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THERMOCHEMICAL DEPOSITION OF REFRACTORY METALS, ALLOYS,
AND COMPOUNDS FOR APPLICATION IN THERMIONIC DEVICES

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by

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THERMOCHEMICAL DEPOSITION OF REFRACTORY METALS, ALLOYS, AND COMPOUNDS FOR APPLICATION IN THERMIONIC DEVICES*

J. I. Federer, R. L. Heestand, F. H. Patterson, and C. F. Leitten, Jr.**

Abstract.—Thermionic devices impose rigid requirements on refractory materials technology in relation to fabrication, purity, and physical characteristics. Thermochemical deposition of refractory metals, alloys, and compounds is being investigated as a technique for solving these materials problems. Tubular and sheet deposits of tungsten, rhenium, molybdenum, and tungsten-rhenium alloys are being prepared by hydrogen reduction of the metal hexafluorides in the temperature range 500 to 1000°C at pressures of 5 to 20 torr. Factors affecting deposition rates, purity, and grain orientation are being studied. High-purity tungsten sheet of uniform thickness has been prepared at about 570°C. Tungsten deposited below 800°C is highly oriented with {100} parallel to the deposition surface. Joining of thermochemically deposited and wrought tungsten by deposition has been demonstrated by the preparation of joints in sheet and tubing. Tungsten-rhenium alloys deposited in a static hot zone at temperatures of 450 to 700°C are inherently nonuniform in composition and deposition rates due to relative differences in the stabilities of WF_6 and ReF_6 . A moving hot-zone technique shows promise of alleviating this problem. Uranium dioxide having oxygen-to-uranium ratios ranging from 2.001 to 2.166 has been deposited by reaction of UF_6 with hydrogen and water vapor or oxygen at 1250 and 1300°C and pressures of 3 to 6 torr. Both bulk solids and extremely fine powder (60 to 200 Å) have been prepared. Stoichiometric silicon carbide has been deposited at a rate of 10 mils/hr by hydrogen reduction of $SiCl_4$ in the presence of methane at 1300°C and a pressure of 100 torr. In general, thermochemical deposition is a unique method for fabricating many refractory materials of high density and purity and requiring a minimum of finishing operations.

Introduction.—The high temperatures to which thermionic components are subjected limit the choice of materials to refractory metals, alloys, and compounds. High-temperature stability and compatibility, purity, and other physical properties further limit the selection. Finally, fabrication and assembly of components constitute an important aspect of materials selection because thermionic devices require many elements fabricated to close tolerances. By analogy with nuclear fuels, fabrication costs will surely exceed materials costs in thermionic elements. Satisfactory fabrication methods are needed, therefore, for materials which appear to be suitable for thermionic application.

Refractory materials fabricated by conventional techniques often possess defects which render them unsuitable for many applications. These defects include high-impurity content, low density, and structural instability. The purpose of this paper is to discuss an alternate fabrication method for refractory materials termed thermochemical, or

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vapor deposition. The process refers to the chemical interaction of gaseous compounds at a heated surface to form a solid deposit and a gaseous by-product. Thermochemical deposition is capable of fabricating refractory materials of high density and purity to required shapes and tolerances, thus minimizing finishing operations. Also, the process is generally conducted at lower temperatures than other fabrication methods, thereby reducing equipment costs and decreasing the possibility for contamination. Many materials suitable for use in thermionic devices may be fabricated by this method.

In order to fully exploit the potential of thermochemical deposition, the ORNL program involves the fabrication of free-standing refractory bodies such as tubing and sheet, coatings, and joints between refractory materials. Included in this program are tungsten, rhenium, molybdenum, tungsten alloys, uranium dioxide, and silicon carbide — all candidate materials for thermionic application. Characterization of the physical and mechanical properties of the products are being correlated with deposition conditions. Through these studies, a greater understanding of the deposition process and control of material characteristics is being achieved. In following sections, the deposition process and characteristics of each product are discussed.

Tungsten. — A refractory metal of prime interest as a structural material and/or emitter is tungsten. Usually the metal is deposited as tubes of round or square cross section, but the process is no less applicable to a variety of geometrical shapes. The apparatus used for depositing tungsten has been described elsewhere¹ and is shown schematically in Fig. 1. Hydrogen and tungsten hexafluoride (WF_6) are metered into a

Fig. 1. Tungsten Deposition Apparatus.

mandrel contained in the main deposition furnace where hydrogen reduction of the halide occurs according to the reaction

The products of this reaction are solid tungsten which forms on the wall of the mandrel and gaseous HF. Other components of the system include a scavenger furnace maintained at 900°C for reducing any halide which passes unreacted through the main deposition furnace, a vacuum pump for maintaining the desired system pressure, a cold trap for condensing pump oil vapor and minimizing carbon contamination of the deposit, and a water scrubber for HF. Deposition equipment for other refractory metals, alloys, and compounds are similar to this system.

Deposition conditions can be readily varied and include temperature, pressure, composition of gases, flow rates, substrate or mandrel material, and geometry. The deposition conditions used in this study are contained within the limits indicated below.

Temperature	500 to 1000°C
Pressure	5 to 20 torr
Hydrogen-to-WF ₆ ratios	15:1 to 70:1
Flow rates	
H ₂	1000 to 3000 cm ³ /min
WF ₆	15 to 200 cm ³ /min
Substrate	Copper, molybdenum
Geometry	3/4-in. OD × 12 in. long or 1 1/2 in. square × 18 in. long

If varied independently, any of these conditions can affect the character of the deposit. The effect of temperature on deposition rates is shown in the data of Fig. 2 which encompass a typical set of conditions. The deposition profiles - deposition rates vs distance from the inlet of the reaction furnace - show that WF₆ is rapidly reduced by hydrogen at 800 and 1000°C, resulting in significantly higher deposition rates near the inlet than farther downstream. At lower temperatures, the gas mixture is not so quickly depleted in WF₆ and the profiles flatten. Thus, uniformity of deposit thickness is favored by lower deposition temperatures for tubular geometry. Similar profiles are obtained for various conditions within the limits previously indicated except as will be discussed.

The process was scaled up to produce sheet material for a mechanical properties evaluation. The tungsten was deposited on the inner surfaces of a 1 1/2- × 1 1/2- × 18-in. long mandrel prepared by sintering packed molybdenum powder. Molybdenum was substituted for copper to minimize distortion due to a larger difference in thermal expansion coefficients between copper and tungsten. Subsequently, the molybdenum is dissolved from the tungsten in a nitric-sulfuric acid solution. The deposition conditions were 555°C at the inlet and 585°C at the outlet of the reaction zone, 200 cm³ of WF₆ per min, 3000 cm³ of H₂ per min, and 5 torr pressure. The gases were preheated to 350°C before entering the deposition zone. Under these conditions, the deposition rate is about 3 mils/hr, and the thickness variation of nominal 1/16-in. thick deposits is ±0.004 in. over a 14-in. length.

An initially high nucleation rate results in numerous small grains in the material adjacent to the substrate. Preferred growth directions are quickly established, however, and growth continues as

Fig. 2. Deposition Rate of Tungsten vs Distance from the Inlet of the Reaction Furnace.

columnar grains having the long axis of each perpendicular to the substrate. Viewed in cross section, that is, parallel to the substrate, the grains have an average diameter of 0.1 mm or less. The deposit faithfully reproduces the smooth surface of the substrate, which is an economic advantage in many applications. The last deposited surface is less smooth because grain growth is not uniform on a microscale. Transmission electron microscopy reveals that the grain structure of material deposited below 800°C is highly oriented with {100} parallel to the substrate. In deposits prepared above 800°C, the {100} orientation is retained locally, but grains having various high index planes parallel to the substrate are mostly observed.

Recent annealing studies by Mills *et al.*² show that the columnar grain structure is thermally stable. If the structure is highly oriented, little or no growth occurs during heat treatments of 100 hr at 1800°C, 15 hr at 2000°C, 15 hr at 2200°C, and 1 hr at 2500°C. In studies of the effect of deposition conditions and heat treatments on orientation, McMurray *et al.*³ found that {100} orientations were stable after 2 hr at 2100°C. Wrought structures, however, recrystallize and undergo extensive grain growth during similar heat treatments.

Chemical analyses show that the material is quite pure. A typical analysis is presented in Table 1. General vacuum tightness and system cleanliness are responsible for the low contents of interstitial impurities, while the low deposition pressures may minimize the fluorine content.

Table 1. Typical Analysis of Thermochemically Deposited Tungsten

Impurity	Concentration (ppm)
H	2
O	7
F	7
N	< 5
C	< 10
B	1-10
Cu	0.1-10
Mg	1-10
Si	1-100

In many applications, refractory materials must be joined to complete an assembly. Preliminary results indicate that satisfactory joints in thermochemically deposited and wrought tungsten can be obtained by deposition of tungsten if the joints are properly designed. The characteristic columnar growth of deposited tungsten affects the integrity of the joint. A 45° plane of weakness occurs at the intersection of columnar grains growing from intersecting surfaces oriented at 90° with each other. Increasing the included joint angle beyond 90° decreases the angle of intersection between columnar grains growing from the mating surfaces and minimizes the tendency for generation of a plane of weakness. Using this principle, sound joints were prepared in wrought tungsten sheet. Metallographic examination of the bonds showed no evidence of voids, fractures, or impurity concentrations. Tube joints were impervious to helium when tested with a helium leak detector set at a sensitivity of 10^{-8} cm³/min.

The results of these experiments have demonstrated that sound joints can be prepared by thermochemical deposition, and that the technique warrants further investigation.

Rhenium. - Rhenium ranks high among the refractory metals considered for thermionic emitters in an atmosphere of cesium vapor. Deposition studies are being extended, therefore, to establish procedures for fabricating rhenium as free-standing shapes, or coatings, on other refractory materials. The deposition reaction is the hydrogen reduction of rhenium hexafluoride. Equipment similar to that shown in Fig. 1 is used for these studies. Good quality rhenium deposits are not as easily obtained as tungsten, although the deposition processes are similar. The deposition conditions that have been studied are included within the limits indicated below.

Temperature	400 to 800°C
Pressure	2 to 50 torr
Hydrogen-to-ReF ₆ ratios	10 to 20
Flow rates	
H ₂	250 to 2000 cm ³ /min
ReF ₆	10 to 25 cm ³ /min
Substrate	Copper
Geometry	3/4-in. OD × 12 in. long

Although the optimum conditions for deposition of uniformly thick, high-density metal have not been determined, ductile coatings of high density metal, 1 to 2 mils thick and 10 in. long, have been deposited at 600°C with a hydrogen-to-ReF₆ ratio of 12:1 and at a system pressure of 10 torr. The flow rates were 250 cm³ of H₂ per min and 20 cm³ of ReF₆ per min. The deposition rate was about 0.5 mils/hr, and the reduction efficiency was 25%.

Preliminary x-ray diffraction data show that the grain structure of rhenium deposits is randomly oriented. The typical grain structure is columnar, although the grains are less needlelike than columnar tungsten grains.

Molybdenum. - Molybdenum also is amenable to thermochemical deposition by hydrogen reduction of molybdenum hexafluoride (MoF₆). Preliminary results indicate that slightly higher temperatures are required for this reaction than for either tungsten or rhenium deposition. The grain structure of molybdenum deposited at 800°C, hydrogen-to-MoF₆ ratio of 40:1, and a system pressure of 10 torr is columnar and similar in appearance to vapor-deposited tungsten.

Tungsten Alloys. - Tungsten alloys also are being prepared by thermochemical deposition. Initial efforts are directed towards alloy systems wherein improved ductility has already been established in material fabricated by conventional techniques. Tungsten-rhenium alloys are being deposited by the simultaneous hydrogen reduction of WF₆ and ReF₆.⁴ The specific objective of these studies is to determine the conditions required to fabricate alloys of any specific composition.

These studies show that the relative ease of reduction of ReF₆ compared to WF₆ results in nonuniformity of composition and thickness in alloy deposits. The deposits are richer in rhenium near the inlet of the reaction zone. In general, deposition rates are also greater near the inlet than for unalloyed tungsten, correlating qualitatively with the higher rhenium content in that region. The axial variation in rhenium content for several deposition conditions is shown in Fig. 3 wherein the rhenium content of the deposits is plotted vs distance from the inlet. The recovery values shown were determined by comparing the metal content of both metered fluorides with the weight of deposit obtained in the main reaction zone.

The curve labeled WRe-16 is typical of deposits prepared at 500 to 700°C - exhibiting a high rhenium content near the inlet followed by a rapid decrease in rhenium with distance from the inlet. The curves labeled WRe-18 and WRe-19 represent deposits prepared under the same conditions as WRe-16 except that argon was included with the reacting gases. Although the mechanism is not understood, argon causes the rhenium to be distributed differently in the deposits. The rhenium content in WRe-19 was substantially increased throughout the deposit and was constant over the distance 5 to 12 in. from the inlet. The presence of argon, however, lowered the metal recovery from 89 to 54%. Lowering the temperature to 450°C also caused a significant change in rhenium distribution as shown by the curve labeled WRe-20. The very high rhenium content near the inlet reflects the greater ease of reduction of ReF₆ compared to WF₆. Analysis of the composition profiles for several deposits reveals that a greater-than-proportionate amount of rhenium was recovered than tungsten.

Fig. 3. Composition of Tungsten-Rhenium Alloys vs Distance from the Inlet of the Reaction Furnace.

To eliminate the axial variation of rhenium content and deposition rates, a moving hot zone has been substituted for the static hot zone. The desired alloy composition is obtained by control of the gas composition and the desired thickness is obtained by control of the rate of movement of the hot zone, temperature, and feed rate. The experimental arrangement utilizes an induction coil about 1 in. long, a gas preheat furnace, and a mechanism for slowly moving the deposition mandrel through the induction coil and preheat furnace. Preliminary experiments were conducted under the following conditions.

Deposition temperature	700 to 750°C
Gas preheat temperature	300°C
Hot zone movement	3/4 in./hr
Flow rates	
H ₂	250 and 500 cm ³ /min
WF ₆	15 and 30 cm ³ /min
ReF ₆	5 and 10 cm ³ /min
Pressure	10 torr

Deposition rates of about 5 mils/hr and 100% efficiency of reduction are obtained under these conditions. A typical analysis of a deposit prepared with a moving hot zone is presented in Table 2. Although the process is not yet optimized, these results indicate the potential for obtaining uniform compositions by this technique.

The interstitial content of alloy deposits is similar to that of unalloyed tungsten (Table 1). The grain structure is typically columnar with a tendency towards more needlelike grains in alloys containing greater than 25% Re. Hardness values increase from 450 DPH for

Table 2. Typical Analysis of Tungsten-Rhenium Deposit Prepared with a Moving Hot Zone

Distance Along Length of Deposit (in.)	Rhenium (%)
1	10.6
2	21.8
3	26.8
4	24.4
5	22.4
6	18.7

unalloyed tungsten to about 1800 DPH for alloys containing 25 to 30% Re. X-ray diffraction results indicate that the hardness increase is associated with an increasing amount of a beta-tungsten type structure (cubic) which coexists with the body-centered cubic solid solution of rhenium in tungsten. The solid solution is the principal phase in low rhenium deposits, while the beta-tungsten type structure is favored by high rhenium contents. The beta-tungsten structure commonly occurs as an intermediate phase in transition-metal systems,⁵ but does not appear in previously reported tungsten-rhenium phase diagrams.^{6,7} These diagrams, however, have not been established below 1200°C, whereas the deposits were prepared in the temperature range 500 to 750°C. Neither sigma phase, which is reported to occur in alloys containing from 26 to 63% rhenium, nor elemental rhenium has been found in any of the as-deposited alloys.

Fuel Compounds. - In addition to the fabrication of structural components, thermochemical deposition of refractory fuel compounds is being investigated.⁸ The main objective of this program is to investigate methods for the direct fabrication of refractory fuel compounds from gaseous metal halides. Initial studies have concentrated on the thermochemical deposition of UO₂ by the reaction of hydrogen and oxygen, or steam, with UF₆ as follows:

This reaction has been proven thermodynamically feasible⁹ when accomplished in steps as shown in reaction paths (3) and (4) or (5) and (6).

The overall reaction (2) is believed to proceed simultaneously by the paths shown in (3) and (4) or (5) and (6). Thermodynamic and kinetic factors favoring direct reduction are (1) low system pressure, (2) temperatures sufficiently high to limit formation and condensation of intermediate compounds, and (3) excess amounts of both hydrogen and oxygen to drive the reactions to completion.

Experiments are conducted in equipment essentially similar to that shown in Fig. 1. A water-cooled injector admits UF_6 directly into the hot zone to prevent condensation of intermediate products UF_4 or UO_2F_2 in the cold zones. Argon is used as a blanket to separate the reactants at the injector tip and to prevent buildup of UO_2 on the injector.

The deposition process has not been optimized in regard to all variables; however, the conditions and results of several deposition experiments presented in Table 3 indicate certain trends. Under the first set of conditions presented in Table 3, a dendritic, crystalline deposit is obtained. Deposits of this type have oxygen-to-uranium ratios of 2.000 to 2.003 and contain less than 10 ppm fluorine. Reducing the pressure to 2 to 3 torr and increasing the UF_6 content of the gases to greater than 3 vol % results in a theoretically dense, uniform deposit. Tubes of this material having a wall thickness of about 40 mils were deposited at a rate of 6 mils/hr.

Stoichiometry in bulk UO_2 can be controlled by adjusting the feed gas composition for a given temperature and pressure. The effect of gas composition on the oxygen-to-uranium ratio at a deposition temperature of 1300°C and pressure of 3 torr is shown in Table 3. The lowest oxygen-to-uranium ratio was obtained for a gas composition relatively high in UF_6 and low in oxygen. The deposits having oxygen-to-uranium ratios of 2.008 and 2.116 contained a second phase identified as U_4O_9 , whereas the deposit having an oxygen to uranium ratio of 2.001 was single phase. Oxygen-to-uranium ratios are constant along the length of a given deposit.

Table 3. Conditions and Results of UO_2 Deposition Experiments

Temperature ($^\circ\text{C}$)	H_2 (vol %)	O_2 (vol %)	UF_6 (vol %)	Pressure (torr)	Oxygen-to- Uranium Ratio	Description
1250	78	20 ^a	2	6	2.000 2.003	Dendritic. Crystallites are red and translucent under intense light. Readily crushed to powder.
1300	78	17 ^a	5	3	2.019	Dense (10.96 g/cm^3), uniform thickness. Deposition surface and fractures appear glassy.
1300	86	10	4	3	2.008	Two-phase structure of UO_2 and U_4O_9 .
1300	81	12	7	3	2.166	Two-phase structure of UO_2 and U_4O_9 .
1300	85	9	6	3	2.001	Single-phase structure.
1300	82	16	2	6	2.001	60 to 200 A powder.

^aAs water vapor.

Uranium dioxide powder is obtained by withdrawing the injector from the hot zone so that gas mixing occurs at a temperature of 900 to 1000°C. UO_2F_2 "snow" is assumed to form on mixing, subsequently passing into the 1300°C hot zone where reduction to UO_2 occurs. Uranium dioxide powder, 60 to 200 A in diameter, was produced at 6 torr with a gas composition of 16 vol % steam, 82 vol % H_2 , and 2 vol % UF_6 . The oxygen-to-uranium ratio of 2.074 was readily reduced to 2.001 by heat treatment in hydrogen at 1000°C for 4 hr.

Within the gas composition limits established for depositing bulk UO_2 , less than 1% of the feed material passes through the system as intermediate or unreacted fuel compounds. Although UO_2 is formed in the hot zone when UF_6 concentration in the feed material is raised above 10%, some intermediate uranium compounds pass through the system into the cold trap. For steam or oxygen concentrations in excess of 40%, trace amounts of intermediate uranium compounds are again found in the cold trap. Raising the system pressure above 20 torr yields UO_2F_2 powder under conditions that produce only UO_2 at lower pressure. Within the current limits of investigation, temperatures above 1300°C give no significant deposition advantage.

Silicon Carbide. — Oxidation-resistant coatings are required for protection of refractory materials in numerous applications, for example, gas-fired thermionic converters. Thermochemical deposition of SiC has been investigated in order to fill this need. Deposition of the carbide has been accomplished by the hydrogen reduction of silicon tetrachloride (SiCl_4) in the presence of methane (CH_4) in equipment similar to that shown in Fig. 1 and described elsewhere.¹⁰ A typical set of deposition conditions follows.

Temperature	1300°C
Pressure	100 torr
Flow Rates	
H_2	2200 cm^3/min
SiCl_4	35 cm^3/min
CH_4	80 cm^3/min

Stoichiometric silicon carbide has been deposited under these conditions at a rate of approximately 10 mils/hr. Small variations in temperature and hydrogen content of the reacting gases do not affect composition. Adherent coatings have been deposited on graphite, tungsten, and alumina.

The grain structure of vapor-deposited SiC is columnar, resembling pyrolytic graphite. A knoop hardness of about 2750 was measured for this material.

These experiments have demonstrated the feasibility of preparing high-density, stoichiometric SiC as coatings or free-standing shapes by thermochemical deposition. Further studies should include deposition of uniform coatings, compatibility with various refractory materials, and oxidation studies.

In summary, these studies have shown that refractory materials having thermionic applications can be prepared by thermochemical deposition. Specific advantages of the process are the lower temperatures that are required as compared to conventional fabrication processes, the smooth surface finish that can be obtained, the applicability to a variety of

geometrical shapes, and the oriented structures that are possible. The relatively inexpensive equipment and simplicity of the process may provide a further economic advantage in the fabrication of the numerous elements required in a thermionic device.

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