Powder metallurgy characterization of thermoelectric materials for selective laser melting

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ABSTRACT

Thermoelectric materials are semiconductors capable of converting heat into electrical energy. Traditional manufacturing of thermoelectric devices involves machining and assembly steps which limit device geometries, result in material loss, and introduce parasitic thermal and electrical resistances. Selective laser melting is an alternative manufacturing method that could eliminate the need for arduous assembly steps, enable unique device geometries, and facilitate effective material-to-device integration. The study discussed here provides the first-ever analysis of thermoelectric material powders for layer-by-layer manufacturing. The results include analysis of two thermoelectric materials, bismuth telluride (Bi_2Te_3) and a half-Heusler compound (ZrNiSn), which are optimal for low and high temperature applications, respectively. Particle size distribution and particle morphology are analyzed using optical and scanning electron microscope images. The ability of the powder to flow and spread in 100-300 µm thick layers is assessed using both blade and roller approaches. Preliminary results on the first-ever, 3D-printed thermoelectric material powder using selective laser melting are described.

Keywords: advanced manufacturing, 3D printing, materials innovation, selective laser melting, thermoelectric, powder metallurgy

1 INTRODUCTION

Selective laser melting (SLM) is an additive manufacturing process that forms solid parts from powdered material. Thin layers of powder are fully melted and fused together with a laser source. The layer-by-layer processing of SLM enables the formation of complex geometries and allows for unique tailoring of the bulk material properties such as grain structure anisotropy.

Thermoelectric devices are currently manufactured using traditional, subtractive manufacturing techniques. These techniques are generally inefficient in terms of bulk material usage, and they limit the achievable part geometries [1]. Selective laser melting of semiconducting materials offers an exciting opportunity to innovate the geometry and assembly of thermoelectric devices while simultaneously manipulating thermoelectric properties [2].

In order to produce a homogeneous part through selective laser melting, it is necessary to optimize both laser processing parameters and powder characteristics. Particle size and morphology of a material powder significantly affect the density of the resulting part.

This study utilizes a commercially available bismuth telluride (Bi_2Te_3) powder, along with a laboratory prepared half-Heusler (ZrNiSn) powder. Previous work discussed melting of compacted bismuth telluride powder using pulsed and continuous wave lasers [2-3]. The melting of metal powders has been well-documented, and this study used stainless steel powder (SS340) for comparison. The particle size distribution and morphology of both thermoelectric material powders were compared to the metal powder, and the ease of powder spreading was evaluated. Finally, laser melting parameters were tested on a prepared powder bed to gain insight into the melting characteristics of these semiconductor material powders.

2 METHODS & RESULTS

2.1 Powder characterization

Large particle sizes allow for effective flowing since smaller particles' larger surface areas lead to increased friction between the particles. However, the presence of only large particles (30-60 µm) in the powder bed leads to a decrease in the achievable packing density. Lower packing density is undesireable in laser powder-bed fusion and can cause melt-pool balling during processing. Additionally, the presence of coarse, irregularly shaped particles can lead to preferred orientation within the powder mass and inconsistent powder melting.

The standard stainless steel (SS340) powder has a low average particle size and a narrow particle size distribution (particle sizes range from 2-15 µm) as show in the scanning electron micrograph of Figure 1. The standard powder particles are predominantly spherical. In contrast, the bismuth telluride and half-Heusler material powders have wider particle size distributions (5-60 µm for the half-Heusler) and irregular particle morphologies (Figures 2-3).

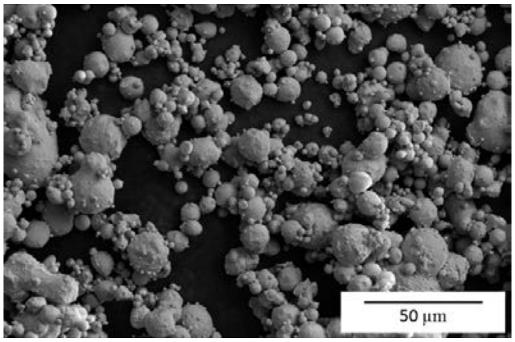


Figure 1. Scanning electron micrograph of standard SS340 atomized powder used in laser powder-bed fusion.

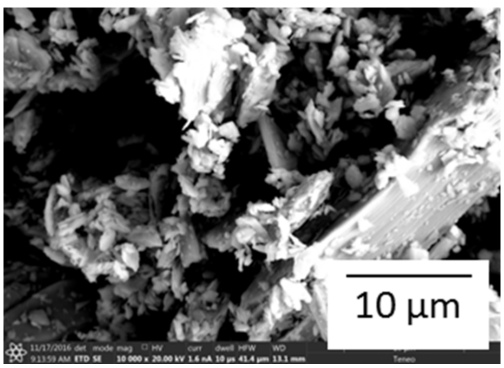


Figure 2. Scanning electron micrograph of a commercial Bi_2Te_3 powder.

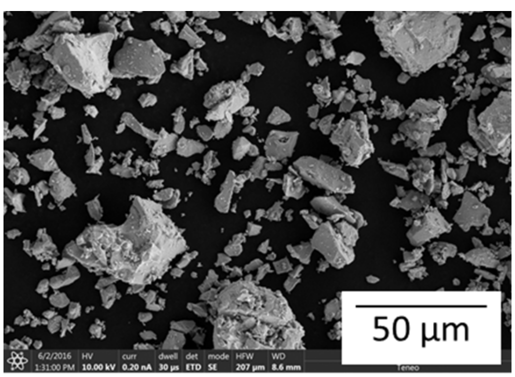


Figure 3. Scanning electron micrograph of half-Heusler (ZrNiSn) powder.

Powder characterization was also conducted using a Malvern Morphologi G3-ID, an automatic morphological imaging device. Two lots of commercially available bismuth telluride (Bi_2Te_3) powder from Alfa Aesar were analyzed. The bismuth telluride powders possess a wider range of particle sizes than the SS340 or half-Heusler powders; they contain particles ranging from smaller than 20 µm to larger than 150 µm. Each lot was sieved (using sieve Nos. 100, 140, 200, 270, 635) to collect particles within size ranges desirable for spreading and melting. Figure 4 shows the frequency of different sized particles within three samples of bismuth telluride powder. Samples 1 and 2 were sieved from powder Lot 1, and the samples' nominal particle sizes were 20-53 µm and 53-75 µm, respectively. Sample 3 was sieved from powder Lot 2 and nominally possessed particles ranging 20-75 µm.

Analysis of particle sizes in each sample of powder reveals the limited efficacy of the sieving procedure. Large frequencies of particles sized below 10 µm were observed in each sample, likely due to particle agglomerates which were not broken up during sieving . There is also a variation of particle sizes observed between each powder lot; Lot 1 possesses a higher frequency of larger diameter particles. The lot-to-lot variation poses processing challenges since a given powder preparation method will not yield consistent results, creating variations in the starting material for the subsequent laser processing step.

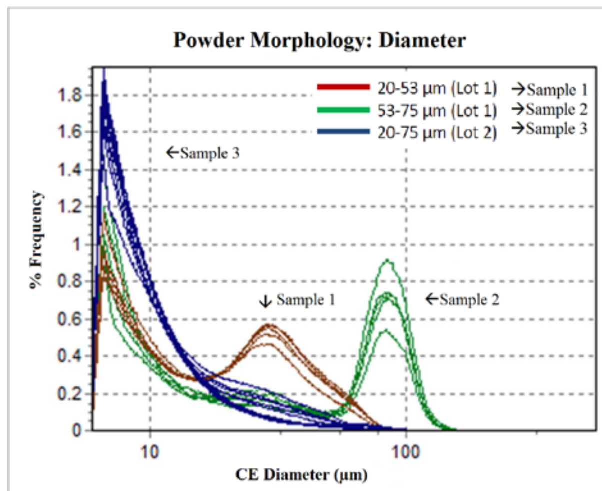


Figure 4. Frequency of particle sizes identified in sieved samples from each lot of Bi_2Te_3 powder.

Figures 5 and 6 show the frequency of particle aspect ratios and circularities in each lot of Bi_2Te_3 powder. Each sample possesses a wide distribution of particle aspect ratios, ranging from 0% to 80-90% elongation where elongation is 1 – aspect ratio. The frequency of particle circularity indicates that the particles are non-spherical. Particle aspect ratio and circularity are consistent between powder lots.

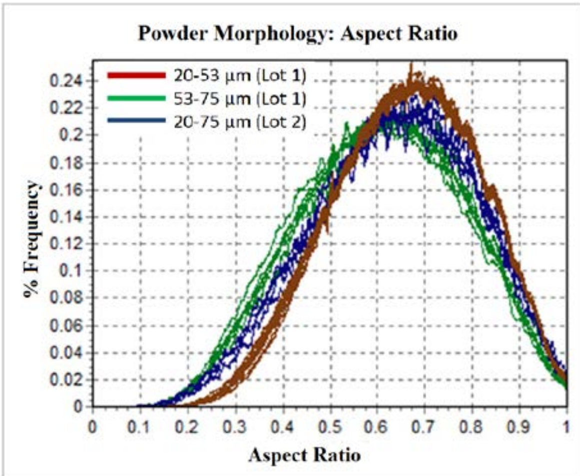


Figure 5. Frequency of particles' aspect ratios in sieved samples from each lot of Bi_2Te_3 powder.

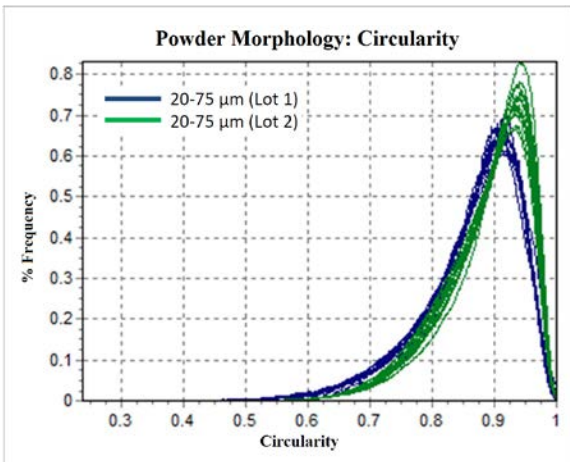


Figure 6. Frequency of particles with specified circularity in sieved samples from each lot of Bi_2Te_3 powder.

2.2 Powder spreading and flowing

Three dimensional parts are formed through selective laser melting by iteratively spreading powder layers to melt and fuse above the previously melted layers. In order to adequately fuse powder layers and form a continuous part during the melting procedure, each layer must be densely packed and of uniform thickness.

The spreading quality of each powder was evaluated prior to melting trials. The powder was spread in 100 µm and 300 µm deep grooves milled into stainless steel plates. Two spreading methods were considered, a blade and a counter-rotating metal roller. Shown in Figure 7, the standard stainless steel powder was easily spread in a 100 µm bed using a single sweep of a blade, indicating good spreading and flowing characteristics due to low inter-particle friction.

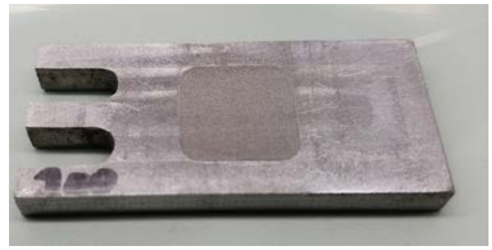
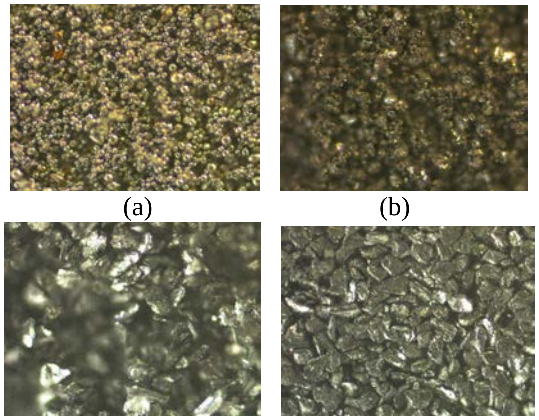


Figure 7. Stainless steel SS340 powder spread in a 100 µm deep groove with a blade.

Attempts to spread both thermoelectric material powders in a 100 µm layer were less successful. Powder spreading was attempted using a blade and roller, and powder was added to fill in gaps after each spreading attempt. The irregularly shaped powder particles can mechanically interlock during the spreading process rather than flowing easily like spherical particles. The powder "lifts-off" during spreading and leaves voids or unfilled areas.

Attempts to spread both thermoelectric materials in 300 µm deep beds were successful using both a blade and counter-rotating roller. The 300 µm depth is larger than the average powder particle size The increase in bed depth allows for additional layers of particles. The increased number of layers does not eliminate the powder particle interlocking, but the powder lift-off is minimized due to the increased area of contact between the top layer of powder and the subsequent layers below. Without tailoring the particle characteristics, spreading trials indicate that a larger powder bed thickness is required to evenly spread the powders. The larger thickness is not ideal and can lead to incomplete melting/fusion between layers and balling during laser processing.

Spreading trials were conducted on sieved bismuth telluride powder to observe the effectiveness of spreading different sized particles. These trials also compared the effectiveness of spreading using a blade versus a counter-rotating roller. Figure 8 shows the top surfaces of 300 µm deep beds of larger bismuth telluride powder particles (44-74 µm) spread by a blade and roller, respectively. The average powder bed surface height is more consistent across surfaces spread by a counter-rotating roller. Beds of smaller bismuth telluride powder particles (20-44 µm) yielded the same result with slightly higher packing density. A spreading trial was conducted using a mix of these particle sizes but resulted in a slightly lower packing density. This could be attributed to the wider range of particle sizes in this particular case or the ratio of smaller to larger particles.



(a) (b)

(c) (d)

Figure 8. Optical microscope images (100x) of (a) SS340 powder spread in a 100 µm depth bed, (b) ZrNiSn powder spread in a 300 µm depth bed, and sieved Bi_2Te_3 powder (44-74 µm) spread in a 300 µm deep bed by (c) blade and (d) roller.

2.3 Preliminary SLM study

An IPG Photonics YLR-100-AC-Y11 series fiber laser with mid-power scanner was used to test melting parameters for thermoelectric material powders. The laser rig is custom designed and built to process small quantities of powder materials. Figures 9 and 10 show a sample prepared using the half-Heusler powder. A single layer of powder was spread with a roller to a thickness of 100 µm. The 50 µm spot size laser was programmed to travel in a vertical hatch pattern (20% overlap) across the bed at 350 mm/s, firing at 20 W. This procedure yielded a solid disc 8 mm in diameter. Figure 9 depicts the melted disc within the surrounding powder bed while Figure 10 shows the resulting melted sample' surface.

Figure 9. Half-Heusler (ZrNiSn) powder bed spread by blade, post laser processing

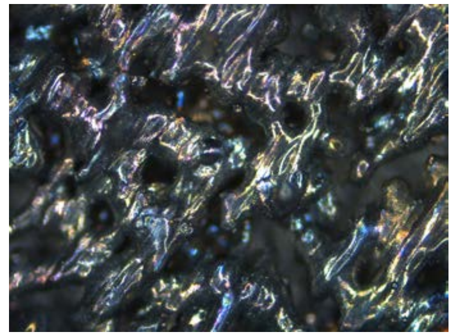


Figure 10. Optical microscope image (100x) of disc formed through laser processing of single layer of half-Heusler material powder.

3 CONCLUSION AND FUTURE WORK

Powder properties relevant for selective laser melting were characterized for two thermoelectric materials. The characteristics were compared to a metal powder currently used for powder-bed fusion. The irregular powder morphology, lot-to-lot variations in powder characteristics, and challenges in particle size separation are critical hurdles for using thermoelectric material powders in selective laser melting. Nonetheless, preliminary laser melting trials show results similar to those obtained in the early days of metals selective laser melting. The promising results show the potential for successfully extending selective laser melting to a broader range of materials once powder metallurgy challenges are overcome.

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REFERENCES

[1] S. LeBlanc, "Thermoelectric generators: Linking material properties and systems engineering for waste heat recovery applications," *Sustainable Materials and Technologies*, 2014.

[2] A. El Desouky, M. Carter, M.A. Andre, P.M. Bardet, S. LeBlanc, "Rapid processing and assembly of semiconductor thermoelectric materials for energy conversion devices," *Materials Letters*, 2016.

[3] A. El Desouky, M. Carter, M. Mahmoudi, A. Elwany, S. LeBlanc, "Influences of energy density on microstructure and consolidation of selective laser melted bismuth telluride thermoelectric powder," *Journal of Manufacturing Processes*, 2017.