Weldability of Three Forms of Chemically Vapor Deposited Tungsten

Electron beam welding can be used to join this inherently brittle material

BY W. A. BRYANT AND R. E. GOLD

ABSTRACT. Methods were developed for electron beam welding several forms of chemically vapor deposited tungsten. Weld ductility was evaluated from the results of ductile-brittle transition temperature determinations. Welds in tungsten produced from WF_6 were considerably more ductile than those associated with WCl_6 produced tungsten. The larger grain size of the latter material was largely responsible for this behavior.

Introduction

The number of uses for chemically vapor deposited (CVD) tungsten has increased dramatically in the last 10 years. One such use is as a thermionic emitter. For this application a material must obviously possess a high electron work function. In addition, however, it must have both high temperature strength (which is closely related to structural stability) and good weldability. There are several forms of CVD tungsten which include fluoride tungsten (produced by deposition from WF_6), chloride tungsten (produced by deposition from WCl_6), and duplex tungsten (chloride tungsten deposited onto a substrate of fluoride tungsten). The high temperature strength and stability of these materials have been studied and compared (Ref. 1). Their weldability is reported here.

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Experimental Procedure

As-deposited 60 mil thick sheets (obtained from a supplier) were ground to 6-32 rms finish on the surfaces perpendicular to the growth direction. At least 4 mils thickness was removed from the surface adjacent to the deposition mandrel by this operation to assure removal of the randomly oriented grain structure usually found there. Following cutting and grinding to test specimen blank size, all material was stress relieved at 1200 C for 1 h in a vacuum of 1×10^{-6} torr or better. Each material possessed the characteristic columnar grain structure and generally contained sublayers resulting from interruptions in the deposition process.

The fluorine content of each material was between 1 and 2 ppm. Slight oxygen additions (less than 20 ppm) were purposely made by the supplier to the fluoride material to provide grain stability that would otherwise possibly require greater and more uncontrollable levels of fluorine. Chlorine contents were determined but are not reported since their validity is in considerable doubt.

Experimental Results

Preliminary Studies

Initially three sets of weld parameters were used on each material type. These parameters are presented in Table 1. Visual inspection revealed all welds to be crack-free with the exception of a small transverse crack in one piece of test set C1-D3. This degree of cracking was confirmed by dye penetrant and radiographic inspection.

Prior to EB welding, test sheets (3¼ in. long × 1¼ in. wide × 0.060 in. thick)* were degreased with M-6 solvent, etched in a 30 lactic-$3HNO_3$-1HF solution (ratio by volume) and consecutively rinsed with flowing hot tap water, boiling distilled water, and

**In the case of duplex material, this total thickness is comprised of 45 mils of fluoride tungsten and 15 mils of chloride tungsten.*

Table 1 — Initial Weld Parameters for CVD Tungsten

Test Set	Type CVD W	Weld Speed, ipm	Preheat, C	Voltage, kV	Current, ma
C1-F1	Fluoride	15	None	30	63
-C1	Chloride	15	None	30	68
-D1	Duplex	15	None	30	68
C1-F2	Fluoride	15	705	30	68
-C2	Chloride	15	705	30	60
-D2	Duplex	15	705	30	55
C1-F3	Fluoride	15	428	30	55
-C3	Chloride	15	428	30	55
-D3	Duplex	15	428	30	55

Table 2 — Bend DBTT (longitudinal/transverse) for CVD Tungsten

Preheat condition	Temperature, deg C, for: Fluoride CVD	Chloride CVD	Duplex CVD
No preheat	370/540	400/540	540/540
428 C preheat	<260/455	>540/>540	315/430
705 C preheat	<430/400	540/480	315/455

Table 3 — Bend DBTT for Welded Fluoride CVD Tungsten

Heat treat condition	Temperature, deg C, for: Longitudinal	Transverse
As-welded	<260	455
Outgassed 50 h at 1800 C	305	360
Outgassed + aged 5000 h at 1540 C	290	340
Outgassed + aged 5000 h at 1700 C	305	305

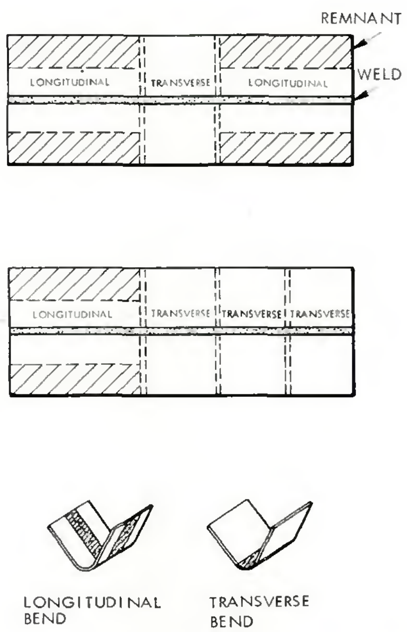

Fig. 1 — Sketches showing sectioning of a set of EB weld specimens into bend test specimens and orientation of the specimens with respect to the weld bead

ethyl alcohol prior to being hot air dried. All handling during cleaning was done using prescrubbed and rinsed latex gloves. The cleaned specimens were then degassed at 1095 C for 1 h at 10^{-5} torr.

Full penetration welds were then made with the first deposited surface facing upward (which is fluoride for the duplex material). During welding, the sheet was fully restrained within a fixture (described in Ref. 2). All welding was performed in vacuum with pressures of 2×10^{-5} torr or lower using a 30 kW Sciaky weld facility.

All welds were visually, dye penetrant, and radiographically inspected. Each of the two test sheets of a given set was sectioned with an abrasive cutoff wheel according to Fig. 1 to provide three longitudinal and four transverse EB weld specimens. Each specimen had the dimensions 1¼ in. long × ⅝ in. wide × 0.060 in. thick. Bend testing was used to determine the ductile-brittle transition temperature (DBTT) of each CVD tungsten type in both the longitudinal and transverse orientations. This testing was done under inert atmosphere and utilized a punch radius of 0.211 in. to give a bend factor of 3.4 t. This produced an outer fiber strain of approximately 13%. The crosshead speed was 1.0 in./min with a span of 1.0 in.

The results of bend tests as influenced by preheat treatment are presented in Table 2. Ductile-brittle transition temperature was taken as the lowest temperature which permitted a bend of 95 degrees. In failed transverse specimens, an obvious preference for weld centerline failures was observed. This behavior is typical of EB welded tungsten and tungsten base alloys where failure has been observed to occur preferentially along the vertical array of grain boundaries. This pattern is produced by solidification in which columnar grain growth follows the direction of greatest rate of heat removal.

In these tests, no postweld conditioning of the weld bead was performed. As a consequence, some variability in the results may have been introduced from the rough weld surface which is intrinsic to electron beam welding. Root defects were not considered critical since they were on the compression side of the bend specimen.

Based on these preliminary results, the welding of specimens for further evaluation utilized a preheat temperature of 428 C for the fluoride and duplex materials and no preheat for the chloride material. In each case, the other parameters attendant with the chosen preheat (Table 1) were used. The desired number of bend specimens (3 longitudinal and 4 transverse) were not obtained from each weld set (Fig. 1) due to weld cracking. Unaccountably, one duplex and 11 chloride (out of a possible total of 21) bend specimens were lost in this manner. In all cases failure was intergranular.

Following outgassing for 50 h at 1800 C, bend specimens were given one of the treatments consisting of aging 5000 h at 1540 C or aging 5000 h at 1700 C. Bend testing was conducted as previously described. The fractures were examined, and the welds metallographically examined to ascertain changes in structure resulting from the heat treatments.

Fluoride Tungsten

The microstructure of the welded fluoride-produced material is shown in Fig. 2, which indicates the extent of the fusion and heat-affected zones. Also seen are top-of-weld and root defects which were generally typical of the bead-on-plate electron beam welds. The entire cross section of the metallographic specimen was checked at 500X magnification for the presence of porosity. Very few pores were noted. Those present were mostly to be found at grain boundaries lying in the heat affected zone. Outgassing (50 h at 1800 C) produced no perceptible change in this material. Where porosity was noted, it was found to occur at grain boundaries either within the fusion or heat-affected zones. Aging the welded material for 5000 h at 1540 C did not alter the microstructure nor increase the incidence of porosity. Aging for 5000 h at 1700 C resulted in minimal grain coarsening outside of the fusion zone. However, the distribution and frequency of porosity was not changed from the as-welded condition.

Bend ductile-brittle transition temperatures of fluoride CVD tungsten were determined for the welded material in each state of heat treatment. These data are given in Table 3. As in the as-welded material, there is a preference in the heat treated materials for weld centerline fractures to occur in the bending of transverse weld specimens.

The DBTT for the transverse welds is lowered by the outgassing treatments. This reduction can likely be attributed to relief of residual stresses which attend solidification. No further significant reduction is affected by the aging treatments.

The increased DBTT value found in the outgassed longitudinally welded material as compared to the as-welded material cannot be accounted for by any marked increase in grain size as the result of the outgassing treatment. If this value for the DBTT is taken as 260 C, it would not be judged to be significantly different from those associated with the heat treated specimens. No further increase in the transition temperature (within the limits of experimental determinations) accompanied either of the aging treatments. The corresponding change in grain size was judged minimal.

Chloride Tungsten

The microstructure of the welded chloride-produced tungsten is shown in Figure 3. Within the heat-affected zone, porosity was noted at one of the interlayer interfaces (Figure 4). The dark band passing through the portion of the specimen unaffected by welding is not a crack but rather is a layer of extremely fine grain size. The remainder of the material is essentially pore free except for the very fine porosity occasionally noted throughout the length of the one layer. No specimens were available with which the effect of outgassing on welded material could be determined. However, it was noted from unwelded material that this heat treatment produced considerable grain growth.

Aging at 1540 C produced grain growth beyond the size (135 microns) found in outgassed material. Within the heat-affected zone several very large pores could be seen at locations once occupied by interlayer interfaces. Fine porosity was noted at these interfaces in regions lying outside the heat-affected region and occurred to a greater extent than it did in as-welded material indicating a coalescence of vacancies.

This interlayer interface porosity was accentuated by the 1700 C aging treatment. Large pores were evident throughout the specimen rather than just within or near the heat-affected zone as occurred during the lower temperature aging treatment. The grain size was equivalent to that produced by the 1540 C age.

The ductile-brittle transition temperatures of chloride CVD tungsten as a function of heat treatment were determined. These results are contained in Table 4. As with the fluoride material a general preference for weld centerline failures for transverse welds was noted. The variability noted in these data is believed to result from the variation in top-of-weld surface topography as discussed earlier. The DBTT values for each heat treatment condition are significantly higher than for their fluoride tungsten counterparts.

Duplex Tungsten

Very little porosity was noted in either the weld zone or outside of it in duplex material prior to any post-weld heat treatment. That which was noted was very fine and generally was found in the heat-affected zone at a location of an interlayer interface. Outgassing the welded material produced considerable grain coarsening of the chloride-produced portion (Fig. 5) but did not appreciably increase the amount of porosity. That porosity which was noted was again found in the heat-affected zone lying along grain boundaries (Fig. 6). Aging at either 1540 or 1700 C had no effect on the extent of porosity but did produce further grain coarsening in the chloride portion.

Since welds were bend tested with the fluoride component being placed in tension, the results on the DBTT determinations (Table 5) would be expected to be similar to those obtained on the fluoride-produced tungsten (Table 3). The data for transverse welds are very similar; the only significant difference being in the DBTT following outgassing, which is considerably higher for the duplex material. As in the other two materials, weld centerline failures were found in the fractured transverse specimens. The DBTT data for the longitudinally welded duplex tungsten are, on the whole, approximately equivalent to the corresponding data for fluoride tungsten. There is essentially no change in DBTT with thermal treatment. This trend is not unexpected since the fluoride portion of the material was very structurally stable.

Table 4 — Bend DBTT for Welded Chloride CVD Tungsten

Heat treat condition	Temperature, deg C, for: Longitudinal	Transverse
As welded (no preheat)	400	540
Outgassed 50 h at 1800 C	(No data - welds cracked prior to testing)	
Outgassed + aged 5000 h at 1540 C	<460	540
Outgassed + aged 5000 h at 1700 C	510	485

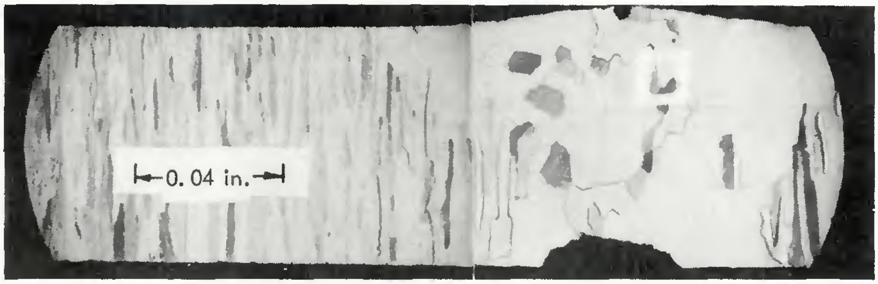

Fig. 2 — Transverse section through fluoride CVD tungsten following EB welding

Fig. 3 — Transverse section through chloride CVD tungsten following EB welding

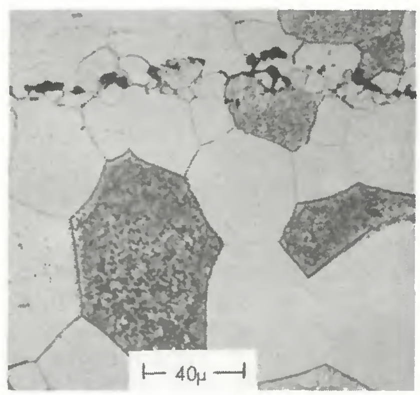

Fig. 4 — Enlarged view of enclosed region of Fig. 3 showing porosity within the heat-affected zone

Summary and Discussion

For longitudinal welds it would be expected that DBTT behavior would closely approximate that of unwelded material. Thus it is not surprising that longitudinally welded chloride CVD tungsten should be more brittle than its fluoride or duplex counterparts since, as shown previously for unwelded material

Fig. 5 — Transverse section through duplex CVD tungsten following EB welding and heat treatment for 50 h at 1800 C

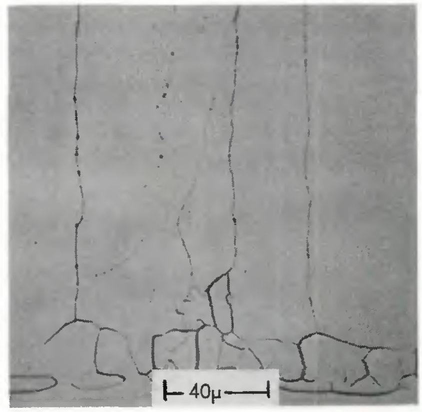

Fig. 6 — Enlarged view of enclosed region of Fig. 5 showing grain boundary porosity found in the heat-affected zone

Table 5 — Bend DBTT for Welded Duplex CVD Tungsten

Heat treat condition	Temperature, deg C, for: Longitudinal	Transverse
As welded	315	430
Outgassed 50 h at 1800 C	315	455
Outgassed + aged 5000 h at 1540 C	<280	315
Outgassed + aged 5000 h at 1700 C	280	330

(Ref. 1), the large grain size of chloride material makes it more susceptible to brittle fracture.

Generally it was found that, considering all material and heat treatments, transverse DBTT was greater than longitudinal DBTT. This is accounted for by the relative ease of weld centerline failures for transverse welds. The inherent relative brittleness of the chloride tungsten was once again evident in the results for transverse welds.

Welded chloride tungsten contained interlayer porosity to a greater extent than was found in the other materials.

Where weldability of CVD tungsten is of primary concern, careful consideration must be given to the brittleness of the chloride-produced form of this material.

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