Application of Ultra-high-vacuum Techniques to study the Phase Diagram and Electrical Resistance of Tantalum Hydride down to 0°

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STUDY of hydrogen–metal solution equilibria by pressure–composition–temperature measurements using conventional vacuum techniques becomes impracticable below a certain temperature because of the slow attainment of equilibrium. This effect is in part due to oxides or other contaminants in the surface layers of the metal.[1,2] By using ultra-high-vacuum techniques to preserve a clean surface it has been found possible to study the tantalum–hydrogen system down to 0°, compared with the previously reported[3] lower limit of 164°, and so explore the region below the critical solution temperature for hydride formation at about 50°.

The tantalum in the form of a filament (purity > 99·5%, 20 cm. long 0·025 cm. diameter) was sealed *via* tungsten pins into a Pyrex glass cell immersed in a constant-temperature bath. The cell was evacuated by an ultra-high-vacuum pumping system. The filament was degassed at 2400° K until the cell pressure was below 1×10^{-9} torr, and allowed to cool to the thermostat temperature. The cell was then isolated from the pump and known quantities of hydrogen (B.O.C. 'Specpure' grade) admitted. Initial absorption was fast (over 99% of each addition absorbed within 1 min.) but was followed by a slower uptake. The final approach to the equilibrium pressure was followed by taking intermittent readings on an ionization gauge with a lanthanum hexaboride filament.[4] The changes in resistance of the filament due to hydrogen absorption were also measured. After each run the filament was again degassed in ultra-high vacuum.

Figures 1 and 2 show the pressure and resistance isotherms. The horizontal parts of the pressure isotherms indicate the formation of a hydride with

a limiting composition corresponding to an approximate formula $TaH_{0.35}$. The heat of formation of the hydride from the hydrogen-saturated metal, deduced from an Arrhenius plot of the plateau pressures, is 23 kcal. per mole of hydrogen. There is a sharp drop in resistance near the above composition.

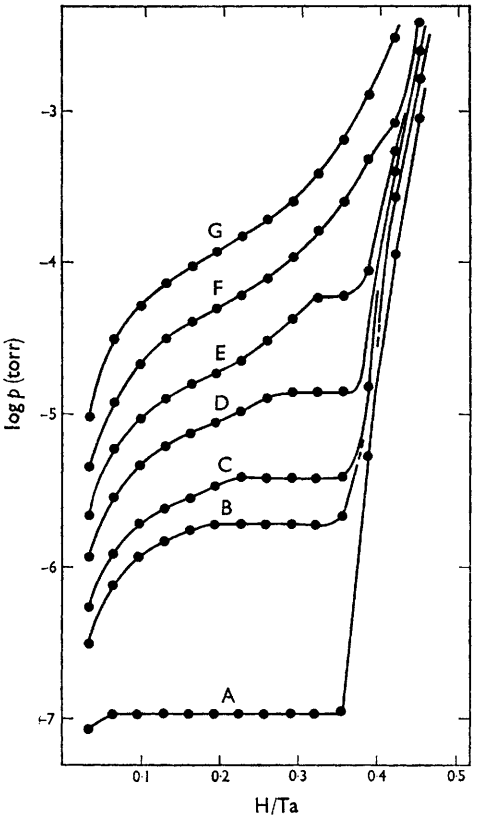

FIGURE 1. *Isotherms of pressure* p (*torr*) *as function of atomic ratio* H/Ta. A = 0°, B = 23·9°, C = 30·2°, D = 40·0°, E = 49·6°, F = 59·8°, G = 69·4°.

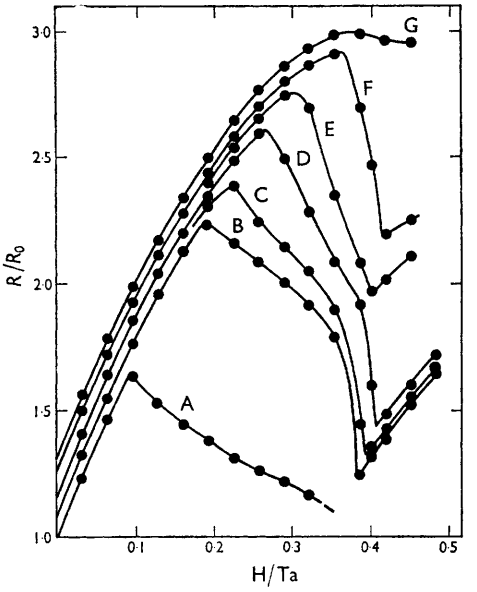

FIGURE 2. *Ratio of resistance of hydrogen-charged filament* (R) *to that of the degassed filament at* 0° (R_0). *Same temperatures as Figure* 1. *Breakage of the embrittled filament occurred during* 0° *run.*

After admission of gas to the cell the longest time taken to reach equilibrium occurs near the completion of the hydride phase. At 0° this required about 1½ hr., but is less at higher or lower concentrations and higher temperatures. No indication of hysteresis phenomena have been observed so far, but one run indicated that slight supersaturation effects may occur in the region of the critical solution point.

No significant uptake of hydrogen by the degassed filament was observed at room temperature under "ordinary" high-vacuum conditions *i.e.*, in the unbaked system with a base pressure of $\sim 10^{-6}$ torr.

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