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Full Length Article

Mechanical properties and microstructures in zirconium deposited by injected powder laser additive manufacturing

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ABSTRACT

The use of laser additive manufacturing based on melting of injected zirconium powder under localized shielding was evaluated in terms of microstructures and mechanical properties of thin wall structures. The material was characterized in both the laser travel and the build directions. The microstructures, tensile properties and fracture behavior were assessed for deposits made using as-received and recycled powder. Electron back-scattered diffraction and transmission electron microcopy revealed a fine structure of Zr-α laths with nano-scale iron-rich precipitates at the lath interfaces. The properties of the fabricated components, which were made using new as-received powder were comparable to a Zr-2.5Nb alloy substrate, with yield strengths of over 569 MPa and uniform strains up to the ultimate tensile stress ranging from 8.5 to 9.9%. However, when recycled powder was used, the ductility dropped with total strains to failure of 1.0–7.5%, as a result of porosity and unmelted powder particles serving as brittle inclusions in the deposited material.

1. Introduction

The development of additive manufacturing methods has enabled a wide range of materials to be deposited with relatively high geometric precision [1]. The use of polymer based additive manufacturing has evolved to provide commercial 3D printing systems which are commonplace for prototyping, however additive manufacturing of metallic components is less common except for more exotic high-value applications. One such area involves nuclear and medical applications where zirconium alloys are commonly used. Since this metal is particularly costly and may be difficult to process using conventional manufacturing processes, this represents great potential for additive manufacturing techniques.

The feasibility of Directed Energy Deposition (DED) laser additive manufacturing using zirconium alloys was examined in a prior study which developed a processing window exploring suitable laser power, travel speed, laser spot size, and powder feed rates to produce defect-free claddings on a zirconium alloy substrate [2]. It was found that pure zirconium powder could be deposited at a rate of 0.80 g/min with a dilution rate of 50% with the substrate, however a low deposition efficiency of only 9.2% was achieved. Considering the high value of the powder material, it is of great interest to consider recycling the un-adhered powder. There also remains a great need to compare the microstructures and mechanical properties in additive manufactured components versus conventional processes. Progress has been made in this regard in the case of titanium alloys where the grain structures and tensile stress-strain behaviour of deposited Ti-6Al-4V alloys have been studied in great detail, including anisotropy of properties in each of the build axes [3].

The present work examines some of these issues during DED laser additive manufacturing (by direct powder injection) of pure zirconium components. The geometry considered here as a test case is a simple planar wall deposited onto a zirconium alloy substrate. Since the laser deposition process is not 100% efficient, the feasibility of recycling the zirconium powder which did not adhere is also studied by evaluating the properties of components made from this reused material.

2. Experimental methods

The system used for laser deposition consisted of a 1.1 kW fiber laser, with a specially designed concentric nozzle to deliver the powder using an Argon carrier gas, along with a supplemental flow of localized auxiliary shielding gas. Details of this nozzle design and its effect on the quality of the deposition are described in a prior work where increasing auxiliary gas flow rates were evaluated to reduce surface oxidation to levels comparable to Zr based welding processes [4]. The deposition

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Table 1
Processing Parameters used for Zr deposition.

Variable	Value
Laser spot diameter	1.4 mm
Laser scan speed	245 mm/min
Powder feed rate	8.9 g/min
Auxiliary shielding gas flow rate	33.0 L/min
Auxiliary shielding gas nozzle type	TIG
Auxiliary shielding gas nozzle discharge diameter	15.9 mm
Argon gas purity	≥99.999%
Angle of nozzle to substrate	45 ± 3°
Distance from the powder delivery nozzle to the deposit	10–11 mm
Distance from tip of shielding gas nozzle to powder delivery nozzle	3 mm
Dwell time between two consecutive passes	5 s

parameters were identified in a previous work which established a recommended processing window to provide a suitable and stable geometry in the deposit [2]. The processing parameters used to deposit thin wall structures are summarized in Table 1.

Cold rolled sheets of Zr-2Nb with dimension of 51 mm × 153 mm × 1.6 mm were used as a substrate for the build. The as-received zirconium powder used for deposition had a composition measured as shown in Table 2, and was produced by spray atomization to achieve nearly spherical powder with a size range such that 96.0% of particles were between 44 to 105 μm. Note, the as-received powder was obtained from a commercial supplier, and represents the state of the art in commercial Zr production considering the difficulties in obtaining low oxygen contents. The recycled powder corresponded to the as-received material which was used during depositions, then collected from the surrounding area of the deposition substrate and sieved to ensure that the size range remained between 44 and 105 μm. This power was reused up to 5 times, and sieved between each deposition trial to maintain consistent size range.

Following deposition of the Zr powder, the oxygen and nitrogen content of the material were measured according to ASTM E1097 and E1409 using a LECO TC-436 system in order to assess the quality of the Ar shielding, based on samples extracted from the middle region of the deposited material. The specimens were examined by Scanning Electron Microscopy (SEM) and energy dispersive x-ray spectroscopy (EDX) using an accelerating voltage of 10–20 kV. Electron Backscattered Diffraction (EBSD) analysis was conducted with a step size of 0.25 μm and a grid size of 400 × 400. The microstructures were also examined by extracting thin foils and twin jet electro-polishing specimens suitable from Transmission Electron Microscopy (TEM) at 200 kV in a JEOL 2010 F instrument. To measure the hardness, a Vickers indenter was used with a 300 g load, and 15 s holding time. The tensile properties of the deposits and the Zr-2.5Nb substrate were measured by extracting specimens with a 6.0 × 2.5 × 1.1 mm gauge dimensions and 8.0 × 2.5 × 1.5 mm gauge dimension respectively. The tensile specimens were loaded to failure at a rate of 0.5 mm/min, using a Tinius Olsen HK10T test frame. X-ray inspection was conducted to ensure no gross defects or large pores were present in the tensile specimens. These dogbone specimens were extracted by Electrical Discharge Machining (EDM) in order to avoid any damage to the surface or microstructures. The strains during tensile testing were measured using digital image correlation using a Correlated Solutions camera system with VIC3D software.

3. Results and discussion

3.1. Fabrication of thin walled sheets

Thin walled sheets with dimensions of approximately 120 × 35 × 1.5 mm were fabricated by cladding repeatedly on the same track in one direction. After deposition of each layer, the powder delivery nozzle stayed at the end of the cladding pass for 5 s before travelling back to the starting point of the deposit. The dwell time was introduced to provide sufficient shielding at the end of the deposit. Prior to creating the thin walled structure shown in Fig. 1, smaller 10 layer deposits were made with varying height increments. The height increment setting that was most successful at maintaining the distance between laser head and the top of the deposition was chosen for creating the thin walled structure. As a result, the CNC machine was programmed to raise the height of the laser head and the powder delivery nozzle by 0.702 mm for a total of 56 layers using the as-received powder, and 0.617 mm for a total of 50 layers using the recycled powder when applying a laser intensity of 800 W. The deposition process was unstable at 400 W, leading to an irregular deposit shown in Fig. 1b, and so further study was only conducted using 800 W. Comparing the recycled and as-received powder deposits in Fig. 1, it can be noted that some irregularities occurred at the beginning of the deposit when using recycled powder. These irregularities and the lower layer thickness of the recycled powder suggests that the recycled powder's melt pool was less stable. The microstructures were analyzed in boxed areas in Fig. 1a and c, and the uniformity of the wall profile was studied by optical microscopy along the dotted cut lines adjacent to those locations.

Chemical analysis was also performed on the 800 W deposit from the vertically middle of the boxed regions shown in Fig. 1. The analysis revealed that the nitrogen and oxygen contents of the deposit, which was made using the as-received powder, were 330 and 4300 ppm respectively. In comparison, the deposit which was made using the recycled powder contained 350 and 4300 ppm of nitrogen and oxygen respectively, which suggests most of the absorption of these elements occurs during the first pass through the deposition process. This also indicates that the bulk material approached an absorption limit after the first processing cycle. The standards for welding of zirconium alloys suggested by AWS [5] state that zirconium turns from silver to straw/gold, to intermediate colours, until finally a powdery white/yellow is produced when it is heated above 427 °C in an open air atmosphere. It also states that acceptable zirconium welds can only be silver or dark straw in colour. The 800 W recycled and as-received powder deposits, as shown in Fig. 1, exhibited purple discolouration towards the top left corner since this location remained at elevated temperatures for the longest time due to heat from previous cladding layers. The presence of purple discolouration in the top left corner indicates that more shielding is required in some regions of the deposit than others, which may be accomplished using a larger gas lens with proportionally increased shielding gas flow rate. Alternatively, a trailing gas shield can also be used. The surface of recycled powder deposit experienced more severe discolouration than the as-received powder deposit, likely due to the higher energy deposition conditions given the smaller build direction height increment between clads.

Fig. 2 shows cross sections of 800 W recycled and as-received powder depositions along the sectioning lines indicated at the arrows in Fig. 1. The figure shows that the successive layers within the deposition

Table 2
Chemical analysis of as-received Zr powder used for deposition.

Element	C	Cr	Fe	H	Hf	Mg	N	Nb	Ni	O	Si	Sn	Zr
Concentration (ppm)	70	< 50	630	19	190	< 1	270	< 50	< 35	2500	17	335	Bal.