Acta Materialia 147 (2018) 24–34

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Microwave flash sintering of metal powders: From experimental evidence to multiphysics simulation

Charles Manière [a], Geuntak Lee [a, b], Tony Zahrah [c], Eugene A. Olevsky [a, d, *]

[a] Powder Technology Laboratory, San Diego State University, San Diego, USA
[b] Mechanical and Aerospace Engineering, University of California, La Jolla, San Diego, USA
[c] MATSYS Inc., Sterling, USA
[d] NanoEngineering, University of California, La Jolla, San Diego, USA

ARTICLE INFO

Article history:
Received 2 June 2017
Received in revised form
23 December 2017
Accepted 2 January 2018

Keywords:
Flash sintering
Microwave
Densification
Electromagnetic-thermo-mechanical model
Ti-6Al-4V

ABSTRACT

Flash sintering phenomena are predominantly associated with ceramics due to thermal runaway of their electric conductivity noticeably represented in materials such as zirconia or silicon carbide. Because of their high electric conductivity, flash sintering of metals is nearly inexistent. In this work, an original metal powder flash sintering method based on a microwave approach is presented. Within the developed approach, an unusually fast (60 s) thermal and sintering runaway of Ti-6Al-4V powder is experimentally revealed under microwave illumination. This phenomenon is simulated based on an electromagnetic-thermal-mechanical (EMTM) model. The developed multiphysics model reveals that the metal powder specimen's runaway does not result from its intrinsic material properties, but results from the resonance phenomenon thermally activated by the surrounding tooling material. The EMTM simulation predicts with a very good accuracy the microwave repartition and the resulting densification and powder specimen's shape distortions observed experimentally. The comparison of the microwave and conventional sintering kinetics indicates an important acceleration of the sintering behavior under microwave heating. The developed sintering approach has a potential of the implementation for time-effective mass production of small metal parts.

1. Introduction

Since the publication of Cologna et al. [1] in 2010, the flash sintering phenomena encounter a great interest in the field of sintering studies. Flash sintering is considered to be one of the most efficient sintering approaches in terms of energy consumption, operating time and productivity for acceptable microstructures. The traditional flash sintering approach [1–4] involves resistive Joule heating of a green sample between two electrodes. In this process, the first incubation stage is observed where the temperature is slowly increased. In the second stage, a thermal runaway happens where the temperatures are quickly increased and accompanied by the fast sintering of the specimen in less than a minute. The thermal runaway profile of this process can be attributed to the negative temperature coefficient (NTC) behavior of the material [5–7]. The materials classically employed for flash sintering, such as zirconia or silicon carbide, require a preheating in a furnace to activate the electric conductivity of the green specimen, which is too low at room temperature [2].

Different alternative approaches have been developed based on the spark plasma sintering (SPS) [8–10], plasma electrode sintering [11], pressure-assisted flash sintering [12], and microwave sintering [13,14]. The flash spark plasma sintering (FSPS) approach using graphite felt as a preheating element has been employed to fully consolidate ZrB_2 [9] and SiC [10] during 17–40 s. Another flash spark plasma sintering (ultra-rapid hot pressing) approach using a copper sacrificial tooling element has been employed and allowed the full densification of SiC in 1 s [8]. A contactless flash sintering based on arc plasma electrode has been also developed enabling the full densification of carbide materials (pure B_4C and SiC:B_4C 50wt%) in about a few seconds of the discharge time [11]. Under microwave illumination, NTC ceramics heating demonstrates hot spot instabilities and exhibits a similar thermal runaway [15,16]. The microwave power dissipation of dielectric ceramics depends on the permittivity imaginary part that can be expressed by the

* Corresponding author. Powder Technology Laboratory, San Diego State University, 5500 Campanile Drive, San Diego, CA, 92182-1323, USA.
E-mail address: eolevsky@mail.sdsu.edu (E.A. Olevsky).

https://doi.org/10.1016/j.actamat.2018.01.017
1359-6454/

material electric conductivity [17,18]. Therefore, for similar reasons, the NTC behavior of some ceramic materials responsible for thermal runaway in traditional resistive flash sintering approaches can also generate a thermal runaway under microwave illumination [13,15]. The microwave flash sintering of ceramics has been demonstrated for Al_2O_3, Y_2O_3, $MgAl_2O_4$, and Yb:$(LaY)_2O_3$ with dense (98–99%) microstructures [13,19,20]. These studies revealed by SEM and AFN characterization the presence of a particle surface softening/melting mechanism that significantly accelerated the sintering for Yb:$(LaY)_2O_3$ [13]. For the same material, transparent specimens have been obtained by the microwave flash method [14]. One of the microwave flash sintering advantages is the volumic and contactless heating of the sample; the flash process can then be entirely controlled by the microwave power.

Most of the materials employed for flash sintering described in the literature are oxides or carbides. A recent flash sintering review [21] estimates the proportion of different materials used for flash sintering as 96% for oxide, carbide and only 3% for conductive high temperature ceramics like $MoSi_2$, ZrB_2. The presence of common metals and alloys among the materials subjectable to flash sintering is nearly inexistent, because their resistivity properties do not manifest the NTC behavior and, in turn, no natural thermal runaway under constant applied voltage. The exception is the flash microwave inkjet techniques of polymer/silver mixtures under antenna microwave illumination that allow fast agglomeration and consolidation of silver particles [22,23].

In this paper, we show that a thermal runaway accompanied by a fast densification similar to the traditional flash sintering method can occur also in metal powders when using a microwave heating approach. It is shown experimentally and demonstrated theoretically how an unusual metal thermal runaway can be obtained based on a resonance phenomenon thermally activated by the surrounding tooling materials. In the present study, the experimental results have been compared to the outcomes of the electromagnetic-thermo-mechanical (EMTM) simulation able to predict the cavity microwave distribution, heating, sample densification and shape distortions [24]. In addition, the conventional sintering densification behavior has been compared to the microwave sintering experiments and showed a considerable densification kinetic acceleration under the metal powder flash microwave sintering mode.

2. Experimental procedures

The microwave sintering experiments have been carried out using the microwave furnace Bloomden PMF15B. The microwave cavity is made of a rectangular waveguide where a magnetron (WITOL 2M343K E625) is connected and a 135 mm diameter cylindrical heating area is located (see cavity Fig. 1). Two SiC susceptors and zirconia balls bed are employed in the heating area. The effective heating area dimensions are 50 mm diameter and 30 mm height, the other areas of the 135 mm cylindrical cavity are filled with fibrous alumina-silica insulation (80%Al_2O_3-20%SiO_2). The samples are pre-consolidated by spark plasma sintering (SPSS DR. SINTER Fuji Electronics model 5015) to a relative density of 40% in order to avoid using a polymeric binder. The sample dimensions after pre-consolidation are 10 mm of diameter and 7 mm height. Ti-6Al-4V powder including 50 μm agglomerates has been employed (see in Fig. 2a the SEM image of the powder). The energy dispersive x-ray spectroscopy (EDS) analysis reveals the presence of a certain level of powder particle surface oxidation (Fig. 2b). The microwave sintering has been conducted utilizing two heating modes. On the one hand, the mode when the Ti-6Al-4V sample is located on the zirconia bed in the center and at the edge of the heating area (called "free surface" FS mode) has been used. On the other hand, the mode when the sample is located in an alumina container and surrounded by a 45–65 nm SiC nano-size powder (called "powder surrounded" PS mode) has been employed. The microwave sintering tests were performed using the constant applied power of 1300 W.

The microwave sintering outcomes have been compared to the results of the conventional sintering using a dilatometer (Unitherm model 1161 dilatometer system) with a cycle of 10 K/min heating rate up to 1480 °C. Both microwave and dilatometer test were conducted under argon atmosphere in order to prevent oxidation of the samples. The sample temperature has been estimated by an S type thermocouple for the dilatometer and by a pyrometer (Mikron infrared MI-P140, temperature range 200–1300 °C) for the microwave sintering. In order to reveal the Ti-6Al-4V sample microstructures, the specimens where cut in half, mirror polished and etched using the Shelton mixture [25] (HF 10 ml, HNO_3 5 ml, H_2O_2 30 ml, H_2O 10 ml).

3. Theory and calculations

3.1. Electromagnetic-thermal-mechanical simulation

The analysis of the microwave heating and sintering behavior requires a comprehensive multiphysics simulation including an electromagnetic part to describe the wave distribution in the cavity, a thermal part for the temperature field repartition, and a mechanical part for the resulting sintering of the sample. Based on our previous work [24], a fully coupled electromagnetic-thermal-mechanical (EMTM) simulation has been employed. This model also includes advanced simulation tools to calculate the surface-to-surface thermal radiation in the internal hot areas. The governing equations of the EMTM model are described in Refs. [15,24]. The external metallic surfaces are subjected to a surface-to-ambient thermal radiation and the convective heat loss is described by an emissivity of 0.79 and the convection heat flux of 5 $W\,m^{-2}$. K^{-1} [26]. The internal boundary emissivity and the electromagnetic-thermal-mechanical material properties are reported in Table A [15,24,27–32].

3.2. Material parameters determination

Some material properties depend on porosity and temperature and require a theoretical approach to be estimated. The dielectric properties of the porous tools like the zirconia balls bed and the nano-SiC powder have been estimated by the effective medium approximation equation (1) [15,18].

$$C_s\left(\frac{\varepsilon_{solid}-\varepsilon_{eff}}{\varepsilon_{solid}+2\,\varepsilon_{eff}}\right)+(1-C_s)\left(\frac{\varepsilon_{gas}-\varepsilon_{eff}}{\varepsilon_{gas}+2\,\varepsilon_{eff}}\right)=0 \quad (1)$$

with, ε_{solid} the solid phase complex permittivity (zirconia or silicon carbide), ε_{gas} the gas permittivity equal 1, ε_{eff} the effective complex permittivity of the whole porous material to be determined, and C_s the solid concentration that is associated with the relative density.

The thermal conductivity of the zirconia balls bed and nano-SiC powder can be determined by another equation (2) that also originates from the effective medium approximation and includes the thermal conductivity of the material and argon gas [18].