THERMAL-EXPANSION METHOD FOR LINING TANTALUM ALLOY TUBING WITH TUNGSTEN

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A differential-thermal-expansion method was developed to line T-111 (tantalum - 8 percent tungsten - 2 percent hafnium) tubing with a tungsten diffusion barrier as part of a fuel-element fabrication study for a space-power nuclear-reactor concept. This method uses a steel mandrel, which has a larger thermal expansion than T-111, to force the tungsten against the inside of the T-111 tube. Variables investigated include lining temperature and decreasing gap size. Liner integrity increased with increasing lining temperature and decreasing gap size. The method should have more general applicability where cylinders must be lined with a thin layer of a second material.
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THERMAL-EXPANSION METHOD FOR LINING TANTALUM ALLOY TUBING WITH TUNGSTEN

by Gordon K. Watson, John D. Whittenberger, and William F. Mattson

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SUMMARY

A differential thermal-expansion method was developed to line T-111 (a tantalum alloy containing 8 percent tungsten and 2 percent hafnium) tubing with tungsten as part of a fuel-element fabrication study for a space-power nuclear-reactor concept. The tungsten liner is necessary to prevent reactions between the T-111 fuel-element cladding and the uranium mononitride fuel. The differential-thermal-expansion liner method uses the difference in the thermal expansions of two different materials to press the liner against the inside of the tube. The tungsten liner, in the form of multiple layers of thin foil, is wrapped around an alumina-coated steel mandrel which is inserted into the T-111 tube. The alumina coating is required to prevent the tungsten from welding to the mandrel.

During high-temperature exposure, the steel mandrel expands more than the T-111 and forces the liner against the T-111 tube. If the time, temperature, and pressure conditions are sufficient, solid-state welding can occur between the liner and the tube.

The variables investigated in this study included the temperature at which the liner was applied, initial assembly gap size, and tube length. The T-111 tubes, 1.91 centimeter (0.75 in.) in diameter, were lined at 1200°C (2200°F) and 1315°C (2400°F). Gap sizes ranged from 0.008 to 0.020 centimeter (0.003 to 0.008 in.). The integrity of the liners tended to increase as the temperature for lining increased and the assembly gap size decreased. Other test results showed that tubes with large length-to-diameter ratios can become barrel-shaped during lining, but this problem can be minimized by control of lining temperature and gap size.

Although this method was developed specifically to line nuclear fuel elements, the technique has general applicability where a cylindrical shape must be lined with a thin layer of a second material.
INTRODUCTION

A concept for a fast-spectrum, lithium-cooled nuclear reactor for space power applications was investigated at the NASA Lewis Research Center (ref. 1). This reactor is designed to operate at 980°C (1800°F) for up to 50,000 hours. The reactor concept is based on the use of uranium mononitride (UN) as the fuel and a tantalum alloy, T-111 (tantalum - 8 weight percent tungsten - 2 weight percent hafnium), as the primary cladding material. The cylindrical fuel pins for the conceptual reactor are about 43 centimeters (17 in.) long and 1.9 centimeters (0.75 in.) in diameter, and the T-111 cladding has a wall thickness of about 0.15 centimeter (0.058 in.).

At the proposed reactor operating temperatures, chemical compatibility problems have been observed when UN is in direct contact with tantalum alloys (ref. 2). Thus, the T-111 cladding and the UN fuel must be separated by a barrier layer to prevent contact and possible reactions. Tungsten was selected as the material for the barrier layer because of its compatibility with UN (ref. 2). The tungsten liner for the proposed fuel elements is to be 0.013 centimeter (0.005 in.) thick. On the basis of diffusion-rate calculations, this thickness of tungsten is considered sufficient to ensure the compatibility of the fuel and cladding for the desired 500,000-hour reactor life (ref. 3). Although the tungsten liner must remain in position to prevent contact between the UN and the T-111, it need not be attached or welded to the T-111 cladding.

Various methods were investigated for applying the tungsten liner to the T-111 cladding (unpublished work at Lewis Research Center). Initial attempts to line T-111 tubing with tungsten by chemical vapor deposition indicated that contamination of the T-111 and accurate control of the liner thickness would be major problems. Several other methods were investigated in which the required 0.013-centimeter (0.005-in.) thickness of the tungsten liner was produced with the use of multiple wraps of 0.0025-centimeter (0.001-in.) thick tungsten foil. One method involved hot isostatic pressing of the tungsten foil around a molybdenum mandrel to produce thin-wall tungsten tubing that could be inserted as a loose liner in the T-111 cladding. Hot isostatic pressing of tungsten foil directly onto the T-111 also was investigated. Another liner method studied (ref. 4) used the creep of an internally pressurized thin-wall tube to press the tungsten foil against the T-111 cladding.

The most promising method investigated for lining the T-111 cladding was a differential-thermal-expansion technique. This method uses a mandrel of a material that has a larger thermal expansion coefficient than T-111 to force the tungsten foil against the T-111 tubing when heated. The development of the differential-thermal-expansion technique and the experimental results are described in this report. The initial experiments were conducted on relatively short lengths of T-111 tubing to assess the usefulness of the technique. Longer tubes then were lined to determine what effects tube length would have on the liner technique.
DIFFERENTIAL-THERMAL-EXPANSION TECHNIQUE

General Description

The differential-thermal-expansion technique for lining a tube uses the difference in the thermal expansions of two different materials to press a liner against the inside of a tube. If the time, temperature, and pressure conditions are sufficient, solid-state welding can occur between the liner and the tube. A schematic of the thermal expansion process is shown in figure 1. In addition to the furnace required for heating the assembly, only three components are required: the tube to be lined, the liner in the form of multiple wraps of thin foil, and a mandrel having a larger thermal expansion coefficient than the tube. In general, the mandrel must be coated with some type of barrier such as alumina (Al₂O₃) to keep the foil from welding to the mandrel. During heating, the mandrel expands at a more rapid rate than the tubing, closes the assembly gap, and forces the liner into intimate contact with the tube so that diffusion welding can occur.

Since the total thermal expansions of the mandrel and the tubing are functions of their respective diameters, the diameter of the tubing to be lined has a considerable effect on the usefulness of the thermal expansion technique. Thus, the technique developed for lining 1.9-centimeter-(0.75-in.)-diameter tubing would probably not be successful for lining tubing 0.6 centimeter (0.25 in.) in diameter, because the minimum gap required for assembly cannot be closed. A more complete description of the relation between assembly gap size, thermal expansivity, and diameter is presented in appendix A.

Mandrel Selection

The selection of the proper mandrel material is one of the most important considerations for the differential thermal expansion method. The mandrel material used for lining T-111 with tungsten must have a larger thermal expansion coefficient than T-111. In addition, the maximum use temperature of the mandrel material should be high in order to promote diffusion welding between the tungsten foil wraps and between the tungsten foil and the T-111. Also, materials that are thought to cause embrittlement of the T-111, such as copper (ref. 5), must be avoided. Based on these considerations, a low-carbon steel was selected as the mandrel material. Low-carbon steel has the additional advantages of being low cost, readily available, and easily machined to close tolerances. Welding of the tungsten foil to the steel mandrel was prevented by plasma spraying the mandrel with alumina. Thermal expansion equations used for the mandrel material and for the T-111 are included in appendix A.
INITIAL STUDIES

The initial investigation of the differential-thermal-expansion liner technique was conducted on short (3.8 cm (1.5 in.)) lengths of commercially produced T-111 tubing. These initial studies were designed to evaluate the quality of the tungsten liner as a function of lining temperature and initial assembly gap size. Two lining temperatures, 1200° C (2200° F) and 1315° C (2400° F), were investigated. Temperatures were limited to 1315° C (2400° F) and below to avoid the possibility of overheating and melting the mandrel. Temperatures much below 1200° C (2200° F) were thought to be inadequate for diffusion welding. The room-temperature assembly gap size was varied from 0.008 centimeter (0.003 in.) to 0.020 centimeter (0.008 in.).

Procedure

Every effort was made during each step of the lining process to minimize contamination of the T-111 in order to prevent the properties of T-111 from being impaired by interstitial contaminants. The T-111 tubing samples, 3.8 centimeters (1.5 in.) long by 1.91 centimeters (0.75 in.) outside diameter by 0.10 centimeter (0.040 in.) wall thickness, were chemically cleaned by the method described in reference 6. Then the tubes were heated in a high-vacuum (1.3×10^-3 N/m² (1×10^-5 torr) or better) furnace at 1090° C (2000° F) for 1 hour to remove residual volatile impurities.

The mandrels for this study were produced by centerless-grinding 5.0-centimeter-(2.0-in.-) long pieces of AISI 1020 steel rod to the desired diameter. After grinding, the mandrels were plasma sprayed with alumina (Al₂O₃) to produce a coating about 0.013 centimeter (0.005 in.) thick. Finally, the mandrels were centerless-ground again to the final diameter. The thickness of the Al₂O₃ coating after grinding was about 0.008 centimeter (0.003 in.).

Tungsten foil, 0.0025-centimeter-(0.001-in.-) thick, was sheared into strips 3.8 centimeters (1.5 in.) wide by 26.2 centimeters (10.3 in.) long. The length of the foil was about 0.15 centimeter (0.060 in.) less than that needed for five full wraps around the inside of the T-111 tubing to prevent overlapping of the foil ends. The tungsten strips then were rinsed successively in water, acetone, and methyl alcohol to remove dirt and grease and were finally hot-air dried.

All of the cleaned components were handled and assembled with the use of nylon gloves to minimize contamination. The tungsten foil strip was wrapped tightly around the mandrel, and the foil-mandrel combination was inserted into the T-111 tubing. The minimum assembly gap size for easy assembly was about 0.008 centimeter (0.003 in.) for the tubing sizes of interest in this study. The outside of the T-111 tube was wrapped with tantalum foil to minimize contamination of the T-111 during subsequent heating. The
entire assembly then was placed upright in a vacuum furnace and was heated for 1 hour at 1200\(^\circ\) C (2200\(^\circ\) F) or 1315\(^\circ\) C (2400\(^\circ\) F) in a vacuum of 1.3\times10^{-3} \text{ newtons per square meter (1x10^{-5} torr)} or better. Upon cooling, the mandrels were easily removed from the tungsten-lined T-111 tubing.

Samples of the lined tubes were examined metallographically. In addition, both surface and bulk chemical analyses were conducted on selected samples of the lined tubing.

Results and Discussion

General comments. - The alumina coating on the mandrels was successful in preventing the liner from welding to the mandrel. In all cases, the mandrels were removed easily after the lining operation.

The first few T-111 tubes lined at the start of this study were not protected with the tantalum foil wrap. After lining, a whitish metallic deposit was observed on the outside surfaces of these tubes. This deposit was probably either iron or an iron compound and was the result of vaporization from the ends of the mandrel, which were not coated with alumina. Wrapping the outside of the T-111 tubing with tantalum foil prevented these surface deposits from forming in subsequent trials. Later in the program, to further reduce the possibility of contamination from the mandrels, the ends of the mandrels, in addition to the cylindrical surfaces, were coated with alumina.

For all gap sizes and bonding temperatures, the tungsten liners appeared to be crack-free, as determined by visual examination. In all cases, the individual wraps of tungsten foil appeared to adhere well to each adjacent wrap and to the T-111 tubing. The foil liners remained intact even when the tubes were cut with a water-cooled cutoff saw.

Several of the lined tubes were flattened to determine the ductility of the lined tubing. The T-111 tubing could be completely flattened at room temperature with no evidence of any cracking. As expected, however, the relatively brittle tungsten liners cracked and spalled during flattening of the T-111.

Dimensional changes. - The dimensional changes in the alumina-coated steel mandrels and in the T-111 tubing as a result of the lining operation are listed in table I as a function of gap size for both lining temperatures. Included in table I is the calculated gap at temperature for each specimen. For these calculations, the thermal expansion of the tungsten liner was neglected. A negative gap indicates that the initial gap was closed and that, at temperature, the tungsten foil was being forced against the T-111. During lining, some of the force on the liner was relieved by plastic deformation of both the mandrel and the T-111. The dimensional changes listed in table I show that deformation of the mandrels was apparently the main stress-relieving process. Of secondary importance was the slight increase observed in the diameter of the T-111 tubing.
The amount of mandrel deformation increased with decreasing initial gap size. For the three lining runs calculated to have negative gaps at 1315°C (2400°F), the mandrel diameters measured after lining were within 0.0003 centimeter (0.0001 in.) of one another. These results indicate that the relatively strong T-111 tubing restrained the radial thermal expansion of the mandrels. The final diameters of the three mandrels were about the same, regardless of the initial mandrel diameter. Once the mandrel was restrained by the T-111, any additional volumetric expansion during heating must have occurred by axial extrusion of the mandrel.

**Metallography.** - All of the lined T-111 tubes were sectioned and examined metallographically. Typical cross sections of the lined tubes are shown in figure 2 for the tubes lined at 1200°C (2200°F) and in figure 3 for the tubes lined at 1315°C (2400°F). (A photomicrograph of the sample which was lined at 1200°C (2200°F) and had a gap of 0.020 centimeter (0.008 in.) is not included in figure 2 because the relatively large spacings between foil layers caused excessive staining of the metallographic sample during etching.)

It can be seen in figures 2 and 3 that as the assembly gap was decreased and the lining temperature was increased, the tungsten foil appears to be pressed more tightly against the T-111. In some cases, grain growth can be seen across the tungsten-foil interfaces. As the lining temperature increased, the amount of recrystallization observed in the tungsten tended to increase. The amount of grain growth, however, was somewhat inconsistent. No contamination of the T-111 tubing was detected metallographically.

**Chemical analysis.** - Samples of as-received T-111, as-received tungsten foil, and lined T-111 tubing were analyzed to determine if the thermal-expansion liner process causes any contamination of the lined tubing. The elements of particular concern were iron, aluminum, and interstitials (carbon, nitrogen, oxygen, and hydrogen) that might be picked up from the mandrel during the lining process. The bulk analyses for the as-received materials and for selected samples of the tungsten-lined T-111 tubing are listed in table II. Some of the lined tubes were analyzed as composites of tungsten and T-111. In other cases, the tungsten liner and the T-111 tubing were analyzed separately.

The test results show that very little, if any, interstitial or aluminum pickup occurred during the lining operation. The analytical results also showed the importance of a protective foil wrap around the T-111 tubing. The unprotected tubing picked up a large amount of iron, probably from the uncoated ends of the mandrels, whereas the iron content of the protected tubing was essentially unchanged from that of the starting material.

In addition to the bulk analyses, the surface compositions of several samples were analyzed by X-ray fluorescence and emission-spectroscopy methods. For the emission-spectroscopy analysis, the surface to be analyzed was etched with a solution of nitric acid and hydrochloric acid. Then, the etching solution was drawn off and analyzed.
The surface analyses of the outside of the lined T-111 tubing samples confirmed that the tantalum foil wrap protected the T-111 from iron contamination. With X-ray fluorescence techniques, the surface analyses of the unprotected tubing showed up to about 50 weight percent iron. No iron was detected on the surfaces of the protected tubes. Analyses of the inside surfaces of the tungsten-lined tubing showed trace amounts of iron contamination. This contamination probably resulted from small cracks formed in the alumina coating of the steel mandrels during heating. Further discussion of this problem is presented in appendix B.

LINING FULL-LENGTH TUBES

Once the feasibility of the differential-thermal-expansion process was demonstrated for lining short lengths of tubing, T-111 tubes of the size required for full-length fuel elements were lined to determine if the process was applicable to longer tubes.

Procedure

The general procedure used for lining the full-size (43-cm (17-in.) long) T-111 tubes with tungsten was basically the same as that used for the initial lining studies on the shorter tubes. The longer tube length, however, made it necessary to change the methods used for component assembly.

To minimize the problems associated with wrapping the relatively large sheets of tungsten foil around the mandrels, the foil was preformed to a cylindrical shape. After the foil was chemically cleaned, it was formed into a cylinder and inserted into a T-111 tube having approximately the same inside diameter as the tube to be lined. The assembly then was heated for 1 hour at 870°C (1600°F) in a vacuum of 1.3×10⁻³ newt·m² per square meter (1×10⁻⁵ torr) or better, which caused the tungsten foil to take a permanent set in a cylindrical shape. Preforming the tungsten foil at temperatures above 870°C (1600°F) resulted in some recrystallization of the tungsten. Because of the associated brittleness, this caused problems during subsequent handling operations. Thus, higher preforming temperatures were avoided.

As preformed, the innermost foil wrap was not circular in shape and interfered with insertion of the close-fitting mandrel. In contrast, the outer wrap was relatively well formed. Therefore, to make the inner wrap more circular, the preformed tungsten foil cylinders were rerolled to reverse the inner and outer foil wraps.

All of the components were cleaned as described in the "INITIAL STUDIES" section of this report. The liner was inserted into the T-111 tube and then was twisted to spiral the liner out against the tube wall. The ends of the liner were squared with the ends of
the T-111 tube, and the alumina-coated mandrel was inserted. Finally, the assembly was wrapped in cleaned tantalum foil and placed vertically in a vacuum furnace having a 60-centimeter- (24-inch-) long hot zone. Temperature uniformity over the length of the assembly was ±5° C (±9° F).

Two full-length, tungsten-lined T-111 tubes were fabricated. The initial assembly gap for both tests was about 0.013 centimeter (0.005 in.). The first tube was lined at 1315° C (2400° F). After evaluation of this tube, the second tube was lined at 1315° C (2400° F).

Results and Discussion

No problems were encountered in the assembly of either full-length tube. Preforming the tungsten liners prior to assembly allowed insertion of the mandrels with a minimum of difficulty. The mandrels were removed easily from both tubes after lining. In both cases, the tungsten liners were well formed, with the individual wraps of foil adhering to each other and to the T-111. One of the lined tubes is shown in figure 4.

After the first full-length tube was lined at 1315° C (2400° F), its diameter was measured every 2.5 centimeter (1.0 in.) along the entire length of the tube. These measurements are compared with measurements of the same tube before lining in figure 5. The results show that the tube deformed nonuniformly during the lining process. The tube diameter near the ends was essentially unchanged; whereas, the diameter near the center of the tube increased about 0.003 centimeter (0.0012 in.) on the average. The mandrel deformed in a similar manner. The diameter of the mandrel was unchanged at the ends and increased about 0.003 centimeter (0.0012 in.) near the center.

The initial lining studies on shorter tubes gave no indication of nonuniform deformation. The reason for the nonuniformity in the full-length tubes may have been that their greater length restricted axial movement of the mandrel except near the tube ends, thereby effectively locking the mandrel in place near the center. Once the mandrel was locked in place, any further expansion took place only in the radial direction.

The lining conditions were changed for the second tube in order to reduce the T-111 deformation. The initial mandrel gap was the same as for the first tube, but the lining temperature was reduced to 1200° C (2200° F). Our thermal expansion calculations indicated that at this lower temperature, the expansion of the mandrel would lack about 0.0013 centimeter (0.0005 in.) of closing the assembly gap; thus, no force would be applied to the T-111 tubing.

The diameter of the second tube also was measured before and after lining. The results of these measurements are presented in figure 6. The tube measurements show a slight increase in tube diameter near the center of the tube as a result of the lining operation, but no diameter change was observed at the tube ends. The nonuniform
deformation observed in this tube, however, was within the diameter tolerance
(±0.003 cm (±0.001 in.)) specified in the fuel-element design. The observed deformation
of both the mandrel and the T-111 tubing indicated that the mandrel closed the initial as-
sembly gap during the lining operation even though the thermal expansion calculations
showed that, at temperature, the gap would not be closed. Apparently, the accuracy of
the data and assumptions used in the thermal expansion calculations were not sufficient
to predict the exact behavior of the mandrel and tubing. The calculations are sufficient,
however, to determine the approximate lining parameters. In practice, the gap size and
lining temperature can be varied slightly until the correct parameters are found to line
tubes with a minimum amount of deformation. Minimizing the deformation by increasing
the gap size or decreasing the lining temperature, however, can have a tendency to re-
duce the diffusion welding that occurs between foil wraps. For our application, it was
not a requirement that the liner be welded to the tube. All that was required was that the
liner remain in position and in contact with the tubing. For applications where diffusion
welding between wraps is necessary, other modifications to the differential thermal ex-
pansion method might be necessary to line long tubes. For example, the tube might be
lined in a furnace having a short, moving hot zone that would allow the mandrel to move
in the axial direction and thus minimize nonuniform deformation.

GENERAL DISCUSSION

Capability of Process

The differential-thermal-expansion technique developed in this study was used suc-
cessfully to apply tungsten liners to T-111 tubing. This technique appears to have sev-
eral advantages over most of the other methods investigated for lining tubes. The tech-
nique should be quite economical because all that is required is a precision-machined
mandrel and a vacuum furnace capable of reaching the required lining temperature. No
problems were encountered in removing the mandrel from a lined tube. In contrast, the
tooling used in some of the other liner methods must be removed by acid leaching. In ad-
dition, the thermal-expansion method was shown to be capable of producing lined tubing
to close dimensional tolerances without the need for any machining after lining. The
liner thickness can be changed easily by changing the number of foil wraps and the asso-
ciated mandrel diameter.

The differential-thermal-expansion method, however, is not without problems. For
example, the use of the steel mandrel resulted in a slight pickup of iron by the liner.
But for our particular application, this trace amount of iron was not thought to be a prob-
lem, since analyses showed that the iron was present only as a very thin surface layer on
the tungsten liner.
Another problem area was encountered in lining tubes having large length-to-diameter ratios. Nonuniform axial movement of the mandrels caused the tubes to be somewhat barrel-shaped after lining. This behavior was minimized by reducing the lining temperature from 1315° C (2400° F) to 1200° C (2200° F). Increasing the initial assembly gap also would accomplish the same objective. Another method might be to use a short, moving hot-zone, which would allow the mandrel to move in the axial direction.

Since the total thermal expansion of the mandrel is a function of diameter, some problems also might be encountered in lining small-diameter tubes, because it might not be possible to close the assembly gap. Although not described in this report, T-111 tubes as small as 0.953 centimeter (0.375 in.) in diameter were lined with tungsten at 1315° C (2400° F) with a 0.008-centimeter (0.003-in.) assembly gap. For the materials used in our study (T-111 tubing and steel mandrels) and a minimum assembly gap of 0.008 centimeter (0.003 in.), we feel that this size of tubing is near the minimum diameter that can be lined by the differential-thermal-expansion method.

The advantages of the differential-thermal-expansion method appear to outweigh the few problem areas. Thus, this method was selected for lining full-length T-111 fuel pins for irradiation studies in the reactor at the Plum Brook Station of the NASA Lewis Research Center. The step-by-step procedures for lining these fuel pins are presented in detail in reference 7. A total of seven full-length fuel pins and 12 shorter (10.7-cm (4.2-in.) long) fuel pins were lined with tungsten. The shorter tubes were lined, three at a time, with the use of long mandrels, as shown in figure 7.

Applicability of Process

Although the differential-thermal-expansion technique was developed to line nuclear fuel pins, the technique has more general applicability. Only three components (mandrel, sheet lining material, and tubing) and a vacuum furnace are required to make use of the technique. It is not necessary to use a solid mandrel; however, the mandrel should possess sufficient strength at the lining temperature to maintain intimate contact between the liner and tubing. In general, it also will be necessary to coat the mandrel with an inert material to prevent the mandrel from welding to the liner. The only condition imposed on the lining material is that it be sufficiently flexible to be tightly wound around the mandrel. Through appropriate adjustment of the assembly gap and annealing temperature, the liner can be completely welded to the tubing with little or no change in the tubing diameter. Furthermore, the differential-thermal-expansion technique can be used to produce lined tubing having a precise liner thickness without additional machining being required.

The possible application of this method for lining pipe, tubing, and other cylindrical shapes is obvious. The differential-thermal-expansion method also might be useful in
the fabrication of more complex geometries. As one example, internal fins might be applied to heat-exchanger tubes through the use of appropriate mandrels.

SUMMARY OF RESULTS

A differential-thermal-expansion technique was developed to line tantalum alloy tubing with tungsten as part of a study of nuclear fuel-element fabrication techniques. The results obtained from the work on the differential-thermal-expansion liner technique are summarized below:

1. The differential-thermal-expansion liner technique was used successfully to line T-111 tubing in the size range of interest for fuel elements in a space-power nuclear-reactor concept. The T-111 tubing, 43 centimeters (17 in.) long by 1.9 centimeters (0.75 in.) in diameter, was lined with a 0.019-centimeter- (0.005-in.-) thick layer of tungsten with the use of an alumina-coated steel mandrel.

2. The integrity of the liner tended to increase with increasing lining temperature and decreasing assembly gap size.

3. Tubes with large length-to-diameter ratios can become barrel-shaped during lining because of nonuniform axial movement of the mandrels. This problem can be minimized by careful selection of the lining temperature and initial assembly gap size.

4. The differential-thermal-expansion method was used to line full-length fuel elements for irradiation studies.

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APPENDIX A

MATHEMATICAL RELATIONS FOR THE DIFFERENTIAL-
THERMAL-EXPANSION LINER METHOD

The general relations involving thermal expansivity, assembly gap size, dimensions, and temperature for the differential-thermal-expansion liner method are discussed in the following paragraphs. A schematic of the tubing-liner-mandrel assembly is shown in figure 8.

In order to press the tungsten foil against the T-111 tubing, the difference between the total thermal expansion of the mandrel \( \Delta d_m \), and the total thermal expansion of the tubing, \( \Delta d_t \), must equal the assembly gap \( g \), or,

\[
\Delta d_m - \Delta d_t = g \quad \text{(A1)}
\]

The total thermal expansions of the mandrel and the tubing can be expressed in terms of the temperature and thermal expansion coefficients by the equations

\[
\Delta d_m = a d_m (T - T_r)
\]

\[
\text{and}
\]

\[
\Delta d_t = a' d_t (T - T'_r)
\]

where \( d_m \) is the diameter of the mandrel, \( d_t \) is the inside diameter of the tubing, \( a \) and \( a' \) are the thermal expansion coefficients of the mandrel and tubing, respectively, \( T \) is the test temperature, and \( T_r \) is the room (or reference) temperature. Equation (A1) can be rewritten in terms of equations (A2) and (A3):

\[
(ad_m - a'd_t)(T - T_r) = g
\]

Thus, if the thermal expansion coefficients and the diameters of the mandrel and the tubing are known, the minimum temperature necessary to close the initial assembly gap \( g \) can be calculated. The preceding equations ignore the slight change in gap size caused by the thermal expansion of the foil liner. The use of equation (A4) is limited by the maximum use temperature of the components in the mandrel-and-tube assembly.

The usefulness of the differential-thermal-expansion technique is limited by the diameter of the tubing to be lined. This behavior can be shown by assuming that the mandrel diameter and the inside diameter of the tubing are essentially identical, or
\[ d_m \approx d_t \approx d \quad (A5) \]

This expression can then be inserted into equation (A4) to give

\[ (a - a')(T - T_r)d = g \quad (A6) \]

or

\[ T - T_r = \frac{g}{a - a'} \left( \frac{1}{d} \right) \quad (A7) \]

Thus, the temperature necessary to close a given gap is inversely proportional to the diameter of the mandrel or to the inside diameter of the tubing. Therefore, the differential-thermal-expansion technique may be useful for relatively large diameter tubing, but it may be inadequate for lining smaller tubes.

For our work, the thermal expansion of the low-carbon steel was assumed to be the same as that of iron. On the basis of the lattice-parameter data from reference 3, the change in the diameter of the steel mandrel, \( \Delta d_m \), can be expressed as

\[ \Delta d_m = d_m \left[ (1.18 \times 10^{-2}) + 23.0 \times 10^{-6} (T - 1000) \right] \quad (for \ T \geq 1000^\circ C) \quad (A8) \]

where \( d_m \) is the diameter of the mandrel at room temperature, and \( T \) is the test temperature.

On the basis of the thermal-expansion data for T-111 in reference 9, the change in the inside diameter \( \Delta d_t \) of the T-111 tubing from room temperature to temperatures above 1000\(^\circ\) C (1832\(^\circ\) F) can be represented by

\[ \Delta d_t = d_t \left[ (6.4 \times 10^{-3}) + (9.0 \times 10^{-6}) (T - 1000) \right] \quad (for \ T \geq 1000^\circ C) \quad (A9) \]

where \( d_t \) is the inside diameter of the T-111 tubing.
APPENDIX B

CONTAMINATION OF LINER

Several additional tests on tungsten-lined T-111 tubes were conducted to determine the extent of the iron contamination found on the inside surfaces of the tungsten liners. The tungsten liners from three different tubes were analyzed. A drop of mixed acid (nitric and hydrochloric acids) was placed on the surface and allowed to remain for a specific length of time. The solution then was drawn off and analyzed by emission spectroscopy. Some of the variables associated with this technique (e.g., etching rate and surface area wetted by the acid drop) make it difficult to obtain absolute values of surface contamination. The results do provide, however, a basis for comparison of liner contamination. In each case, a significant amount of iron was detected on the surface of the foil liner adjacent to the mandrel (i.e., the inside surface of the liner). The amount of surface iron was about 10 to 30 times the amount of surface iron detected on control samples of the tungsten foil. To obtain some indication of the depth of iron contamination, the inside surface of the foil layer adjacent to the foil layer in contact with the mandrel (i.e., the inside surface of the second layer of tungsten foil) also was analyzed for iron. The analytical results show that no iron was picked up by this layer of foil. In each case, the surface-iron analysis of the second layer was identical to the surface-iron analysis of the tungsten-foil control samples. Thus, the iron contamination appears to be concentrated on the inside of the innermost tungsten-foil layer, while the remaining four layers of foil appear to be free of iron.

To provide some indication of the thickness of the iron-contaminated layer, an entire tungsten liner was dissolved in acid, and the solution was analyzed for iron by emission spectroscopy. A total iron content of less than 100 parts per million was detected. Since the iron content of the tungsten before lining was 50 parts per million, the total iron pickup that can be attributed to the lining process is less than 50 parts per million. By using this value for iron pickup and assuming all of the iron is concentrated on the surface of the tungsten, the thickness of the iron layer was calculated to be only about 50 atom-layers thick. If this iron were to become uniformly distributed throughout the T-111 and the tungsten, the iron concentration in the T-111 would only increase about 5 parts per million, which is well within the T-111 specification limits. In either case, the small amount of iron should have little, if any, effect on the performance of the fuel pins. A few tests were conducted, however, in an effort to reduce the amount of iron contamination.

Various types and thicknesses of coatings were tried on the steel mandrels to reduce the amount of iron picked up by the tungsten liner. One approach was to increase the thickness of the alumina coating. Another approach was to use a duplex coating
consisting of a layer of plasma-sprayed tungsten between the steel mandrel and the alumina coating. Tubes were lined with the use of various coated mandrels and then the inside surfaces of the tungsten liners were analyzed for iron by emission spectroscopy. The type of coating on the mandrel and the analytical results are listed in table III. It can be seen from the analyses that the type and thickness of coating had very little effect on the surface iron contamination. Apparently, the transport of iron through the coatings is not diffusion controlled. A much more likely explanation for the iron contamination is that as the mandrel expands during heating, small cracks form in the relatively brittle coating on the mandrel, thereby permitting vapor transport of the iron from the mandrel to the tungsten liner. Thus, simply increasing the thickness of the mandrel coating will not reduce the liner contamination. The use of a mandrel coating which has about the same thermal expansion as the mandrel would probably be the most effective method for preventing the liner contamination. This approach, however, was not investigated in this study.
REFERENCES


### TABLE I. - MANDREL AND T-111 TUBING DIMENSIONAL CHANGES FOR INITIAL LINER STUDY

<table>
<thead>
<tr>
<th>Lining temperature (°C)</th>
<th>Room-temperature gap size (cm)</th>
<th>Calculated gap size at lining temperature (cm)</th>
<th>Measured decrease in mandrel diameter (cm)</th>
<th>Measured increase in T-111 tube diameter (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0°</td>
<td>32°F</td>
<td>cm</td>
<td>in.</td>
</tr>
<tr>
<td>1200</td>
<td>0.008</td>
<td>0.003</td>
<td>-0.0056</td>
<td>-0.0022</td>
</tr>
<tr>
<td>1200</td>
<td>0.010</td>
<td>0.004</td>
<td>-0.0030</td>
<td>-0.0012</td>
</tr>
<tr>
<td>1200</td>
<td>0.015</td>
<td>0.006</td>
<td>+0.0020</td>
<td>+0.0008</td>
</tr>
<tr>
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<td>0.020</td>
<td>0.008</td>
<td>+0.0071</td>
<td>+0.0028</td>
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<tr>
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<td>0.003</td>
<td>-0.0084</td>
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<tr>
<td>1315</td>
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<td>0.004</td>
<td>-0.0058</td>
<td>-0.0023</td>
</tr>
<tr>
<td>1315</td>
<td>0.015</td>
<td>0.006</td>
<td>-0.0005</td>
<td>-0.0002</td>
</tr>
<tr>
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<td>0.020</td>
<td>0.008</td>
<td>+0.0046</td>
<td>+0.0018</td>
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</table>

### TABLE II. - BULK ANALYSES OF TUNGSTEN-LINED T-111 TUBING AND AS-RECEIVED MATERIALS

<table>
<thead>
<tr>
<th>Lining temperature (°C)</th>
<th>Initial protective foil wrap</th>
<th>Material analyzed</th>
<th>Element, ppm by weight</th>
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</thead>
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<tr>
<td></td>
<td></td>
<td></td>
<td>C</td>
</tr>
<tr>
<td>1200</td>
<td>No T-111/W composite</td>
<td>84</td>
<td>&lt;5</td>
</tr>
<tr>
<td>1315</td>
<td>No T-111/W composite</td>
<td>66</td>
<td>25</td>
</tr>
<tr>
<td>1315</td>
<td>Yes T-111 IV</td>
<td>18</td>
<td>--</td>
</tr>
<tr>
<td>1315</td>
<td>W c</td>
<td>28</td>
<td>--</td>
</tr>
<tr>
<td>1315</td>
<td>Yes T-11 only</td>
<td>45</td>
<td>26</td>
</tr>
<tr>
<td>1315</td>
<td>T-111/W composite</td>
<td>55</td>
<td>--</td>
</tr>
<tr>
<td>1315</td>
<td>As-received T-111</td>
<td>39, 104</td>
<td>18</td>
</tr>
<tr>
<td>1315</td>
<td>As-received W</td>
<td>30</td>
<td>&lt;10</td>
</tr>
</tbody>
</table>
TABLE III. - EFFECT OF MANDREL COATING ON SURFACE IRON CONTAMINATION OF TUNGSTEN LINER

[Amounts of surface iron on control samples of tungsten foil were 0.001, 0.003, and 0.03 µg/cm².]

<table>
<thead>
<tr>
<th>Coating</th>
<th>Thickness</th>
<th>Surface iron on tungsten liner, µg/cm²</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>cm</td>
<td>in.</td>
</tr>
<tr>
<td>Alumina</td>
<td>0.013</td>
<td>0.005</td>
</tr>
<tr>
<td>Alumina</td>
<td>0.013</td>
<td>0.005</td>
</tr>
<tr>
<td>Alumina</td>
<td>0.025</td>
<td>0.010</td>
</tr>
<tr>
<td>Duplex</td>
<td>Tungsten</td>
<td>0.013</td>
</tr>
<tr>
<td></td>
<td>Alumina</td>
<td>0.003</td>
</tr>
<tr>
<td>Duplex</td>
<td>Tungsten</td>
<td>0.013</td>
</tr>
<tr>
<td></td>
<td>Alumina</td>
<td>0.013</td>
</tr>
</tbody>
</table>

Figure 1. - Schematic drawing of the differential-thermal-expansion liner technique.
Figure 2. - Cross-section microstructures of T-111 tubing lined with tungsten at 1200° C (2192° F) by the differential-thermal-expansion method. Etchant for T-111 alloy, solution of 50 milliliters of nitric acid, 20 milliliters of water, and 30 grams of ammonium bifluoride. Etchant for tungsten, Murakami's reagent. X150.
Figure 3. - Cross-section microstructures of T-111 tubing lined with tungsten at 1315°C (2400°F) by the differential-thermal-expansion method. Etchant for T-111 alloy: solution of 50 milliliters of nitric acid, 20 milliliters of water, and 30 grams of ammonium bifluoride. Etchant for tungsten: Murakami's reagent. X150.
Figure 4. T-111 tube lined with tungsten by the differential-thermal-expansion lining technique for use as cladding for full-length fuel element.
Figure 5. - Variation in diameter of full-length tube lined with tungsten at 1315° C (2390° F).

Figure 6. - Variation in diameter of full-length tube lined with tungsten at 1200° C (2192° F).
Figure 7. - Three T-Ill tubes lined simultaneously with the use of a single mandrel.

Figure 8. - Schematic drawing of mandrel and T-Ill tubing assembly used in the differential-thermal-expansion liner technique. The symbol \( d_m \) is the mandrel diameter; \( d_I \) is the inside diameter of the T-Ill tubing; and \( g \) is the total assembly gap between the tungsten-wrapped mandrel and the T-Ill tube.