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Microstructural evolution and mechanical properties of differently heat-treated binder jet printed samples from gas- and water-atomized alloy 625 powders

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ABSTRACT

In this study, we investigate the effect of powders resulting from different atomization methods on properties of binder jet printed and heat-treated samples. Air-melted gas atomized (GA) and water atomized (WA) nickel-based alloy 625 powders were used to binder jet print samples for a detailed comparative study on microstructural evolution and mechanical properties. GA printed samples achieved higher sintering density (99.2%) than WA samples (95.0%) due to differences in powder morphology and chemistry. Grain sizes of GA and WA samples at their highest density were 89 ± 21 μm and 88 ± 26 μm, respectively. Mechanical tests were conducted on optimally sintered samples and sintered plus aged samples; aging further improved microstructure and mechanical properties. This study shows that microstructural evolution (densification, and carbide, oxide and intermetallic phase formation) is very different for GA and WA binder jet printed and heat-treated samples. This difference in microstructural evolution results in different mechanical properties with the superior sintered and aged GA specimen reaching a hardness of 327 ± 7 $HV_{0.1}$, yield strength of 394 ± 15 MPa, and ultimate tensile strength of 718 ± 14 MPa which are higher than cast alloy 625 values.

1. Introduction

Alloy 625 is a nickel-based alloy broadly utilized in aerospace, chemical and petrochemical applications as a high temperature structural material due to its high temperature mechanical and corrosion resistance properties [1–5]. Alloy 625 is a solid-solution strengthened face-centered cubic (fcc) Ni–Cr–Mo alloy; however, precipitation hardening also plays a significant role in hardness, tensile and creep strength [6,7]. Additionally, intermetallic and carbide precipitates may form during aging at 550–750 °C. Various precipitates such as γ′ [Ni_3(Al, Ti)], γ″ [Ni_3(Nb, Al, Ti)], δ [Ni_3Nb] and different carbides including $M_{23}C_6$, MC, M_6C ('M' is Cr, Ni, Nb or Mo) as well as Laves phase (Cr, Fe, $Ni)_2$(Ti, Nb) and Ni_2(Cr, Mo) form in Ni-based alloys depending on composition, aging time and temperature [7–10]. Casting is a major conventional part fabrication method for Ni-based alloys; however, elemental segregation, shrinkage defects and formation of undesirable phases are serious issues encountered in large parts [1]. Reducing these defects is a time consuming and expensive process, and forming and machining of the cast parts are difficult due to high hardness, mechanical strength ductility and work hardening [11].

Powder bed binder jet printing (BJP), also referred to as binder jetting, is a fast, low-cost additive manufacturing (AM) process enabling the manufacture of complex internal and external geometries [12]. During BJP, powder is deposited layer-by-layer and selectively bonded in each layer with binder [13,14]. The powder bed is then heat treated for curing, and finally, the resulting "green" parts are sintered to fully densify the low density cured parts [15]. Detailed studies are needed to find optimum powder and processing parameters resulting in similar or improved microstructure and properties of BJP alloy 625 parts compared to cast parts.

In general, two common techniques are used to produce nickel-based alloy powders: (1) gas atomization (GA), leading to spherical particles that pack to higher density, and (2) water atomization (WA), leading to irregular particles with better shape retention ability and lower cost [16]. Although GA powder is most commonly

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used for manufacturing of Ni superalloy components, higher costs are a consideration. To achieve required material characteristics such as ductility, strength and corrosion resistance, microstructural evolution during sintering is crucial. For injection molded WA and GA stainless steel powder, sintering results in 97% density for WA powder samples, while GA powder parts sintered to near-full density due to particle morphology, initial green density and particle chemistry [16]. For WA and GA Ni-based alloy powder, a denser microstructure was reported for GA samples than WA samples because of lower oxygen content and higher packing density of GA powder [1].

To our knowledge, there has been no systematic comparison of the properties of WA and GA Ni-based alloy parts made by AM. We therefore binder jet printed samples with WA and GA alloy 625 and compared the effects of powder and sintering and aging processes on microstructure evolution, densification and mechanical behavior. This information enables the use of suitable numerical models to further improve processing and will result in a better understanding and development of microstructure and mechanical properties of binder jet printed alloy 625 WA and GA parts.

2. Materials and methods

In this study, the nitrogen gas and water atomized alloy 625 powders were supplied by Carpenter Technology Corporation and HAI Advanced Material Specialists, Inc., respectively. The nominal, provided and measured chemical compositions are given in Table 1.

Powder morphology and elemental composition analyses were conducted with a JEOL (JSM 6510) scanning electron microscope (SEM) equipped with energy dispersive X-ray spectroscopy (EDS). X-ray diffraction (XRD) was performed on a PANanalytical EMPYREAN diffractometer with a Co K_α radiation source (λ = 1.79 Å, 40 kV, 40 mA) and 2θ scans from 35° to 120°. The particle size distribution (1 g each suspended in isopropyl alcohol to prevent agglomeration) by volume was analyzed with a Microtrac S3500 tri-laser diffraction particle analyzer via spherical and non-spherical particle algorithms. Phase transformation temperatures (solidus, solvus) were measured with a TA Instruments Q 600 differential scanning calorimeter (DSC) by heating powder samples to 1350 °C at 10 °C/min in ultra-high purity Ar (100 mL/min).

General morphology and internal porosity of powder particles and as-printed samples were visualized with a Bruker SkyScan1272 micro-computed tomography scanner (μCT) at 100 kV and 100 μA and a 0.11 mm Cu filter, averaging of 10 frames, and angular range of 0°–180° with 0.2°–0.3° steps. Powder samples were filled into a low absorbance 1.5 mm plastic straw, compacted to reduce particle movement and scanned without random movement.

To manufacture parts, an M-Flex ExOne binder jet printer (BJP) was used as detailed in Ref. [15]. After printing, the fragile GA and WA parts ("green parts") were cured at 175 °C in a JPW Design & Manufacturing furnace and then sintered in a Lindberg tube furnace in an alumina powder bed under vacuum with the following heating profile: heating at 5 °C/min from room temperature to 600 °C, 3.2 °C/min to 1000 °C, 2.8 °C/min to the holding temperature (1225, 1240, 1255, 1270, 1285 or 1300 °C), holding for 4 h and then cooling at 1 °C/min to 1200 °C, 3.1 °C/min to 500 °C and finally cooling down to room temperature [15]. Five samples for each combination of sintering temperature and powder type were prepared. After sintering, samples with highest density were solution treated at 1150 °C for 2 h and aged at 745 °C for 20 h.

Density and volume changes (shrinkage) of the as-printed and sintered samples were measured via the Archimedes principle with an OHAUS AX324 precision balance (0.1 mg resolution). Density of the sintered samples was also determined from sample cross-sectional optical micrographs with the ImageJ image analysis software [17]. For microstructural examination of the sintered coupons, cross sections were cut from the specimens, mounted, ground and polished using a Struers Tegramin-25 automatic system according to [18]. After surface preparations, samples were etched with a Kalling solution (ASTM E407, # 94) and micrographs were taken with a Keyence digital optical microscope (OM) (dark field Z20 lens and multi-diffused adapter). Microstructural characterizations, compositional analysis and fractography were conducted with a JEOL JSM 6510 SEM equipped with EDS. The *d*-spacing values and lattice parameters were determined with the same XRD and scan parameters used for as-received powder. Carbon and oxygen contaminations were detected with LECO TC600 Oxygen/Nitrogen and CS844 Carbon/Sulfur Analyzers.

Vickers microhardness tests were performed on the cross sections of samples with a Leco LM 800 microhardness tester (100 gf for 10 s). Rate controlled tensile tests at 5 mm/min were performed using an MTS 880. Dimensions of the as printed sample were 125 mm long, 12.5 mm wide, 7.5 mm thick, gage length of 29 mm following ASTM standards for metallic tensile test specimen.

3. Results and discussion

3.1. Powder feedstock characterizations

Powder particle micrographs showed irregular WA and spherical GA powder particles (Fig. 1). Both types of particles had a dendritic microstructure typical for rapid solidification. μCT scans of as-received powders shown in Fig. 1c and f confirm the SEM-observed powder particle morphology (irregular WA, spherical GA particles) and, additionally, revealed porosity within powder particles. Internal porosity that forms during rapid solidification in the powder atomization process was also seen in cross-sectional SEM micrographs (Fig. 1b and e).

Chemical compositions (as provided by manufacturer and as measured with EDS, Table 1) are all close to nominal alloy 625 composition. The main differences between the powder types were morphology, carbon and oxygen impurity content which was higher in WA (1.90 and 0.09 wt%) vs GA powder (0.04 and 0.01 wt%, Table 1), and the average powder size and particle size distribution

Table 1
EDS composition of WA and GA nickel-based alloy 625 powders and oxygen and carbon content of as-received powders and printed/sintered samples [wt.-%].

	Composition in atomic weight percent [wt-%]													
Elements	Ni	Cr	Fe	Nb	Mo	Al	Ti	Co	Mn	Si	O	C	O	C
Nominal composition [18]	>58	20–23	<5	3.1–4.1	8–10	<0.4	<0.4	<1.0	<0.5	<0.5	As-received		Printed and	
WA powder (provided)	Bal.	21.8	3.6	3.5	9.3	<0.4	<0.4	<1.0	0.4	–	powder		cured	
WA powder (EDS)	Bal.	16.9	3.7	3.7	8.7	0.05	0.16	0.62	0.8	0.7				
GA powder (provided)	Bal.	21.5	4.2	3.3	8.9	–	–	–	0.4	0.4				
GA powder (EDS)	Bal.	21.8	4.4	3.6	8.0	0.02	0.02	0.22	0.4	0.5				
WA											1.90	0.09	1.43	0.65
GA											0.04	0.01	0.28	0.44