STRUCTURAL STUDIES OF MATERIALS

MECHANICAL RESONANCE SPECTROSCOPY OF INTERPARTICLE BOUNDARIES IN HIGH-DENSITY IRON POWDER COMPACTS

A. V. Vdovichenko,[1,2] Yu. N. Podrezov,[1] and V. V. Skorokhod[1]

Characteristics of mechanical resonance are measured on powder iron samples with a porosity of 0.06 produced by die compaction and sintering at 573 to 1373 K. Young's modulus and parameter C_N that describes how the resonant frequency varies with increasing amplitude of oscillation are calculated. It is established that C_N decreases with increasing temperature of sintering. Comparing the resonance characteristics with results from fracture toughness tests and structural analysis shows that C_N correlates with fracture toughness and failure mode of samples. The results indicate that C_N is related to the evolution of the interparticle boundaries and may be used as a highly sensitive criterion to quantitatively assess the quality of sintered powder materials.

Keywords: *powder iron, sintering, interparticle boundaries, mechanical resonance.*

INTRODUCTION

Sintering is an essential powder metallurgy process ensuring needed mechanical properties. This process may be divided into the following stages: formation and growth of interparticle boundaries; decrease and obliteration of pores; subsequent recrystallization. There are different diffusion models developed to describe microscopic processes occurring in sintering, which are reviewed, for example, in [1], and rheological models developed to describe the macroscopic behavior of a powder body in sintering [2, 3]. However, the mesostructural state of a powder body, i.e., contacts between particles in sintering, needs to be known. For example, this is important in the sintering of powder parts for which strong bonds between particles must be ensured but grain growth must be prevented. To control the structure is problematical. On the other hand, widely applied dilatometric methods are not sensitive to the state of interparticle boundaries. At the same time, macrocharacteristics determined with conventional resistometric and ultrasonic methods are known [4, 5] to be sensitive not only to imperfect contacts but also to other factors (porosity, presence of oxide films, geometry of porous space, microstructure of powder phase). Hence, the results of such measurements can hardly be used to determine the state of boundaries.

Resonance mechanical methods have recently been developed to reveal defects of different types.

The objective of this paper is to determine the resonance characteristics of powder iron samples sintered at different temperatures and to compare the results with structural analysis data to establish quantitative parameters characterizing the quality of sintered boundaries between powder particles based on physical acoustics.

[1]Institute for Problems of Materials Science, National Academy of Sciences of Ukraine, Kiev, Ukraine.
[2]To whom correspondence should be addressed; e-mail: vdovyche@ipms.kiev.ua.

EXPERIMENTAL PROCEDURE

The test samples were made from PZh3M2 iron powder produced by scale reduction. Powder particles were equiaxial, almost spherical, and 50 to 100 μm in size. The samples were produced from the starting powder mixed with 0.5 wt.% of zinc stearate by single-action compaction in a rigid die mold at room temperature under 1020 MPa. After compaction, some part of the samples was sintered in hydrogen at 573 to 1373 K for 1 h. To prevent their damage, the samples after sintering were not subjected to additional mechanical treatment. The samples were formed into rectangular bars of $55 \times 10 \times 5$ mm.

Two resonance spectroscopy methods were used. The first was intended to measure the natural frequency of longitudinal oscillation of a free-ended bar and then determine Young's modulus of the material. The other method was to determine how the natural frequency of transverse oscillation of a cantilever bar depended on amplitude.

To examine the natural frequencies of longitudinal oscillation, the sample with a rubber sheet being fixed at the mid-section of its long-side faces was placed between two wideband piezoelectric transducers so that small gaps remained between the sample ends and transducers. To ensure acoustic contact, a small amount of mineral oil was introduced into the gaps. A variable-frequency electrical signal from the generator caused mechanical oscillation in one of the transducers that was transmitted to the sample through an immersion liquid. The mechanical response of the sample to the oscillation was transmitted to the other transducer and then the electrical signal went through amplifier to recording instruments. Since the node of oscillation is located in the mid-section in longitudinal oscillation in the first mode (and all odd modes), i.e., this section has zero displacement, the type of fixation in this case minimally affects the resonance frequencies of such oscillation. The resonance characteristics were used to calculate Young's modulus [6]:

$$E = \rho \cdot (2 \cdot l \cdot f_b)^2, \tag{1}$$

where ρ is the experimental density of porous material; l is the bar length; f_b is the frequency of the first mode of natural longitudinal oscillation. The maximum relative cyclic strains of the sample for longitudinal oscillation were of the order of 10^{-6}. The resonance frequencies were of the order of 10^4 Hz.

The change in the natural frequency of the first mode of cantilever bending oscillation with increasing amplitude was the second resonance characteristic to be determined. The measurement was performed over a kilohertz frequency range and the maximum relative strain was 10^{-5} to 10^{-3}. The method was employed as follows. One end of the sample was secured in a fixture placed on the movable table of a VÉDS-400 electrodynamic shaker. The vibration of the shaker table excited oscillations in the sample. The oscillation frequency was measured using a sinusoidal generator within the sample resonance range; the amplitude of the sample free end was determined at different excitation frequencies using a measuring microscope, and a resonance curve was plotted. Then the signal strength was increased and measurement was repeated. Preliminary studies showed that the resonance frequency
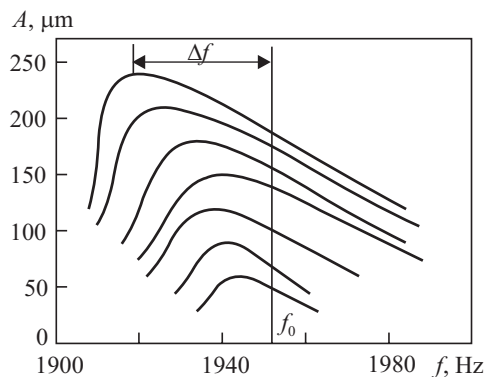

Fig. 1. Resonance curves of the first bending oscillation mode for the 0.06-porosity powder iron sample sintered at 1373 K at seven different excitation levels: A is amplitude of the free end of the cantilever bar; f_0 is resonance frequency at minimal strain; Δf is resonance frequency shift

decreases with increasing amplitude of oscillation; moreover, it changes to much greater extent for powder metals than for compact ones. As an example, Figure 1 shows resonance curves for a sample sintered at 1373 K at different excitation levels. The resonance curves were used to determine how the resonance oscillation frequency depended on the amplitude of the maximum cyclic strain ε calculated with the following formula of elasticity theory:

$$\varepsilon = \pi A f \sqrt{\frac{3\rho}{E}} \,, \qquad (2)$$

where A is the amplitude of the free end; f is the frequency of the first mode of resonance transverse oscillation of the bar; ρ is the sample density; E is Young's modulus determined from Eq. (1) using the frequency of longitudinal oscillation.

The density ρ of the samples was determined with hydrostatic weighing. The porosity was assessed as $\Theta = 1 - \rho/\rho_0$, where ρ_0 is the theoretical density of iron equal to 7860 kg/m^3.

The samples were tested for fracture toughness K_{Ic} in plane deformation by three-point bending of a notched sample as described in [7] at 77 K and a vacuum of $2 \cdot 10^{-7}$ Pa. The microstructure and fracture surface were examined using optical and scanning electron microscopes.

RESULTS AND DISCUSSION

The powder particles have spongy surface (Fig. 2). After compaction under 1020 MPa, the sample contained both bulk pores and a developed network of planar pores (shown with a white arrow in Fig. 2b). These planar defects remain unchanged after low-temperature sintering and disappear after sintering at 773 K (Fig. 2c), while bulk pores remain in place. A further increase in sintering temperature to 1373 K does not change the state of interparticle boundaries.

The mean porosity of a green compact was 0.08 and of all sintered samples 0.06, which remained unchanged even after recrystallization at high temperature. The practically unchanged porosity of the samples indicates that sintering over the temperature range in question was promoted by surface diffusion. There was practically no shrinkage, which usually accompanied bulk mass transfer, and consolidation resulted from improvement of interparticle contacts.

The fracture pattern changes with increasing sintering temperature (Fig. 3). After sintering to 773 K inclusive, the samples are fractured at interparticle contacts (Fig. 3a, b). Brittle transcrystalline fracture prevails at high-temperature sintering (Fig. 3c, d).

It is shown that the resonance frequency of transverse oscillations of all samples decreased with increasing amplitude; this dependence remained linear. However, the inclination of lines changed with sintering temperature. Figure 4 represents plots showing relative changes in resonance frequency depending on the amplitude of the maximum
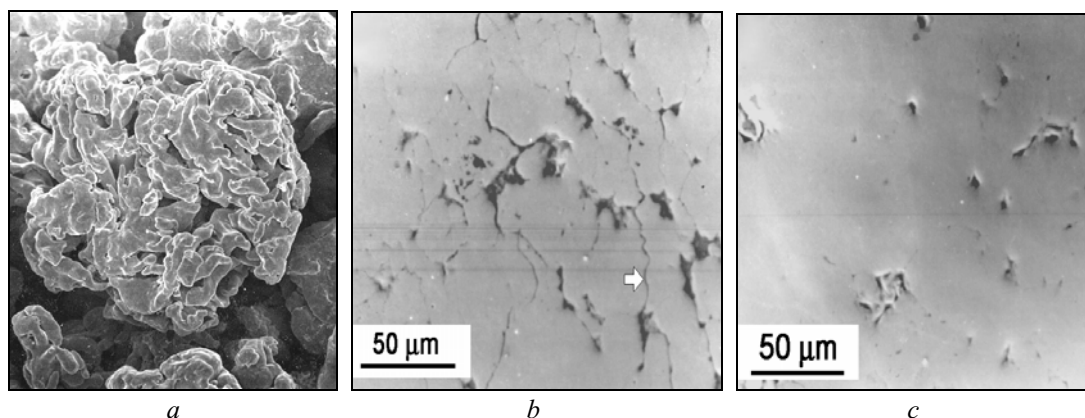

a   *b*   *c*

Fig. 2. Microphotographs of starting powder particle (*a*) and sections of powder iron after compaction (*b*) and after sintering at 773 K (*c*)
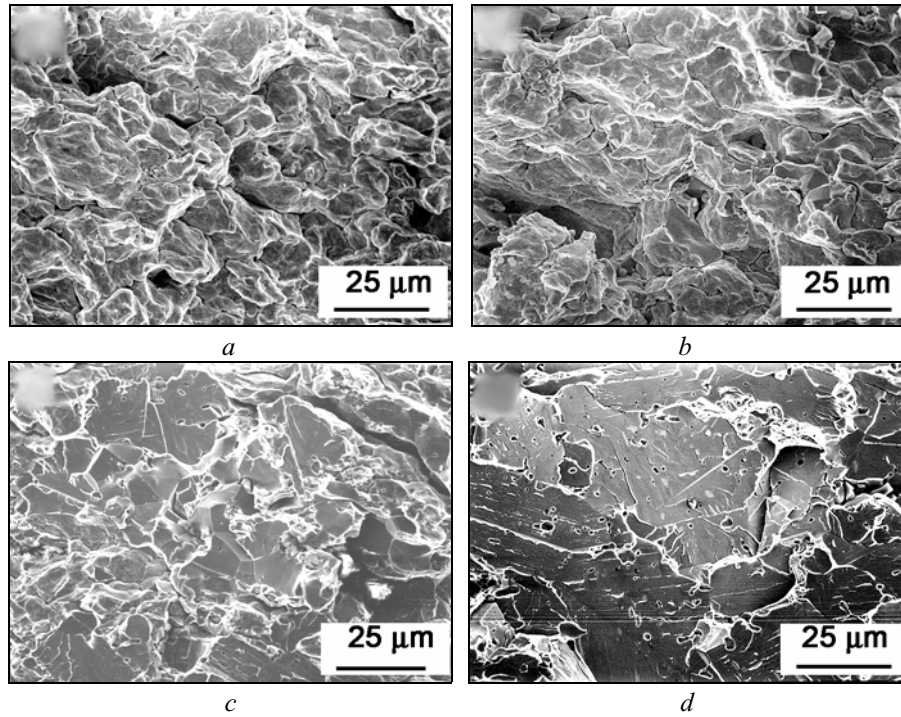
Fig. 3. Fracture surfaces of powder iron samples compacted (*a*) and sintered at 973 (*b*), 1173 (*c*), and 1373 K (*d*)

cyclic strain determined with Eq. (2). The relative shift of resonance frequency was determined as follows:

$$\frac{\Delta f}{f_0} = \frac{f_0 - f}{f_0},$$ (3)

where f_0 is the resonance frequency at minimal strain and f is the current resonance frequency measured at a given level of excitation signal. The f_0 values were determined by extrapolating the lines fitting the experimental amplitude dependences of the resonant frequencies to zero amplitude.

For all structural states in question (Fig. 4), the natural frequencies of oscillation linearly decrease with increasing amplitude of the maximum relative cyclic strain ε and can be described by the following empirical formula:

$$\frac{\Delta f}{f_0} = C_N \varepsilon,$$ (4)

where C_N is the line inclination coefficient.

Note that the amplitude dependence of the resonance oscillation frequency for powder iron samples differs from similar dependences for materials produced with conventional methods. The most substantial difference is greater change in the resonance frequency of powder materials than of compact ones. The above-described experiments show that the resonance frequency of a steel 20 sample decreased by 0.5% ($\varepsilon = 4.7 \cdot 10^{-4}$) with increasing maximum cyclic strain from 4 to 100 MPa. The literature data show [8] that the resonance oscillation frequency of an armco-iron sample decreased by 0.2% only at maximum cyclic strain amplitude $\sigma = 200$ MPa. Moreover, this change tended to increase with greater σ and was attributed by the authors to fatigue damage accumulation. The resonance frequency of powder iron sintered at 1373 K changed by 2% at $\sigma = 100$ MPa ($\varepsilon = 5.8 \cdot 10^{-4}$) and that of powder iron annealed at 573 K exceeded 8% even at $\sigma = 46$ MPa ($\varepsilon = 3.6 \cdot 10^{-4}$). We see that the resonance frequency of powder iron changes to a grater extent than that of compact materials even after high-temperature sintering when there is no impact of plastic deformation on material structure. We observe that the
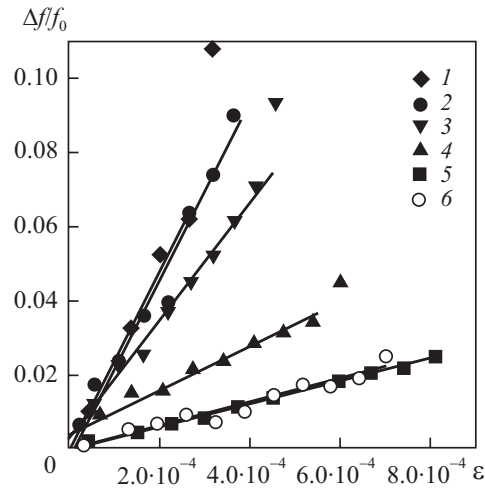
Fig. 4. Relative resonance frequency shift as a function of the maximum cyclic strain amplitude measured in oscillations in the first mode of cantilever powder iron samples sintered at different temperatures (K): *1*) without sintering, *2*) 573, *3*) 773, *4*) 973, *5*) 1173, *6*) 1373

dependence of resonance frequency on deformation deviates from linearity at large strains (Fig. 4); this may result from additional cracks originating from fatigue damage, which, as shown in [8, 9], decreases resonance frequency.

Comparing data for powder iron with results for compact materials permits a conclusion that the abrupt decrease in the natural oscillation frequency of sintered materials results from their structural features. The authors of [10] modeled the elastic longitudinal oscillation of bars from materials with plane pores; these materials are linearly deformed in tension and compression cycles but Young's modulus is different for tension and compression. The result [10] indicates that such two-dimensional defects decrease the resonance oscillation frequency. It seems that plane pores cause the substantial dependence of the natural oscillation frequency on the amplitude of maximum cyclic strain.

It should be noted that the substantial nonlinearity of oscillation makes one treat with caution the ε values calculated with Eq. (2) based on assumptions of linear elasticity theory. To establish strict dependences, the oscillation nonlinearity should be further experimentally and theoretically studied. It is essential that parameter C_N is sensitive to structural changes that occur in sintering.

In terms of structurization, sintering temperature for powder iron can be divided into three ranges: the first is to 573 K inclusive, the second above 573 to 1173 K, and the third 1173 K and above.

Sintering over the first temperature range does not produce any noticeable structural changes: interparticle plane pores (imperfect boundaries) remain, which indicates that there are no metal bonds between powder particles. This is evidenced by the fracture pattern of samples. After fracture toughness testing, fracture occurred at particle boundaries, and K_{Ic} agreed with the fracture toughness of green compacts and constituted 5 MPa · m$^{-1/2}$. The characteristics determined with resonance methods after sintering over this range also practically do not change as compared with green compacts (Fig. 5a, b). Young's modulus of compacts (128 GPa) constituted almost 70% of the value for iron with a porosity of 0.06 without plane defects and with spherical pores (this value was calculated using generalized self-consistent scheme and constituted 187 GPa [4]). Parameter C_N was approximately equal to 240 after holding of samples at 573 K.

The second temperature range is most interesting in terms of structurization since sintering process that causes interparticle surfaces to disappear starts at these temperatures. On the one hand, interparticle plane pores present in compacts and samples heated at 573 K are not observed in samples sintered at 773 K (Fig. 2). On the other hand, the samples sintered at this temperature fractured mainly at interparticle boundaries and fracture toughness increased only insignificantly. Hence, it is concluded in [11] that fracture pattern is influenced by the segregation of admixtures at the interparticle boundary. The results of resonance analysis show that Young's
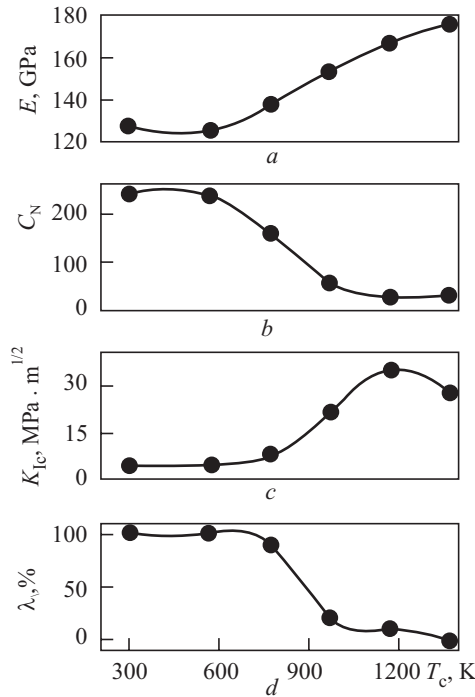
Fig. 5. Mechanical resonance characteristics and fracture characteristics of powder iron depending on sintering temperature: *a*) Young's modulus E; *b*) coefficient C_N; *c*) fracture toughness K_{Ic}; *d*) fraction of interparticle fracture in the total area of fracture surface λ_i

modulus increases after sintering at 773 K and continues increasing with sintering temperature (Fig. 5a). At the same time, parameter C_N noticeably decreases after sintering already at 773 K and continues decreasing with increasing temperature to 973 K. Abrupt transfer from interparticle to prevailing transcrystalline fracture (Fig. 3) and associated increase in fracture toughness occur over this temperature range (773–973 K). The substantial nonlinearity of oscillations of the samples sintered at 773 K may indicate that sintering does not obliterate plane pores but only decreases their area in spite of the absence of plane defects in photographs of metallographic sections. This may be due to the absence of bonds between areas of particles with a large amount of admixtures, though there are bonds at boundaries at the level of iron lattice. In terms of mechanics, such areas are plane pores that weaken the sections of samples and lead to interparticle fracture. According to resonance tests, we conclude that perfect boundaries are completely formed only at 973 K. This is confirmed by fractographic tests: samples sintered at this temperature fractured mainly by transcrystalline mechanism.

After sintering over the third temperature range (1173 K and above), perfect boundaries are finally formed between particles. The samples are recrystallized, and grain boundaries leave the area of particle interfaces, and increase in sintering temperature leads to grain growth. Young's modulus over this temperature range continues increasing and reaches 176 GPa, and C_N somewhat decreases (to 30) with increasing temperature to 1173 K and then remains unchanged. Therefore, the collective crystallization that decreases K_{Ic} does not affect C_N, which confirms the assumption that this parameter depends on the state of interparticle boundaries.

Analysis of the data shows that the resonance oscillation frequency resulting from changes in elastic characteristics and resonance frequency shift characterizing oscillation nonlinearity are sensitive to mesostructural changes associated with the state of interparticle boundaries. The formation of metal bonds increases Young's modulus and decreases C_N. However, note that the second parameter is more sensitive to the formation of mesostructures since Young's modulus increased by 38% in complete sintering of compacts while the nonlinearity parameter decreased by one order of magnitude. In addition, elastic characteristics of powder materials are known to depend on the porosity and microstructure of the powder phase and on geometric characteristics of the powder

371

phase and porous space. Therefore, changes in Young's modulus indicate not only perfect interparticle boundaries but also processes accompanying sintering such as compaction, pore spheroidizing, diffusion of admixtures from interparticle boundaries, change in grain sizes and dislocation structure, etc. At the same time, we assume that resonance frequency shift, which indicates nonlinear response to mechanical effect, results from the presence of mesostructural defects, i.e., plane imperfect boundaries. These defects also make powder bodies tend to interparticle fracture, when mechanical properties of the powder phase are not completely used. The experimental data show that the parameters of nonlinear response of materials with defects to mechanical effect should be further examined to establish new performance criteria, including those for nondestructive examination and monitoring of the consolidation of powder materials.

CONCLUSIONS

It is established that imperfect boundaries in powder iron cause the substantial dependence of the resonance oscillation frequency on cyclic strain amplitude.

The resonance frequency linearly depends on amplitude for all states: from green compacts to samples sintered at 1373 K. The inclination of the C_N (ε) line depends on sintering temperature. Changes in this characteristic correlate with fracture toughness K_{Ic} and fraction of interparticle fracture λ_i. Therefore, it is assumed that C_N is sensitive to the state of interparticle boundaries and can be a promising characteristic used in nondestructive examination of sintered materials.

REFERENCES

1. F. B. Swinkels and M. F. Ashby, "A second report on sintering diagrams," *Acta Metall.*, **29**, 259–281 (1981).
2. V. V. Skorokhod, *Rheological Basis of Sintering Theory* [in Russian], Naukova Dumka, Kiev (1972), p. 151.
3. E. A. Olevsky, "Theory of sintering: from discrete to continuum," *Mater. Sci. Eng. R.*, **23**, 41–100 (1998).
4. A. V. Vdovichenko and Yu. N. Podrezov, "Evolution of dynamic Young's modulus and damping ability of porous iron," *Metallofiz. Noveish. Tekhnol.*, 27, No. 11, 1429–1440 (2005).
5. O. V. Roman, V. V. Skorokhod, and G. R. Fridman, *Ultrasonic and Resistometric Inspection in Powder Metallurgy* [in Russian], Vysshaya Shkola, Minsk (1989), p. 182.
6. L. D. Landau and E. M. Lifshits, *Theoretical Physics. Vol. VII: Elasticity Theory* [in Russian], Nauka, Moscow (1987), p. 248.
7. A. S. Drachinskii, A. E. Kushevskii, and Yu. N. Podrezov, "Effect of the scale factor on the results of determinations of the mechanical properties of iron-base P/M materials," *Powder Metall. Met. Ceram.*, **22**, No. 3, 233–237 (1983).
8. V. A. Kuz'menko (ed.), *Fatigue Tests at High Loading Frequencies* [in Russian], Naukova Dumka, Kiev (1979), p. 336.
9. A. V. Vdovichenko, Yu. F. Lugovskii, and V. A. Nazarenko, "Fatigue failure of rolled high-porosity materials made of copper fiber at resonance bending oscillations," *Powder Metall. Met. Ceram.*, **30**, No. 1, 87–89 (1991).
10. O. V. Vdovichenko, V. V. Skorokhod, and M. B. Shtern, "Modeling elastic oscillations of rods from powder materials in the presence of 2D defects," in: *Mathematical Models and Computational Experiment in Materials Science*, Issue 9, Inst. Probl. Materialoved. NANU, Kiev (2007), pp. 3–7.
11. S. A. Firstov and M. Shlesar (ed.), *Structure and Strength of Powder Materials* [in Russian], Naukova Dumka, Kiev (1993), p. 174.