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Microwave resonant sintering of powder metals

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ABSTRACT

Microwave sintering of metal powder compacts is simulated within a simple semi-analytical multiphysics model taking into account the effective dielectric and magnetic properties. It is demonstrated that microwave heating exhibits resonant behavior which facilitates rapid densification of powder metals in a way similar to flash sintering. Rapid, stable and efficient microwave sintering regimes can be implemented by using adaptive control over the intensity and/or frequency of the microwave radiation.

Microwave sintering is an innovative method of consolidation that offers a number of advantages, such as shorter processing time, lower sintering temperature, reduced energy consumption etc. [1–3]. Microwave sintering of ceramic materials has been widely investigated since the 1980s [1]. In the end of the 20th century it was demonstrated experimentally that microwave sintering of powder metals is also possible [4]. The experimental research was complemented by theoretical studies aimed on the description of microwave absorption in powder metals [5–7]. Recently, a description of the effective microwave dielectric [8] and magnetic [9] properties of compacted powder metals taking into account the non-uniform structure of the electromagnetic field inside the particles has been suggested. This paper reports the results of a simulation of microwave sintering of metal powder compacts accomplished within a framework of a simple model based on this description.

The power W absorbed in a body subjected to a high-frequency electromagnetic field is

$$W = \frac{\omega}{2}\int\left(\varepsilon_0\varepsilon''\mathbf{E}^2 + \mu_0\mu''\mathbf{H}^2\right)dV, \tag{1}$$

where ω is angular frequency of the field, ε_0 and μ_0 are the electric and magnetic constants, $\mathbf{E}$ and $\mathbf{H}$ are local amplitudes of the electric and magnetic fields, ε'' and μ'' are imaginary parts of the complex dielectric permittivity ε and magnetic permeability μ of the material, respectively, and the integration is performed over the volume of the body.

When the material under consideration is inhomogeneous, it can be characterized by the effective properties, ε_{eff} and μ_{eff}, which are obtained by averaging within mathematical models that reflect the geometry and packing configuration of the particles. While bulk metals reflect the microwave radiation due to very high electric conductivity, metal powders in which electric contacts between individual particles are impeded (e.g., by native oxide layers) can be partially penetrated by microwaves [5]. An important role of the native oxide layers in the microwave absorption has been confirmed experimentally [10]. The absorption of radiation in most metal powders at a standard microwave frequency of 2.45 GHz is mostly due to magnetic-type losses (μ''_{eff}) associated with the eddy currents induced in the particles by the magnetic component of the microwave field. However, at higher frequencies and at higher values of metal's electric conductivity, when the electromagnetic wavelength within individual particles becomes comparable to or smaller than the particle size, the dielectric-type losses described by ε_{eff}'' may also become significant [8].

The effective medium models accounting for the wave structure of the electromagnetic field within individual particles have been developed recently for the effective dielectric [8] and magnetic [9] properties of compacted metal powders with insulating native oxide layers on the particles. While it is also possible to extract ε_{eff} and μ_{eff} from direct simulation of electromagnetic wave propagation in an array of metal particles [7], the effective medium expressions make it easy to calculate the varying properties repeatedly as the density of the compact and the (temperature-dependent) conductivity of metal evolve during sintering. For example, during sintering nickel particles with a radius of 1 μm and a thickness of oxide layers 2.5 nm from 50% to full density, the calculated real part of the effective dielectric permittivity of the compacted metal powder, ε'_{eff}, at a frequency of 2.45 GHz varies from 1200 to 4800, whereas the imaginary part, ε''_{eff}, varies from 0.05 to 1.4. In the same process μ'_{eff} is little different from 1, and μ''_{eff} varies in the range 0.003–0.014. The dielectric and magnetic loss tangents, $\tan\delta_e = \varepsilon'_{eff}/\varepsilon''_{eff}$ and $\tan\delta_m = \mu'_{eff}/\mu''_{eff}$, remain small throughout the process, which means that with respect to microwave heating the

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compacted metal powder acts as a dielectric material with relatively low microwave losses. A similar "dielectric-like" behavior should be observed if the metal particles are not fully insulated from one another but have high-resistance contacts of any kind between them.

Simulation of microwave sintering is generally a complex task that requires solving equations of electrodynamics, thermal physics, and continuum mechanics. For particular configurations such "multiphysics" simulations can be performed by finite element methods [11]. However, general features of microwave sintering of metal powder compacts can be analyzed within a framework of a simple semi-analytical model described below. The model uses the Mie solution of the electromagnetic problem for a spherical body exposed to a plane electromagnetic wave [12]. The microwave power absorbed in a spherical powder compact is

$$W = -\pi \frac{\mathbf{E}_0^2}{{k_0}^2} \sqrt{\frac{\varepsilon_0}{\mu_0}} \sum_{n=1}^{\infty} (2n+1) \left[\operatorname{Re}(a_n + b_n) + |a_n|^2 + |b_n|^2 \right], \tag{2}$$

$$a_n = -\frac{j_n(Nr)[rj_n(r)]' - j_n(r)[Nrj_n(Nr)]'}{j_n(Nr)\left[rh_n^{(1)}(r)\right]' - h_n^{(1)}(r)[Nrj_n(Nr)]'},$$

$$b_n = -\frac{j_n(r)[Nrj_n(Nr)]' - N^2 j_n(Nr)[rj_n(r)]'}{h_n^{(1)}(r)[Nrj_n(Nr)]' - N^2 j_n(Nr)\left[rh_n^{(1)}(r)\right]'},$$

where $\mathbf{E}_0$ is the electric field amplitude in the incident plane wave, $k_0 = \omega\sqrt{\varepsilon_0\mu_0}$ is the wavenumber in vacuum, $r = k_0 R$, R is radius of the compact, $N = \sqrt{\varepsilon_{\mathrm{eff}}\mu_{\mathrm{eff}}}$ is the refraction index of the inhomogeneous material of the compact, $j_n(x) = \sqrt{(\pi/2x)}J_{n+1/2}(x)$, $h_n^{(1)}(x) = \sqrt{(\pi/2x)}[J_{n+1/2}(x) + iY_{n+1/2}(x)]$, J and Y are Bessel functions of the first and second kind, respectively.

For the simulation described here $\varepsilon_{\mathrm{eff}}$ and μ_{eff} are calculated within the effective medium models [8,9], and they depend on the relative density of the powder compact ρ and temperature T. Spatial variation of all variables in the model (ρ, T, $\varepsilon_{\mathrm{eff}}$, μ_{eff}) within the compact is neglected.

The energy balance is determined by the microwave absorption and heat removal from the compact surface via thermal radiation (all other heat loss mechanisms are neglected):

$$\frac{4}{3}\pi R^3 c_m \rho_m \rho \frac{dT}{dt} = W - 4\pi R^2 \varepsilon \sigma^{\mathrm{gray}}{}_{SB}\left(T^4 - T_0^4\right), \tag{3}$$

where c_m is specific heat capacity of metal, ρ_m is absolute density of metal, $\varepsilon_{\mathrm{gray}}$ is gray-body emissivity, σ_{SB} is the Stefan-Boltzmann constant, and T_0 is the ambient temperature.

The change in the relative density of the compact, ρ, is calculated by the master sintering curve method [13]. Master sintering curves are the experimentally obtained dependencies Φ of the relative density on the cumulative value of the temperature evolution function Θ:

$$\rho = \Phi\{\Theta[t, T(t)]\}, \tag{4}$$

$$\Theta[t, T(t)] \equiv \int_0^t \frac{1}{T} \exp\left(-\frac{E_a}{RT}\right) dt',$$

where the value of the activation energy, E_a, is chosen to obtain the best fit to the experimental data.

The radius of the sample changes due to dilatation in inverse proportion to the cubic root of the relative density:

$$R = R_0 \sqrt[3]{\rho_0/\rho}, \tag{5}$$

where R_0 is the initial radius, and ρ_0 is the initial relative density.

The described model (2)–(5) has been applied to numerical simulation of microwave sintering of nickel powder compacts. The master sintering curve for the nickel powder was approximated by a polynomial based on the data from [13].

If the properties of the compact were not changing due to sintering, then under microwave heating its temperature would grow until the absorbed power equals the power lost by thermal radiation. At that point the temperature growth would saturate and a steady state would be reached. However, as the density of the compact increases due to sintering, its size and properties change and this modifies the conditions of microwave absorption. Therefore, the temperature may exhibit variations throughout the sintering process.

Fig. 1 shows an example of a simulated microwave sintering process with a spherical nickel powder compact. The microwave intensity in this particular case was picked so that the maximum temperature would almost reach the melting point but would not exceed it. It can be seen that the temperature behavior is not monotonic. The compact first heats to a temperature of only about 650 °C at which the sintering proceeds extremely slowly. However, after a certain time period, the temperature exhibits several large oscillations, almost reaching the melting point. The densification advances significantly during each of these oscillations. Finally, the temperature returns to the previous level and the densification stalls.

The explanation of such an unstable character of microwave heating at 2.45 GHz lies in the resonant nature of microwave absorption. The absorbed microwave power (2) exhibits resonant peaks when the ratio of the diameter of the compact to half the wavelength in the material approaches an integer number. The wavelength, λ, is determined by the effective dielectric and magnetic properties of the powder material: $\lambda = \operatorname{Re}[2\pi/(k_0\sqrt{\varepsilon_{\mathrm{eff}}\mu_{\mathrm{eff}}})]$. The diameter of the compact, $2R$, decreases due to densification, and at the same time the value of the wavelength in the powder material, λ, changes due to the variation of $\varepsilon_{\mathrm{eff}}$ and μ_{eff} with temperature and density. During sintering the wavelength in the powder compact considered in the above example changes gradually from 3.53 to 1.77 mm, while the radius decreases from 1 to 0.79 cm. An analysis shows that the first temperature peak seen in Fig. 1 actually consists of two peaks separated by an 8 s time interval. The diameter to half-wavelength ratio, $4R/\lambda$, for these peaks is approximately equal to 12 and 13. The second temperature peak corresponds to $4R/\lambda \approx 14$, and the third peak to $4R/\lambda \approx 15$.

Unstable microwave heating of metal powder compacts is frequently observed in experiments, with the sudden increases in the temperature leading to melting and destruction of the samples. For example, it has been demonstrated [14] that this phenomenon may result from the fact that the densifying metal compact is gradually "tuning" the microwave cavity and at some point brings it to resonance. These temperature jumps are of a different nature than the thermal

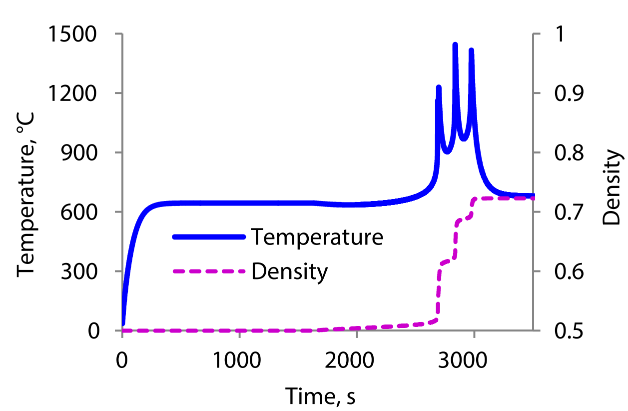

Fig. 1. Simulated temperature and relative density vs. time during microwave heating of a nickel powder compact. Frequency 2.45 GHz, initial radius of compact 1 cm, radius of nickel particles 1 μm, thickness of the oxide layer on particles 2.5 nm. The intensity of the incident microwave radiation 510 W/cm^2.