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Study of Densification Progress During Sintering of Doped Tungsten

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ABSTRACT :

Tungsten has the highest melting point and lowest vapour pressure amongst all metals. In addition, it has excellent high temperature strength and hardness of the four common refractory metals. These unique combinations of properties are tailor-made for certain applications. One such application is for the manufacture of lamp filaments. Tungsten lamp filaments must operate at high temperature for long times without significant distortion of the initial coil geometry. To meet these requirements, a bubble strengthening method is applied by a special alloying procedure called AKS-doping referring to the three dope elements of Aluminium (Al), Potassium(K) and Silicon(Si). The non-sag (NS) tungsten is produced via conventional powder metallurgical process. It is the only process that readily affords critical control over the content and dispersion of the dopant elements, which generate special non-sag characteristics. In the present work, experiments were conducted to know the progress of Sintering of NS Tungsten ingots at different points in Sintering cycle. It was observed that the density of the sintered ingot at different point in the sintering cycle do not vary linearly. During the beginning of sintering cycle the percentage change in density is more and at the end of the cycle, the change in density is marginal.

Key Words: NS tungsten, Sintering, Potassium bubbles, Sintering cycle.

1. INTRODUCTION

The starting material for the production of non-sag tungsten is Ammonium Para Tungstate (APT). Ammonium Para Tungstate is converted to Tungsten Blue Oxide during First reduction, which is subsequently doped with Al, K & Si to impart non-sag properties. The doped Tungsten Blue Oxide is then converted to the doped Tungsten metal powder during second reduction by Hydrogen in a furnace. About 10% of the doping potassium is necessary in the tungsten metal to produce the non-sag effect in bubble form. Potassium bubbles are required to produce the interlocking grain structure that gives the tungsten lamp filament a long life [1]. Tungsten powder obtained after second reduction is acid washed to remove excess dopants present on the surface of the tungsten particles which serve no purpose. After acid wash, powder is washed repeatedly with distilled water and dried in an oven. The dried powder is sifted to remove agglomerates and blended to get a homogeneous blend. This powder is pressed to the required size and presintered to give enough strength for handling during sintering.

The common technique for sintering NS-doped tungsten is direct sintering. In direct sintering, large electric current is passed through the pressed ingot in order to self-resistance heat the ingot to temperatures as high as 3000^0C [2]. A water jacketed bottle is used to contain the hydrogen atmosphere. One end of the ingot was clamped using water cooled copper contacts with tungsten inserts. The other end of the ingot was held by a tungsten clip which is free to move as the bar contracts as shown in Fig.1 [3].

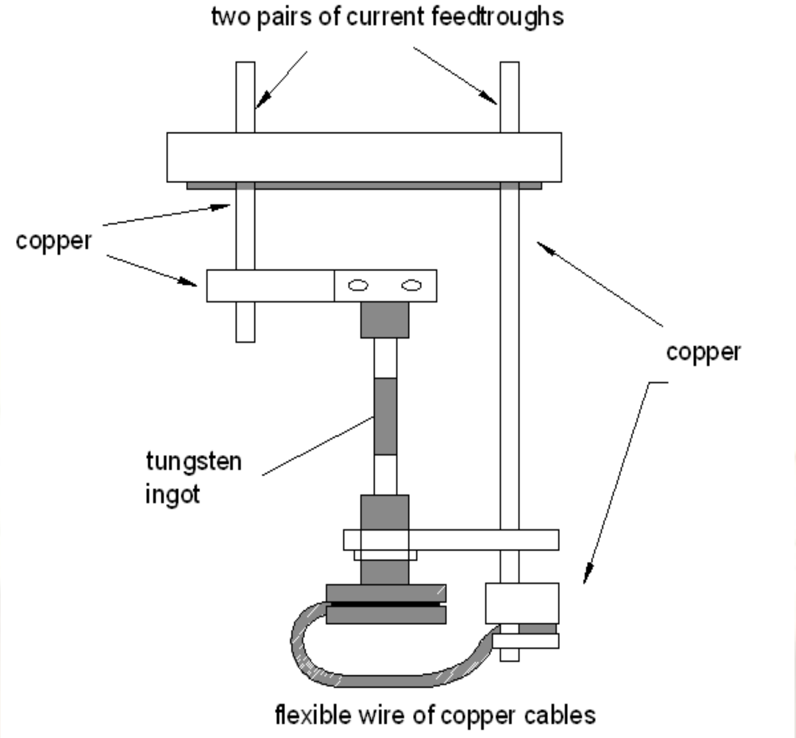


Figure 1. Sample holder for sintering tungsten ingots with direct current

Sintering of NS doped tungsten performs three functions[2]. First, sintering densifies the ingot in order to provide a density and grain structure which is suitable for thermo mechanical processing. Second, Sintering adjusts the chemistry of the ingot, including Potassium, Aluminium and Silicon and interstitial concentrations, so that the ingot is amenable to thermo mechanical processing. Third, Potassium bubbles are generated during sintering which are required to produce interlocking grain structure. Potassium is virtually insoluble in tungsten, and it remains in the sintered ingot in small, roughly spherical pores [4,5,6]. Sintering is the last point in the wire making process where the bulk chemistry of the NS doped ingot can be adjusted [2].

2. EXPERIMENTAL SET UP AND PROCEDURE

Pressed and presintered ingots were used for sintering. Experiments were conducted to check the density of the ingots at different points in the sintering cycle. Six ingots pressed from same batch of the doped Tungsten powder were taken for the experiment. At the end of each trial, density of the ingot was measured and samples of these ingots were taken for characterization. Fig. 2 shows Sintering cycle used for the experiment. It contains three ramps and three holds.

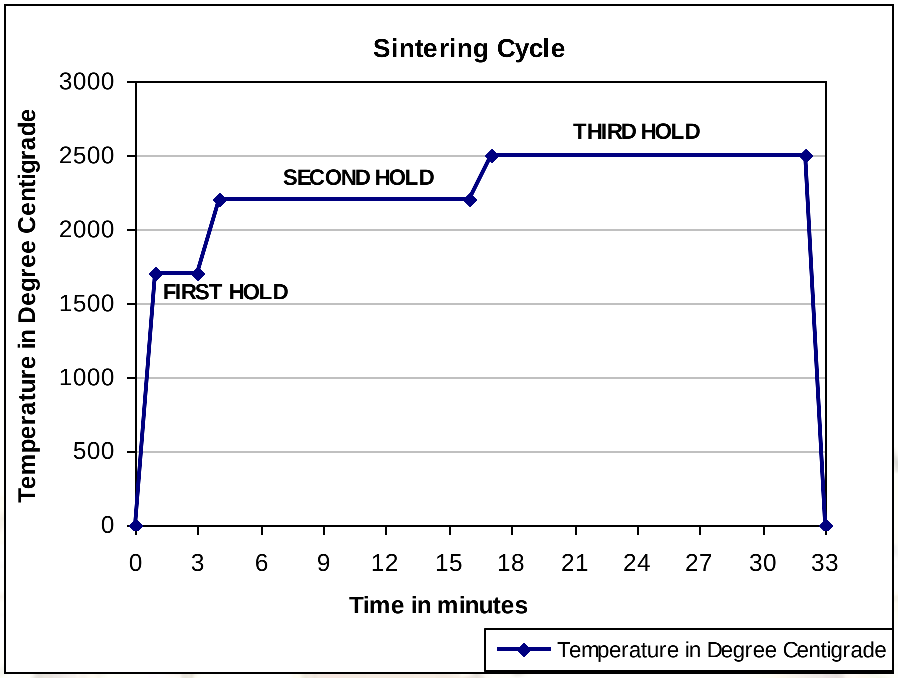


Figure 2. Sintering Cycle

3. RESULTS & DISCUSSIONS

During sintering, temperature is controlled by the current transmitted through the ingot. Control of both ramps and holds is important to get uniform good quality lamp wire. The ingot density and Potassium concentration are sensitive to both the ramp rates & hold currents, since these control the temperature history of the ingot during sintering[7,8,9].

From the experiments conducted, it was observed that the density of the sintered ingot at different point in the sintering cycle do not vary linearly. Percentage change in density is more during the first 10 minutes after start of the sintering cycle and after 22 minutes; the change in density is marginal.

During the beginning of sintering, the ingot contains open porosity filled with oxygen and closed porosity containing potassium. As sintering process starts, hydrogen carries away the oxygen from the open porosity, thereby reducing the open porosity. This is the reason for more percentage change in density during the beginning of sintering.

R.L.Coble [10] describes the geometric structure of particles and pores during sintering in three stages. In the first stage, the inter particle contact area increases to ~0.2 of the cross sectional area of the particle, leading to neck growth and gives rise to an increase in the relative density of the compact to ~0.6. In the second stage, inter particle necks lose their identity and the pore phase approximates to a continuous channel coincident with three grain edges throughout the matrix. All the pores are situated on grain boundaries. In the third stage, continuous porosity is pinched off forming closed isolated pores within the matrix and these pores spheroidize during sintering. Bewlay [11] reports that during third stage of sintering, potassium diffuses from sub micron pores into larger pores, its vapour pressure opposes further shrinkage of these pores; this restricts further densification. Hence the change in density is marginal after 22 minutes of sintering.

While sintering, the first hold is to let the contacts seat firmly on the ingot. Rapid volatilization of impurities takes place in the second ramp and hold and slow densification starts. During the third hold, impurities left out are removed and rapid densification occurs. During this period, the temperature gradually increases due to smoothening of the surface and complete densification (88-92% of theoretical density, theoretical density of tungsten is 19.3g/cc) takes place. Moon et al [9] concluded that densification of AKS-doped tungsten is controlled by lattice diffusion, but studies made by B.P.Bewlay [11] indicate that densification is controlled by grain boundary, not lattice diffusion.

Fig. 3 shows relation between density of sintered ingot with sintering time and sintering kinetics. The degree of sintering is described by the densification parameter 'ϕ' (1) which is defined as-

$$\phi = (\rho_1 - \rho_0)/(\rho_T - \rho_0) \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(1)$$

Where, ρ_1 is the density of the sintered ingot, ρ_T is the theoretical density of the ingot (19.3 g/cc) and ρ_0 is the pressed density (10.76 g/cc). At 1700^0C after 3.33 minutes the densification parameter is 0.26 and corresponding sintered ingot density is 13 g/cc. At 2200^0C after 10.16 minutes the densification parameter is 0.51 and corresponding sintered ingot density is 15.12 g/cc. At 2200^0C after 16.16 minutes the densification parameter is 0.62 and corresponding sintered ingot density is 16.02 g/cc. At 2500^0C after 22 minutes the densification parameter is 0.73 and corresponding sintered ingot density is 17.02 g/cc. At 2500^0C after 25 minutes the densification parameter is 0.76 and corresponding sintered ingot density is 17.29 g/cc. At 2500^0C after 32 minutes the densification parameter is 0.77 and corresponding sintered ingot density is 17.39 g/cc.

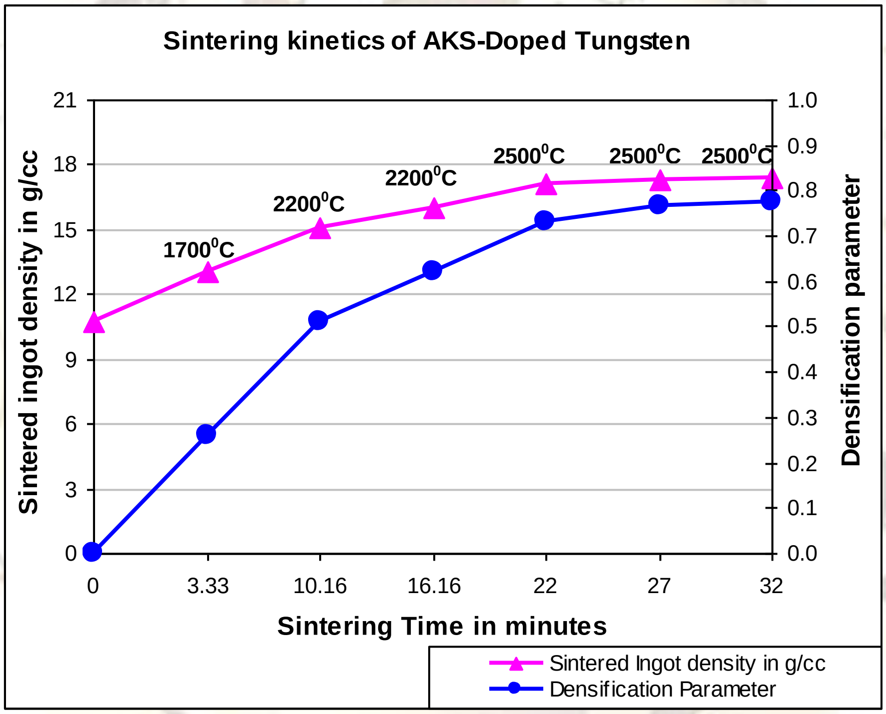
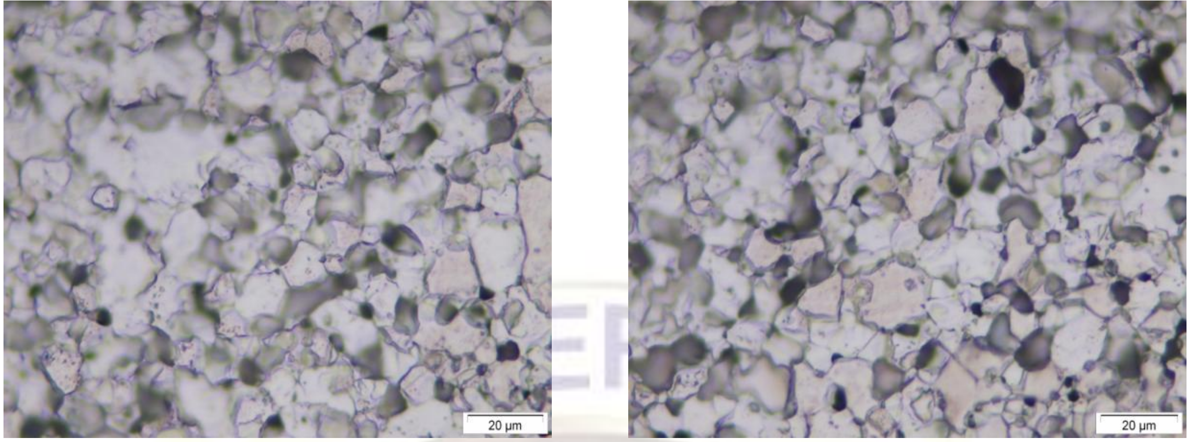


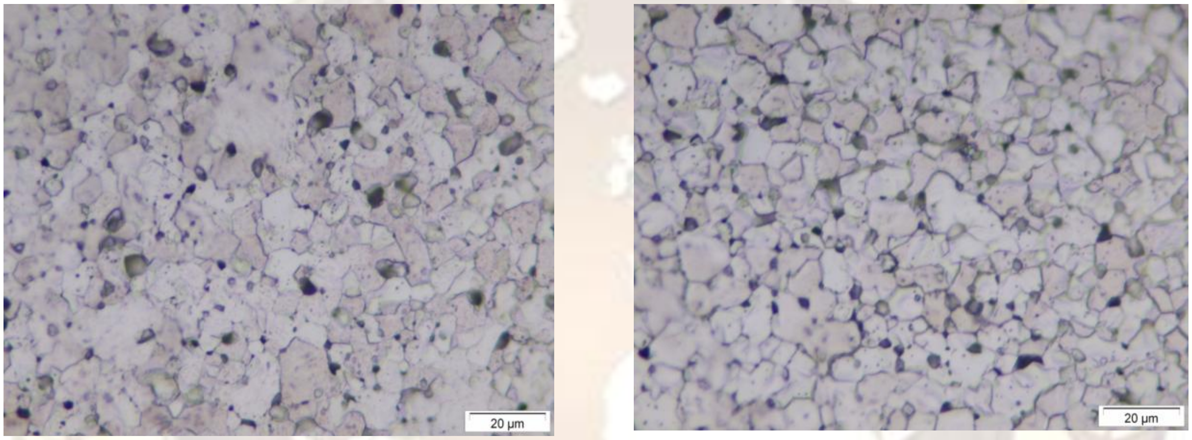
Figure 3. Sintering kinetics of AKS-Doped Tungsten Powder

Fig. 4(a-d) shows microstructure of ingots sintered with different sintering time. Inter particle neck growth is observed after 10.16 minutes at 2200^0C as shown in Fig. 4(a). During this stage most of the grain boundaries within the individual tungsten particles prior to sintering have migrated either to the surface of the particle or the interparticle necks. After 16.16 minutes at 2200^0C a small number of micro pores are observed within the grain and inter particle necks have completely lost their identity and the porosity is largely disconnected and located on grain boundaries as shown in Fig. 4(b). At 2500^0C after 22 minutes and till the end of the cycle, spheroidization of the porosity occurs and grain growth significantly occurs as shown in Fig. 4(c-d). Potassium present in the bubbles also pin the grain boundaries and restricts grain growth during sintering. Since potassium is present in gaseous form, vapour pressure opposes densification [2].



a. Sintering time 10.16 minutes

b. Sintering time 16.16 minutes



c. Sintering time 27 minutes

d. Sintering time 32 minutes

Figure 4. Microstructure of the ingots sintered at different sintering time

4. CONCLUSION

- The density of the sintered ingot at different point in the sintering cycle does not vary linearly.
- Percentage change in density is more during the first 10 minutes after the start of sintering cycle. This is due to reduction in open porosity which is filled with oxygen. As sintering progresses, hydrogen carries away the oxygen from the open porosity, thereby reducing the open porosity. Also, increase in inter particle contact area of the particle leading to neck growth and gives rise to an increase in the relative density of the compact.
- At the end of sintering cycle, the change in density is marginal. This is because of diffusion of potassium from sub micron pores into larger pores during third stage of sintering, its vapour pressure opposes further shrinkage of these pores; this restricts further densification. Also Potassium present in the bubbles pin the grain boundaries and restricts grain growth. Hence the change in density is marginal.

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