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The relationship of microstructure to mechanical properties of Al-, Si-, K-doped tungsten wire

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Tungsten wire, commercially doped with aluminium, silicon, and potassium, was obtained from six different manufacturers with a diameter of 0.0063 cm. The tensile strength at 1620 °C, a measurement of the quality of the wire used by the General Electric Company, was determined and found to be different for each wire. Additional samples of wire were annealed at 1620 °C under the same conditions employed for the tensile test. To determine the microstructure, these samples were thinned for transmission electron microscopy along with the wires in the as-drawn condition. The widths of from 75 to 125 subgrains were measured for each sample. In addition, in the annealed samples, the spacing of the strings of potassium bubbles was also determined. It was found that the 1620 °C tensile strength was related to the increase in width of the subgrains during the 1620 °C anneal and that the amount of subgrain widening was determined by the spacing of the strings of bubbles. Hence, the 1620 °C tensile strength was related to the bubble string spacing; the closer together were the strings, the higher was the 1620 °C tensile strength. It was also shown that the grain morphology of the wire, as measured by the stretch or "sag" of a filament during a stress test, is also determined by the spacing of the strings of potassium bubbles.

1. Introduction

Tungsten to be used for incandescent lamp filaments is always doped with potassium, aluminium, and silicon. The dopants are added as aqueous solutions of potassium silicate and aluminium chloride to the oxide which is then reduced to tungsten in hydrogen. The useful potassium is contained in pockets inside the reduced tungsten crystals [1]. The tungsten powder is then pressed and sintered to form ingots [2] which are unidirectionally rolled, swaged, and drawn to wire. In this way, the pockets of potassium originally inside the crystals are elongated and spheroidized with each successive reduction and anneal, respectively, to form strings of very fine bubbles of potassium in the wire [3–7]. An example of the strings of bubbles is given in Fig. 1, which will be discussed later.

The strings of bubbles set the grain morphology of the filament by retarding lateral motion of sub-grain and grain boundaries while grain growth occurs parallel to the bubble strings and to the wire axis. In this way, the interlocking boundaries of large area are formed which reduce grain-boundary sliding and increase filament life over that of undoped wire. The latter wire would have only transverse grain boundaries (the so-called "bamboo structure") and would, therefore, be subject to early failure by grain-boundary sliding. Thus, the mechanical properties of tungsten wire at elevated temperatures are related to the microstructure and grain morphology of the wire. The high-temperature tensile strength of as-drawn wire will depend upon the stability of the microstructure, i.e. the rate of decrease of number of dislocations and the mobility of the subgrain and grain boundaries, which, by migrating, sweep out dislocations during primary nucleation and recrystallization.

Although microstructural variations can be caused by varying the processing, from rolling of the ingot through swaging of rod and wire drawing (all at elevated temperatures), the final distribution of the potassium bubbles in the wire in terms of the spacing between the strings of bubbles depends primarily on the distribution of the pockets of potassium in the ingot. Thus, when aluminium, silicon and potassium-doped lamp filament wire of the same diameter, manufactured by six different commercial suppliers, became available (provided by the Chemical and Metallurgical Products Department of the General Electric Co., Cleveland, Ohio), it presented an opportunity to characterize the microstructure by means of transmission electron microscopy and to relate the microstructure to the high-temperature tensile strength of the wire.

2. Experimental procedure

As a quality control measure, the Chemical and Metallurgical Products Department subjects all wire to a high-temperature tensile test. The tensile test is performed by heating the wire in a tungsten tube furnace

Figure 1 Transmission electron micrograph of wire F after 1 min anneal at 1620 °C in hydrogen.

placed on an Instron tensile machine. The tube and wire are brought to 1620 °C in an atmosphere of hydrogen, at which temperature the wire is held for 30 sec prior to the test. The samples are then pulled in tension to fracture at a cross-head rate of 1 in. min^{-1}. The entire test takes about 1 min, after which the wire and tube are cooled to room temperature. The six wires, approximately 0.0063 cm diameter, and labelled, arbitrarily, A to F (manufactured by Osram GMbH, General Electric Co., Toshiba Corp., GTE Products Corp., Luma Lampen AB, and Nippon Tungsten Co.), had different tensile strengths at 1620 °C and, therefore, the wires were subjected to transmission electron microscopy to relate microstructural features to fracture strength as measured by this test.

Rather than using the tensile tested wires for TEM, additional samples of wire were heated for 1 min at 1620 °C under the same conditions used in the tensile test. This was done to eliminate the confusion that would result from the deformation caused by the tensile test. The transmission electron microscope (TEM) samples were prepared by inserting the wire into a 0.15 cm o.d. platinum tube to a depth of 0.5 cm. The tube was flattened to encase the wire and the flattened sides were ground on 600-grit paper until the tungsten wire was about 0.002 cm thick. The mechanically thinned specimen was then electropolished in 4% sodium hydroxide in distilled water at about 30 V until a small hole appeared.

Twelve to fifteen transmission electron micrographs at magnifications of × 50 000 to × 200 000 were taken of both the as-drawn wires and the wires that were annealed at 1620 °C. From the micrographs, the widths of from 75 to 125 subgrains were measured for each sample along with the number and spacing of the strings of potassium bubbles which were visible in TEM only in the annealed wires.

3. Results

Fig. 2 shows an example of the microstructure of as-drawn wire B. The subgrains in wire B are well defined, contain only a few dislocations, and are separated by sharp low-angle (a few degrees of misorientation) boundaries. There are variations in the density of dislocations in the as-drawn wires as may be seen by comparing wire F in Fig. 3 with wire B in Fig. 2. Variations also occur from place to place in the same wire.

On annealing for 1 min at 1620 °C, the dislocation density decreases, the subgrain width increases, and strings of potassium bubbles become visible. Fig. 1 shows an area of the annealed wire F as an example. In general, the potassium bubbles are very small, the smallest in Fig. 1 being about 5 nm. Because the contrast of the bubbles in TEM may depend upon the orientation and the smoothness of the surface of the subgrain in which they reside, not all of the bubbles will be visible in all of the subgrains in the micrographs of all wires. In addition, some of the strings of bubbles may be hidden by the grain or subgrain boundaries: boundaries were not counted as strings of bubbles. However, because these effects hold true for all samples, and as many as 125 subgrains were examined for each sample, any relationship of bubble string spacing to mechanical properties should be evident. String spacing is the term used for the distance between lines of bubbles and not the spacing between individual bubbles in any one line.

Table I lists the width of the subgrains in the as-drawn wires, the width after the 1620 °C anneal, and the spacing of the strings of bubbles within the subgrains after the anneal.

Of particular interest in this work is the effect of the bubble strings on the ability of the subgrain boundaries to migrate during the 1620 °C anneal because migration will reduce the dislocation content and,

Figure 2 TEM of as-drawn wire B showing subgrains.

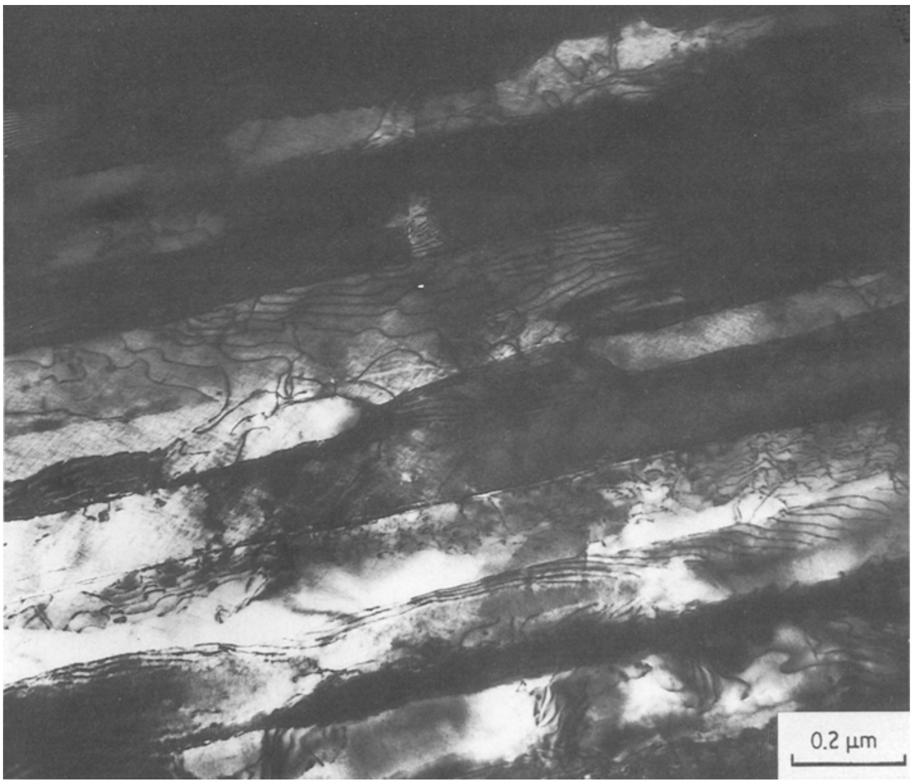

Figure 3 TEM of as-drawn wire F.

therefore, the strength of the wire. Fig. 4 shows the relationship between bubble string spacing and the change in width of the subgrains during the 1620 °C anneal; the further the bubble strings are apart, the greater the distance the subgrain boundaries can move. Thus, the amount the subgrains widen depends upon the spacing of the bubble strings.

Because potassium is not soluble in tungsten [8], the potassium-filled bubbles are immobile and the distance between the strings of bubbles will not change during the anneal. Thus, the 1620 °C tensile strength should be related to the distance the subgrain boundaries move during the anneal. The effect of the spacing of the strings of bubbles on the 1620 °C tensile strength is plotted in Fig. 5 and listed in Table II along with the room-temperature tensile strength. From Fig. 5, it is clear that the 1620 °C tensile strength is related to the bubble string spacing; the further apart are the bubble strings, the lower the tensile strength. It follows then, from Fig. 4, that the 1620 °C

TABLE I

Wire	Subgrain width, μm		Increase in width during anneal (μm)	Bubble string spacing (μm)
	As-drawn	Annealed[a]		
A	0.18	0.52	0.34	0.45
B	0.14	0.43	0.29	0.37
C	0.125	0.35	0.225	0.28
D	0.19	0.37	0.18	0.25
E	0.22	0.33	0.11	0.21
F	0.19	0.30	0.11	0.21

[a]1 min at 1620 °C.

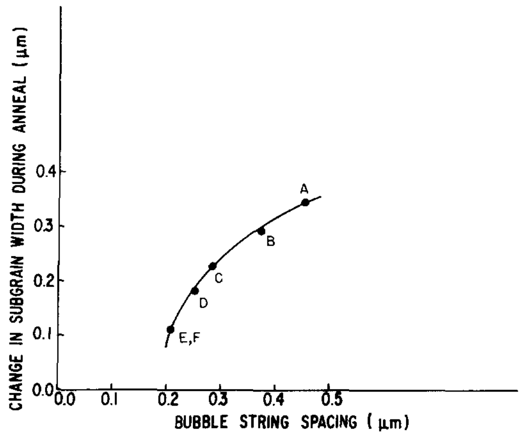

Figure 4 Plot of change in subgrain width during the 1620 °C anneal as a function of the bubble string spacing.

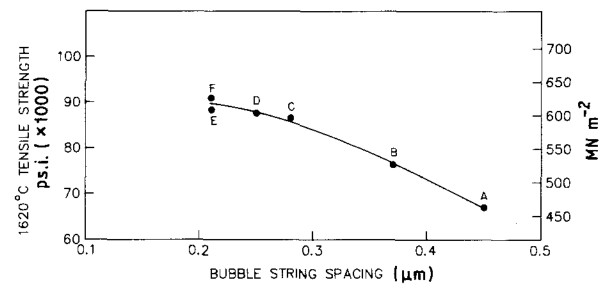

Figure 5 Tensile strength at 1620 °C as a function of the bubble string spacing.

TABLE II Mechanical properties

Wire	Tensile strength			
	Room Temp		1620 °C	
	p.s.i.	$MN\,m^{-2}$	p.s.i.	$MN\,m^{-2}$
A	315 337	2172.7	67 035	461.9
B	391 964	2700.6	76 627	528.0
C	427 171	2943.2	86 764	597.8
D	325 474	2242.5	87 745	604.6
E	344 767	2375.4	88 290	608.3
F	366 894	2527.9	92 105	634.6

tensile strength of the wire is related to the ease with which the subgrain boundaries can move during the anneal. Fig. 6 shows that the tensile strength at 1620 °C decreases in the same order as the subgrains increase in width during the anneal.

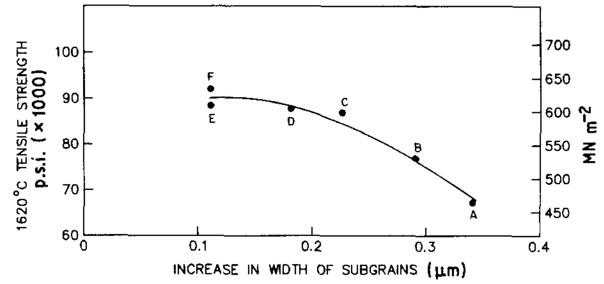

Figure 6 Tensile strength at 1620 °C as a function of the increase in width of subgrains during the anneal.

Because the strings of bubbles impede lateral motion of the subgrain and grain boundaries during recrystallization, the spacing of the strings might be expected to relate to the aspect ratio of the growing grains [3, 6] and, thereby, to the fracture strength of the recrystallized wire. To test the possibility, lamps with coiled-coil filaments and a standard atmosphere were made using wires A to F. A measure of the strength of the wire was obtained by spinning the lamps on a table shown in Fig. 7 with the filaments normal to the diameter of the table and parallel to the plane of the table [5]. Power was supplied to the lamps through slip rings mounted on the shaft of the table; the filaments were at operating temperature of approximately 2900 K during the stress test and were, therefore, fully recrystallized. The elongation or "stretch" of the filament under stress, caused by grain-boundary sliding, is described as "sag" which can be measured after the test; the greater the sag, or creep, of the wire, the lower the strength of the recrystallized wire. Fig. 8 shows a plot of the sag of filaments of wires A to F after being stressed for 5 h at 4 g on the spinning table. It is clear that the sag of the recrystallized wire is related to the spacing of the strings of potassium bubbles in the wire which determines how far the subgrain boundaries move during the early stages of recrystallization.

4. Discussion

The relationship between sag and bubble string spacing arises from the grain morphology and its effect upon grain-boundary sliding. That is, without bubble strings, lateral grain growth would produce a classic "bamboo" structure in the wire, and, at filament operating temperatures, the grains would readily slide apart along their boundaries. On the other hand, the doped wire forms grains with interlocking boundaries, the so-called "non-sag" structure which provides greatly increased grain-boundary area. The greater the area of the individual grain boundaries, the lower the tendency for sliding on those boundaries and the lower the sag or creep of the filament [5].

While the material processing is known in detail for only one of the wires, it is reasonable to assume that all manufacturers use similar processes, from doping through wire drawing. The critical element in the process is the distribution of potassium in the ingot. The spacing of the pockets of potassium in the ingot will determine the spacing of the strings of bubbles in

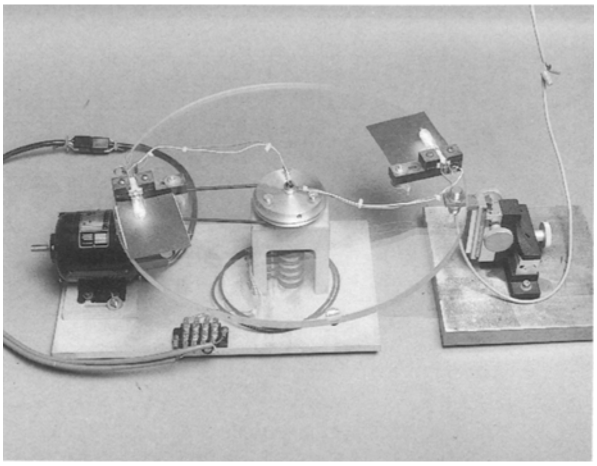

Figure 7 The spinning lamp stress tester.

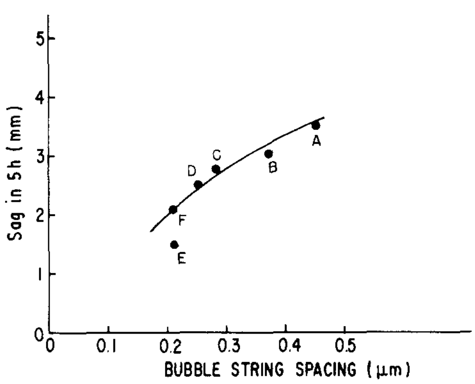

Figure 8 "Sag" of the filaments from wires A to F in the spinning stress test as a function of the bubble string spacing.

the final wire. Because the potassium bubbles do not move during recrystallization anneals, the spacing of the strings will not be changed by the anneals. The widening of the subgrains is determined by the spacing of these strings because the migration of the subgrain boundaries is limited by the strings of bubbles and the bubbles do not move during the anneal. The tensile strength at 1620 °C is then also determined by the spacing of the strings of bubbles because they determine the degree of widening of the subgrains and, therefore, the reduction in the number density of dislocations. The greater the widening of the subgrains, the lower the 1620 °C tensile strength; the tensile strength at 1620 °C is a measure of the spacing of the strings of bubbles in the wire.

The effect of the bubble string spacing on the sag of the filaments, which obviously relates to the grain morphology of the filament, was not expected. Because the sag relates to the spacing in the same way that widening and 1620 °C tensile strength does for the six wires, it follows that the string spacing also determines the grain morphology of the wire.

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