Journal of Manufacturing Processes 25 (2017) 411–417

Contents lists available at ScienceDirect

Journal of Manufacturing Processes

journal homepage: www.elsevier.com/locate/manpro

Influences of energy density on microstructure and consolidation of selective laser melted bismuth telluride thermoelectric powder

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ARTICLE INFO

Article history:
Received 5 October 2016
Accepted 6 December 2016

Keywords:
Selective laser melting
Thermoelectrics
Bismuth telluride

ABSTRACT

Selective laser melting is a well-established additive manufacturing technique for metals and ceramics, and there is significant interest in expanding the manufacturing capability of this technique by enabling processing of more materials. Notably, selective laser melting has not been demonstrated on semiconducting materials which are significant for energy conversion technologies. Thermoelectric materials are semiconductors that can convert heat into electrical power. The traditional thermoelectric manufacturing process involves many assembly and machining steps which lead to material losses, added time and cost, and performance degradation. Utilizing selective laser melting in the manufacturing of thermoelectric modules can minimize assembly steps and eliminate machining processes. In this study, a standard bismuth telluride thermoelectric powder was processed for the first time in a commercial Prox™ 100 selective laser melting system under different energy densities. The surfaces of Bi_2Te_3 specimens were successfully melted under all processing conditions, and the entire thickness of specimens processed at higher laser power inputs was mostly melted. The increase in laser power also reduced the presence of porosities on the surface of the specimens. However, cross-sectional examination showed that internal pores were present within the molten region and at the interface between the molten region and the unmelted powder under all investigated laser power inputs. The consistency of these results with early results for select laser melting of metals demonstrates the promise for expanding the materials processing capabilities of this additive manufacturing technique.

1. Introduction

Thermoelectric devices convert waste heat into electrical power, creating opportunities for waste heat recovery in high-temperature systems from industrial power plants to automobiles [1,2]. A thermoelectric device typically consists of multiple junctions of n- and p-type semiconducting thermoelectric legs. The legs are connected electrically in series and thermally in parallel as shown in Fig. 1. When a temperature gradient is applied across the thermoelectric material, electric charge carriers move from the hot side to the cold side, ultimately resulting in a voltage drop—a phenomenon known as the Seebeck effect.

The performance of a thermoelectric material is evaluated through the dimensionless figure of merit $ZT = \left(S^2\sigma/k\right)T$, where S is the Seebeck coefficient, σ is the electrical conductivity, k is the thermal conductivity, and T is the absolute temperature. A material with good thermoelectric properties has a high Seebeck coefficient, high electrical conductivity, and low thermal conductivity to maintain the temperature gradient across the material. Chalcogenide materials are commonly used in thermoelectric devices that operate over a wide range of operating temperatures (100–1000 K) [3,4]. Bismuth telluride (Bi_2Te_3) and its solid solutions are among the most commonly used chalcogenides in off-the-shelf thermoelectric devices operating near room temperature [3,5,6]. Although noteworthy progress in enhancing the thermoelectric properties of Bi_2Te_3 and its alloys through microstructural and compositional changes has been recently reported [7–10], challenges related to device manufacturing and assembly continue to slow the progress of thermoelectric power generation [4,11].

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http://dx.doi.org/10.1016/j.jmapro.2016.12.008
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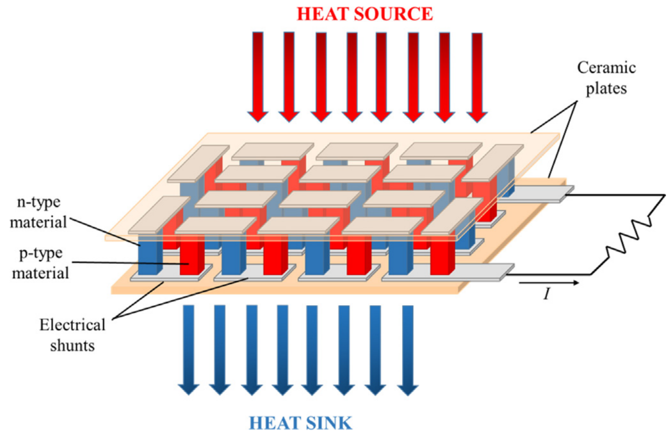

Fig. 1. Schematic of a thermoelectric device showing multiple n- and p-type leg couples connected electrically in series and thermally in parallel. A typical thermoelectric device can consist of hundreds of legs.

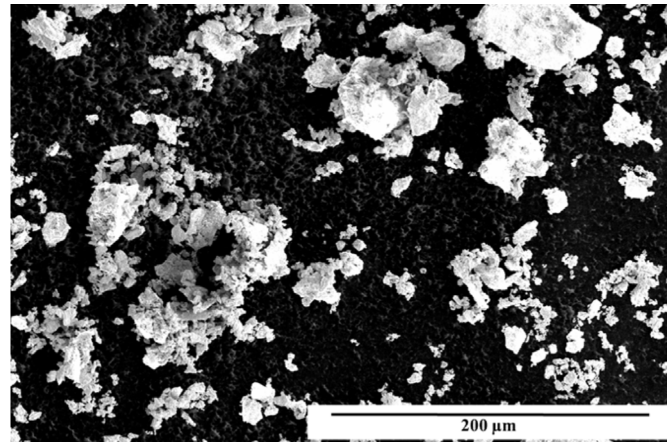

Fig. 2. Scanning electron micrograph of off-the-shelf Bi_2Te_3 powder.

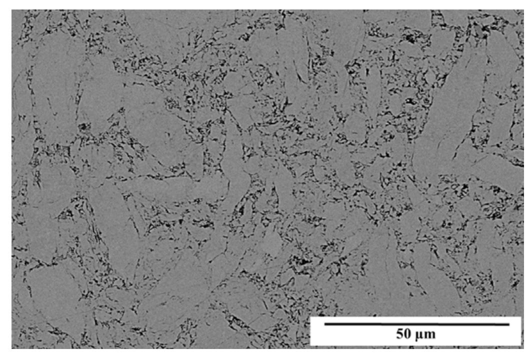

Fig. 3. Scanning electron micrograph of a polished cross-section of a Bi_2Te_3 powder compact.

The traditional thermoelectric device manufacturing process involves multiple steps including processing and assembly. First, the constituents of the thermoelectric compound are alloyed using mechanical milling processes. The alloyed powder is then consolidated through methods such as hot pressing or spark plasma sintering. The resulting ingots are diced into the leg shape, and finally the legs are generally picked and placed into the final assembly. In contrast, additive manufacturing represents an attractive manufacturing alternative for thermoelectric devices. A recent workshop by the U.S. Department of Energy indicates that labor accounts for a significant portion of the cost of these devices [12]. Additive manufacturing technologies such as selective laser melting (SLM) are capable of producing 3-dimensional objects directly from a digital CAD model [13] without the need for customized tooling or manual assembly. This capability can potentially be leveraged to eliminate the assembly of many components and reduce the time and cost involved in producing complex thermoelectric modules. For example, Crane et al. [14] present a simulation case study using a prototype thermoelectric system to highlight the potential capability of additive manufacturing in automated self-assembly and integration. Co-authors of this work recently provided an overview of challenges in thermoelectric device manufacturing and highlighted the potential of additive manufacturing technologies in thermoelectric materials processing [15].

Selective laser melting (SLM) has been used in numerous studies in the literature to produce complex customized parts from metallic materials and alloys including stainless steels [16–20], titanium alloys [21–24], and nickel-based super alloys [25–28] among others. Preliminary findings on the formation of single melt tracks of Bi_2Te_3 powder compacts using a low repetition pulsed laser [29,30,15] were previously reported. In the current work, an initial investigation is conducted on the full consolidation of bismuth telluride (Bi_2Te_3) powder using SLM. Bi_2Te_3 powder is the only commercially available thermoelectric pre-alloyed powder. Off-the-shelf Bi_2Te_3 powder lacks the physical characteristics that would allow it to spread into sequential thin layers which is essential for the SLM process. The irregular shape of the particles and the large particle size distribution (visible in Fig. 2) result in increased friction between the particles and leads to poor flowability and spreadability of the powder. This initial investigation is focused on processing Bi_2Te_3 in powder compact form primarily to provide insight into the effect of laser energy density on the microstructural evolution and densification behavior of laser-irradiated Bi_2Te_3. These insights will serve as a foundation to enable subsequent layer-by-layer SLM processing of thermoelectric modules.

Table 1
Selective laser melting processing parameters for the 16 powder compacts processed in a ProX100.

Specimens	Power (W)	Speed (mm/s)	Energy density (J/mm^2)
1–4	10	350	0.57
5–8	15	350	0.86
9–12	20	350	1.14
13–16	25	350	1.42

2. Experiments

Bi_2Te_3 powder (−325 mesh, 99.99% trace metals basis, Sigma Aldrich) was compacted using a hydraulic press and a hardened steel 6 mm die. The final compacts are 6 mm diameter and ~500 μm thick. Fig. 3 shows a scanning electron micrograph of a representative Bi_2Te_3 powder compact used in this study. Next, the powder compacts were processed on a commercial ProX^{TM} 100 SLM system. The ProX^{TM} 100 uses a 50-W fiber laser with a Gaussian profile and approximately 100 μm spot size to selectively melt metallic powder in a 3.94 × 3.94 × 3.15 in. build envelope. This study is the first of its kind in processing thermoelectric powder on this SLM system.

The powder compacts were placed on the build plane and processed under an argon atmosphere with the oxygen level kept below 700 ppm. Sixteen specimens in total were processed. The scanning speed of the laser and the hatch distance were kept constant at 350 mm/s and 70 μm, respectively, and the laser energy density was varied by adjusting the laser power as shown in Table 1.

For subsurface analyses, the specimens were cleaved normal to the build direction. The specimens were then cast in epoxy resin, and an Allied High Tech Multiprep polishing unit was used to grind and polish the cross-sectional surface down to 0.04 μm