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THE INFLUENCE OF HEAT-TREATMENT AT 1300°C ON THE INTERNAL MORPHOLOGY OF TANTALUM, TANTALUM - 10 w/o TUNGSTEN, AND TUNGSTEN FOILS, AND THE ADDITIONAL EFFECT OF IMMERSION IN LIQUID URANIUM

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June 1988

כולל כותרת ותקציר בעברית

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ABSTRACT

The internal morphology of 0.3-mm-thick foils of tantalum, tantalum - 10 w/o tungsten, and tungsten is investigated by optical microscopy and a scanning electron microscope capable of microanalysis by EDAX. Comparison is made between foils: as-received, heat-treated at 1300°C for 20 hours (for tantalum also for 40 hours), and after immersion and reaction with liquid uranium for 20 hours at 1300°C. The heat-treatment does not influe ce the internal morphology (grain size) in tantalum and tantalum - 10 w/o tungsten, and the multilayer structure of these foils following the immersion experiments is solely due to the effect of liquid uranium. This effect depends on prior annealing. In tungsten the heat-treatment changes the dominant flow-line character of the foil into a grain structure, and the foil structure following the immersion experiment is due to the combined effect of heat-treatment and uranium penetration.

השפעת הטיפול בחום ב-1300°C על המורפולוגיה הפנימית של פחיות מטנטלום, מטנטלום - w/o 10 טונגסטן, ומטונגסטן, והאפקט הנוסף של טבילה באורניום נוזלי

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תמוז תשמ"ח - יוני 1988

תקציר

המורפולוגיה הפנימית של פחיות בעובי 0.3 mm מטנטלום, מטנטלום - w/o 10 טונגסטן, ומטונגסטן נחקרת באמצעות מיקרוסקופ אופטי ומיקרוסקופ אלקטרוני סורק המסוגל לבצע מיקרואנליזה בשיטת EDAX (*ניתוח נפיצת האנרגיה של קרני-x*). נערכת השוואה בין פחיות כפי שהתקבלו, פחיות לאחר טיפול בחום ב-1300°C במשך 20 שעות (עבור טנטלום גם במשך 40 שעות), ופחיות לאחר טבילה ותגובה עם אורניום נוזלי במשך 20 שעות ב-1300°C. הטיפול בחום אינו משפיע על המורפולוגיה הפנימית (גודל הגרעינים) בטנטלום ובטנטלום - w/o 10 טונגסטן, והמבנה הרב-שכבתי של הפחיות הללו בעקבות ניסויי הטבילה הוא אך ורק בשל האפקט של האורניום הנוזלי. אפקט זה תלוי בהרפייה מוקדמת. בטונגסטן הטיפול בחום משנה את אופי קווי הזרימה הדומיננטיים של הפחית למבנה גרעיני, והמבנה של הפחית בעקבות ניסויי הטבילה הוא תוצאה של האפקט המשולב של טיפול בחום וחדירת אורניום.

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1. Introduction

Recently results have been reported on immersion of 0.3-mm-thick foils of tantalum metal (refs. 1,2), of tantalum - 10 w/o tungsten alloy (ref. 2), and of tungsten metal (ref. 3) in liquid uranium in the temperature range of 1160°C to 1350°C for times up to 20 hours. The first immersion experiments, of tantalum in liquid uranium (ref. 1), were initiated to investigate and characterize the failure of tantalum crucibles in long-containment melting of uranium. The experiments were carried out in zirconia crucibles in a smelting furnace adapted for immersion experiments of solid metals in liquid metals (ref. 4), and the resulting diffusion of liquid uranium into the foils was studied by optical microscopy and by a scanning electron microscope capable of microanalysis by energy-dispersion-analysis-of-x-rays (SEM-EDAX). A multilayer structure was observed in liquid-uranium-reacted foils of tantalum (refs. 1,2) and of tantalum - 10 w/o tungsten (ref. 2). Such a structure, however, did not develop in the liquid-uranium-reacted tungsten foils (ref. 3).

Layers identified in tantalum that had been immersed in uranium are (refs. 1,2): outer uranium, uranium-tantalum (U/Ta ~ 1), precipitated columnar tantalum (<1 w/o U), inner uranium, and inner tantalum (with growing grains and uranium along grain boundaries). In the tantalum foil which underwent reaction with liquid uranium at 1300°C for 20 hours, layer thicknesses are: precipitated columnar tantalum - 90 μm, inner uranium - 10 μm, and inner tantalum - 130 μm (increase in total thickness). The average grain size in the inner tantalum layer is 45 μm, with typical grown grains of 65 μm.

Layers identified in tantalum - 10 w/o tungsten that had been immersed in uranium are (ref. 2): outer uranium, uranium-tantalum (U/Ta ~ 1, 0.3 w/o W), precipitated tantalum (<1 w/o U, down to 1-2 w/o W), and inner Escher-type grains of tantalum-tungsten (up to 18 w/o W) and of uranium (<2 w/o Ta, <0.4 w/o W). In the tantalum - 10 w/o tungsten foil which underwent reaction with liquid uranium at 1300°C for 20 hours, the layer thicknesses are: Uranium-tantalum - 25 μm, precipitated tantalum ~ 55 μm, and inner - 140 μm. The Escher-type tantalum - 15 w/o tungsten grains are 20÷50 μm in size.

For tungsten immersed in uranium (ref. 3), the penetration of liquid uranium into the tungsten foil takes place along grain boundaries forming tungsten grains bathed in liquid uranium. For higher temperatures and longer reaction times the local uranium bath tends to pull off tungsten grains into the original uranium liquid. In the tungsten foil which reacted with liquid uranium at 1300°C for 20 hours, a local uranium-bath layer, about 50 μm thick, is observed with 15-μm tungsten grains; the average size of the inner tungsten grains is 30 μm.

In order to separate the effects of temperature and of liquid uranium on the internal morphology of foils in the three cases, microscopy methods (including SEM-EDAX) have been employed. A comparison is made between as-received foils, on the one hand, and heat-treated foils and uranium-reacted foils, both after 20 hours at 1300°C, on the other hand.

2. Experimental details

Metallurgical-grade, 0.3-mm-thick foils of tantalum metal, tantalum - 10 w/o tungsten alloy, and tungsten metal, containing less than 0.1 w/o impurities, were used in the measurements. For the immersion experiments uranium chunks, containing less than 0.3 w/o impurities, were used.

The tantalum samples were cut from 600×200 mm^2 sheets. The as-received and heat-treated samples were flat, weighing ~6 grams. A cylindrical sample, 44 mm long, weighing 5.5 grams, was used for the immersion in liquid uranium.

The tantalum - 10 w/o tungsten samples were cut from 600×400 mm^2 sheets. The as-received and heat-treated samples were flat, weighing ~6.4 grams. A scoop-shaped sample, 44 mm long, weighing 5.5 grams, was used for the immersion in liquid uranium.

The tungsten samples were cut from 450×300 mm^2 sheets. The as-received and heat-treated samples were flat, weighing ~5.4 grams. Due to the poor workability of tungsten (at room-temperature), a flat sample, 65 mm long, weighing ~4.3 grams, was used for the immersion in liquid uranium.

The heat-treatment and the immersion in liquid uranium were carried out in a smelting furnace adapted for experiments on the immersion of solid metals in liquid metals (ref. 4). Zircoa magnesia-stabilized zirconia crucibles (50-mm high, with 40-mm inner diameter, and a 5-mm-thick wall, standing in protective graphite crucibles) were used. The foils were placed in empty crucibles for heat-treatment. These crucibles contained about 750 grams of uranium for the immersion experiments.

The samples used for immersion were attached with a tantalum wire (1 mm in diameter) to a molybdenum rod (8 mm in diameter) prepared according to the sample shape. The molybdenum rod was connected to a stainless-steel feed-through (6 mm in diameter) of a Wilson seal positioned on top of the resistance-heated vacuum furnace.

Heating the furnace to 1300°C lasted 5-8 hours. Cooling from this temperature took at least 7 hours. The immersion of the foils in liquid uranium was rapid (1 s); the pull-out following the reaction was slower (45 min, at a rate of 1 mm/min), to drain off the excessive liquid. Heat-treatments and reactions were performed for 20 hours. For tantalum foils, heat-treatment for 40 hours, as well as 20-hour annealing followed by immersion for 20 hours, were also performed.

The temperatures were read from the W - 5% Re/W - 26% Re thermocouple that controlled the furnace. Further details of the setup are given elsewhere (ref. 4).

Foil samples were examined:

(a) in the as-received state;

(b) after heat-treatment at 1300°C for 20 hours (and tantalum also for 40 hours);

(c) as-received and immersed in liquid uranium at 1300°C for 20 hours;

(d) annealed for 20 hours and immersed for 20 hours in liquid uranium at 1300°C.

The samples were cut perpendicularly to the foil surface. The reacted foils were generally cut in regions covered by uranium due to immersion and climb. After mounting in plastic, and polishing and etching, the samples were examined under an optical microscope and a scanning electron microscope with EDAX microanalysis capability. Samples were examined parallel and perpendicular to the rolling directions. No noticeable differences were found.

3. Results

3.1. As-received foils

Optical micrographs of sections of 0.3-mm-thick, as-received foils of tantalum, of tantalum - 10 w/o tungsten, and of tungsten are shown in Figs. 1, 2, and 3, respectively.

As-received tantalum contains no flow lines, and its average grain size is 20 μm (Fig. 1).

As-received tantalum - 10 w/o tungsten has numerous flow lines and its average grain size is 30 μm (Fig. 2).

As-received tungsten is dominated by numerous flow lines and its grains cannot be discerned (Fig. 3).

Figure 1. Tantalum foil, 0.3-mm-thick, as-received - optical micrograph (×200).
Average grain size: 20 μm.
No flow lines.

Figure 2. Tantalum - 10 w/o tungsten foil, 0.3-mm-thick, as-received - optical micrograph (×200).
Average grain size: 30 μm.
Numerous flow lines.

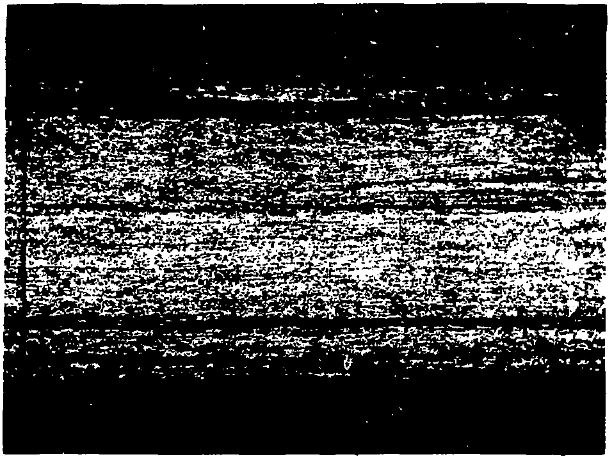


Figure 3. Tungsten foil, 0.3-mm-thick, as-received - optical micrograph (×200).
Grains: undistiguishable.
Numerous, dominant flow lines.

3.2. Heat-treated foils

Optical micrographs of sections of 0.3-mm-thick foils of tantalum, of tantalum - 10 w/o tungsten, and of tungsten, heat-treated at 1300°C for 20 hours, are shown in Figs. 4, 6, and 7, respectively. An optical micrograph of a section in a tantalum foil, 0.3-mm-thick, heat-treated at 1300°C for 40 hours, is shown in Fig. 5.

Tantalum, after heat-treatment at 1300°C for 20 hours, is similar to as-received material: it has no flow lines and its average grain size is 20 μm (Fig. 4). Extending the heat-treatment at 1300°C to 40 hours does not change the internal morphology (Fig. 5).

Tantalum - 10 w/o tungsten, after heat-treatment for 20 hours at 1300°C has only few flow lines but its average grain size, 30 μm (Fig. 6), is similar to that in as-received material.

Tungsten, after heat-treatment for 20 hours at 1300°C, looses all flow lines, which are so dominant in as-received material. The grains show up and they are rich in deformation twins. The average grain size following the heat-treatment is 15 μm (Fig. 7).

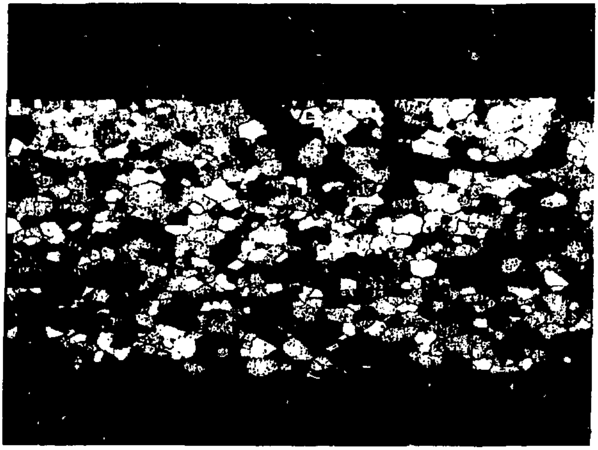


Figure 4. Tantalum foil, 0.3-mm-thick, after heat-treatment at 1300°C for 20 hours - optical micrograph (×200).
Average grain size: 20 µm.
No flow lines.

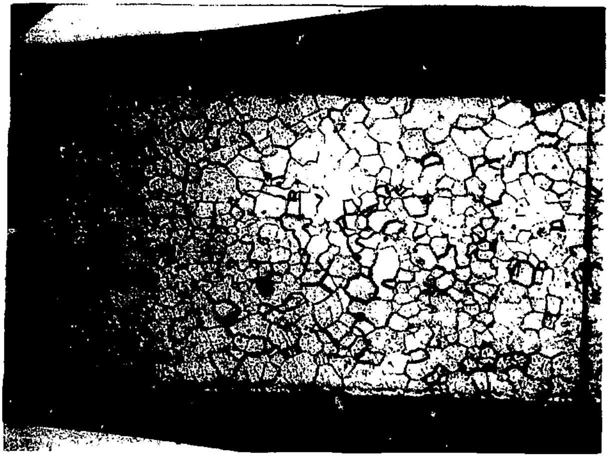


Figure 5. Tantalum foil, 0.3-mm-thick, after heat-treatment at 1300°C for 40 hours - optical micrograph (×200).
Average grain size: 20 μm.
No flow lines.

Figure 6. Tantalum - 10 w/o tungsten foil, 0.3-mm-thick, after heat-treatment at 1300°C for 20 hours - optical micrograph (×200).
Average grain size: 30 μm.
No flow lines.

Figure 7. Tungsten foil, 0.3-mm-thick, after heat-treatment at 1300°C for 20 hours - optical micrograph (×200). Grains are rich in deformation lines. Average grain size: 15 μm. No flow lines.

3.3. Foils immersed in liquid uranium

SEM photographs of sections of 0.3-mm-thick foils of tantalum, of tantalum - 10 w/o tungsten, and of tungsten, following immersion in liquid uranium at 1300°C for 20 hours, are shown in Figs. 8, 10, and 11, respectively. A SEM photograph of a section of a tantalum foil, initially 0.3-mm thick, annealed at 1300°C for 20 hours and then immersed in liquid uranium at 1300°C for 20 hours, is shown in Fig. 9.

Following the immersion in liquid uranium the as-received tantalum foil acquires a multilayer structure (Fig. 8), characterized by EDAX as follows (refs. 1,2):

(a) precipitated columnar tantalum layer (contacted by the bulk liquid) - 90 μm thick;

(b) inner uranium layer - 10 μm thick;

(c) inner tantalum layer - 130 μm thick, with a continuous, 10 μm thick intra-grain uranium matrix, and tantalum grains averaging 45 μm.

The tantalum foil, annealed at 1300°C for 20 hours before immersion in liquid uranium, acquires a different multilayer structure (Fig. 9), characterized by EDAX as follows:

(a) precipitated columnar tantalum layer - extremely thin, less than 10 μm thick;

(b) inner uranium layer - about 10 μm thick;

(c) inner tantalum layer - close to 0.5 mm thick, with rather large amount of the continuous intra-grain uranium metrix, and tantalum grains averaging 45 μm, as in the unannealed tantalum foil.

The foil thickens to 0.5 mm as a result of the massive penetration of liquid uranium.

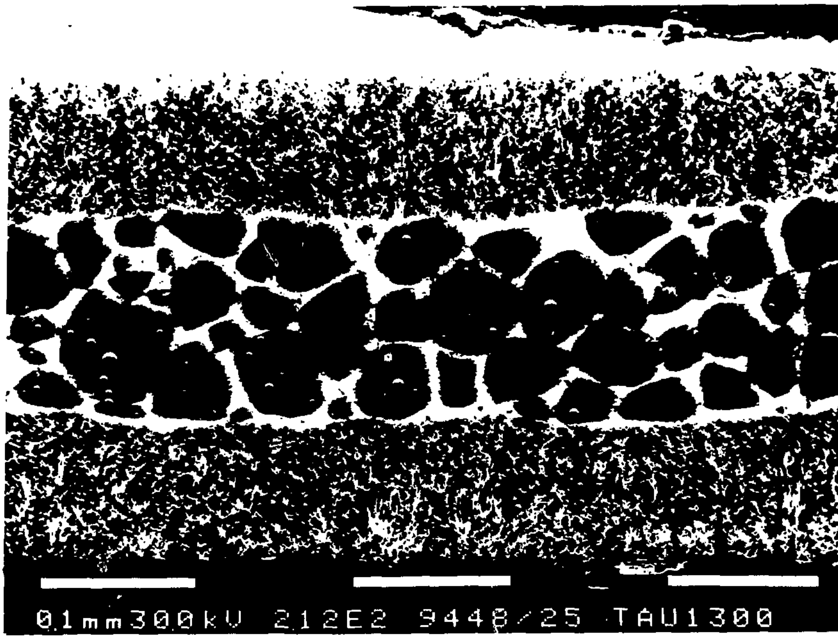


Figure 8. Tantalum foil, 0.3-mm-thick, after immersion in liquid uranium at 1300°C for 20 hours - SEM photograph.
The white bar represents 0.1-mm length.
Multilayer structure, inwards.
outer uranium;
precipitated columnar tantalum (rather thick, ~90 μm);
inner uranium;
inner tantalum, with continuous intra-grain uranium and average grain size of 45 μm.

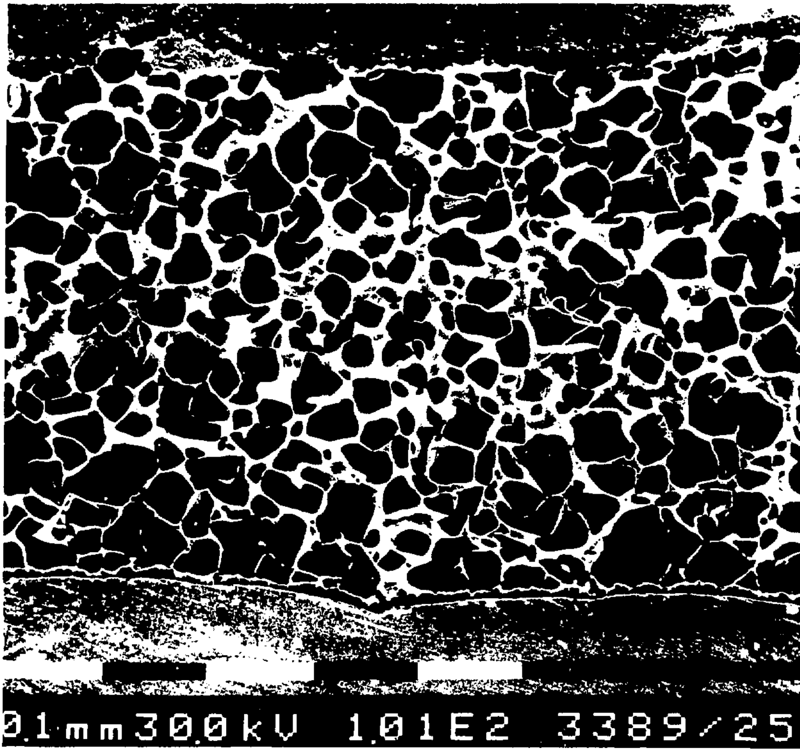


Figure 9. 0.3 mm tantalum foil, heat-treated at 1300°C for 20 hours in vacuum and then immersed in liquid uranium at 1300°C for 20 hours - SEM photograph.
The white bar represents 0.1-mm length
The swollen foil is ~0.5 mm thick.
Multilayer structure, inwards:
outer uranium;
precipitated columnar tantalum (relatively thin, <10 μm);
inner uranium;
inner tantalum, with large amount of intra-grain uranium and tantalum grains averaging 45 μm.

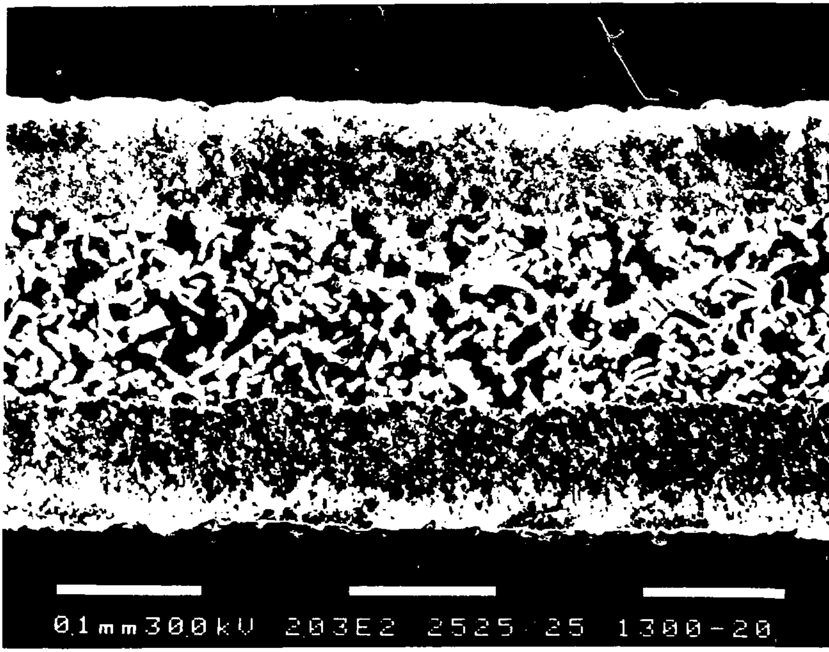
Following the immersion in liquid uranium also the tantalum - 10 w/o tungsten foil acquires a multilayer structure (Fig. 10), characterized by EDAX as follows (ref. 2):

(a) Uranium-tantalum layer (U/Ta ~ 1, 0.3 w/o W) - 25 μm thick;

(b) precipitated tantalum layer (<1 w/o U, down to 1-2 w/o W) - 55 μm thick;

(c) inner layer, 140 μm thick, with Escher-type tantalum - 15 w/o tungsten grain, 20-50 μm in size, and comparable uranium puddles (<2 w/o Ta, <0.4 w/o W).

Following the immersion in liquid uranium, the tungsten foil, contrary to the above mentioned cases, does not acquire a multilayer structure (Fig. 11). Penetration of liquid uranium takes place, along grain boundaries, and a 50-μm thick local uranium bath is seen to contain tungsten grains, 15 μm in average size. Inner tungsten grains, grown to 30 μm, are observed (ref. 3).



<u>Figure 10.</u> Tantalum - 10 w/o tungsten foil, 0.3-mm-thick, after immersion in liquid uranium at 1300°C for 20 hours - SEM photograph.
The white bar represents 0.1-mm length.
Multilayer structure, inwards:
outer uranium;
uranium-tantalum (0.3 w/o W);
precipitated tantalum (1-2 w/o W);
inner, with Escher-type Ta - 15 w/o W grains in uranium matrix.

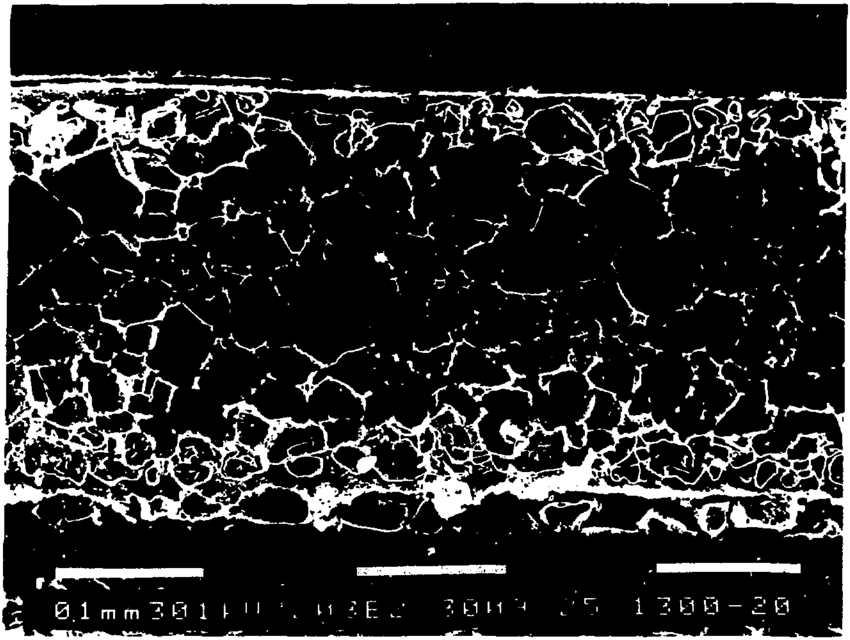


Figure 11. Tungsten foil, 0.3-mm-thick, after immersion in liquid uranium at 1300°C for 20 hours - SEM photograph.
The white bar represents 0.1-mm length.
No multilayer structure.
A continuous uranium phase lines all the grain boundaries.
Average grain size: 30 µm.
No flow or deformation lines.

3.4. Summary

The results concerning average grain size and flow line density in tantalum, in tantalaum - 10 w/o tungsten, and in tungsten are summarized in Table 1.

Table 1. The average grain size and flow line density in foils of tantalum, tantalum - 10 w/o tungsten, and tungsten: as-received, heat-treated for 20 hours at 1300°C, and after immersion in liquid uranium at 1300°C for 20 hours.

Metal	As-received foils		Heat-treated foils after 20 hours at 1300°C		Foils after immersion in liquid uranium for 20 hours at 1300°C	
	Average grain size (µm)	Flow lines	Average grain size (µm)	Flow lines	Average grain size (µm)	Flow lines
tantalum	20	none	20[a]	none	45[b]	none
Ta - 10 w/o W	30	numerous	30	few	20-50[c]	none
tungsten	-[d]	dominant	15	none	30	none

a. No change after 40-hour heat-treatment at 1300°C.

b. No change for immersion succeeding annealing at 1300°C for 20 hrs.

c. Escher-type grains.

d. Undistinguishable.

4. Discussion

From Figs. 1-2, 4-6, 8-10, and Table 1 it follows that the effect of heat-treatment at 1300°C (for 20 hours) on the internal morphology of tantalum metal and tantalum - 10 w/o tungsten alloy is negligible (with reduction in flow line density in the latter case); in tantalum it is found to be negligible even after 40 hours. Therefore the multilayer structures in foils of tantalum and tantalum - 10 w/o tungsten after immersion for 20 hours in liquid uranium at 1300°C are caused by the diffusion of liquid uranium, and not by the healing of the substrate. The above probably holds for the entire 1160-1350°C temperature range for times up to 20 hours (refs. 1,2). The prior state of annealing of the foil is extremely important, as shown by "as-received" tantalum at 1300°C.

A mechanism for the formation of the multilayer structure is proposed elsewhere (ref. 2). The liquid uranium penetrated to the intergranular boundaries in these foils is an intermediary agent, dissolving the original material and precipitating it in a more stable, unstrained crystallographic structure, having a lower surface energy. In the tantalum foil this entails the growth of a columnar tantalum layer and the inner tantalum grains, with a liquid uranium film engulfing them. In the Ta - 10 w/o W foil, in addition to the layer formation, a differentiation of the dissolved tungsten occurs. The precipitated tantalum is strongly depleted and the inner layer is heavily enriched (up to 15 w/o) in tungsten, with the formation of Escher-type Ta-W grains separated by similar uranium puddles. As the effect of temperature alone on the grain size in the foils is rather minimal, the drastic changes in grain size and internal morphology are the results of their reaction with liquid uranium.

From Figs. 3, 7, 11 and Table 1 we conclude that in the case of the tungsten foil the internal morphology depends on temperature. Heat-treatments at other temperatures also affect the internal morphology of the foils (ref. 3).
The effect of temperature adds to the effect of the penetrating uranium in establishing the quite regular internal morphology of the tungsten foil.

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בהוצאת מה"ל - פרסומים