Studies on Commercially Pure Titanium (CP-Ti) Fabricated by Electron Beam Melting (EBM)

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論 文 内 容 要 旨

Chapter 1

Titanium is the 7th most abundant element in earth's crust. Titanium and its alloys are widely used in aerospace, power generation, and biomedical industries due to their high strength-to-weight ratio, outstanding corrosion resistance, and excellent biocompatibility. Particularly, there is less toxic or allergic elements in commercially pure titanium (CP-Ti) compared with other common titanium alloys such as Ti-6Al-4V alloys, making it extremely suitable for biomaterials. However, because titanium has a strong tendency to react with oxygen and nitrogen due to their reactive nature when exposed at high temperature atmospheric environment, the fabrication process is required to be conducted under protective environment, such as vacuum, and protective inert-gas atmosphere. Furthermore, the relatively low thermal conductivity and work hardening effect for titanium alloys also increase the difficulty to the machining process, so the cutting tools with good qualities are highly required, and usually service life of the cutting tool is reduced. Therefore, the production cost of titanium alloys stays at a high level, and accordingly the prices of the titanium alloy products are difficult to be lowered which makes it one of its limitation for widespread application. Hence, innovative manufacturing methods should be developed for titanium alloys to reduce the cost of the production.

Additive manufacturing (AM) is a newly developed revolutionary manufacturing technology that is still under rapid development in the recent years. AM technology represents process to fabricate components with desired shape directly based on the 3D model controlled through computer, usually by depositing layers of materials. Thus, it is a kind of near-net shape manufacturing method that provides high freedom in design regardless of the restrictions from the conventional manufacturing methods, for which the shapes should be designed only if it can be fabricated, giving advantages in fabricating components with complex structures or even realizing customized designs efficiently. Accordingly, AM is well applied to materials with poor machinability like titanium, and it's such a promising manufacturing method that has broad application prospects in many industries such as aerospace, biomedical, and power generation industries. Electron beam melting (EBM) is one of the powder-bed-fusion AM technologies using electron beam as the energy source. It was developed by ARCAM Company in Sweden. Because EBM proceeds under highly vacuum environment, it prevents the reaction between the materials and the atmosphere, which makes it suitable for fabricating titanium and its alloys. As one of the promising materials for EBM application, the studies of CP-Ti fabricated by EBM are quite limited. The process window of for acquiring dense CP-Ti components with good geometrical morphologies fabricated via EBM has not yet been obtained. Systematical studies with respect to the microstructure evolution and the mechanical properties of the as-built specimens under different build parameters are crucial to CP-Ti for further applications, especially when regarding it as one of the adequately applied materials for EBM.

The objective of the study is to systematically investigate the microstructure and mechanical properties of the CP-Ti fabricated by EBM under various built parameters, such as, beam current, scan speed, preheat temperature, and geometric structure. Furthermore, based on the studies of CP-Ti, the fabrication process of EBM could be further understanding.

Chapter 2

Process map is an approach to map out the outcomes in terms of the combining effects of variables based on the simulation or experimental results especially for thermally based AM application, in which it is very useful in achieving the optimal processing window for AM process. Among all the processing parameters during EBM process, the scan speed and the power of electron beam have a significant influence on the input energy and the geometry of melt pool which determine the final microstructure and properties of outcomes. Therefore, CP-Ti blocky specimens were fabricated under different beam currents and scan speeds through EBM. The microstructural and mechanical properties were examined through optical microscope (OM), scanning electron microscope (SEM), electron backscattered diffraction (EBSD) observation, and tensile tests.

The process map was established based on the relationship between the geometrical morphologies (porous, even, and uneven) of as-built specimens and the corresponding build parameters, and the preferred process window for dense specimens with good geometrical morphology (even specimen) was with line energy of about 300-600 J/m at relatively low scan speed. And as the scan speed increased, the process window narrowed down owing to the increasing instability of the melt pool.

The microstructural morphology of as-built CP-Ti could be divided in to columnar, granular, and mixed microstructure under different built parameters. The diversity of microstructure was not only affected through the various building conditions during melting procedure, but also the thermal experiences after the solidification at the following fabrication process of EBM. Generally, higher line energy was inclined to increase the tendency of forming granular microstructure.

The mechanical property of as-built specimens was examined through tensile test at room temperature. The tensile strengths for porous specimens were reduced by the large irregular incomplete melted pores inside, and those for dense specimens were comparable to the CP-Ti components fabricated through conventional methods. The tensile strength for dense specimens didn't varied obviously with the increase of line energy. Nevertheless, the specimens with fully columnar microstructure exhibited slightly higher tensile strength because of relatively higher strain within the microstructure.

Chapter 3

The preheat process is a specific and necessary procedure in EBM process, which provides an elevated thermal environment of the components during the fabrication (typically 650 °C to 900 °C). It is performed before the melting process through scanning the whole pre-laid powder layer by defocused electron beam, and the main purpose of it is to prevent the smoking during the fabrication process. In addition, owing to the preheat procedure, the elevated thermal environment could also reduce the residual stress of the as-built components. To date, the preheat temperature during the EBM process is generally chosen based on experiences. And the difference in preheat temperatures could probably have influence on the outcomes of the EBM-build components, such as the preferable processing window, microstructure, and mechanical properties. Consequently, the influence of preheat temperature on the properties of the as-built components should be considered.

Blocky CP-Ti specimens were fabricated under different preheat temperatures, which were from 550 to 750 ℃ with the interval of 50 ℃. 9 blocks were fabricated together at same preheat temperature with different scan speeds and beam currents. The influence of preheat temperature on the microstructure and mechanical properties of as-built CP-Ti was examined through checking the condition of unmelted powder, EBSD observation, heat treatment, and tensile tests.

The unmelted powder was too loose at low preheat temperature, which caused smoking during the fabrication process. The unmelted powder was extremely stiff and severely sintered at high preheat temperature owing to high input energy, which would affect the quality of the recycled powder. As the increase of preheat temperature, the microstructure became coarser slightly and there were large recrystallized grains appeared at high preheat temperature of 750 ℃. Correspondingly, the tensile strength was reduced slightly because of the coarsened microstructure and the recrystallized grains. The appropriate method to determine a suitable preheat temperature is to firstly determine the range of it based on the conditions of unmelted powders. Then, within the certain range, the relatively low preheat temperatures could be considered, which could result in higher strengths, and energy saving.

Chapter 4

Since EBM could fabricate components directly from the 3D model, it is capable of fabricating products with complex structures, which would have additional properties such as light weight, reduced elastic modulus, and improved biocompatibility. For the EBM-built components with conventional structure, which are continuously melted along the build direction, the heat produced during the following melting process could conducted downward owing to the large thermal conductivity of the solid material. If the components are not fully melted continuously along the build direction, such as components with irregular structures, and piles of products for mass production, because of the thermal conductivity of powder is lower than that of the solid material, the powders will hinder the heat from conducting downward, and lower the temperature at the bottom, which could attribute to the microstructural homogeneity. Therefore, the components with different geometric structures were fabricated, and investigations were conducted to compare the properties between the different structures as well as along their build direction respectively.

Specimens with layered and solid structures were fabricated with EBM under same build parameters. The microstructure was examined through SEM, EBSD, electron probe micro-analyzer (EPMA), chemical composition analysis, and transmission electron microscope (TEM). The mechanical properties were examined with micro hardness, and tensile tests.

Owing to the texture developed from EBM process that could grow stronger continuously along the build direction, the solid specimens exhibited stronger texture comparing with that of the layered specimens. Consequently, the tensile properties for solid specimens showed relatively anisotropic along various loading directions, and the tensile properties for layered specimens were homogeneous along various loading direction as well as along the build direction. Generally, layered specimens exhibited more homogeneous microstructure and mechanical properties than those of the solid specimens, which indicated that the EBM-built components with complex structures could possess more homogeneous properties.

Chapter 5

Although EBM performs well in manufacturing with CP-Ti, there are still limitations of this kind of technology, such as the resolution, maximum size of the components, high costs, etc. Consequently, EBM-built CP-Ti, as one of the excellent biomaterials, is quite appropriate to be applied in biomedical industries in the near future, especially for high value-added parts, such as customized implants, etc. And several studies have already successfully fabricated the CP-Ti implants using EBM. The biocompatibility is one of the important characteristics for biomaterials. The biocompatibility of material is not only related with its alloy contents, but also its microstructure, thermal experience, surface roughness, etc. For the EBM fabricated components, they usually exhibit rather rough surfaces. Hence, biocompatibility of the as-built surface would be important for whether the as-built implants could be applied directly. Therefore, the biocompatibility of EBM-built CP-Ti was examined through MTT assay.

Specimens prepared for MTT assay were divided into polished surface with three kinds of microstructural morphologies (columnar, granular, and mixed microstructure), and as-built middle surface of CP-Ti fabricated through EBM. In addition, CP-Ti specimens fabricated through conventional method (rolled-annealed) were also prepared for comparison. The MTT assay were conducted using BHK-21 fibroblast cell. The toxicity of the specimens were evaluated through the cell viability that calculated based on the measurements with the microplate reader (ELISA reader). And the attachment and spreading of cells onto the specimens were investigated through SEM observation.

The polished surface of as-built CP-Ti exhibited excellent biocompatibility, which is comparable or even higher comparing with that of the specimens fabricated through the conventional methods. However, the biocompatibility for the as-built surfaces with large surface roughness was insufficient for direct biomedical application owing to the extremely high surface roughness (Ra = 78 μm), which hindered the spreading of cells on the surface of the specimens. Therefore, Extra post processing procedure should be required prior to the application of the EBM-built implants, such as blasting, machining, etc.

Conclusions

In summary, the study showed that the EBM-built CP-Ti exhibited good performance at microstructure and tensile properties, which makes CP-Ti a promising material for EBM technology, especially in fabricating high value-added products.

論文審査結果の要旨

チタンおよびその合金は、高い比強度および優れた耐食性・生体適合性を有するため、航空宇宙・生体医療分野で広く使用されている。工業用純チタン（CP-Ti）は生体毒性やアレルギー性を示す元素を含まないため高い生体適合性を示すが、高温での製造プロセスにおける酸化による特性劣化や、結晶構造に由来して加工性が低い点が課題である。本論文は、近年大きな注目を集めている Additive Manufacturing 技術のうち、真空環境にて造形を行い、複雑形状のネットシェイプ成形も可能な電子ビーム積層造形（EBM）の CP-Ti への適用について検討したもので、全 6 章より構成されている。

第 1 章は本論文の序論であり、本研究の背景や目的について示している。

第 2 章では、種々の電子ビーム出力および走査速度にて作製した試料を基にプロセスマップを構築し、緻密かつ形状精度に優れた造形物を得るためのプロセスウインドウを決定している。また、得られた試験片の組織と室温引張特性について、プロセスパラメータの影響を明らかにした。

第 3 章では、EBM プロセスにおける予熱温度の影響について検討している。他の造形条件が同じであっても予熱温度の低下により入熱量が低下し、造形物中に残存した未溶融粉末が強度・延性に影響を及ぼすことを明らかにした。

第 4 章では、積層方向に緻密層と粉末層を交互に積層させた造形物を作製し、粉末層の存在が緻密層の組織および機械的特性に与える影響について調べている。粉末層が存在することで緻密層の集合組織形成が抑制され、力学特性の面内異方性が低減されることを見出した。

第 5 章では、EBM を用いて作製した CP-Ti の生体適合性について調べている。表面を研磨した試験片では既存材と同等の良好な生体適合性を示したが、造形ままの表面では低い値が得られた。この理由として、造形ままでは表面粗さが大きいためと結論付けている。

第 6 章は本論文の結論である。

以上、本論文は、EBM を用いた CP-Ti の造形について、電子ビーム出力、走査速度、予熱温度を考慮してプロセス最適化を行うとともに、組織や力学特性に及ぼすプロセスパラメータの影響について基礎的に明らかにした。また、生体医療分野への応用に向け、作製した造形物の生体適合性についても評価を行っており、材料システム工学の発展に寄与するところが少なくない。

よって、本論文は博士(工学)の学位論文として合格と認める。