PECULIARITIES OF Fe POWDER CONSOLIDATION IN CONDITIONS OF SPARK-PLASMA SINTERING

ОСОБЕННОСТИ КОНСОЛИДАЦИИ ПОРОШКА ЖЕЛЕЗА В УСЛОВИЯХ ИСКРО-ПЛАЗМЕННОГО СПЕКАНИЯ

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Abstract: Theoretical and experimental data on the impact of heating rate during spark-plasma sintering on densification kinetics, grain size and capillary pressure in powder compacts, based on Fe, are given. It is found out, that an increase of heating rate in range from 10 °C/s to 20 °C/s leads to acceleration of process of obtainment of non-porous compacts and decrease of structure grain size.
KEYWORDS: HEATING RATE, SPARK-PLASMA SINTERING, DENSIFICATION, POROSITY, GRAIN SIZE

1. Introduction

It is known that physical-mechanical properties of powder compacts are largely dependent on their porosity and grain structure [1, 2], and method of spark-plasma sintering (SPS) provides accelerated consolidation of powder materials, which influences the process of compacts densification [3, 4]. Fe-based composites are the most common among metal-matrix composite materials, used in industry [5]. Thus, studies of impact of heating rate on densification kinetics and structure of pure Fe compacts are of high scientific urgency.

The goal of present work is to study the impact of heating rate during SPS on densification kinetics and structure of Fe compacts.

2. Preconditions and means for resolving the problem

Studies were performed on PZhR-3 (GOST 9849-86) (ПЖР-3 (ГОСТ 9849–86)) Fe powder with mean particle size of 60 μm.

Consolidation of Fe powder was performed by SPS method on "GEFEST" ("ГЕФЕСТ") experimental complex [6], which allows powders consolidation at mechanical loading in vacuum by passage of superposition of direct and pulsing currents of 10 kHz frequency and total amplitude of 1.1 kA.

Consolidation mode, considered in [7], was taken as base: heating rate was 10 °C/s, isothermal holding temperature was 1100 °C, isothermal holding time was τ = 180 s, cooling rate was 10 °C/s. Change of heating rate was due to increase of current amplitude rise rate from 14 A/s to 30 A/s.

Porosity of consolidated specimens was studied according to GOST 9391-80 (ГОСТ 9391-80). Theoretic evaluation of grain size change was performed using MS Excel software.

Experimental studies of compacts grain structure were performed using methods of computer metallography according to DSTU ISO 643:2009 (ДСТУ ISO 643:2009) and [8] using "Biolam-I" ("Біолам-І") optic microscope and ImageJ software.

3. Results and discussion

In order to define the possibility of impacting structure formation process in Fe compact during heating, an SPS process model, based on views of continuum sintering theory [3], which describes macroscopic behavior of porous body during consolidation and connects external pressure and deformation rate tensor components, was considered. On its base, authors [3, 9] created a system of differential equations, which describes changes of pores shape, porosity and grain size depending on heating temperature at constant pressure. Yet, in order to use equations, given in [3] for case of Fe powder SPS, coefficients A_0, m, which characterize system viscosity and presence of liquid phase, must be known, as well as Fe diffusion characteristics at SPS conditions, which, according to papers [10 – 16] lie in wide range. Therefore, it was decided to find values of A_0, m coefficients for powder consolidation in presence of liquid phase [4] experimentally in order to be able to prognose Fe powder behavior during SPS, and evaluations from paper [3] were written in such form:

$$\begin{cases} G' = \begin{cases} 6{,}67 \times 10^{-4}\omega \cdot \ln\dfrac{235}{\omega} \cdot G_0 \cdot \left(\dfrac{G_0}{G}\right)^3 \cdot \theta^{-1{,}14}, \; if\; T > 733\,K; \\ 0, if\; T \le 733\,K \end{cases} \\ \theta' = (1-\theta) \cdot \left\{ \left(\dfrac{3\cdot\theta}{2}\right)^{\frac{m+1}{2}} \cdot \dfrac{\frac{3\cdot\alpha}{2\cdot G}\cdot(1-\theta) - \sigma x}{A_0 \cdot exp\left(\frac{Q_{cr}}{R\cdot T}\right)(1-\theta)^{\frac{m+1}{2}}} \right\}^{\frac{1}{m}} \end{cases} \quad (1)$$

where G' – change of grain size, G_0 – initial grain size, ω – heating rate, θ' – change of porosity, α – surface tension, σ_x – external stresses, A_0, m – creep law constants, Q_{cr} – dislocation crawl activation energy, R – universal gas constant, T – temperature.

In order to check the possibility of these dependences usage as well as to find A_0, m coefficients, series of experiments, aimed at determination of porosity θ and grain size G in consolidated specimens of initial clean Fe powder, were performed.

Variation of heating rate from 10 °C/s to 20 °C/s was due to change of current amplitude rise rate in range from 14 A/s to 30 A/s. Changes of current value during Fe powder SPS are shown on Fig.1

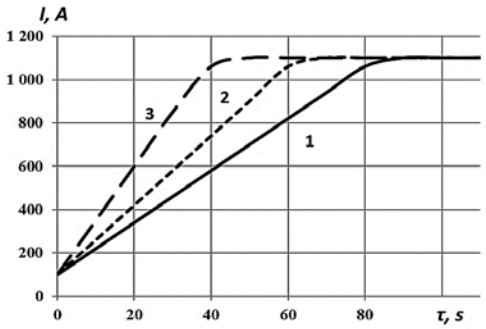


Fig. 1 *Experimental curves of changes of current amplitude during SPS with different heating rate*
1 – 10 °C/s; 2 – 15 °C/s; 3 – 20 °C/s

Curves of heating and porosity change are shown at Fig. 2 and Fig. 3 respectively. Temperature sensor was installed on matrix surface, which leads to offset of temperature curves by time axis. During the studies, it was found out that specimens, consolidated without further isothermal holding at heating rate higher, than 20 °C/s, are destroyed after they are taken out of matrix.

Analysis of obtained data shows that an increase of heating rate from 10 °C/s to 20 °C/s leads to decrease of densification time from 60 s to 40 s (see Fig. 3). Optical microphotographs confirm significant porosity decrease after 30 s of sintering with heating rate of 20 °C/s (see Fig. 4).

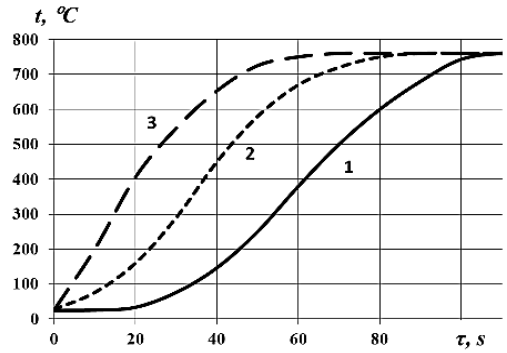


Fig. 2 *Experimental curves of changes of temperature during SPS with different heating rate*
1 – 10 °C/s; 2 – 15 °C/s; 3 – 20 °C/s

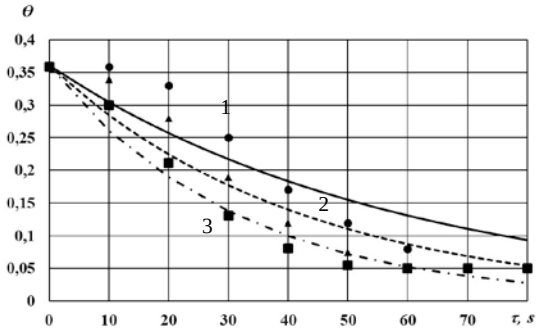
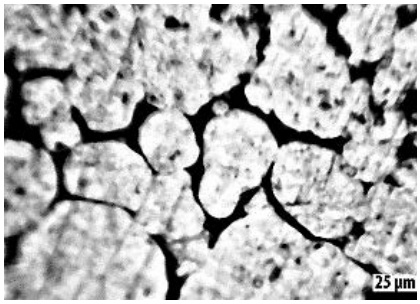
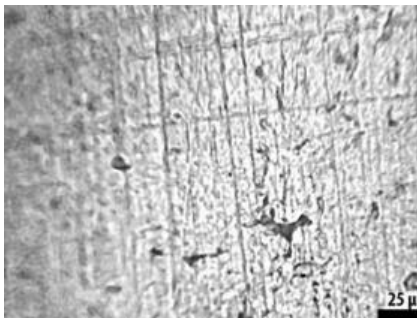


Fig. 3 *Experimental curves of changes of porosity of consolidated Fe powder specimens after SPS with different heating rate*
1 – 10 °C/s; 2 – 15 °C/s; 3 – 20 °C/s



a



b

Fig. 4 *Optical microphotographs of consolidated Fe powders, holding time 30 s, magnificantion ×250, pores are dark*
a – heating rate of 10 °C/s; b – heating rate of 20 °C/s

Obtained experimental data was approximated with 90 % accuracy by exponential curves:

$$\theta_{10} = 0{,}36 \cdot e^{-0{,}017 \cdot \tau}, \quad (2)$$

$$\theta_{15} = 0{,}36 \cdot e^{-0{,}024 \cdot \tau}, \quad (3)$$

$$\theta_{20} = 0{,}36 \cdot e^{-0{,}032 \cdot \tau}, \quad (4)$$

Analysis of grain size changes (see Fig. 5) during SPS at heating up to 1100 °C shows, that at heating rate of 10 °C/s mean grain size of specimens increases from 2.4 µm to 16 µm, and use of heating rate of 20 °C/s leads to almost two times decrease of grain growth (mean grain size is 10 µm), which is confirmed by optical microscopy (see Fig. 6) and leads to increase of specimens hardness from 60 HRB to 85 HRB.

In order to avoid negative values and characterize absence of grain growth in Fe before recrystalliza5tion begins, experimental data was approximated with 80 % accuracy by exponential curves:

$$G_{10} = 1{,}8663 \cdot e^{0{,}0293 \cdot \tau}, \quad (5)$$

$$G_{15} = 1{,}91128 \cdot e^{0{,}0379 \cdot \tau}, \quad (6)$$

$$G_{20} = 3{,}1594 \cdot e^{0{,}0453 \cdot \tau}. \quad (7)$$

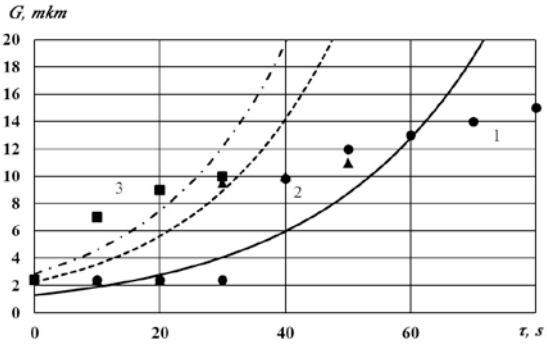
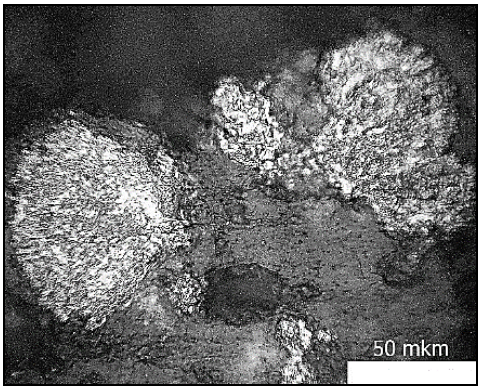
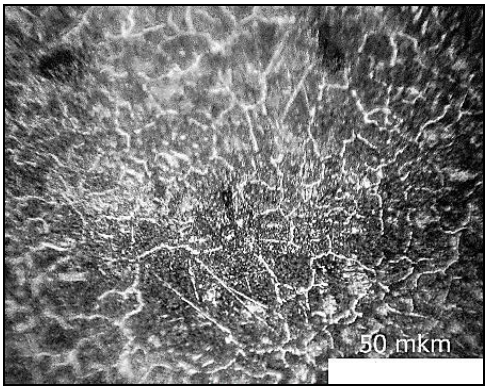


Fig. 5 *Experimental values of grain size in consolidated Fe powder specimens after SPS with different heating rate and curves of their approximation*
1 – heating rate of 10 °C/s; 2 – heating rate of 15 °C/s; 3 – heating rate of 20 °C/s

Obtained approximation evaluations allowed determination of A_0 and m coefficients at value of surface tension of α = 1.2 J/m^2 and activation energy of Q_{cr} = 200 kJ/mol for case of liquid phase presence ($m \rightarrow 1$) [4, 13,16]. Found values are A_0 = 240 MPa·s and m = 0,9. This leads to a solution of differential equations system (1), which allows description of Fe powder behavior during SPS with different heating rates with 80 % accuracy (see Fig. 7 and Fig. 8).



a



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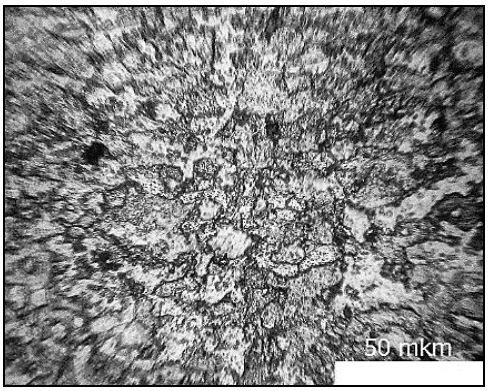

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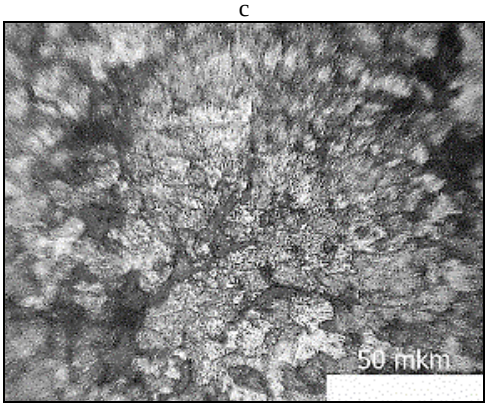

d

Fig. 4 *Microstructures of initial Fe powder and consolidated Fe powder specimens after SPS with different heating rates, magnificantion ×450*
a – initial Fe powder; b – consolidated specimen, heating rate of 10 °C/s; c – consolidated specimen, heating rate of 15 °C/s; consolidated specimen, heating rate of 20 °C/s;

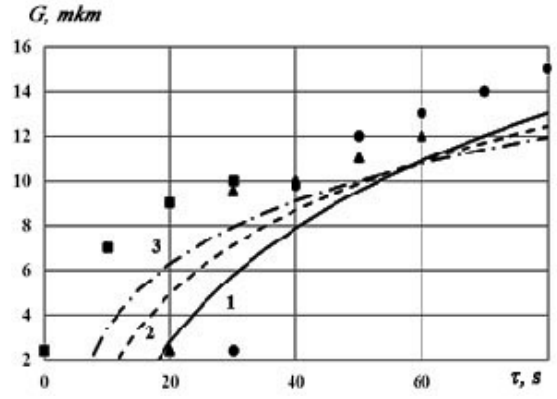

Fig. 7 *Experimental values and theoretical curves of grain size in consolidated Fe powder specimens after SPS with different heating rate*
1 – heating rate of 10 °C/s; 2 – heating rate of 15 °C/s; 3 – heating rate of 20 °C/s

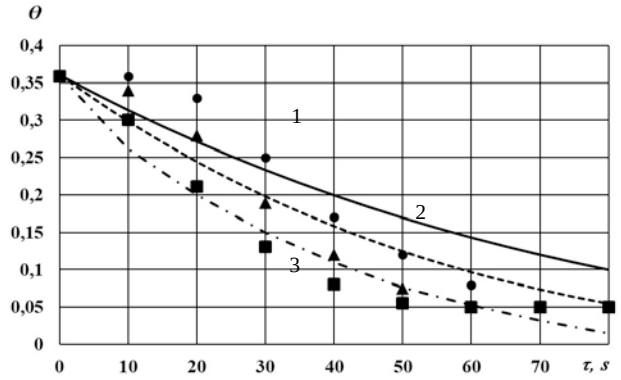

Fig. 8 *Experimental values and theoretical curves of changes of porosity of consolidated Fe powder specimens after SPS with different heating rate*
1 – 10 °C/s; 2 – 15 °C/s; 3 – 20 °C/s

Dependences of porosity changes during SPS with different heating rate, shown on Fig. 8, and A_0 coefficient allowed theoretical determination of changes of capillary pressure between Fe powder particles dependence on specimens density, based on continuum sintering theory. A model of densification of cylindric specimen in hard matrix under external pressure of 30 MPa was considered (see Fig. 9). Material was considered as having pores and linearly viscous non-porous phase.

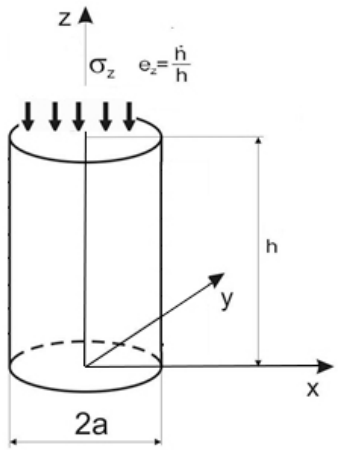

Fig. 9 *Model of densification of cylindric specimen in hard matrix under external axial pressure*

This model was described using evaluations (8–10) [3, 4, 17]:

$$\sigma_r = 2\eta\left(e_r - \frac{1}{3}e\right) + \varsigma e + P_l, \tag{8}$$

$$\sigma_z = 2\eta\left(e_z - \frac{1}{3}e\right) + \varsigma e + P_l, \tag{9}$$

$$e = \frac{1}{3}(e_z + 2e_r)\cdot \tag{10}$$

where η and ζ – coefficients of shear and volumetric viscosity respectively;

P_l – capillary pressure;

e – rate of volume change.

Behavior of consolidated material can be described by the following evaluations:

$$\eta = \eta_m (1-\theta)^2, \quad (11)$$

$$\zeta = \frac{4}{3}\eta_m \frac{(1-\theta)^3}{\theta}. \quad (12)$$

where η and ζ –shear and volumetric viscosities respectively;

$\eta_м$ – coefficient of shear viscosity, which, according to [4], can be represented as:

$$\eta_m = \frac{A_0}{2}. \quad (13)$$

Theoretical dependences of change of capillary pressure on porosity change during SPS with heating rates of 10 °C/s, 15 °C/s and 20 °C/s were obtained as a result (see Fig. 10). It is found out, that during Fe powders consolidation capillary pressure impacts their densification process only when total specimens density is higher than 80 %. Values of capillary pressure depend on heating rate and are 0.5 MPa for heating rate of 10 °C/s and 2 MPa for heating rate of 20 °C/s, which is due to formation of liquid phase and decrease of pores size, rate of excretion of which depends on heating rate.

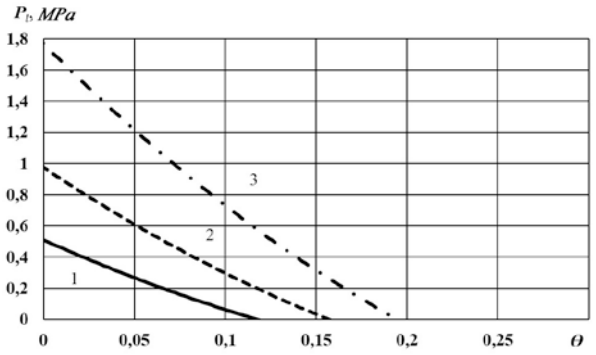

Fig. 10 *Theoretical curves of changes of capillary pressure depending on porosity of consolidated Fe powder specimens after SPS with different heating rate*
1 – 10 °C/s; 2 – 15 °C/s; 3 – 20 °C/s

4. Conclusions

Regularities of impact of heating rate during SPS in range from 10 °C/s to 20 °C/s on Fe compact densification kinetics are found.

Basing on evaluations od continuum sintering theory, it is shown and experimentally confirmed that an increase of heating rate during Fe powders SPS from 10 °C/s to 20 °C/s leads to a decrease of densification time from 60 s to 40 s, increase of capillary pressure from 0.5 MPa to 2 MPa and increase of consolidated specimens hardness from 60 HRB to 85 HRB due to obtainment of more fine-grain structure.

5. Literature

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