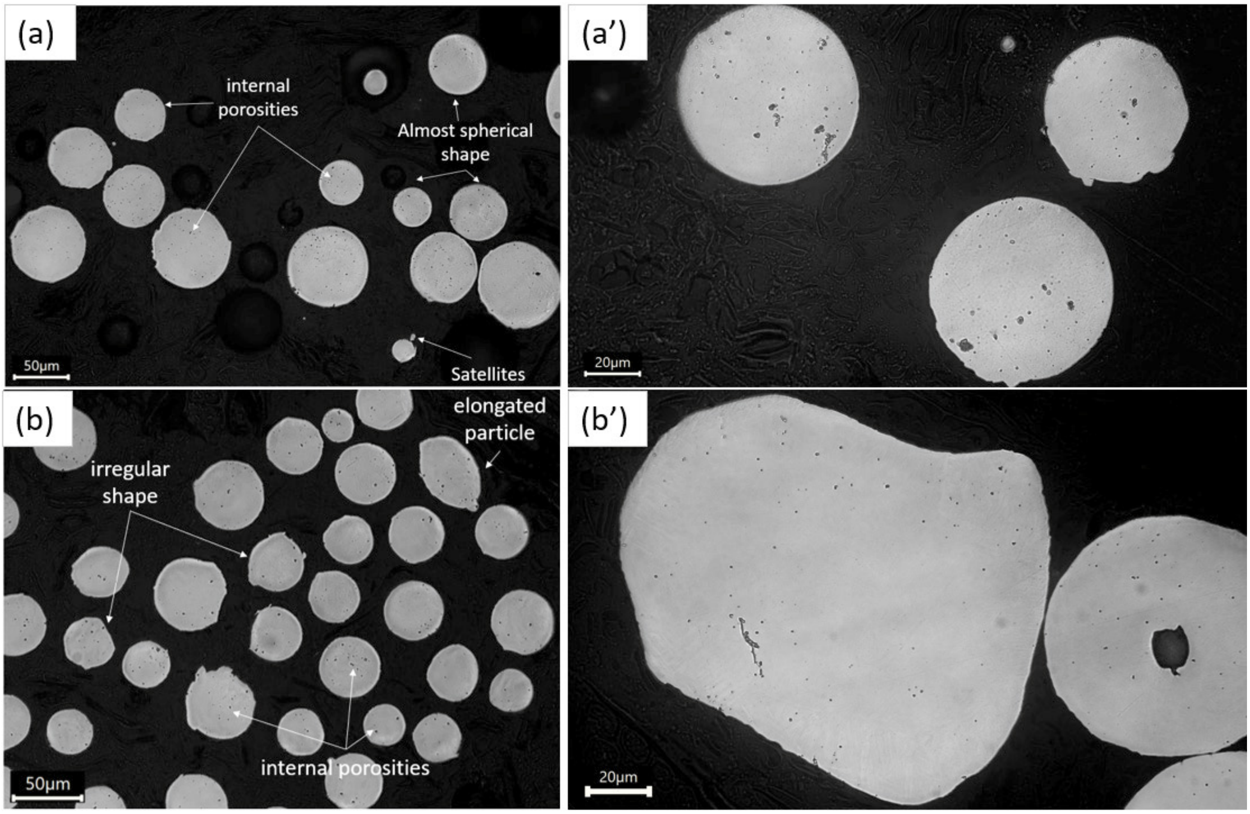

Fig. 2. Optical images of (a) virgin powders low magnification; (a’) virgin powders high magnification; (b) recycled powders low magnification; (b’) recycled powders high magnification.

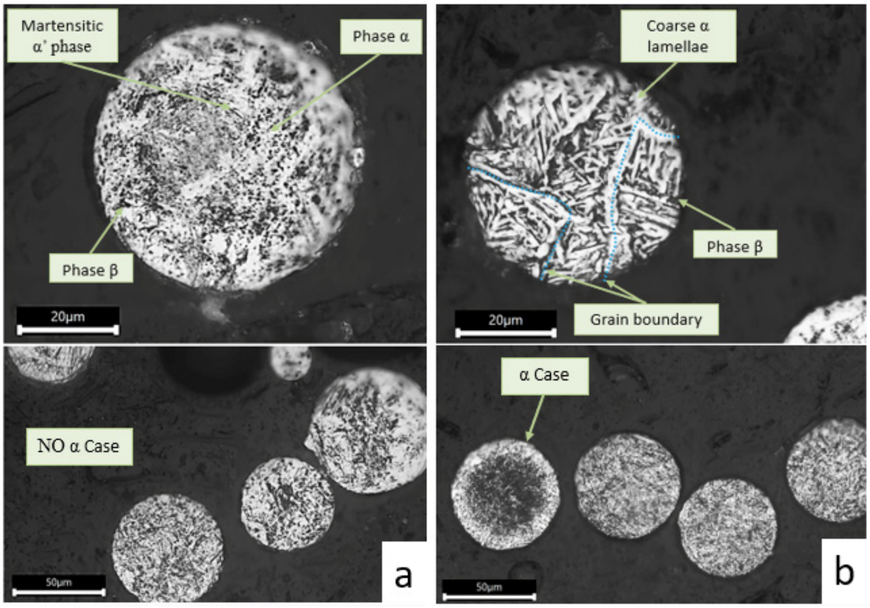

Fig. 3. Optical micrographs showing the microstructure of virgin (a) and recycled (b) powders.

The etching process revealed that the microstructure of virgin powders was predominantly characterized by a fine and martensitic structure. This microstructure was primarily comprised of the α' phase, which was distinguished by finely dispersed acicular grains of the α phase. It was also noted that this unique microstructure was a direct consequence of the rapid cooling of the β phase that occurred during the atomization process.

Upon examining the powder that had undergone recycling for five cycles, it was found that the microstructure of most particles closely resembled that of the virgin powder exhibiting a martensitic structure. However, a few particles displayed coarser grains in comparison to the original microstructure due to the slower cooling rate during the production cycle.

Furthermore, the recycled powders were found to exhibit the so-called alpha-case phenomenon, which refers to an oxygen-rich layer formed around the particles at high temperatures due to the diffusion of oxygen into the metal, resulting in the creation of a hard interstitial layer. This layer is harder than the particle core, and thus more brittle, often becoming the site of micro-cracks that can degrade the corrosion and fatigue resistance of the metal.

Importantly, this oxygen-rich layer was exclusively observed in the recycled powders and not in the virgin ones.

4. Conclusions

This study aimed to investigate the impact of the recycling process on powder morphology, internal defects, and powder microstructure, analyzing two batches of Ti-6Al-4V powder particles. One batch consisted of virgin powders produced by plasma atomization, while the other batch consisted of powders recycled five times after the Electron Beam Melting process, with ELI powders (Ti-6Al-4V grade 23) added at each cycle.

The results of the investigation revealed the following:

- The virgin powders were found to have a considerable number of satellites, which are known to reduce the flowability of the particles. On the other hand, the powders that had undergone five cycles of recycling showed a reduced number of satellites, possibly due to pre-heating inside the Electron Beam Melting chamber, resulting in improved flowability. Nonetheless, it is important to note that the recycling process did cause some particle damage such as broken particles and increased roughness surface.
- The virgin powders were found to have a high number of internal micro porosities, specifically 3.72%, which was attributed to the entrapment of gas during the atomization process. Such porosities are known to be detrimental to the properties of components manufactured from these powders. On the other hand, the powders that had undergone five cycles of recycling showed a lower number of internal voids, equal to 2.76%. This could be due to pre-heating inside the EBM chamber.
- The microstructure analysis revealed that the virgin powders had a martensitic microstructure, which was a result of the fast cooling rate during the plasma atomization process. On the other hand, the 5-times recycled powders exhibited a mixed microstructure, where some particles had a similar microstructure to the virgin ones, while others had a coarser microstructure. Furthermore, an oxygen-rich layer, commonly known as Alpha case, was observed in the recycled powders.

5. References

Bellini, C., Berto, F., Cocco, V. Di, Franchitti, S., Iacoviello, F., Mocanu, L. P., & Javad Razavi, S. M. (2022). Effect of recycling on internal and external defects of Ti-6Al-4V powder particles for electron beam melting process. Procedia Structural Integrity, 41(June), 175–182. https://doi.org/10.1016/j.prostr.2022.05.019

Bellini, C., Borrelli, R., Di Cocco, V., Franchitti, S., Iacoviello, F., Mocanu, L. P., & Sorrentino, L. (2021). Failure energy and stiffness of titanium lattice specimens produced by electron beam melting process. Material Design and Processing Communications, 3(6). https://doi.org/10.1002/mdp2.268

Capus, J. (2017). Titanium powder developments for AM – A round-up. Metal Powder Report, 72(6), 384–388. https://doi.org/10.1016/j.mprp.2017.11.001

Carrion, P. E., Soltani-Tehrani, A., Phan, N., & Shamsaei, N. (2019). Powder Recycling Effects on the Tensile and Fatigue Behavior of Additively Manufactured Ti-6Al-4V Parts. Jom, 71(3), 963–973. https://doi.org/10.1007/s11837-018-3248-7

Chandra, S., Tan, X., & Wang, C. (2018). Additive manufacturing of a single crystal nickel-based superalloy using selective electron beam melting Conference. May. https://doi.org/10.25341/D4V30K

Chen, G., Zhao, S. Y., Tan, P., Wang, J., Xiang, C. S., & Tang, H. P. (2018). A comparative study of Ti-6Al-4V powders for additive manufacturing by gas atomization, plasma rotating electrode process and plasma atomization. Powder Technology, 333(2017), 38–46. https://doi.org/10.1016/j.powtec.2018.04.013

Cooper, F. (2016). Sintering and additive manufacturing : '" additive manufacturing and the new paradigm for the jewellery manufacturer ."' Progress in Additive Manufacturing, 85–87. https://doi.org/10.1007/s40964-015-0003-2

Emminghaus, N., Bernhard, R., Hermsdorf, J., & Kaierle, S. (2022). Residual oxygen content and powder recycling: effects on microstructure and mechanical properties of additively manufactured Ti-6Al-4V parts. International Journal of Advanced Manufacturing Technology, 121(5–6), 3685–3701. https://doi.org/10.1007/s00170-022-09503-7

Emminghaus, N., Hoff, C., Hermsdorf, J., & Kaierle, S. (2021). Residual oxygen content and powder recycling: Effects on surface roughness and porosity of additively manufactured Ti-6Al-4V. Additive Manufacturing, 46. https://doi.org/10.1016/j.addma.2021.102093

Filipovic, V. P. (2016). Powder recyclability in electron beam melting for aeronautical use Aircraft Engineering and Aerospace Technology : An International Journal Article information : January. https://doi.org/10.1108/AEAT-11-2013-0212

Foti, P., Mocanu, L. P., Razavi, S. M. J., Bellini, C., Borrelli, R., Di Cocco, V., Franchitti, S., Iacoviello, F., & Berto, F. (2022). Effect of recycling powder on the fatigue properties of AM Ti6Al4V. Procedia Structural Integrity, 42, 1436–1441. https://doi.org/10.1016/j.prostr.2022.12.183

Gardan, N., & Schneider, A. (2015). Topological optimization of internal patterns and support in additive manufacturing. Journal of Manufacturing Systems, 37, 417–425. https://doi.org/10.1016/j.jmsy.2014.07.003

Gatto, M. L., Groppo, R., Bloise, N., Fassina, L., Visai, L., Galati, M., Iuliano, L., & Mengucci, P. (2021). Topological, mechanical and biological properties of Ti6Al4V scaffolds for bone tissue regeneration fabricated with reused powders via electron beam melting. Materials, 14(1), 1–20. https://doi.org/10.3390/ma14010224

Ghods, S., Schultz, E., Wisdom, C., Schur, R., Pahuja, R., Montelione, A., Arola, D., & Ramulu, M. (2020). Electron beam additive manufacturing of Ti6Al4V: Evolution of powder morphology and part microstructure with powder reuse. Materialia, 9(February), 100631. https://doi.org/10.1016/j.mtla.2020.100631

Gordeev, E. G., & Valentine, P. (2018). An In-Depth Review on Direct Additive Manufacturing of Metals An In-Depth Review on Direct Additive Manufacturing of Metals. https://doi.org/10.1088/1757-899X/328/1/012005

Guschlbauer, R., Momeni, S., Osmanlic, F., & Körner, C. (2018). Materials Characterization Process development of 99 . 95 % pure copper processed via selective electron beam melting and its mechanical and physical properties ☆. Materials Characterization, March, 0–1. https://doi.org/10.1016/j.matchar.2018.04.009

Hann, B. A. (2016). Powder Reuse and Its Effects on Laser Based Powder Fusion Additive Manufactured Alloy 718. https://doi.org/10.4271/2016-01-2071

Ishfaq, K., Rehman, M., Khan, A. R., & Wang, Y. (2022). A review on the performance characteristics, applications, challenges and possible solutions in electron beam melted Ti-based orthopaedic and orthodontic implants. Rapid Prototyping Journal, 28(3), 525–545. https://doi.org/10.1108/RPJ-03-2021-0060

ISO/ASTM. (2015). ISO/ASTM 52900-Standard Terminology for Additive manufacturing–general principles–terminology. i, 1–9.

Milberg, J., & Sigl, M. (2008). Electron beam sintering of metal powder.

Mostafaei, A., Zhao, C., He, Y., Reza Ghiaasiaan, S., Shi, B., Shao, S., Shamsaei, N., Wu, Z., Kouraytem, N., Sun, T., Pauza, J., Gordon, J. V., Webler, B., Parab, N. D., Asherloo, M., Guo, Q., Chen, L., & Rollett, A. D. (2022). Defects and anomalies in powder bed fusion metal additive manufacturing. Current Opinion in Solid State and Materials Science, 26(2). https://doi.org/10.1016/j.cossms.2021.100974

Nie, Y., Tang, J., Huang, J., Yu, S., & Li, Y. (2021). A study on internal defects of prep metallic powders by using x-ray computed tomography. Materials, 14(5), 1–11. https://doi.org/10.3390/ma14051177

Opatová, K., Zetková, I., & Kučerová, L. (2020). Relationship between the size and inner structure of particles of virgin and re-used ms1 maraging steel powder for additive manufacturing. Materials, 13(4). https://doi.org/10.3390/ma13040956

Powell, D., Rennie, A. E. W., Geekie, L., & Burns, N. (2020). Understanding powder degradation in metal additive manufacturing to allow the upcycling of recycled powders. Journal of Cleaner Production, 268, 1–29. https://doi.org/10.1016/j.jclepro.2020.122077

Seyda, V., Kaufmann, N., & Emmelmann, C. (2012). Investigation of Aging Processes of Ti-6Al-4 v Powder Material in Laser Melting. Physics Procedia, 39, 425–431. https://doi.org/10.1016/j.phpro.2012.10.057

Shanbhag, G., & Vlasea, M. (2021). Powder reuse cycles in electron beam powder bed fusion— variation of powder characteristics. Materials, 14(16). https://doi.org/10.3390/ma14164602

Strondl, A., Lyckfeldt, O., Brodin, H., & Ackelid, U. (2015). Characterization and Control of Powder Properties for Additive Manufacturing. Jom, 67(3), 549–554. https://doi.org/10.1007/s11837-015-1304-0

Sun, Y., Aindow, M., & Hebert, R. J. (2018). The effect of recycling on the oxygen distribution in Ti-6Al-4V powder for additive manufacturing. Materials at High Temperatures, 35(1–3), 217–224. https://doi.org/10.1080/09603409.2017.1389133

Sutton, A. T., Kriewall, C. S., Leu, M. C., & Newkirk, J. W. (2016). Powders for additive manufacturing processes: Characterization techniques and effects on part properties. Solid Freeform Fabrication 2016: Proceedings of the 27th Annual International Solid Freeform Fabrication Symposium - An Additive Manufacturing Conference, SFF 2016, 1004–1030.

Tang, H. P., Qian, M., Liu, N., Zhang, X. Z., Yang, G. Y., & Wang, J. (2015). Effect of Powder Reuse Times on Additive Manufacturing of Ti-6Al-4V by Selective Electron Beam Melting. Jom, 67(3), 555–563. https://doi.org/10.1007/s11837-015-1300-4

Yusuf, S. M., Choo, E., & Gao, N. (2020). Comparison between virgin and recycled 316l ss and alsi10mg powders used for laser powder bed fusion additive manufacturing. Metals, 10(12), 1–18. https://doi.org/10.3390/met10121625