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Experimental and numerical analysis of the particle size effect on the densification behaviour of metal injection moulded tungsten parts during sintering

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ABSTRACT

The sintering behaviour of metal injection moulded tungsten components using fine (FSSS 0.4 to 1 μm) and coarse (FSSS 5.0 to 7.0 μm) powder has been investigated via dilatometric and three-point bending tests in a Setaram© analyser under a pure hydrogen atmosphere at temperatures up to 1700 °C. The experimental results have been applied to determine the material parameters in the viscoplastic constitutive law for continuum sintering model, which has been implemented in a finite element code ABAQUS software via the user subroutine UMAT for predicting the final shrinkage and density distribution of the sintered components. The effect of particle size of the powder on the densification behaviour, sintering activation energy, and sintering stress has been analysed.

1. Introduction

Metal injection moulding (MIM) is a near net-shaping technology for manufacturing small and complex components in large quantities, which includes mixing of metallic powder with plastic binder to form a feedstock, shaping the green parts by injection moulding of the feedstock into the cavities with desired shapes, removing the binder using thermal, solvent or catalytic methods to obtain brown parts with a porous structure, and sintering to a nearly full dense material and final dimensions of the parts [1]. Sintering, as a final step of MIM determining the final density and grain size, has an important effect on the mechanical and physical properties of the MIM components [2]. In general, the relative density of the sintered MIM components in materials such as iron alloys, stainless steel, copper, and ceramics is higher than 95% [3]. However, this high dense level is difficult to achieve at sintering temperatures below 1650 °C for pure tungsten (W), which has the highest melting temperature (3420 °C) among the metals [2]. Because of the very high fusion point, the consolidation of conventional W powders is difficult and generally requires a temperature in excess of 2700 °C through solid state sintering in a self-resistance sintering furnace under hydrogen (H_2) atmosphere [4]. The low as-sintered density is an obstacle to the production of pure W components by MIM for high temperature applications such as electrodes of high-intensity discharge lamps, plasma facing components for fusion reactors, and furnace parts.

Many efforts have been reported to improve the sinterability of W for the purpose of decreasing the sintering temperature. Hayden et al. [5] proposed to reduce the activation energy of sintering through the addition of small amounts (<1 wt.%) of Group VIII transition metals. German et al. [6] reported that the sintering temperature of W can be decreased from 2800 °C to 1400 °C by using less than 1 wt.% addition of transition metals, such as palladium and nickel. Yu et al. [7] investigated the effect of 0.4 wt.% Ni additives on the densification of MIM W–1.5% Al_2O_3 alloy. Ni had an important effect on promoting the densification process, which reduced the sintering temperature greatly. The residual Ni element in sintered parts of approximately 0.008 wt.%, however, would not be acceptable for some applications. Another important approach to activate the sintering of W is through the selection of a nano-sized precursor W powder. However, this powder is expensive and prone to contamination [8]. Malewar et al. [9] reported that the sintering temperature of nano-sized W produced by high-energy mechanical milling can be decreased from the conventional temperature 2500 °C to 1700 °C. El-Atwani et al. [10] performed sintering tests using fine-grained, hard, and ductile pure W powders for future fusion reactor applications. The bottom-up approach via powder consolidation by spark plasma sintering (SPS) was used under different temperature (1300–1800 °C) and pressure (90–266 MPa) conditions. Pure W powder, with an average particle size of approximately 1 μm was sintered to high density, approximately 94% of the theoretical one, with almost

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List of symbols

λ	shrinkage during sintering
Δl	change in length
l_0	original length of pre-sintered components
l	length at any time during sintering process
P	external load at the centre of the specimens for three-point bending testing
$\dot{\delta}$	deflection rate
$\Delta\delta$	change in deflection
ΔT	change in temperature
Δt	change in time
β	heating rate
T_1	temperature at sintering time t
T_2	temperature at sintering time $t + \Delta t$
v_1	isothermal shrinkage rate recorded at a temperature T_1
v_2	isothermal shrinkage rate recorded at a temperature T_2
$K(T)$	Arrhenius constant
t	time
n	constant that depends on the sintering mechanism
γ	free surface energy
Ω	atomic volume
G_c	size of the crystallite
k	Boltzmann's constant
R	universal gas constant
D_0	pre-exponential term of the diffusion coefficient
m	constant that depends on the geometric shape selected for the particles
ψ	constant that depends on the geometric shape selected for the particles
A	constant that depends on the geometric shape selected for the particles
$\dot{\varepsilon}$	total strain rate
$\dot{\varepsilon}_e$	elastic strain rate
$\dot{\varepsilon}_{th}$	thermal strain rate
$\dot{\varepsilon}_{vp}$	viscoplastic strain rate
D_e	elastic stiffness matrix for the isotropic materials
$\dot{T}$	incremental temperature rate
σ'	deviatoric stress tensor
σ_m	mean stress
$\mathrm{tr}(\sigma)$	stress tensor trace
I	second-order identity tensor
G_p	shear viscosity of the porous material
K_p	bulk viscosity of the porous material
σ_s	sintering stress
B	material parameter of sintering stress
C	material parameter of sintering stress
$\dot{\varepsilon}_z^{vp}$	vertical strain rate
σ_z	applied external stress
ν_p	Poisson's ratio
T	absolute temperature
G	grain size
V_a	atomic volume
D_{b0}	grain boundary diffusion frequency
δ_b	thickness of grain boundary
Q_b	activation energy for grain boundary diffusion
η_p	uniaxial viscosity
ρ_0	initial relative density after pre-sintering step
ρ	relative density
g	gravity acceleration
b	width of the beam-binding component
h	thickness of beam-binding component
Q	sintering activation energy
Q_g	activation energy for grain growth
α	coefficient of thermal dilatation
$G(x)$	objective function
λ^{exp}	experimental uniaxial shrinkage
λ^{num}	numerical uniaxial shrinkage

no grain growth at a temperature below 1400 °C and an applied pressure up to 266 MPa. Chanthapan et al. [11] performed an experimental investigation using W powder (0.6–0.9 μm) sintered by field-assisted sintering technology (FAST) at various processing conditions. The sample sintered with in-situ hydrogen (H_2) reduction pretreatment and pulsed electric current during heating contained the lowest amount of oxygen. The maximum relative density achieved was 98.5%, which was for the sample sintered at 2000 °C, 85 MPa for 30-min holding time. Recently, Wang et al. [12] reported that nanocrystalline W powder can be sintered to near full density at a temperature as low as 1100 °C under a H_2 atmosphere without external pressure. Prabhu et al. [13] led an experimental investigation with anther sintering heating mode; microwave sintering has been studied using as-received W and activated W powder. These authors concluded that the relative density of the sintered part using as-received powder was 85% and that using activated powder was 93%. Because MIM is a process for producing small parts in complex shapes, innovative sintering approaches such as SPS, FAST and microwave sintering have not been applied in MIM industries to date. Piotter et al. [14] performed a comprehensive study on the conventional sintering of W samples with a powder particle size of 2 μm in a dry H_2 atmosphere at temperatures higher than 2000 °C. Thermal treatment resulted in a grain size of approximately 18 μm and a final density of 95% measured by a helium pycnometer. To reduce the grain size of the sintered W parts, one solution is to use fine powders (e.g., an average particle size of 0.7 μm), which can be sintered at lower temperature (e.g., 1650 °C) to obtain closed residual pores necessary for the subsequent hot isostatic pressing (HIP) treatment. With the HIP cycle at 1600 °C under 250 MPa for 3 h, the final MIM W parts can reach a near full density (relative density of 98.6–99%) and fine grain structures (grain size of approximately 5 μm) [15,16]. In addition to the application for near fully dense parts, MIM has also been used to produce porous W skeletons [17]. In this case, the relatively coarse powders were crucial to the performance of the final parts.

In this study, conventional sintering experiments in a dilatometer is used to investigate the densification behaviours of different components made of fine (e.g., the Fisher Sub Sieve Sizer (FSSS) particle size of 0.4, 1.0 and 3.0 μm) and coarse (e.g., the FSSS particle size of 5.0, 6.0 and 7.0 μm) W powders, with a maximum sintering temperature 1700 °C. Three-point bending testing is employed to measure the viscosity of the components during the sintering process. The continuum sintering model with a viscoplastic constitutive law and the identified material parameters has been used to predict the shrinkage, distortion and density distribution of the sintered parts. The numerical simulation is implemented by using the finite element code ABAQUS and the user subroutine UMAT.

2. Experimental analysis

2.1. Materials and sample preparation

Fine and coarse W powders of various particle sizes (Xiamen Honglu, China) were used in this study. To obtain the MIM powders with good flowability, deagglomeration was performed first via rod milling. For each type of W powder, 5 kg of powder was placed into a 5-litre polyurethane bottle with 15 kg of W rods, which has the diameter of 8 mm and length of 10 mm. The powder was milled for 8 h at a rotation speed of approximately 28 rpm. Fig. 1a and b presents scanning electron microscope (SEM) images of the fine (FSSS 0.4 μm) and coarse