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Metallurgy and Ceramics

BATTELLE MEMORIAL INSTITUTE

JACKETING OF URANIUM FOR HIGH-TEMPERATURE SERVICE

II - Barrier Coat Materials

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SUMMARY

The effectiveness of various barrier metals between uranium, uranium-molybdenum alloys, or uranium-columbium alloys and a low-carbon steel was measured by sandwich-type diffusion-to-failure tests. Failure resulted when an iron-uranium alloy was formed by diffusion of one, or both, of the constituents through the barrier metal. The efficacy of the barrier coat was judged on the basis of the time and temperature required for failure versus the thickness of the barrier-coat metal. Most of these evaluations were made between 900 C and 1100 C over times ranging up to at least 100 hours at temperature.

The following metals were tested against pure uranium: silver, tantalum, tungsten, molybdenum, columbium, vanadium, zirconium, titanium, beryllium, and palladium. The most effective barriers at 1100 C under the conditions of test and in the order of their effectiveness were found to be tungsten, tantalum, and columbium or molybdenum. When the uranium was alloyed with small amounts of molybdenum or columbium, the barriers did not function satisfactorily. In the case of molybdenum-uranium alloys, it was found that the molybdenum content had to be increased to above five weight per cent before tantalum barrier coats were again as effective at 1100 C as they were with pure uranium. At higher molybdenum percentages, columbium, molybdenum, and tungsten barriers also appeared to function satisfactorily. With additions of up to five weight per cent columbium in uranium, irregular failures occurred with

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tantalum and columbium barriers. Double barriers of zirconium with tungsten or tantalum, the latter on the steel side of the sandwich, functioned well against both of the uranium alloys.

INTRODUCTION

The work covered in this report is a portion of the experimental research undertaken for the design of a uranium-containing metallic fuel rod for operation in air at 1090 C. (See also "Jacketing of Uranium for High-Temperature Service, I. High-Temperature Fuel Rods", H. A. Saller and J. T. Stacy, Report BMI-T40, September 30, 1950). This report deals with that phase which involved the testing and evaluation of various metals as barrier coats between uranium alloys, and low-carbon steel. The work is of a preliminary nature and is concerned more with a comparison of the various barrier metals than with the acquisition of numerical data, e.g., diffusion coefficients. It is fundamental for the design of certain types of all-metal fuel elements for high-temperature operation.

Essentially, a barrier coat is a material which insulates one material from another and prevents diffusion between the two substances. An attractive method of shielding and supporting uranium core material at temperatures in the range of 900 to 1100 C (1650 - 2010 F) is by jacketing the core with a high-temperature alloy. However, those high-temperature alloys most useful for such jacketing contain two, or more, of the following elements: Cr, Co, Fe, or Ni. Without a barrier coat, uranium in contact with such a jacket would form a molten eutectic with

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EXPERIMENTAL WORK

Types of Tests

An attempt was made to determine the diffusion constants for the diffusion of uranium into silver at 600, 700, 800, and 900 C.

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Silver-plated uranium cylinders were exposed for 100 hours at temperature in vacuo, after which successive layers of silver were analyzed chemically for uranium. From the distribution of concentration, the diffusion constants were determined by Matano's method.* However, the growth and warpage of the uranium rods were so extensive that results were very erratic. In view of these results and in order to secure more practical values for the efficacy of barrier action, further diffusion-coefficient determination tests were omitted in favor of the simpler and more useful (for the purpose intended) method of evaluating the effectiveness of the barrier coat by diffusion-to-failure tests.

The two types of diffusion-to-failure tests used to determine the effectiveness of barrier coat materials were the sandwich type and the short open-end tube type. Practically all of the barrier-coat data presented in this report were secured from the sandwich-type test in which the uranium, or uranium alloy, was insulated from a sandwich of low-carbon steel pieces by intervening layers of barrier-coat metal. The whole assembly was held together with a steel clamp.

The open-end tube test was designed to simulate possible service use in which the uranium core and barrier coat would be contained in a jacket of high-temperature alloy. Small tubes were machined from three different austenitic steels of superior high-temperature corrosion resistance and strength (15 Cr - 35 Ni, 25 Cr - 20 Ni, and 25 Cr - 12 Ni).

* F. N. Rhines and R. E. Mehl, "Rates of Diffusion in Alpha Solid Solutions of Copper", Trans. AIME, 128, 1938, page 185.

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Cores of uranium, wrapped with various barrier foils, were inserted in the tubes and upset until the outside diameters of the tubes were expanded enough to insure that good contact had been established between the three metals. In general, the open-end tube test gave results similar to the simpler sandwich-type test and consequently was discontinued in favor of the latter.

Discussion of Tests

Several factors other than temperature could logically influence the time required for a barrier to fail by affecting the diffusion rate through the barrier. For example, in a fuel element during actual operation, such diffusion might be greatly influenced by irradiation, an effect not considered in present tests, although its importance is recognized. Grain size, diffusion anisotropy, cold work, and pressure might also influence diffusion rates.

The effect of grain size on diffusion rates is related to the rapidity of transgranular versus intergranular migration of the foreign atoms. According to Mehl*, this mechanism is not clearly understood. In these tests, it was felt that this factor might be circumvented by using materials either commercially available or prepared in a manner similar to that which might be used in actual fuel-rod construction. The effect of diffusion anisotropy likely would be insignificant, inasmuch

* R. F. Mehl, Trans. AIME, 156, page 325, 1944.

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as over the temperature range of interest (900 to 1100 C) all of the metals under test (except beryllium) which were noncubic at lower temperatures would have transformed to cubic structures.

In these tests, it was thought that in such a high-temperature range the influence of previous cold working on diffusion would largely be eliminated by recrystallization and grain growth. To determine the effect of cold work on diffusion, sandwich-type samples were made up using either annealed or 50 per cent cold-rolled tantalum as a barrier in combination with either alpha annealed or 62.5 per cent cold-rolled uranium. After 100-hour exposure at 1090 C in vacuo, the uranium penetration into the tantalum was measured microscopically. Upon averaging the results for similar samples, it was found that the maximum uranium penetration was within the range of 18 to 24 per cent of the tantalum thickness (0.005 inch) for the four series, the lower rate being exhibited by the annealed specimens. This variation did not seem to be important in these tests.

With regard to the effect of pressure on the rate of diffusion, it has been reported that the diffusion constant for a given temperature increases logarithmically with pressure.* To determine the effect of clamp pressure on diffusion in sandwich tests, four similar series were run in which pressures higher than necessary were used to close the steel clamps containing the diffusion specimens. The results of these tests

* A. B. Kinzel, Welding Journal, Vol. 23, 1944, pages 1124-1143.

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closely agreed with those previously discussed in regard to the effect of cold work. Apparently, the excess clamping pressures were dissipated either before, or soon after, the steel clamps reached 1090 C.

Materials

In selecting barrier metals for immediate investigation, the melting temperature and ready availability were used as the first criteria. On the availability basis, metals of the platinum and rare earth groups were eliminated from consideration, although most of them were satisfactory from the melting-temperature viewpoint (palladium was later tested). A study of uranium binary-alloy constitution diagrams served to eliminate from consideration those metals which would form low-melting eutectics, e.g. iron, chromium, nickel, cobalt, etc. Attention was given to beryllium and vanadium, however, because of the high melting points of their eutectics with uranium. After such an appraisal, the following metals were investigated as barrier coats in the range 900 to 1100 C: tantalum, tungsten, molybdenum, columbium, vanadium, zirconium, titanium, beryllium, and palladium. Although silver melts at 960 C, it was tested also as previous information indicated that it was an excellent barrier.

In general, all materials were procured commercially where possible. Tantalum, tungsten, molybdenum, and columbium were secured from the Fansteel Metallurgical Corporation and the zirconium was purchased from the Foote Mineral Company. The silver foil was rolled from stock

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on hand and both the silver and palladium plating was done at Battelle. For most of the sandwich-type tests, a regular grade of 1010 carbon steel was used. The vanadium was obtained from the Knolls Atomic Power Laboratory and analyzed as follows:

C, 0.18%

Al, Si, Mo, Cr, Ni, each less than 0.05%

Mg and Mn, each 0.01 to 0.10%

Fe, 0.05 to 0.15%

Ca, less than 0.005%

A small amount of 0.005-inch beryllium foil was fabricated by hot rolling extruded stock in a welded steel jacket at 870 C (1600 F). After rolling, the jacket was removed by pickling in dilute nitric acid. This foil was quite brittle, but with proper precautions, a few tests were made using the foil as a barrier. The titanium was forged and rolled from an arc-melted ingot. It analyzed as follows: 0.4% C and 0.02% N.

Procedure

In the sandwich-type diffusion-to-failure tests, which the short open-end tube test closely approximated, the uranium, or uranium alloy, was sandwiched between barrier foils (or plated with the barrier metals, as in the cases of silver and palladium). This pack, in turn, was placed between pieces of low-carbon steel and the entire assembly introduced into a welded steel clamp. (See Figure 1.) The clamp was then compressed until the components of the sandwich were in good contact.

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Clamp

Assembled Pack

FIGURE I. DIFFUSION PACK

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Considerable care had to be taken when compressing the clamp, inasmuch as too much pressure would deform the barrier coat and possibly cause the uranium or steel to shear through the barrier foil, even though those edges which were next to the foil were beveled. In practice, pressure was gradually applied until the sandwich components resisted movement by a knife blade pressed against the side of the pack. This generally required from 40,000 to 50,000 pounds pressure on the clamp itself, depending upon the amount of clamp deformation necessary to contact various sandwich thicknesses, etc.

Prior to assembly, the uranium and low-carbon-steel pieces were polished on abrasive papers through grit Number 400A and cleaned in carbon tetrachloride and acetone. It was especially necessary to grind the pure uranium on the final grit immediately before pressing to avoid the formation of an oxide layer before pack assembly.

In testing the barrier properties of various metals against uranium, a low-carbon steel was used on the opposite side of the barrier for two reasons: First, iron is the principal constituent of the stainless steels which possess desirable high-temperature properties for jacketing materials. Second, iron forms a low-melting eutectic with uranium similar to those formed by nickel, chromium, and cobalt, which elements are all important constituents of high-temperature alloys.

After assembly, the samples were held at temperature for predetermined times, quenched, and sectioned for examination. If the failure had been extensive, metallographic inspection was not necessary, inasmuch

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as the presence of the iron-uranium alloy was easily detectable. Except for some of the molybdenum barrier-coat samples and the uranium-alloy specimens, which were sealed in either evacuated or argon-filled quartz tubes, all specimens were run in vacuo in two Globar-heated vacuum furnaces. One of these was controlled with a Tagliabue "Celectray" controller and the other by a Leeds and Northrup two-point controller-recorder. Temperature fluctuation in the former seldom exceeded ± 2 C, while in the latter it varied from ± 3 C up to ± 7 C, depending upon the closeness of the cycle during "on" and "off" periods for which the current could be set. Those specimens sealed in quartz were run in small laboratory muffles where temperature variation during cycling might run as high as ± 12 to ± 17 C in the air adjacent to the quartz tubes. However, the deviation in specimen temperature was undoubtedly less. Sample temperatures were taken with a Leeds and Northrup Semi-Precision Potentiometer and Chromel-Alumel thermocouples. Occasional checks were made with a platinum-platinum 10% rhodium thermocouple to determine the accuracy of the Chromel-Alumel readings.

Barrier Coats for Uranium

Data on the effectiveness of various barrier-coat materials between uranium and low-carbon steel were particularly desired in regard to the maximum temperature at which the material would function as a barrier for 100 hours. To determine these data, different barriers in various thicknesses of foil or electroplate were made up in sandwich-type

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packs and tested at different temperatures for 100 hours. Additional specimens run at a variety of temperatures and times served to locate the time-temperature limits for a particular thickness of barrier to function as such. When these data were plotted, a logarithmic relationship was found to exist between the temperature of test and the time to failure over the limited temperature and time ranges used. A typical plot is shown in Figure 2.

In many cases, the points did not fall in such perfect order as to be separated neatly by a line such as shown in Figure 2. In the instances of the 0.004- and 0.006-inch silver-foil barrier coats, where effective barrier action occurred near the melting point of silver, the test results were very irregular and the lines delineating the limit of barrier action had to be approximated. Undoubtedly, some of this behavior was caused by volatilization of the silver in vacuo at these temperatures. Further mention of these difficulties will be made and formulae for the approximate limits of barrier action will be given in the discussions of the various barrier metals.

Table I lists the maximum temperatures at which these various barrier metals (except palladium) may be expected to function between uranium and low-carbon steel for 100 hours. Figure 3 summarizes the effectiveness of some of these materials over a range of temperatures and times. These limit tracings are shown within the parameters of the experimental work. Whether or not these lines may be extrapolated is open to question, but it would seem likely that they might be used to

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FIGURE 2. METHOD OF DETERMINING THE LIMITING TIME AND TEMPERATURE FOR FAILURE

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TABLE 1. MAXIMUM TEMPERATURES FOR 100-HOUR BARRIER ACTION
BETWEEN URANIUM AND LOW-CARBON STEEL⁽¹⁾

Barrier Material	Maximum Temperature, C, for Barrier Thickness, In.					
	0.0025	0.005	0.010	0.002	0.004	0.006
Tungsten	(2)	(2)	-	-	-	-
Tantalum	-	(2)	(2)	-	-	-
Columbium	1025	1060	(2)	-	-	-
Molybdenum	-	1055	(2)	-	-	-
Vanadium	-	975	1035	-	-	-
Beryllium	-	950	-	-	-	-
Titanium	-	900	925	-	-	-
Zirconium	(3)	(3)	(3)	-	-	-
Silver, Electroplated	-	-	-	900	905	910
Silver, Foil	-	-	-	910	920	935

- (1) A maximum test temperature of 1100 C. Sandwich-type specimens.
(2) Over 100 hours at 1100 C.
(3) No barrier against steel.

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FIGURE 3. LIMITS OF EFFECTIVE BARRIER ACTION BETWEEN URANIUM AND LOW-CARBON STEEL

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determine data for other limits in this manner, as it is well known that the logarithm of the diffusion constant yields a linear relationship when plotted against the reciprocal of the absolute temperature. However, an examination of the slopes of the 0.0025-inch and the 0.005-inch columbium-foil lines indicates that, if these lines were extrapolated toward lower temperatures, they would intersect. A similar situation would occur in the case of the molybdenum tracings. Such a condition, i.e., one in which the thinner foil would be equivalent to one of double its thickness, appears to be an unlikely possibility. Consequently, either the lines must be very short arcs of large curves, or the plotting and data are not of sufficient accuracy to allow for extrapolation.

Plated - Ag, Pd

The effectiveness of silver as a barrier was determined for both foil and electroplated deposits in thicknesses of 0.002, 0.004, and 0.006 inch for times up to 250 hours at temperature. Equal thicknesses of silver foil appeared to function at somewhat higher temperatures than plated silver, but the results obtained became more erratic as the melting point of silver was approached. Formulae approximating the limiting relationships between time, temperature, and thickness for silver to function as a barrier between uranium and low-carbon steel are shown in Table II.

Figure 4 shows a typical specimen in which the barrier had failed to prevent diffusion between the low-carbon steel and the uranium. The

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TABLE II. FORMULAE⁽¹⁾ RELATING LIFE OF BARRIER TO TIME AND TEMPERATURE
(ALL BARRIERS BETWEEN URANIUM AND LOW-CARBON STEEL)

Barrier Metal	Thickness of Barrier, Inch	Limiting Relationship T = Temperature, C; t = Time, Hours	Test Limits	
			C	Hours
Ag, Electroplated	0.002	$T = -19 \log t + 942.5$	-	5-250
	0.004	$T = -21 \log t + 950$	-	5-250
	0.006	$T = -23 \log t + 956$	-	5-250
Ag, Foil	0.002	$T = -13.5 \log t + 942$	-	5-100
	0.004	$T = -9.5 \log t + 953^{(2)}$	-	5-100
	0.006	$T = -8 \log t + 955^{(2)}$	-	5-100
Mo, Foil	0.0025	$T = -134 \log t + 1231$	1025-1100	-
	0.005	$T = -164 \log t + 1384$	1025-1100	-
	0.010	$T = -309 \log t + 1767$	1025-1100	-
Cb, Foil	0.0025	$T = -61 \log t + 1148$	1025-1100	-
	0.005	$T = -125 \log t + 1312$	1060-1100	-
V, Foil	0.0025	$T = -234 \log t + 1225$	950-1050	-
	0.005	$T = -85 \log t + 1145$	975-1050	-
	0.010	$T = -31 \log t + 1099$	1037-1075	-

(1) 0.0025-inch tungsten, 0.005-inch tantalum, and 0.010-inch columbium foils did not fail in 100 hours at 1090 C. No further tests made.

(2) Erratic test results. Formulae only rough approximations.

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FIGURE 4. FAILED SILVER BARRIER COAT SPECIMEN
SHOWING THE LOW-CARBON STEEL (Fe), THE
IRON-URANIUM ALLOY, AND THE REMAINS OF
THE SILVER BARRIER COAT

FIGURE 5. THE CONDITION OF A 0.004-INCH SILVER
PLATE BARRIER COAT AFTER 233 HOURS AT
900 C

FIGURE 6. TANTALUM BARRIER COAT AFTER 100 HOURS
AT 1080-1090 C

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alloy formed is dendritic and typical of those observed in all "failed" specimens. On the other hand, Figure 5 illustrates a silver barrier which did not fail to function.

Electroplated palladium of 0.005-inch thickness was tested for 100 hours at 1090 C between uranium and low-carbon steel. It did not function as a barrier under those conditions.

Refractory Metals - Ta, W, Mo, Cb

Both tantalum and tungsten were found to be excellent barriers between uranium and low-carbon steel or nickel-chromium-iron alloys of the stainless steel types. A tantalum barrier of 0.005 inch or a tungsten barrier of 0.0025 inch were found suitable for more than 100 hours at 1100 C (2012 F). Figure 6 shows typical diffusion results using a tantalum barrier. Upon cooling sandwich-type specimens, it was found that the tantalum generally adhered to the iron or stainless steel more firmly than to the uranium core. When the uranium piece was appreciably thick (probably 0.100 inch minimum), a considerable fracture zone or even a void was observed between the tantalum barrier and the uranium after cooling and sectioning the sandwich. Figure 7 shows such a fracture zone.

Molybdenum, while not so effective a barrier between steel and uranium as tantalum or tungsten, appeared to be a suitable barrier for 100 hours at 1100 C when used in sufficient thickness, e.g., 0.010 inch. The limiting time and temperature relationships for molybdenum foil to function as a barrier over the range of tests made are shown in Table II.

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FIGURE 7. FRACTURING OF TANTALUM-URANIUM INTERFACE
UPON COOLING AFTER 100 HOURS AT 940 C

FIGURE 8. APPEARANCE OF A 0.005-INCH COLUMBIUM
BARRIER COAT AFTER 100 HOURS AT 1080-
1100 C. FAILURE APPEARS IMMINENT

FIGURE 9. TYPICAL VANADIUM BARRIER-COAT DIFFUSION
AFTER 8 HOURS AT 1008 C. NOTE WIDE DIF-
FUSION BAND BETWEEN VANADIUM AND LOW-
CARBON STEEL AND SECOND PHASE IN
VANADIUM

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Difficulty was encountered in getting consistent results with columbium barriers in diffusion-to-failure tests. Early tests indicated that a 0.005-inch columbium foil was a good barrier between uranium and low-carbon steel for 100 hours at 1100 C, but later tests did not always confirm this. Figure 8 shows a 0.005-inch columbium barrier foil which had not failed during 100 hours at about 1090 C, although, on the basis of other tests, failure should have occurred. Very extensive diffusion is observable between the columbium and the uranium, while a fairly heavy diffusion layer can be seen between the columbium and the steel. A similar sample run at the same time failed. In view of such inconsistent results, limits were approximated for columbium-foil barrier action between low-carbon steel and uranium by running numerous duplicates. These limits are shown in Table II.

Others - V, Zr, Ti, Be

Vanadium was found to function quite successfully as a barrier between low-carbon steel and uranium in the thickest gage tested, i.e., 0.010-inch sheet. Above 1075 C (1965 F), all barrier action ceased except for very short periods of time. This was not unexpected, inasmuch as a vanadium-uranium eutectic forms at 1040 C. Diffusion between the vanadium and the steel appeared to occur to a considerably greater extent than that between the vanadium and the uranium. This may be seen in Figure 9, where the characteristic wide diffusion band is located on the steel side of the interface. In specimens where this wide band was

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found, a rather large amount of second phase was also observed in the vanadium barrier. The appearance of this wide diffusion band in the steel concurrently with the increase of second phase in the vanadium might also be ascribed to carbon depletion in the steel with the formation of carbide in the vanadium. However, no effort was made in this survey to determine the method of formation or the compositions of diffusion bands. A summary of the effectiveness of vanadium barrier action is given in Table II.

Over the temperature range 950 - 1050 C (1740 F - 1925 F), zirconium did not function as a barrier between uranium and low-carbon steel in thicknesses up to 0.010 inch. In this range, failures occurred with great rapidity, e.g., in less than six hours at 950 C for a barrier of 0.010-inch. Metallographic inspection of diffusion specimens in which a foil of tantalum, tungsten, or columbium was used to separate the zirconium from the steel shows little evidence of zirconium-uranium diffusion (see Figure 10). This evidence indicates that barrier failures occurred because of diffusion between zirconium and iron rather than between zirconium and uranium.

A series of 100-hour exposures were made using 0.005- and 0.010-inch titanium foil as a barrier between uranium and low-carbon steel. The 0.005-inch foil functioned up to 900 C (1650 F) and the 0.010-inch sheet up to 925 C (1700 F) for 100 hours at temperature.

A similar series of 100-hour exposures at temperatures from 900 to 1100 C were made using 0.005-inch beryllium foil. No failures occurred at 900 C or 950 C, while failures occurred at 975 C and above.

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FIGURE 10. A DOUBLE BARRIER OF TANTALUM AND ZIRCONIUM SHOWS LITTLE DIFFUSION BETWEEN ZIRCONIUM AND URANIUM AFTER 24 HOURS AT 1000 C

FIGURE 11. AFTER 100 HOURS AT 1090 C, A TANTALUM BARRIER IN CONTACT WITH A MOLYBDENUM-URANIUM ALLOY CONTAINING 4.1 WEIGHT PER CENT MOLYBDENUM SHOWS THE PRESENCE OF A YELLOW INTERGRANULAR PHASE

FIGURE 12. A DOUBLE BARRIER OF TUNGSTEN AND ZIRCONIUM AFTER 100 HOURS AT 1082-1117 C. THE MOLYBDENUM-URANIUM ALLOY CONTAINS 0.5 WEIGHT PER CENT MOLYBDENUM

FIGURE 13. A TANTALUM BARRIER AFTER 100 HOURS AT 1072-1109 C. THE MOLYBDENUM-URANIUM ALLOY CONTAINED 8.9 WEIGHT PER CENT MOLYBDENUM

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Barrier Coats for Uranium Alloys

Molybdenum-Uranium Alloys

The dimensional stability on thermal cycling imparted to uranium by small additions of molybdenum or columbium suggested that such alloys might be used advantageously as cores for high-temperature canning experiments. However, when uranium alloys, low in molybdenum or columbium, were canned using a 0.005-inch tantalum barrier coat, unexpectedly early failures occurred at 1090 C (2000 F). When a molybdenum-uranium alloy containing 8.9 weight per cent molybdenum was similarly canned, no failure occurred in 100 hours at the same temperature. In view of this behavior, a series of molybdenum-uranium alloys varying from 0.1 to 8.9 weight per cent molybdenum were prepared for sandwich-type diffusion-to-failure tests in packs containing a barrier and SAE 1020 steel.

A series of these molybdenum-uranium samples, in which the barrier coat consisted of 0.005-inch tantalum foil, were run for 100 hours in vacuo or in argon at temperatures between 1040 and 1090 C. (1900 - 2000 F). Figure 14 summarizes the results of these tests. While these data are not conclusive, the trend indicates that barrier failures do not occur when the molybdenum content is increased above about 5 weight per cent. It is likely that molybdenum segregation in some of these alloys was a factor in not securing more positive results.

After diffusion, the tantalum barriers in contact with molybdenum-uranium alloys frequently showed a lemon-yellow phase, apparently molten

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FIGURE 14. EFFECTIVENESS OF 0.005-INCH Ta BARRIER COAT BETWEEN Mo-U ALLOYS AND LOW-CARBON STEEL AT 1040 TO 1090 C (1900-2000F) FOR 100 HOURS

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at test temperature (1090 C), which was not seen when diffusion occurred only with pure uranium. Figure 11 shows this phase as a network in the tantalum barrier.

Other contact diffusion-to-failure specimens, using barriers of molybdenum, columbium, or tungsten instead of tantalum between molybdenum-uranium alloys and a low-carbon steel, showed a similar effect in that failures occurred when the uranium alloy was low in molybdenum. An exception to this general behavior was demonstrated by a sample containing a double barrier of tungsten and zirconium, the tungsten lying next to the steel in the sandwich (see Figure 12). The molybdenum-uranium alloys used in these specimens contained either 0.5 or 8.9 weight per cent molybdenum.

Columbium-Uranium Alloys

Considerably less data were secured on the effectiveness of various barriers between columbium-uranium alloys and low-carbon steel than in the previously discussed case of molybdenum-uranium alloys. However, 100-hour tests were made in the range 1055-1090 C (1922 - 2000 F) using alloys of 0.8, 1.3, 2.3, and 4.9 weight per cent columbium against the following barriers: columbium, tantalum, tungsten, and double barriers of zirconium-tantalum and zirconium-tungsten, wherein the zirconium was protected from diffusion with the steel by either tantalum or tungsten.

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The results of these tests are presented in Table III. It will be seen that tantalum barriers failed earlier than would have been expected, while columbium barriers failed as would have been anticipated. The double barriers with zirconium ran well. In a few instances, heavy oxidation occurred which possibly interfered with test accuracy (see Note 2). Further barrier studies using columbium-uranium alloys would appear to be profitable considering the erratic results obtained.

Comment

It is possible that increased diffusion rates can account for the sudden decrease in the effectiveness of tantalum barriers when small amounts of molybdenum or columbium are added to uranium. In this regard, Rhines and Mehl⁽¹⁾ have shown that the diffusion coefficients between copper and its alloys vary markedly with increases of the solute in the copper alloy. A similar phenomenon may be occurring in this case. At higher concentrations of the solute, i.e. molybdenum, it was found that the diffusion between the tantalum barrier and the molybdenum-uranium alloy decreased. At 8.9 weight per cent molybdenum, practically no diffusion between the barrier and the molybdenum-uranium alloy was discernible after 100 hours at 1072 - 1109 C (see Figure 13). This anomalous behavior with increasing molybdenum content is difficult to understand. While it is known that the activation energy for diffusion

(1) F. N. Rhines and R. F. Mehl, Trans AIME, 128, 185 (1938).

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TABLE III. SANDWICH-TYPE DIFFUSION TESTS OF VARIOUS
BARRIERS VS. COLUMBIUM-URANIUM ALLOYS

Weight Per Cent, Cb	Barrier		Average (1) Temperature, C	Time, Hr	Results
	Type	Thickness, In.			
0.8	Cb	0.005	1090	100	Good
0.8	Cb	0.005	1080	100	Failed
0.8	Ta	0.005	1080	100	Failed
0.8	W	0.0025	1090	100	Failed?
	Ta	0.005			
0.8	Zr	0.005	1055	100	Good
	W	0.0025			
0.8	Zr	0.005	1055	100	Good
1.3	Cb	0.005	1090	100	Failed
1.3	Ta	0.005	1072	100	Good (2)
1.3	Pd (plate)	0.005	1089	100	Failed
2.3		0.005	1083	100	Good (3)
2.3	Ta	0.005	1083	100	Failed
4.9	Cb	0.005	1069	100	Good (2)
4.9	Cb	0.005	1074	100	Good (3)
4.9	Ta	0.005	1083	100	Good (2)
4.9	Ta	0.005	1074	100	Failed

- (1) In argon-filled quartz tubes. Variations up to ± 17 C in some cases.
- (2) Heavily oxidized.
- (3) Microscopic examination revealed extensive diffusion and failure appeared imminent.

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is proportional to the melting temperature, (1) the increase in the melting point of molybdenum-uranium alloys up to 10 weight per cent molybdenum does not appear to be great enough to account for the marked change in the diffusion characteristics over this range. This variation may, however, be associated with other pronounced changes which have been noted in these alloys with increasing molybdenum content, e.g., increasing dimensional stability on thermal cycling, increasing resistance to oxidation, and increasing difficulty in removing the oxide by pickling.

(1) F. Seitz, "The Physics of Metals", McGraw-Hill Book Co., New York, 1943, page 180.

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