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FABRICATION AND PROPERTIES OF SEAMLESS TUNGSTEN TUBING

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Fabrication and Properties of Seamless Tungsten Tubing

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SUMMARY

A process for the production of seamless tungsten tubing is described. The process consists of the extrusion of sintered powder sleeves at elevated temperatures to sizes close to the final dimension. The tubing is finished by warm drawing. A size range of .100" diameter to 1-1/2" diameter has been fabricated. A preliminary evaluation of properties of the extruded tubes is given. This work was sponsored by the Atomic Energy Commission.

I. INTRODUCTION

The need for high quality tungsten tubing is appearing in a number of advanced nuclear and space applications.

Wrought tungsten made from powder is presently available commercially in the form of rods, wire and sheet. A government-sponsored program is under way to develop the extrusion of structural shapes. Seamless tungsten tubing is presently obtained by electro-discharge machining from solid bars. Tubing has also been made by vapor deposition. One producer has made tungsten tubing by the cold extrusion of a tungsten slurry followed by a high temperature sintering operation. In addition, seamless tubing is available from one commercial source in the W 26% Re alloy.

The process which will be described here is the hot extrusion of seamless tubing and the subsequent warm drawing of the extruded tubing. The work was aimed at a target size of 3/8" Outside Diameter and .030" Wall, although both smaller and larger tubes were fabricated. Typical tubes are shown in Figure 1. The work was sponsored by the Division of Reactor Development of the Atomic Energy Commission under Contract Number AT (30-1)-2784.

II. EXTRUSION OF SEAMLESS TUNGSTEN TUBING

A. Discussion of Problem Areas

At the inception of this work, it appeared that it would be necessary to extrude the tubing close to the final size and that drawing should be considered only as a finishing operation. This assumption and the inherent properties of tungsten raised a number of problems in the development of a successful extrusion process:

1. Extrusion Temperature

Metals in general are extruded in the plastic range at temperatures above the temperature of recrystallization. In the case of tungsten, this meant an extrusion temperature of between 2700°F and 3600°F. These high temperatures created considerable technical problems.

2. Contamination

Tungsten at elevated temperatures is subject to severe atmospheric contamination. Solutions had to be found for the problem of protecting the metal during heating for extrusion, during the extrusion, and after the extrusion.

3. Stiffness

In order to develop a practical and economical extrusion process, high reductions in area are needed. The high strength of tungsten was expected to create problems with excessive extrusion pressure.

4. Extrusion Mandrels

The conventional method of extrusion for tubing uses a die to define the outside diameter of the tube and a mandrel to define the inside diameter. In the case of high temperature extrusion of tungsten tubing of the target size, it appeared impossible to use the conventional mandrel extrusion technique. The high temperature of the tungsten billet and the high unit pressure combined with the small size of the mandrel and eliminated the possibility of using the conventional mandrel and posed the problem of developing a different method for forming the inside diameter of the tube.

B. Principal Techniques Used in Solution of Problems

It was possible to solve most of the problems discussed above by the application of an extrusion technique developed in earlier work aimed at the hot extrusion of ceramic nuclear fuel elements and sponsored by the Atomic Energy Commission. This is the so-called "filled billet" technique, the principle of which is described in Figure 2. In this technique, the material to be extruded (in this case, a tungsten sleeve) is surrounded on all sides by a filler material. The assembled billet is then extruded and all parts of the billet are reduced equally in cross-sectional area. In order to maintain geometrically similar cross-sections from the extrusion billet to the extruded rod, it is necessary to use "streamlined flow" extrusion techniques, as contrasted with the turbulent flow method used for many conventional metals (Figure 2). In this technique, the outside and center of the billet become the outside and center of the extruded section. Streamlined flow is obtained by the use of a conical entrance die and by good lubrication of the extrusion billet.

The "filled billet" technique provided a solution of the mandrel problem. The extrusion stiffness problem could be alleviated by choosing a sufficiently soft filler material. Atmospheric contamination could be eliminated completely by this method if the filler material surrounding the tungsten sleeve is welded tight. The choice of a relatively soft filler material eliminated one problem of high temperature extrusion - that of chilling the tungsten. Some chilling of the filler material was permissable.

Although the filled billet method eliminated most of the problems which were described earlier, it introduced one new problem - that of the removal of the filler core from the extruded tube. The resistance of tungsten to acid attack permitted various metallic fillers to be removed chemically. The chemical attack on the filler material is slow and makes impractical the removal by etching of cores of more than a few inches in length. It is necessary to obtain a starting hole through the entire length of the inside filler in order to permit circulation of acids and economical removal of the filler. For tubing of 1/2" diameter or larger, such starting holes could be obtained by deep hole drilling. This is a relatively expensive and risky operation and requires very straight extruded tubes. A more practical method was developed during this program. This consists of co-extruding an easily removable material - in this case, graphite - at the center of the filler core. After extrusion, the graphite is poked out by a long drill. Graphite cores as small as .050" in diameter have been removed for lengths up to one foot. For graphite cores over 1/8" diameter, lengths up to ten feet can be removed without too much difficulty.

C. Description of Present Technique

Two distinct techniques were developed in this work. In one case, the extrusion is carried out at 3300°F; in the other case, at 2200°F. The lower temperature technique gives better dimensional control, and is more economical. It is presently used exclusively for the production of commercial quantities of tungsten tubing.

1. High Temperature Extrusion Technique (3300°F)

The assembled extrusion billet is shown in Figure 4. The tungsten starting material is a sintered powder cylinder. Such cylinders of commercial purity powders having a density of better than 90% are available from two commercial sources (General Electric Lamp Metals and Components Department and Wah Chang Smelting and Refining Company). Inside and outside surfaces of the tungsten cylinders should have a smooth machined or ground finish.

The filler material is sintered powder molybdenum. The only specification for the molybdenum components is that they should be vacuum tight. The filler consists of an outer can, an inner core and a graphite center. An end plug is placed at one end of the outer can, and is welded to the can so as to form a vacuum-tight assembly. The assembled billet is heated to 3300°F in an induction furnace, using a graphite inductor under an inert atmosphere. It is then extruded at relatively high extrusion speeds (100"/minute minimum) using a conical die and a glass lubrication technique licensed under the Ugine Sejournet process. Extrusion reductions (cross-sectional area of billet/cross-sectional area of extruded rod) from 5X to 26X have been used. The extrusion constant* for billets containing over 75% of molybdenum filler material at 3300°F is approximately 20 TSI.

After extrusion, the rod is cropped so as to expose the graphite core which is then removed mechanically. The molybdenum is removed by chemical attack in nitric acid, which is circulated through the inside of the tube by the use of a pump.

2. Low Temperature Extrusion Technique (2200°F)

The early investigations of extrusion temperatures had shown that tungsten can be extruded at temperatures as low as 2200°F using a molybdenum filler. At this low temperature it was possible to modify the extrusion technique and to produce tubing having better dimensional tolerances by the use of a low carbon steel filler in conjunction

* Extrusion constant K in $P=K \ln R$ where P is the unit pressure and R is the reduction in area.

with the molybdenum. Figure 5 shows a typical billet used at the lower temperature. The inside of the tungsten sleeve is filled with a sleeve of molybdenum surrounding a graphite core. The tungsten sleeve is surrounded by a relatively thin (1/8" minimum) molybdenum sleeve. This assembly is placed within a heavy low carbon steel can, which is closed by a steel end plug provided with an evacuation tube. The assembly is evacuated and the evacuation tube sealed off.

The billet is heated in a resistance furnace to 2200°F under an inert atmosphere to prevent excessive scaling of the steel can. Extrusion is carried out through conical entrance dies using conventional oil-graphite lubricants.

The amount of permissible reduction in area depends to a large degree on the relative proportion of low carbon steel present. By proper billet design, extrusion ratios as high as 40X can be obtained. The extrusion constant for these extrusions is 15 - 20 TSI. After extrusion, the rods are processed in the same fashion as the high temperature extrusions.

Several attempts have been made to extrude tungsten sleeves using low carbon steel filler material only. At 2200°F, it appears that there is some reaction or penetration of the steel with the tungsten sleeve and with the graphite. The latter can be prevented by wrapping a thin sheet of molybdenum around the graphite in order to prevent contact with the steel. Attempts have been made to lower the extrusion temperature still further. A limited number of extrusions were made at temperatures as low as 1850°F. Although no complete evaluation of tungsten tubes produced at such low temperature has been performed at this time, all indications are that the tubes are sound. Such a process would lead to a considerable reduction in cost by the complete elimination of molybdenum components.

3. Extrusion of Larger and Smaller Tungsten Tubes

Techniques were developed to permit the fabrication of tungsten tubing considerably larger and smaller than the target size.

For tubing having an inside diameter of 3/4" or larger, it is possible to use conventional mandrels to form the inside surface in conjunction with the lower temperature extrusion technique. Figure 6 shows a typical billet for such an extrusion.

Both the inside and outside of the tungsten are surrounded by molybdenum sleeves. The tungsten-molybdenum assembly is then enclosed entirely by a low carbon steel inner and outer can which is provided with an evacuation tube. After extrusion over a mandrel, the central hole permits easy circulation of the acid for removal of the filler material. Extrusion over a mandrel has the additional advantage that the starting tungsten sleeve is easier to produce than sleeves needed for filled billet extrusions with a co-extruded central core. As an illustration, the sleeve needed to produce a .875" outside diameter,

.030" wall tube by filled billet and co-extruded core would be 3.500" outside diameter, .120" wall, while the sleeve needed for a mandrel extrusion would have an outside diameter of 1.320" and a wall of 1/4". The latter sleeve is considerably easier to produce by powder metallurgy methods.

Tubing having diameters below 1/4" are usually produced by a re-extrusion method. In this case, the first extrusion would yield a rod containing the filler material on outside and inside, including the graphite center. This rod is cut into short lengths which are reinserted in a heavy low carbon steel billet. This assembly is re-extruded yielding the small diameter tubing. When the diameter of the final graphite center becomes smaller than 1/8", its removal becomes tedious and difficult for long lengths. Graphite cores as small as .050" have been removed successfully for lengths up to one foot. Further development of better core removal techniques may eliminate this problem.

III. DRAWING OF TUNGSTEN TUBING

Although a technique for drawing of tubing has been developed, it is not as advanced as the technique for extrusion of tubing. Considerably more work will be needed to permit large amounts of reduction in area and in size by drawing. The present technique uses warm drawing, ($800^{o}F$ - $1000^{o}F$) through conventional carbide dies over hardened steel mandrels.

The most difficult problem which had to be solved in order to permit successful drawing was that of providing a point for the tubing, which can pass through the die and through which the draw pull is transmitted. Attempts at swaging the tubing, the use of starting shoulders and other conventional methods were unsuccessful, resulting in splitting of the tubes and slipping of the draw bench jaws on the hard tungsten. The method finally adopted is illustrated in Figure 7. It consists of etching the outer surface of the tungsten to allow about one inch of the tube to pass through the die. A short tungsten sleeve is then brazed to the inside of the reduced section of the tube. The hardened steel mandrel has a long thin section which passes through the sleeve with the mandrel shoulder resting against the brazed sleeve. The drawing jaws grip the threaded part of this reduced section of the mandrel.

The tubes to be drawn are prelubricated on the outside only by a coating of a mixture of graphite suspended in sugar syrup. This is baked on the tube before drawing. The tube with inserted mandrel is heated to $800^{o}F$ - $1000^{o}F$ in a furnace placed directly in front of the drawing die. The die itself is placed in a heated die holder maintained at the drawing temperature. The drawing proceeds when tools and tube have reached temperature. Slow draw speeds (10"/minute) have been used; faster speeds may be possible, but have not been explored.

After drawing, it is possible to remove the tube from the mandrel by applying a light pull. Favorable differential thermal contraction and spring back avoided the need of expanding the tungsten tube before mandrel removal for tubing in the size range of 3/8" to 1/2". For smaller tubes and for longer lengths, (over 3 feet), such an expansion method will probably have to be developed.

The starting material for the more successful drawing operation was tubing extruded by the lower temperature (2200°F) method and given a recrystallization treatment at 2600°F. Tubing extruded by the high temperature method was considerably out of round and broke invariably in the first rounding draw, where the mandrel gave relatively little support.

Draws giving a reduction in area up to 15% per draw and total reductions of up to 35% between anneals have been performed, although it is believed that these values do not represent actual limits of the process. After drawing, the tubing has an excellent finish and very close tolerances on outside diameter and inside diameter.

IV. TUBE EVALUATION

The main purpose of the evaluation program was to guide the fabrication development. Evaluation of properties of interest to potential users is presently underway by the users and will not be reported here. The evaluation program therefore was limited mainly to the "as-extruded" tubing in an effort to determine optimum conditions for further drawing. The ductility of the extruded tubing and the brittle-to-ductile transition temperatures were determined by ring-squash tests and by bend tests. The chemistry of extruded tubing was compared to that of the starting sleeves. In addition, density measurements were obtained as well as dye penetrant and vacuum tightness data for presence of cracks or porosity. Outside diameter, inside diameter and wall dimensions were measured. All phases of fabrication were followed by metallography.

A. Ductility

1. Ring Squash Tests

In order to determine the optimum extrusion conditions and heat treatments for drawability and possible drawing temperatures, short rings were cut from extruded tubes. These were squashed in a direction perpendicular to the tube axis at various temperatures.

Figure 8 shows the results of these simple tests for tubing extruded at 2200°F. Tests were carried out at temperatures between 700°F and 1000°F and most of the test data fell within the shaded band of the graph.

Some earlier tests carried out on material extruded at 3300°F had shown that this material exhibits ductility similar to that of 2200°F extrusions after a 2600°F anneal. Further evaluation of this material was not carried out because of difficulties in drawing high temperature extrusions.

From this data it was determined that 2500°F - 2600°F anneal would be necessary for further drawing of the 2200°F extruded material. It was also concluded that the drawing temperature is not critical within the range of squash test temperature. All of the material tested showed negligible ductility at room temperature.

2. Bend Tests

A limited number of bend tests was carried out to determine the brittle-to-ductile transition temperature for extruded tungsten tubing. The specimens were strips cut longitudinally from the tubing. Three types of tubes were tested: 2200°F as-extruded tubes; 2200°F extruded and annealed at 2600°F tubes; and 3300°F as-extruded tubes. The data (Figure 9) indicates the lowest transition temperature of approximately 130°F for the 2200°F as-extruded tubing and the highest transition temperature of approximately 550°F for the 3300°F as-extruded tubing.

B. Density and Leak Tightness

Density measurements performed on material extruded at 3300°F and 2200°F showed theoretical density in all cases. A large number of tubes were tested by helium leak tests and found to be tight at room temperature. Metallography confirmed the absence of cracks and voids. Dye penetrant tests showed that all tubing produced during the later phases of this work were free of surface cracks or pin-holes.

C. Chemistry

Analysis of material extruded both at high and low temperatures showed that no significant changes in the chemistry occur during the extrusion by the filled billet method.

D. Metallography

Some of the more important conclusions of the extensive metallography carried out during this work will be given here.

1. Effect of Extrusion Temperature

Extrusions carried out at 3300°F show a completely recrystallized fine grained structure with a grain size of 10 ASTM (Figure 10). Extrusions carried out at 2200°F show a heavily worked structure with very little, if any, evidence of recrystallization (Figure 11).

2. Effect of Annealing

When annealing low temperature extruded tubing, no visible changes occur below 2200°F. At 2200°F and above, partial recrystallization is observed. Almost complete recrystallization is achieved at 2900°F for one hour.

3. Effect of Reduction Ratio

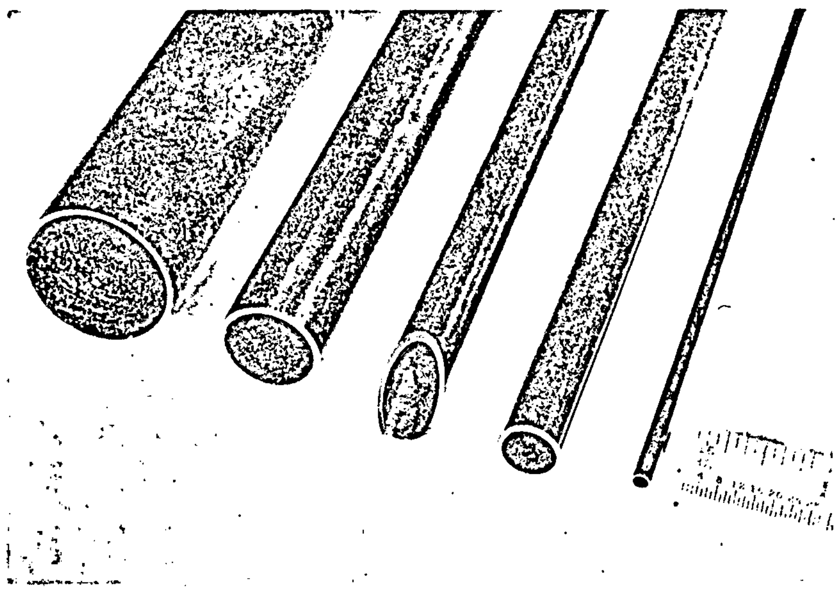
The elongated cold worked structure obtained by an extrusion at 2200°F has been shown in Figure 11. Re-extrusion of such tubing gives further refinement to this structure. After warm drawing, the structure is fibrous and similar to that seen in highly worked rolled sheet.

V. CONCLUSION

In this work, a practical method for the extrusion of seamless unalloyed tungsten tubing has been developed. Further work will be needed to optimize the variables and to make the process economically more attractive.

Preliminary experiments have shown that extruded tungsten tubing can be warm drawn to close tolerances and high surface finishes. The limits of the drawing process have not yet been established.

The combination of extrusion and drawing makes available to industry high quality seamless tungsten tubing in a size range from .100" upward.



RF 9050

Fig. 1 - Typical Extruded Tungsten Tubing

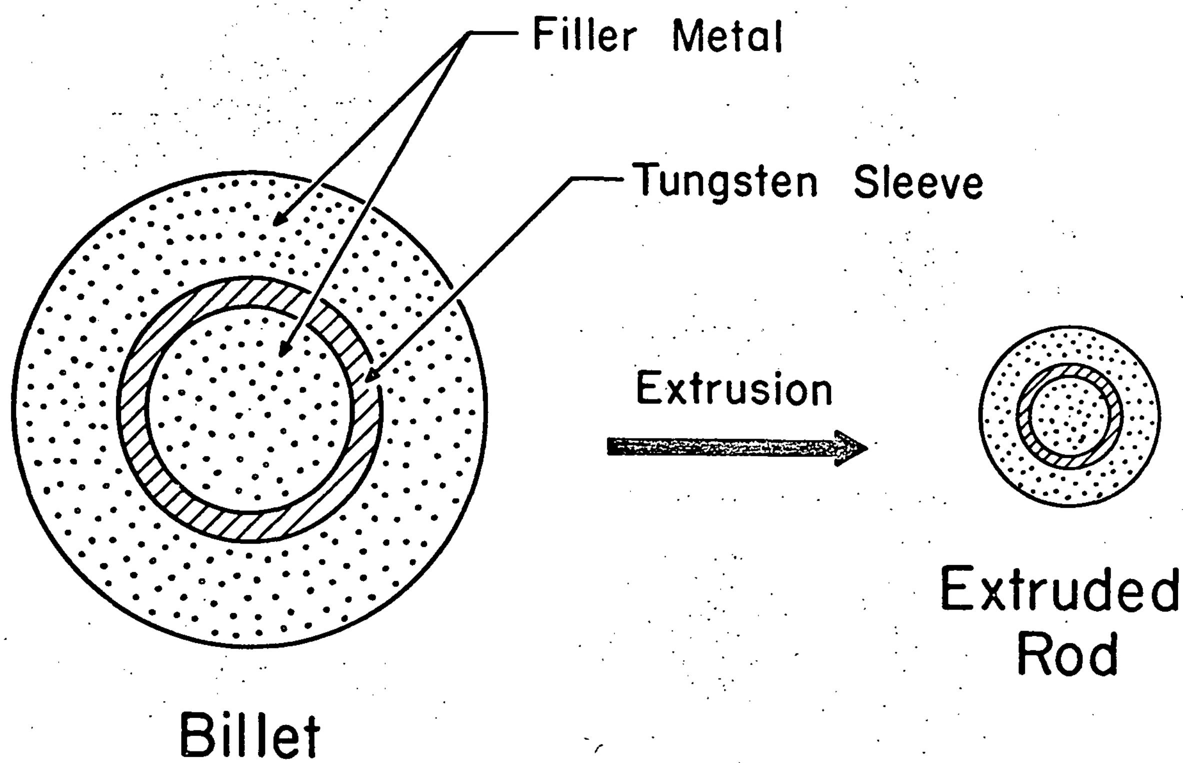
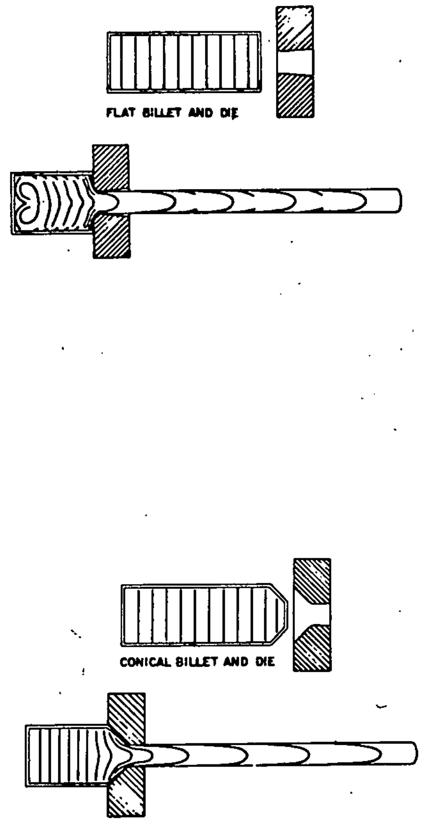


Fig. 2 - Filled Billet Method of Extrusion



RF 1532

Fig. 3 - Turbulent Flow (Top)
Streamlined Flow (Bottom)

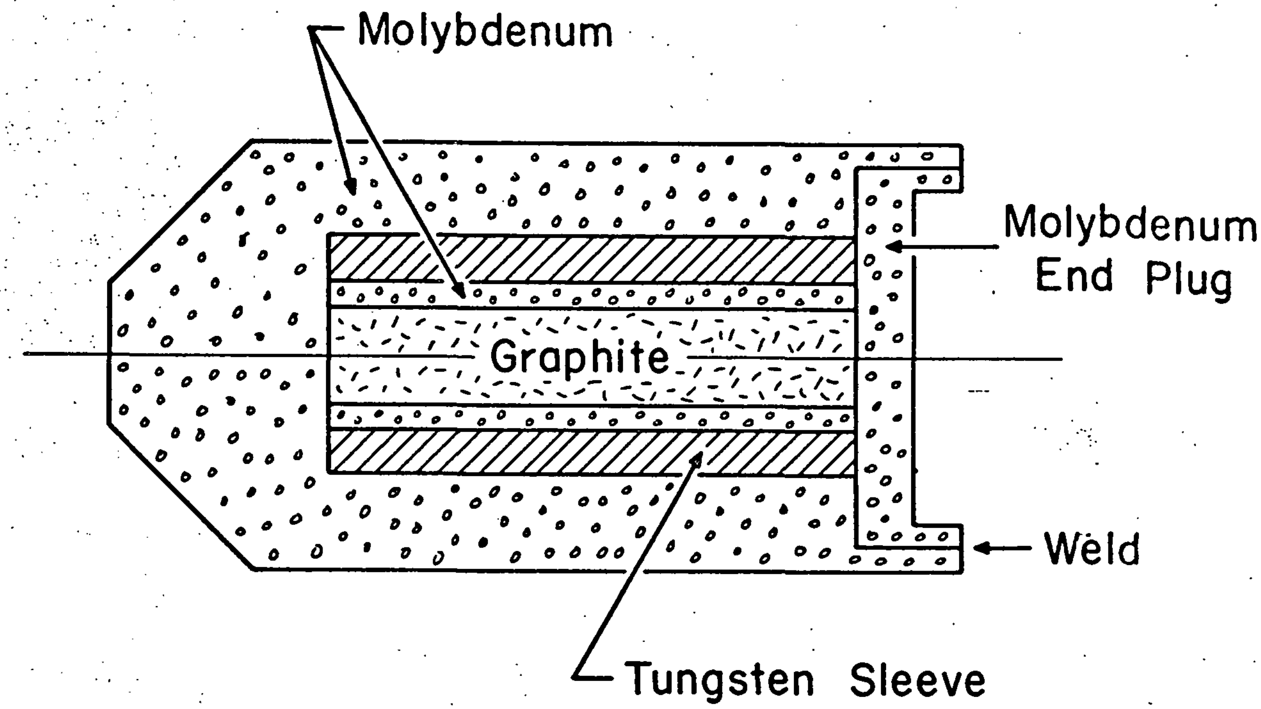


Fig. 4 - Billet for 3300°F Extrusion of Tungsten Tubes

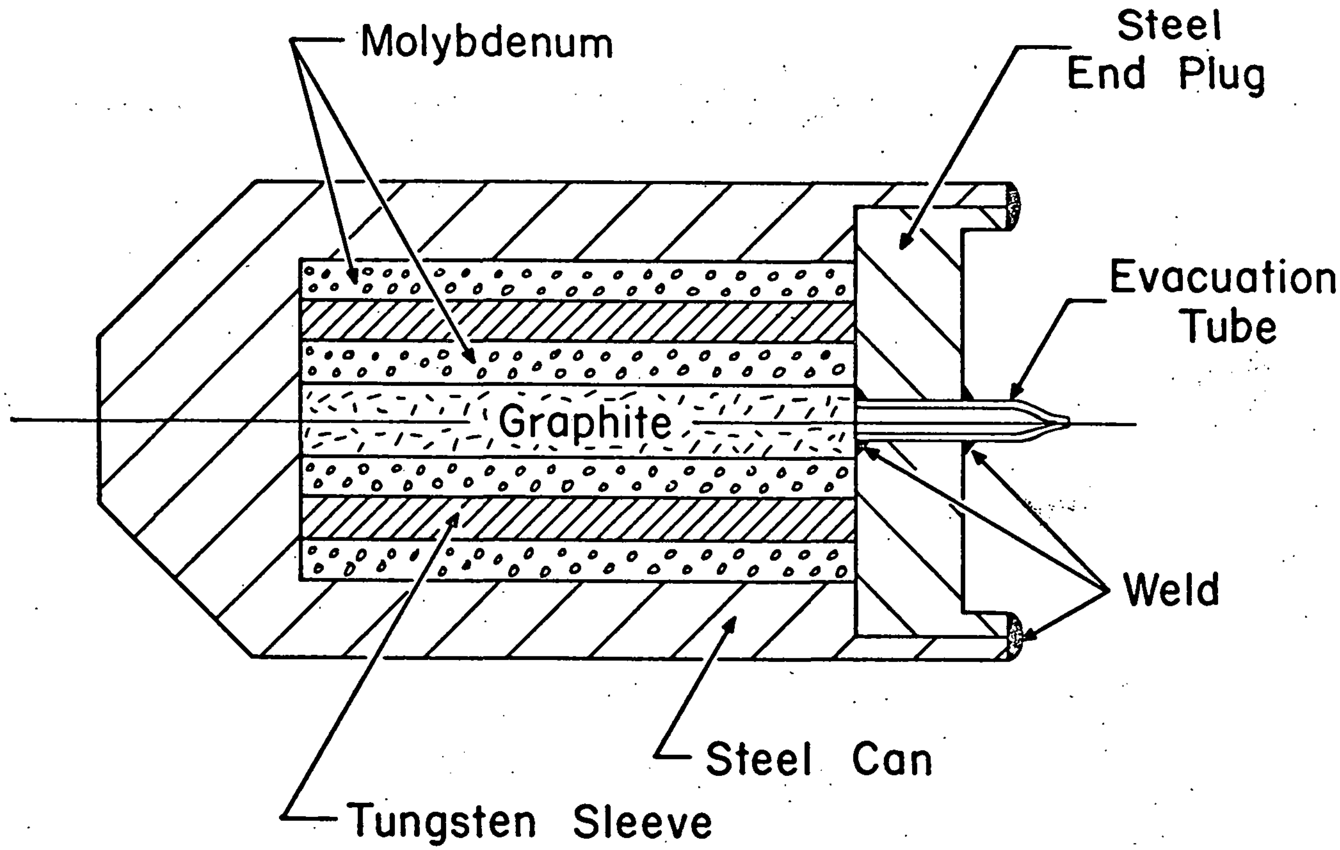


Fig. 5 - Billet for 2200°F Extrusion of Tungsten Tubes

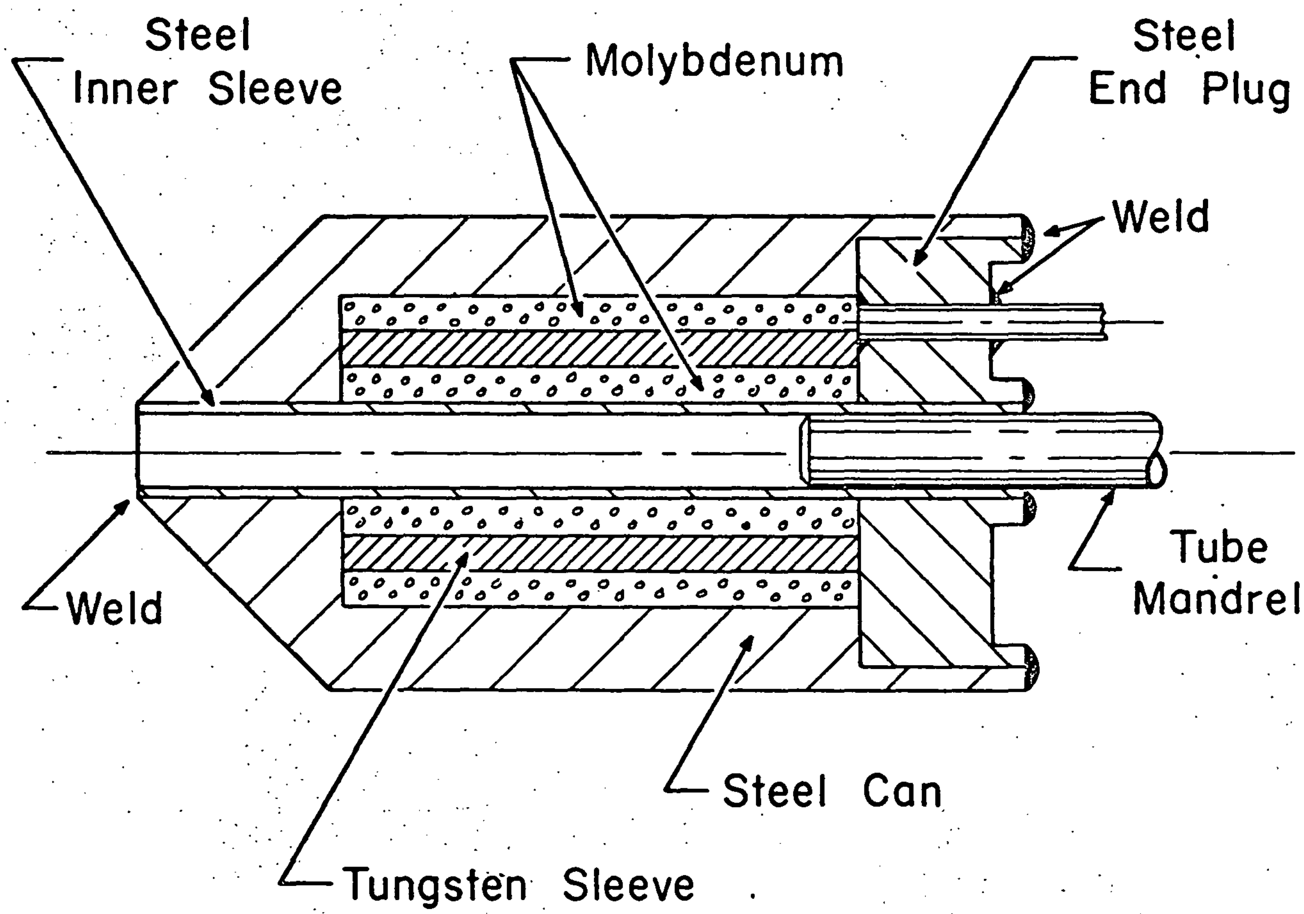


Fig. 6 - Billet for 2200°F Extrusion of Large Tungsten Tubes Using a Mandrel

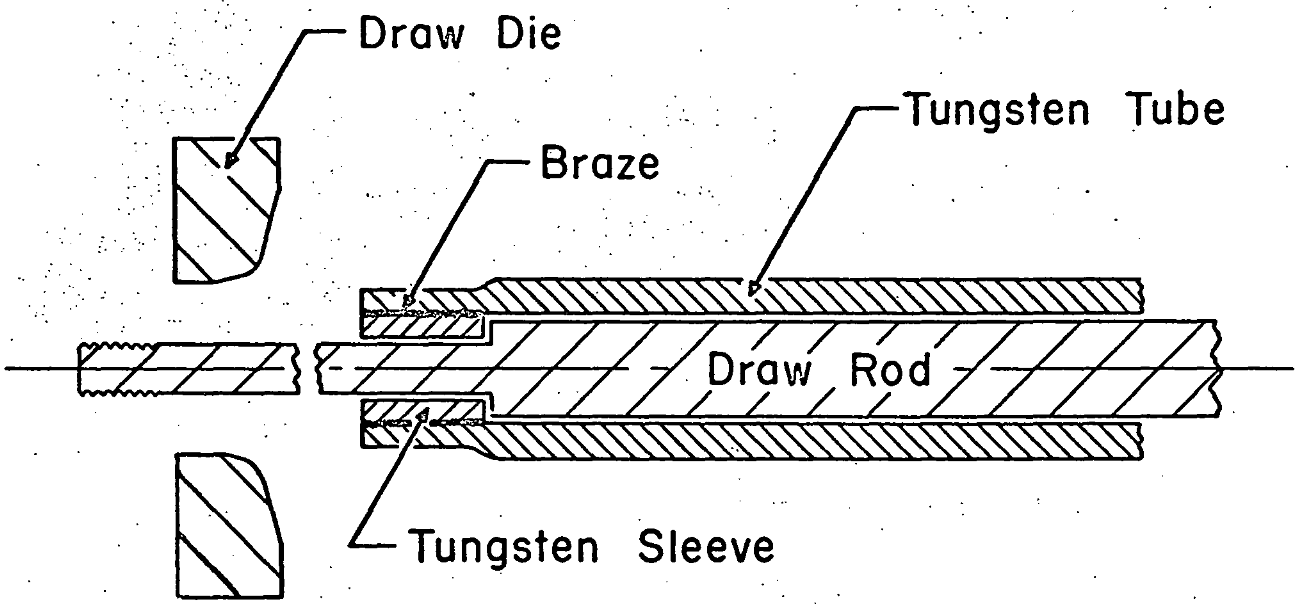


Fig. 7 - Technique Used for Drawing of Tungsten Tubing

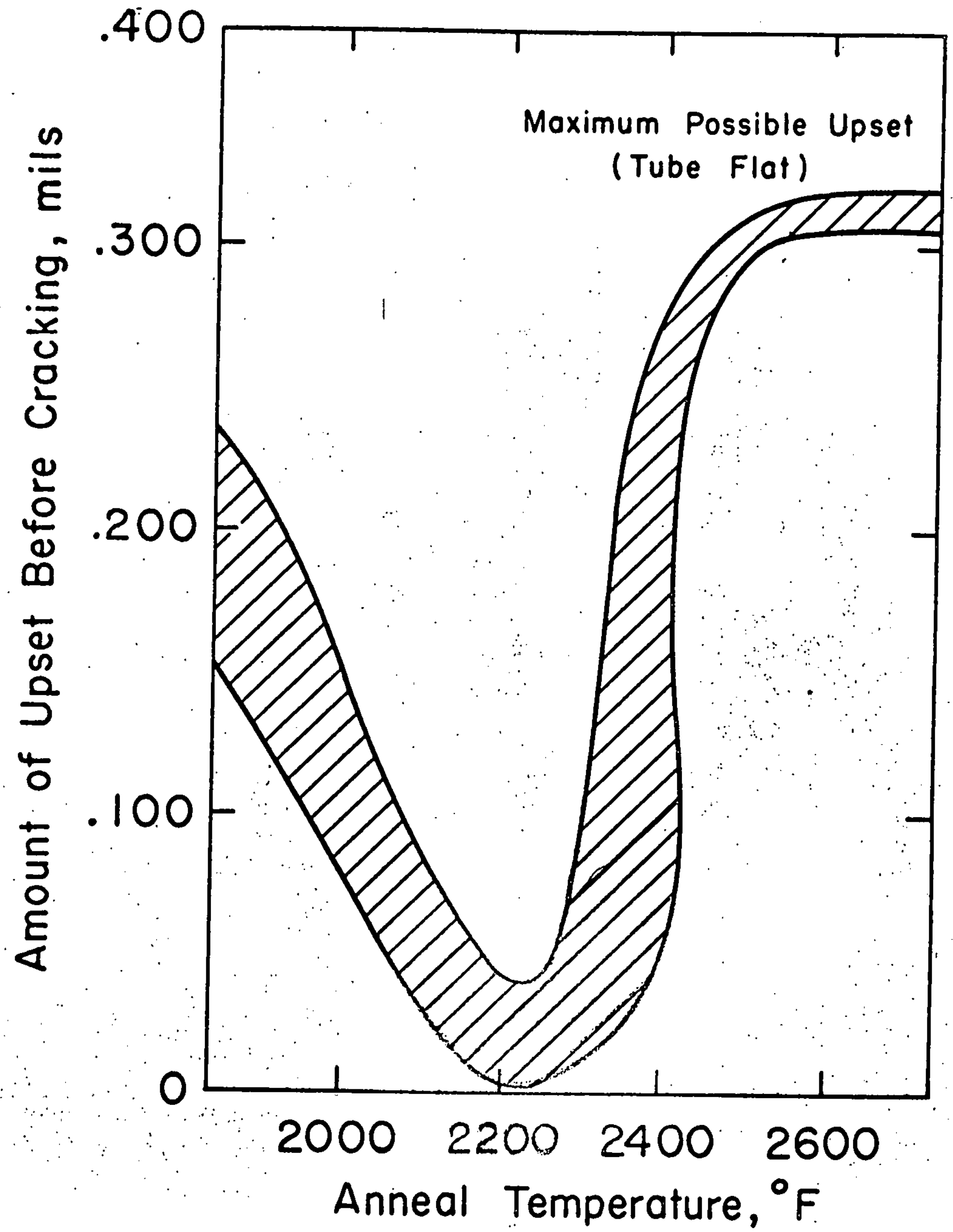


Fig. 8 - Upset before cracking of tungsten rings (0.440 inch OD, 0.035 inch wall) vs. temperatures of annealing for one hour. Tungsten tubes extruded at 2200°F. Test temperature 700°-1000°F.

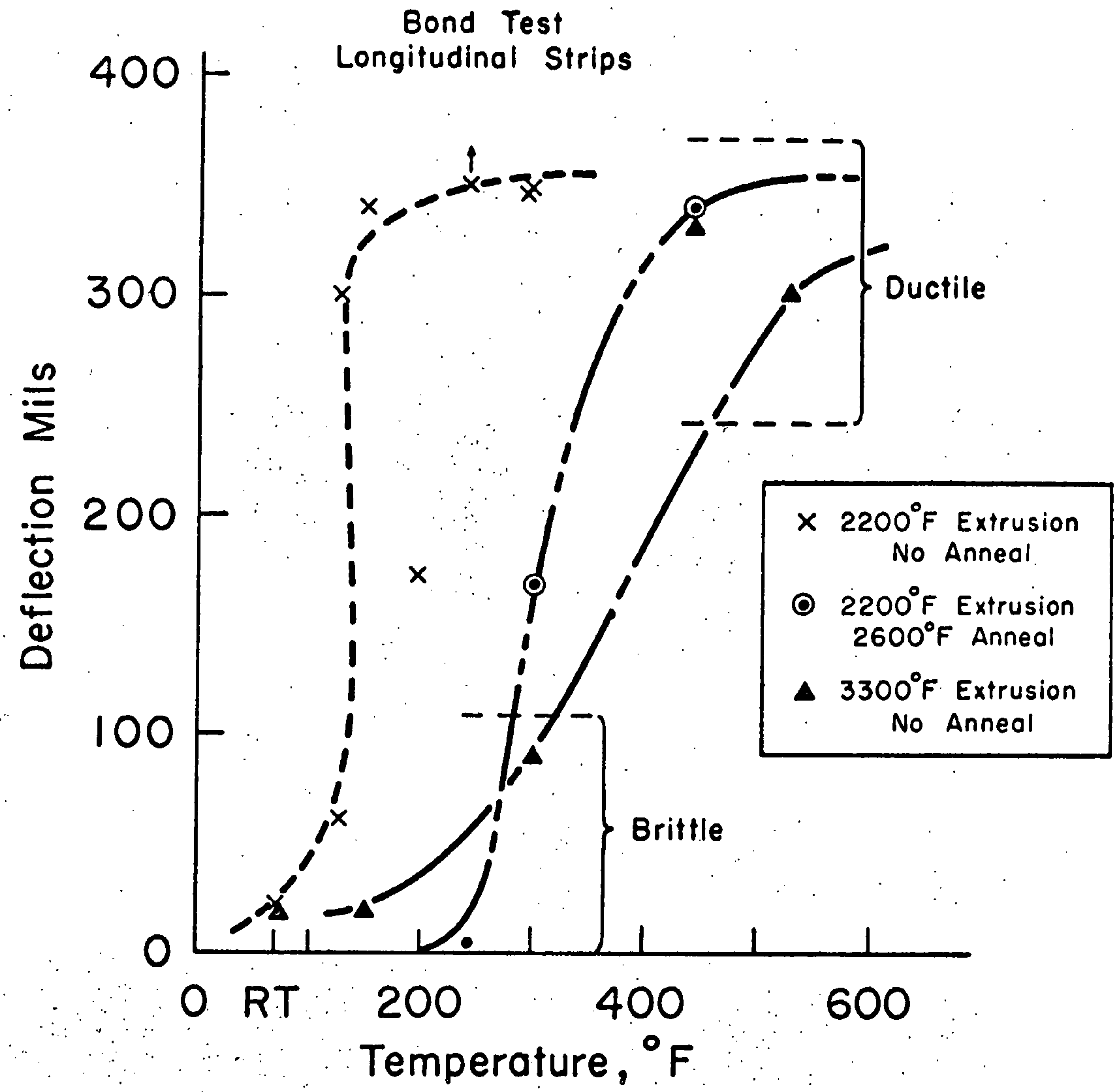
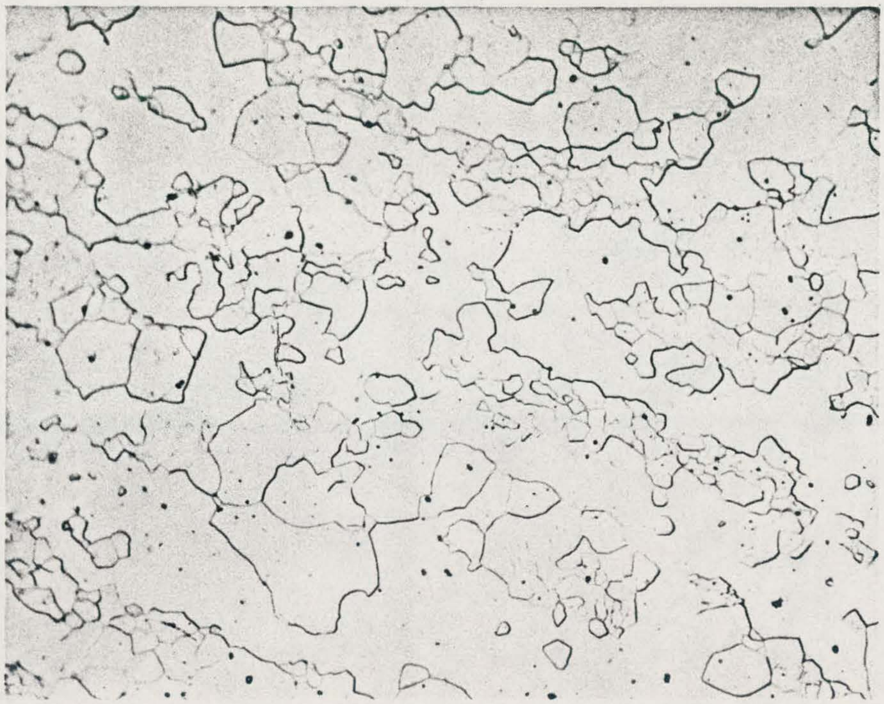


Fig. 9 - Bend test results, tungsten tubing, longitudinal strips 2 x 0.125 x 0.045 inch, plotted for specimens in three different conditions.

2200°F as-extruded; 2200°F extruded, 2600°F anneal; 3300°F as-extruded



500X Bt Lt A-4215-5C

Fig. 10 - Photomicrograph of longitudinal section of tungsten tube extruded at 3300°F 18x5X Reduction

500X Bt Lt A4309-1f

Fig. 11 - Photomicrograph of longitudinal section of tungsten tube extruded at 2200^{o}F 18x5X Reduction