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Radiation processing of powders for improved fusion structural materials

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Abstract

Radiation processing of metal powders for improved sintering and recrystallization processes leads to better metal structures and property improvements. Structure changes in radiation-modified metals obtained using powder metallurgy methods are discussed.

1. Introduction

Experiments conducted for a wide range of metal powders have shown that powder processing using 2–4 MeV electron or X-ray irradiation gives metals and alloys (Mo, Fe, W, Mo, Al, Ni, Cu, stainless steels, etc.) with homogeneous fine grain structures and enhanced steady-state deformation properties (samples densification is improved by 2–2.5 times, wear resistance is increased by an order of magnitude, rupture strength increases by 30–45%, corrosion resistance of stainless steels is improved by 2–2.5 times, and conditions of sintering can be improved considerably) [1,2]. This paper considers the improvements resulting from radiation processing.

2. Powder grinding

Similar to ultra sound treatment of powders, high-energy electron or X-ray radiation processing causes changes in particle distributions that improves conditions for sintering. Particle size distributions for W and Mo powders are shown in Table 1. Powders were processed using X-rays with à photon energy of 1 MeV at temperatures of 20–80°C for the dose and dose rate ranges of 1–100 Mrad and 0.1–4.0 Mrad/s, respectively.

Data in Table 1 show that midrange particle size fraction is reduced after irradiation, and small sizes are increased. Irradiation makes it possible to eliminate conglomerates and to make the load rate much lower.

Changes in the tungsten powder particle size distribution are shown in Table 2 versus irradiation dose. There is the optimum irradiation dose D_{opt} for which the maximum concentration of small fractions is observed.

Similar changes that lead to higher resistance of the powder to recrystallization and improvement in pressing conditions are observed after electron and gamma-irradiation of all metal powders studied.

3. Chemical refinement

Powder radiation processing removes chemical impurities by breaking them into components and allowing release [1,2]. Chemical reactions stimulated by electron or gamma irradiation lead to impurity concentration changes in powders that results in enhanced physical and chemical properties of the processed metal. For example, chemical contents variations in powder composition after irradiation for tungsten acid salts using 1 MeV electron-induced X-rays are shown in Table 3.

4. Changes in powder particle surface state

Irradiation affects the sintering processes in the earliest stages of contact between powder particles.

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Table 1
Particle size distribution for W and Mo powders

Powder type	Technology	Fraction particle size (mcm)					
		>20	20–16	16–12	12–8	8–5	5–3
		Particle fraction concentration (%)					
Molybdenum	Traditional	9.4	17.4	34.6	25.7	8.4	4.5
	Radiation	8.9	7.4	15.5	29.8	15.8	22.6
Tungsten	Traditional	28	34.8	21.4	10.8	3.6	1.4
	Radiation	17.8	24.8	34	14.8	6.4	2.2

Table 2
Dependence of tungsten powder particle size distributions on gamma-irradiation dose

Dose, D/D_{opt}	Fraction particle size (mcm)					
	>20	20–16	16–12	12–8	8–5	5–3
	Particle fraction concentration (%)					
0	34.2	30.9	21.2	15.1	2.3	1.3
0.88	21.5	34.6	29.3	11.8	1.5	1.3
1.00	17.3	32.1	32.3	13.3	3.6	1.4
1.47	33.5	33.6	21.0	8.2	1.8	1.9
2.00	33.7	32.1	20.3	10.3	1.7	1.9

Table 3
Tungsten powder chemical impurities

State	Impurity concentration (%)			
	Fe_2O_3	NH_3	MoO_3	O_2
Initial	0.008	0.009	0.005	14.2
Irradiated	0.001	0.003	0.001	11.2

Experimental studies of initial sintering behaviour using electron microscopy and positron annihilation techniques have shown that differences in the structure of the samples of stainless steel powders irradiated to different doses are similar to pressed powder structural evolution during annealing and demonstrate considerable sintering acceleration caused by alterations in the particle surface state [1].

Structural alterations during the initial sintering stages correlate with data on the angular distribution of positron annihilation gamma-radiation in pressed samples obtained from electron irradiated niobium and tungsten pressed powders [3]. Some parameters of the angular distribution spectra are adduced in Table 4 for these metals, where the parameter D characterizes the redistribution of positron annihilation probability between conductivity and bound electrons; P is spectrum width at half maximum (mrad); $N(0)$ is the spectrum maximum normalized to one spectrum area at à zero angle value; ΔD, ΔP, ΔN are corresponding relative variations of annihilation parameters percent.

The D value for pressed powders is higher by 15.5% than that for annealed polycrystals. The parameter D decreases for pressed powders by 22% at a fluence value of $\Phi = 5 \times 10^{16}$ cm^{-2} and then slowly grows with fluence increase. Other annihilation parameters correlate with D behaviour. Similar variations of annihilation radiation angular-distribution were observed in tungsten pressed powders.

These results are in a good agreement with data from a positron lifetime study of copper and nickel powders irradiated by electrons and X-rays [4–6]. It was shown that two lifetime components can be distinguished in ultra disperse pressed powders. Electron irradiation has a small effect on positron annihilation in free powders, since initial radiation-induced vacancy concentrations decrease due to their annihilation on the metal particle surface. It was established [7,8] that vacancy diffusion in an elastic stress field leads to their concentration fast growth in near-surface layers, short-living component intensity growth and increase of the parabolic part share in positron radiation angular distribution spectrum, responsible for annihilation at conductivity electrons. On the other hand, it was shown in Refs. [4–6] that long-living component of the positron lifetime spectrum decreases as irradiation time or pressing load grow because

Table 4
Parameters of angular distribution functions for positron annihilation radiation in niobium and tungsten samples

Material	State	Annihilation parametrs					
		D	ΔD	P	ΔP	N	ΔN
	Annealed polycrystal pressed powder	1.55		12.48		38553	
		1.79	15.48	11.43	8.41	39689	2.95
Nb	Pressed sample obtained of electron irradiated powder, $\Phi = 5\ 10^{16}$ cm^{-2}	1.40	9.68	13.08	4.81	35953	6.74
	The same, $\Phi = 10^{17}$ cm^{-2}	1.55		12.45	0.24	37447	2.87
	Neutron irradiated polycrystal, $\Phi = 10^{18}$ cm^{-2}	2.53	63.23	10.13	18.83	43686	13.31
	Pressed powder	1.78		11.70		39758	
W	Pressed sample obtained of electron irra-diated powder, $\Phi = 10^{17}$ cm^{-2}	2.00	12.36	10.60	9.40	42123	5.95
	Errors	±0.05	±0.05	±0.05	±0.05	±200	±0.05

of the micro-pores dispersal and curing due to diffusion processes in elastic stress fields. As irradiation dose grows the trend of increase for average single vacancies concentration and intensity of annihilation at free electrons become prevailing.

The mentioned above competing processes can proceed in pressed samples obtained by electron-irradiated powders pressing, too. The non-monotonic behaviour of electron-positron annihilation parameters dose dependences obtained in our experiment, indicates that both single vacancies formation in powder particles volume and micropore curing in their near-surface layers are proceeding during irradiation.

5. Theoretical estimate

Kinetics of point defects accumulation in the near-surface layer of metal powder particles can be described by the following system of differential equations (vacancies are considered to be immovable):

$$\begin{aligned} \tfrac{\partial C_i}{\partial t} &= K(1 - mC_v) - \tfrac{D_i}{a^2\gamma} mC_i C_v + D_i \tfrac{\partial^2 C_i}{\partial x^2}, \\ \tfrac{\partial C_v}{\partial t} &= K(1 - mC_i) - \tfrac{D_i}{a^2\gamma} mC_i C_v, \end{aligned} \quad (1)$$

with the following initial and boundary conditions

$$\begin{aligned} &C_i(x,0) = 0;\ C_v(x,0) = 0, \\ &\tfrac{\partial C_i}{\partial t}|_{x=0} = K(1 - mC_v^0) - K_1 C_i^0 + D_i(1 - mC_v^0)\tfrac{\partial^2 C_i}{\partial x^2}|_{x=0}, \\ &\tfrac{\partial C_v}{\partial t}|_{x=0} = K(1 - mC_i^0) - D_i mC_v^0 \tfrac{\partial^2 C_i}{\partial x^2}|_{x=0}, \end{aligned} \quad (2)$$

where C_i and C_v are atomic concentrations of interstitials and vacancies; K is the point defect volume generation constant; m is the number of atoms in the recombination zone; g is a geometrical factor depending on the lattice type; D_i is the coefficient of interstitial diffusion; K_1 is the factor taking into account interstitials surface absorption, $C_i^0(t)$ and $C_v^0(t)$ are surface defect concentrations.

The first term in the system of Eq. (1) defines point defects generation and accounts for recombination. The second term takes account of the probability for interstitials to annihilate in the course of thermal migration, and the last describes macroscopic diffusion under a concentration gradient.

Eq. (1) was solved approximately for low irradiation doses by expansion at early times. The qualitative picture obtained in the second approach is shown in Fig. 1.

The results of calculations correlate with the data for annihilation of photon angular distribution studies in irradiated pressed powder samples.

The appearance of excess interstitials concentration in near-surface layers due to irradiation causes micropore curing, and thus creates favourable conditions for contact formation between particles during pressing. Availability of branched boundaries appearing as a result of micropore curing, enhances surface diffusion and accelerates the sintering process.

6. Ready product structure

The advantage of powder radiation processing is that a more perfect metal structure formed upgrading final product quality.

Photos of the structure of the tungsten for samples obtained using traditional and radiation technologies are shown in Fig. 2.

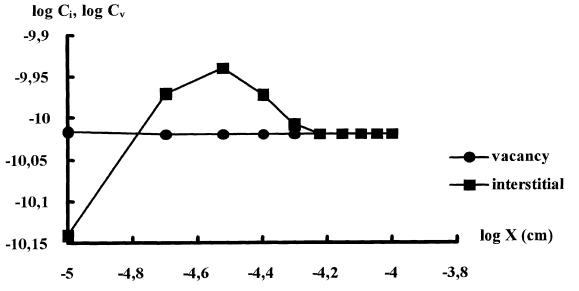

Fig. 1. Vacancy and interstitial concentration distribution through the depth of the sample during irradiation.

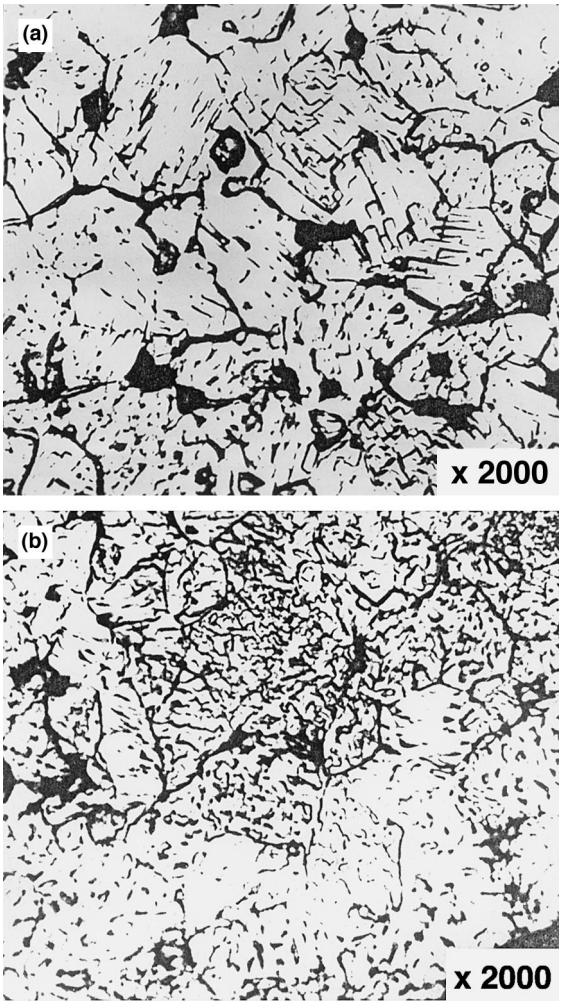

Fig. 2. Structure of tungsten samples (a) traditional technology, (b) radiation technology.

Fig. 2 demonstrates that tungsten samples obtained from irradiated powders are characterized by an absence of big pores and small grains. To determine other pecularities of radiation-modified tungsten structure we have conducted experiments on electric explosive destruction of tungsten wires.

Tungsten wires 45 mm in length and 14.5 mm in diameter were subjected to rapid heating (10^{-5} s) by means of pulse electric currents of high density (10^7 A/cm^2) with reserve energies of 10 kJ in a vacuum chamber at a pressure of 5×10^{-5} Pa. The high velocity of vapour-dispersed liquid flow prevented possible phase transition effects during tungsten wire evaporation.

The wire obtained by traditional powder metallurgy technology was entirely evaporated (a). In the comparison the wire made of irradiated powder retains a dense framework shown in Fig. 3 has been left. Therefore the

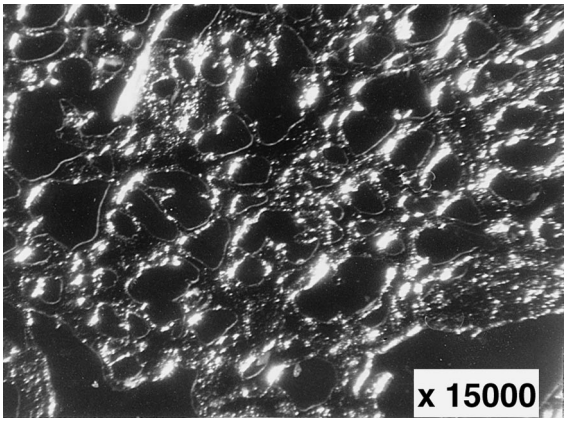

Fig. 3. Tungsten microstructure after electric explosive destruction for tungsten wire obtained by radiation technology.

results show the spatial superstructure remaining for the radiation-modified metal.

The observed local erosion and superstructure formation for the metal made from irradiated powder can be explained by the enhanced diffusion contributions to the sintering and recrystallization processes in irradiated powders.

We suppose that the formation of a structure with higher density and greater perfection as in metal samples obtained by radiation technology is due to the surface state alterations of the powder as a result of high energy electron or gamma-irradiation. Microvoids curing in the powder particle surface layers leads to formation of branched system of boundaries that can work as probable tracks of enhanced diffusion and accelerate sintering.

Recrystallization process as well as sintering are characterized by availability of uniformly distributed closely spaced recrystallization centers and tracks for enhanced diffusion. Metal densification decreases with the distance from recrystallization centers. Enhanced diffusion in the maximum densification direction corresponding to the nearest distance between crystallization centers is considered to be the main cause for superstructure formation that contributes to mechanical property improvements.

Therefore a number of experimental results demonstrate the high efficiency of radiation methods applied to a wide range of fusion metals and alloys for improved service properties.

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