BEHAVIOR OF IRON POWDER UNDER CRYOGENIC CONDITIONS

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Abstract

A sample of commercial atomized DP 200 iron powder was subjected to a cryogenic investigation by low temperature X - ray diffraction. It was established that the lattice parameter has a linear decrease as temperature decreases meanwhile the powder density features linear increasing. The thermal linear expansion coefficient varies for different temperature ranges, having a slow decrease at lower temperatures. Based on the observed variation we found an average thermal linear expansion coefficient of $13.77 \cdot 10^{-6}$ K^{-1} for all the investigated temperature range of 123 -298 K. The resulting absolute value for lattice parameter is 285.14 pm and the absolute density result is 7.99 g/cm^3.

Keywords: iron powder, low temperature, cryogenic behavior

INTRODUCTION

The cryogenic application in metallurgy proves to be an interesting research field of a wide range of applications such as magnetic materials development and even the food industry [1-3]. Also, many alloys were tested in cryogenic conditions such austenitic and ledeburitic stainless steels due to their applications as cutting tools [4, 5]. Some of current research uses the cryogenic treatment for the improvement of wear resistance of cutting tools by immersion in liquid nitrogen [6]. There followed the austenitizing temperature, cooling rate and holding time in order to improve wear resistance of the cutting tools.

The cryogenic treatment fits bulk alloy applications in various scientific directions. It is interesting to fit cryogenic treatments to metallic powder. Some of recent research uses low temperature combined with ball milling in order to achieve nano-powders instead of nanocrystalline powders [7 - 9]. Some reports mention that cryo-milling achieves particle diameter around 8 nm instead of the usual high energy ball milling which achieves only a 100 nm average particle diameter [10]. The cryogenic temperature induces several variations at crystal lattice level which affects the powder state. The frozen metal powders are more susceptible to a brittle milling, meanwhile cold welding is avoided. Considering all mentioned aspects, the aim of present article is to figure out the cryogenic behavior of iron elemental powder at several low temperatures.

EXPERIMENTAL PROCEDURE

Samples of DP 200 iron (0.02% C) elemental powder were investigated initially by optical microscopy and sieving analysis to establish the grain size distribution. Samples were investigated by X-ray diffraction at room temperature and at several cryogenic temperatures: 223, 173, and 123 K.

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The X-ray diffraction investigation was performed on a DRON 3 X-ray diffractometer equipped with data acquisition module and Matmec VI.0 soft. All X-ray diffraction patterns were obtained with $Cu_{k\alpha}$ radiation.

The cryogenic investigation was performed with a special device, the UNRT 180 coupled on the DRON 3 diffractometer with cooling in liquid nitrogen. The DP 200 powder samples, having 2 grams each, were deposed on the X-ray sample holder. The UNRT 180 device was coupled to the special electronic temperature control. This device controls the liquid nitrogen flow directed on the X-ray sample in order to maintain the temperature at the set value, assuring a constant value of the temperature during the investigation.

Each sample was cooled for 15 minutes in order to achieve a constant value of the temperature (223, 173, and 123 K). After that, the X-ray cryo investigation was started with a speed of 1 degree 2θ/min resulting in an exposure time of 40 minutes for each sample. The total exposure time of each sample at cryo temperatures is 55 minutes.

Samples treated at 173 and 123 K were also investigated by optical microscopy. Optical microscopy was performed on a Carl Zeiss Jena transmitted light microscope using a Samsung 8 Mpx. digital capture.

The sieving analysis was performed on a vibrator table using the following sieve mesh: 200, 160, 125, 100, 80, and 64 μm.

RESULTS AND DISCUSSION

DP 200 is one of the most commonly used iron elemental powders for wide powder metallurgy (PM) applications. It is produced by atomization by Ductil Powder Buzau – Romania. The resulting microphotograph for this powder is presented in Fig.1a. We observe the typical shape for atomized powder having irregular particles formed by several iron spheres welded together before solidification. The observed morphology is very important for cryogenic application because of fragility of the necks at low temperature. Most evidenced particles feature an average diameter under 200 μm and only few particles feature a prolonged shape over 200 μm.

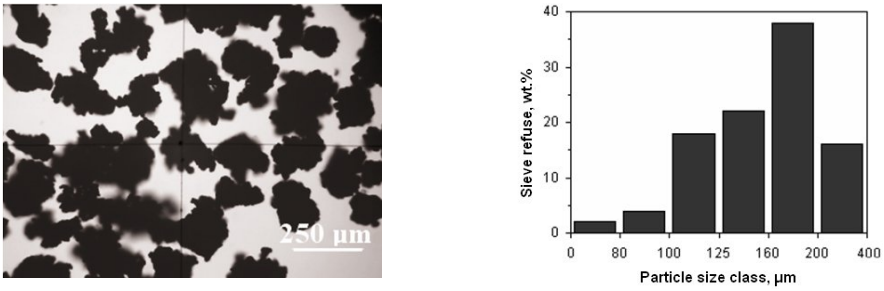


Fig.1. Initial DP 200 iron powder investigation: a) transmitted light microphotograph, b) particle diameter distribution histogram.

The grain size distribution analysis agrees to optical microscopy investigation, Figure 1b. Particles over 200 μm grain size are only 18 wt. % of all powder the other fraction being situated under this value. We notice a maximum of distribution at 160 μm, sieve refuse representing an amount of 38 wt. %, a situation in full agreement with microscopic inspection. As well, we noticed significant power amount for 100 and 125 μm fractions situated around 20 wt. %. Finally, results that DP 200 iron powder is not uniform, presenting a simple Gaussian distribution. The average grain size is around 160 μm.

The presence of several different fractions in the total amount of powder is important for further investigation because of their wide range. A wide range of cryogenically tested particles provides average resulted values, avoiding possible influences related to the grain size.

The resulted X-ray patterns for DP 200 powder investigated, at several low temperatures, are presented in Fig.2. At room temperature (298 K) Figure 2a we observe broad diffraction peaks for iron (110), (200), and (211), which could be compared to the bulk bronze peaks resulted from the sample holder. The iron powder peaks` broadening are related directly to the grain structure inside of the powder particles related to their diameter.

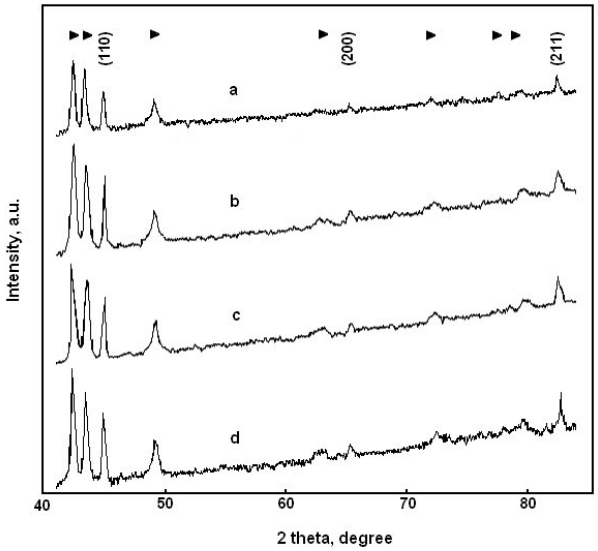


Fig.2. The X-ray diffraction patterns for DP 200 iron elemental powder subjected to cryogenic treatment: a) 298 K, b) 223 K, c) 173 K, and d) 123 K. There are pointed in brackets Miller indices for Fe peaks, the other ones (►) are bronze peaks from specimen holder.

The X-ray pattern at 223 K is presented in Fig.2b. The observed iron peaks (110), (200), and (211) are situated at slightly greater 2 theta angles than in the initial state and their shape become slighter and narrower. Also we observe a diminishing of the baseline spreading. Both observations are related to the diminishing of the decrease of thermal noise at crystal lattice level. The observed tendency is more obvious for X-ray patterns obtained at 173 and 123 K.

As observed roughly from the X-ray diffraction patterns evolution with decreasing of temperature the cryogenic cooling affects the crystal lattice and properties of powder articles. For a more accurate observation we calculate lattice parameter, thermal linear expansion coefficient (TEC) and powder density. The evolutions of the mentioned parameters with temperature are presented in Fig.3.

Lattice parameter was calculated by the least square method considering an α Fe (CVC) structure and an extrapolation equation of type:

$$a_i = a_0 + bf \; ; \qquad (1)$$

where: a_0 – extrapolated lattice parameter at $2\theta = 180^o$, a_i – resulted lattice parameter for each diffraction peak, f – extrapolation function [11]:

$$f = (\cos^2\theta)/\sin\theta + (\cos^2\theta)/\theta; \quad (2)$$

where: θ - is half of diffraction angle 2 theta.

We found that lattice parameter a_0 depends on temperature. Temperature decreasing to cryogenic level induces a severe decreasing of lattice parameter due to the thermal contraction.

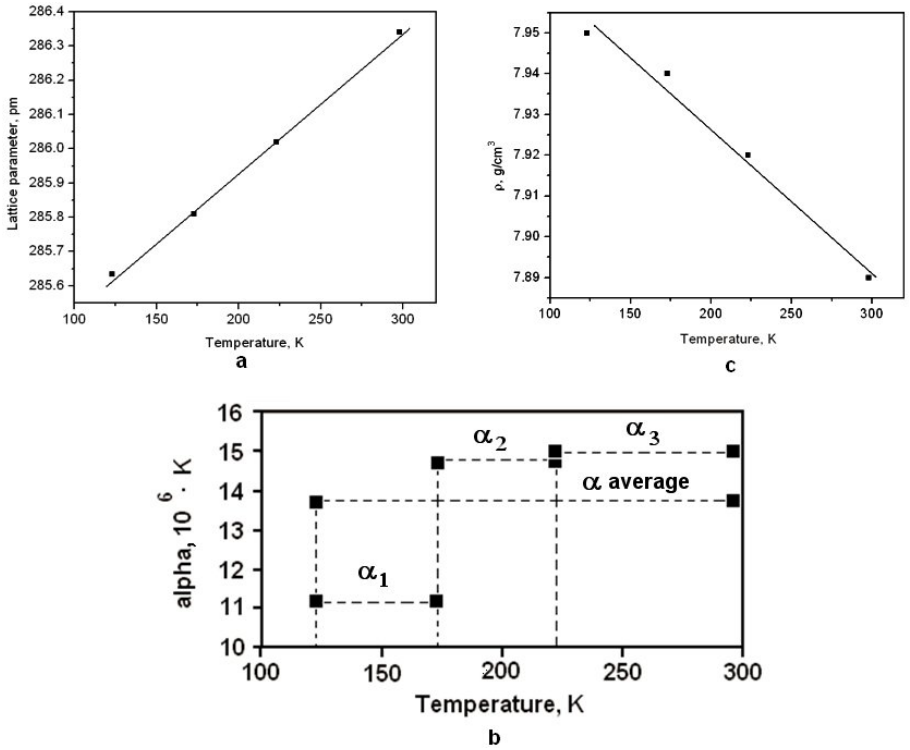


Fig.3. Iron powder properties versus temperature: a) lattice parameter, b) thermal expansion coefficient, and c) density.

On the variation in Fig.3a we observe a linear decreasing of lattice parameter with temperature described by straight line fitted to experimental points. Considering the linear variation, the lattice parameter variation is described by following equation:

$$a_0 = 285.14 + 0.004T \quad (3)$$

Applying the condition to the limit, respectively absolute temperature of 0 K results in a constant value of lattice parameter at 285.14 pm. This value represents the absolute cryogenic reference for the crystal lattice of iron powder.

The thermal linear expansion coefficient could be calculated from resulted lattice parameters values at considered temperatures according to the relation [12]:

$$\alpha_m = a_f - a_i / a_0 (T_f - T_i); \quad (4)$$

where: a_f and a_i – lattice parameter value at T_f and T_i temperatures, and a_0 – reference lattice parameter – the value obtained for 298 K.

In Figure 3b is plotted the diagram of TEC versus temperature. We observe that TEC is strongly influenced by the temperature range generally decreasing with temperature. The lower value of TEC, α_1, was found for the 123 – 173 K temperature range. The other TEC values are α_2 for 173 – 223 K and α_3 for 223 – 298 K. We calculate the average TEC for DP 200 iron powder, the resulting value is $13.77 \cdot 10^{-6}$ K^{-1}. Obtained TEC average value is sensibly greater than value reported for bulk iron [13], a fact sustained by powder morphology related to the possibly remaining traces of elements from powder elaboration such 0.02% C. However, this value is proper for rough cryogenic application, for more precise applications the diagram in Figure 3b is welcomed.

Density is another important technological property for bulk materials and for metallic powders. The effective density of iron powder could be calculated from X-ray diffraction data considering the CVC model having two atoms / unit cell. For the first being density, ρ, the relation is described:

$$\rho = m/V = m/a_0^3, \quad (5)$$

where: V – unit cell volume, m – weight of atom in unit cell. The weight, m, could be written as follows:

$$m = N \cdot A_{Fe}/N_A, \quad (6)$$

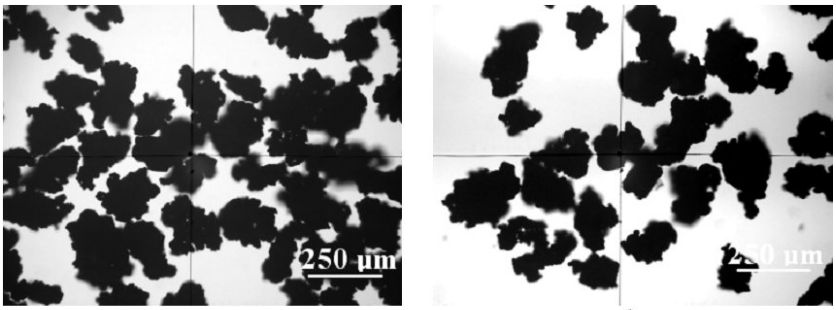
where: A_{Fe} – iron atomic weight, N – number of atoms per unit cell, and N_A - Avogadro number.

We calculate the DP 200 iron powder effective density at each temperature resulting in the plot, presented in Fig.3c. At room temperature there resulted a density of 7.89 g/cm^3 comparative to the values reported [14]. In Figure 3c we observe a pronounced increasing of powder effective density due to the severe contraction of the crystal lattice observed by the shrinkage of lattice parameter. This increasing presents a linear trend described by the relation:

$$y = 0.0004x + 7.9968 \quad (7)$$

Considering the condition to the limit, respectively the absolute temperature of 0 K, results in the absolute density of iron powder of 7.99 g/cm^3. This value could be considered for more precise cryogenic applications involving iron powder.

The cryogenic investigation was performed without mechanical stress on the powder particles. The deep cryogenic treatment applied to iron particles for over 55 minutes (time for a correspondent diffraction test) could affect the morphology of particles. It may involve some cracks over the microscopic necks of the powder particles and possibly fragmentation due to the fast increasing of lattice parameter and decreasing of density during fast recovering to room temperature. To investigate this aspect we perform an optical microscopy inspection for the powder exposed at 173 and 123 K, see Fig.4a and 4b.



a b

Fig.4. Optical microphotographs for iron powder after cryogenic investigation: a) at 123 K and b) at 173 K.

In Figure 4a we observe powder particles subjected to fast warming form 123 K to the room temperature 298 K. Basically there are no major changes in particles morphology and dimensions are quite the same with initials. In minor details we observe some small alteration of the particle border and we notice the presence of more small particles than in the initial state (compared with the image in Fig.1a). Similar aspects are observed for the powder which was cooled at 173 K with a mention that the minor differences in morphology are slightly diminished compared with the powder cooled at 123 K. All these aspects prove that DP 200 iron powder is morphologically and dimensionally stabile after cryogenic treatment performed in a no-stress condition.

Iron powder is a ductile component during conventional ball milling due to the particle deformation until cold hardening as reported in literature [15,16]. The nanocrystalline state of iron powder is achieved after several milling times around to 20 hours in a planetary ball mill having a 20g acceleration field [17,18]. Temperature decreasing causes a constriction of lattice parameter and an increase of density, Figure 3. Strong contraction of lattice parameter relating to an increased density affects the behavior of particles under milling conditions. The minor changes of DP 200 particles after cryogenic treatment, previously observed, could act as cracking promoters during cryo-milling due to a faster cold hardening of particles. According to our observation, DP 200 iron powder will act as a brittle component in cryo-milling conditions [9, 10] instead of ductile behavior during conventional milling, promoting the formation of individual nanoparticles rather then nanocrystalline ones.

CONCLUSIONS

A sample of atomized iron powder DP 200 (0.02% C) was subjected to a cryogenic investigation by low temperature X - ray diffraction. The initial powder grain size distribution is not uniform and presents a simple Gaussian distribution. The average of grain size is around 160 µm. It was established that the lattice parameter has linear decreases with temperature, and powder density features a linear increase. The thermal linear expansion coefficient varies for different temperature ranges, having a slow decrease at lower temperatures. Based on the observed variation, we found an average thermal linear expansion coefficient (TEC) of $13.77 \cdot 10^{-6}\ K^{-1}$ for all investigated temperature ranges of 123 - 298 K. There was realized a diagram of a TEC versus temperature suitable for more precise cryogenic applications. The resulting absolute value for crystal parameter is 285.14 pm and the absolute density results 7.99 g/cm^3. Optical microscopy inspection proves the stability of DP 200 iron powder under no stress during cryogenic treatment.

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