The evolution history of superalloy powders during hot consolidation and plastic deformation

Liming Tan, Yunping Li, Chenze Liu, Chuan Yang, Hanhui Ding, Lan Huang, Feng Liu*, Zijun Qin, Liang Jiang*

State Key Laboratory of Powder Metallurgy, Central South University, Changsha 410083, China
Powder Metallurgy Research Institute, Central South University, Changsha 410083, China
High Temperature Materials Research Institute, Central South University, Changsha 410083, China

ABSTRACT

Powder metallurgy superalloys are widely used to produce critical rotating parts in turbine engines. However, the issues like what is the driving force of precipitation on prior particle boundary (PPB) during consolidation and how the PPB structure evolves during plastic deformation still remain to be resolved. In this study, with the characterization of the microstructure evolution from powder, consolidation, to plastic deformation, the segregation of alloying elements on the particle boundaries at elevated temperature was analyzed by equilibrium thermodynamic calculations, and the distortion of PPB structure under large plastic deformation was quantitively simulated by finite element (FE) methods. The results indicated that surface energy serves as the main driving force of PPB precipitation and hot extrusion could distort the PPB structure and yield more uniformly refined grains.

1. Introduction

Due to the excellent comprehensive properties at elevated temperatures, superalloys have been widely adopted as high temperature materials for aircraft engines [1–8]. In terms of polycrystalline superalloys, powder metallurgy (P/M) is used to produce critical turbine discs with complex compositions and less elemental macrosegregation [3,9–11]. Generally, typical powder metallurgy routes include powder preparation and consolidation, some plastic deformation methods like hot extrusion and forging are also employed at specific conditions [12,13]. Among them, atomization process like argon atomization (AA) and plasma rotation electric pole (PREP) are the common techniques to yield clean pre-alloyed powders [14]; hot isostatic pressing (HIPping) is the a widely used method to consolidate superalloy powder [15–18]; plastic deformation after HIPping, such as hot extrusion and isothermal forging, are mostly utilized to obtain disc parts with desired microstructure [19], for example, hot extrusion after consolidation is used to break up ceramic inclusions which can adversely impact low cycle fatigue (LCF). There is no doubt that direct hot isostatic pressing (as-HIP) would dramatically save costs of producing PM superalloys, but the frequent precipitation on prior particle boundary (PPB) during HIPping detriments the mechanical properties [17,20–26].

There have been numerous studies on the precipitates on PPB, which were found to consist of oxides, carbides, oxy-carbides, and γ' precipitates [18,24,27–32]. Some results also indicated that there were some optional ways to relatively reduce the detrimental effects of PPB precipitation, such as HIPping at higher temperature and subsequent supersolvus heat treatment, but these could induce excessive grain coarsening [18,25,33–35]. Hence thereafter hot deformations are broadly used to modify the PPB structure and refine the grains [36]. Nevertheless, few of them reported what is the driving force of PPB precipitation and how the PPB structure evolves during plastic deformation, even though much attention has been paid to the elemental segregation and grain boundary engineering in many alloy systems recently [37–40].

In this study, the characterization of the microstructure evolution from powder, HIPping, to plastic deformation was performed, and we tried to deal with the abovementioned two issues experimentally and theoretically. In specific, the segregation of alloying elements on the powder surface at elevated temperature was analyzed by equilibrium thermodynamic, and finite element (FE) simulation was performed to estimate the distortion of PPB structure under large plastic deformation.

2. Materials and Methods

The P/M superalloy FGH96 with chemical composition of Co 13, Cr

Table 1
Processing routes of FGH96 specimens.

	Atomization	Powder size, μm	HIP		Hot extrusion
			Temperature, °C	Pressure, MPa	
1#	AA	< 74	1170	150	Yes
2#	PREP	< 150	1150	150	No

16, Mo 4, W 4, Al 2.1, Ti 3.7, Nb 0.7, B 0.015, Zr 0.03 and C 0.03, rest Ni (wt%) was studied in this work. The γ' solvus temperature of FGH96 was around 1107 °C. In practice, the powders of FGH96 were prepared by argon atomization (AA) and plasma rotation electric pole (PREP) techniques, the screened powder was then loaded into a steel container which was degassed to 10^{-3} Pa at 400 °C and sealed. After that, the powder was consolidated by HIPping under different conditions as indicated in Table 1. During HIPping, the temperature and pressure were increased simultaneously, and held at set conditions for over 2 h, then the billets were gradually cooled within furnace. To investigate the PPB evolution under plastic deformation, one of the HIPped billets were hot extruded at 1070 °C with extrusion ratio of 7:1.

In order to investigate the elemental changes with the depth of superalloy powder, auger electron spectroscopy (AES) measurement was undertaken by spectrometer PHI 680, and the surface was moved layer by layer, via sputtering under 2 KeV positive Ar ions for 10 min. Based on the sputtering rates of AES on SiO2, about 30 Å/min, the sputtered depth of FGH96 powder can be roughly estimated. Moreover, Time-of-flight secondary ion spectrometry (ToF-SIMS) installed at FERA3 microscope was used to detect the segregation or distribution of alloying elements in powder, HIPped and extruded samples.

To characterize the microstructure evolution after HIPping and hot extrusion, scanning electron microscope (SEM) observation was performed under a field emission gun SEM FEI Quanta 650 equipped with electron backscatter diffraction (EBSD) detector. Additionally, grain structure in HIPped and extruded superalloys was studied by EBSD, the samples were polished by abrasive papers, followed by vibration polishing for over 8 h. EBSD scan step size for all samples was set as 0.5 μm to guarantee enough pixels in each detected grain. The EBSD data were analyzed by HKL CHANNEL5 software.

Transmission electron microscope (TEM) site-specific specimens were prepared with a FEI HELIOS NANOLAB 600i dual beam focused ion beam (FIB) milling technique, and TEM observation was carried out on FEI TEM instruments Tecnai G2 F20 with accelerating voltage of 200 KV to identify the precipitates on PPB.

3. Results

3.1. The Element Distribution of Powders

The results of AES analysis on a $50 \times 50\,\mu m^2$ PREP powder surface are shown in Fig. 1. Significant Ni, O, and C signals were detected at the powder surface prior to sputtering. As the powder was sputtered away layer by layer, other elements, such as Ti, Al, Cr, Mo, etc., started to be detected gradually. From Fig. 1, it can be judged that the changes of element concentrations, especially for C, O, and Ti, distinctly varied with the sputtering depth within 10 nm. C and O segregated at the surface or near-surface within 1.5 nm depth of the PREP powder were thought caused by surface adsorption and non-equilibrium phases.

Since AES can only present average element concentrations at a relatively wide area, the ToF-SIMS was used as a complement to characterize the element distributions on powder surface and interior, which are presented in Figs. 2 and 3 respectively. ToF-SIMS is not very sensitive to the light element ions (like O and C ions) and heavy element ions (like W and Ta ions), hence we just showed some selected alloying elements in this work. Considering the fact that edge effect may cause

some inaccuracy as the powder surface was not flat enough, it is more conclusive to state that Ti, Zr, and Nb were segregated at interdendrite or intercellular regions in FGH96 powder, based on ToF-SIMS analysis of powder section, as shown in Fig. 3.

3.2. The Microstructure After HIPping

The microstructure of PPB networks in HIPped FGH96 alloys are shown in Fig. 4, the average grain sizes of HIPped 1# and 2# FGH96 were around 4.8 μm (ASTM 12.5) and 10 μm (ASTM 11.2), correspondingly. PPB was obvious in both of the HIPped alloys, and it was denser in 1# specimen consolidated by smaller AA powders.

The ToF-SIMS results indicating element distribution at PPB in HIPped 1# FGH96 are shown in Fig. 5, which illustrates that Al, Ti, Zr, and some Nb were segregated along PPB.

TEM was adopted to further identify the kinds of precipitates along PPB. As indicated in Fig. 6, ZrO_2, Al_2O_3, MC-type carbide, and γ' were found existed along PPB in HIPped 2# specimen. Specifically, the oxides and carbides were much smaller than γ' precipitates, which was large enough to envelop the tiny ones. In consistent with this result, the presence of oxides, carbides, and γ' were also identified in HIPped 1# FGH96 [41].

3.3. The Microstructure After Hot Extrusion

As a complement to above part, the grain structure after HIPping and hot extrusion are presented in Fig. 7. It can be judged from Fig. 7(a) that some part of PPB acts as grain boundary after HIPping, particularly for the large PPB with high sphericity outlining the shape of initial powder, and a series of tiny grains were located along PPB. After extrusion, the PPB structure were distorted. Specifically, the PPB was elongated along extrusion direction to be rugby-ball like or oval shaped. Moreover, the grain size distribution was more homogeneous and smaller comparing with that of HIPped one.

To further identify the existence of distorted PPB structure, the distribution of alloying elements in 1# FGH96 after hot extrusion was characterized by ToF-SIMS, as illustrated in Fig. 8. Generally, the elements, such as Al, Cr, Ni, Ti, and Nb were randomly dispersed, but the Zr corresponding to stable ZrO_2 still remained at the distorted PPB.

4. Discussions

4.1. Thermodynamics of Precipitation on PPB

The atomized powders consist of non-equilibrium precipitates formed at extreme undercooling conditions, as the powder atomization is a process with very fast cooling rate approximate to 10^4–10^5 °C/s [42]. There exists segregation of element at dendrite-interdendrite, cellular-intercellular, or surface-interior regions in a single powder particle. Specifically, the detected O indicates the presence of oxides formed by reaction with the oxygen during atomization or oxygen absorbed from external atmosphere, the C element is related to the carbides in consistence with other similar works [31,42,43]. In addition, Ti, Nb, Zr are preferred to stay at interdendrite or intercellular area, as the γ phase with higher undercooling degree solidifies as dendrite or cellular firstly, while the left liquid with amount of γ'-forming elements Ti, Nb, and carbide or oxide forming elements Zr starts to solidify thereafter.

Even though there were some C, O and their compounds accumulated at powder surface, but the degree of the segregation after HIPping was much higher than the original powder statues, regarding the characterization of precipitates at PPB in HIPped. For example, the Zr existed in powder majorly migrate onto the PPB during HIPping. What is the driving force of this segregation? In order to explain that, let us try to solve it based on thermodynamic, mainly according to the methods developed by Luthra and Briant [44].