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PHYSICAL CHEMISTRY

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Abstract

Full Text

PHYSICAL CHEMISTRY

V. I. RAKHOVSKII, A. P. LYUBIMOV, and V. M. GARMASH

ON THE QUESTION OF THE PENETRATION OF SILVER INTO TUNGSTEN

(Presented by Academician P. A. Rebinder, 22 VI 1960)

The service life of contacts in high-current electrical apparatus is determined chiefly by the severity of the operating thermal regime. The latter is determined to a considerable extent by the thermal conductivity and heat resistance of the contact material, as well as by the rate of erosion of the contacts, which depends strongly both on the indicated parameters and on the melting temperature of the contact material. Therefore attempts have more than once been made to create complex multicomponent contact compositions that would combine high thermal conductivity with a high melting point. Silver and tungsten are usually used as the principal elements of such compositions. In this case the question of the possibility of silver penetrating into tungsten, both by diffusion and as a result of the occurrence of some other processes taking place at a temperature of the order of 1000° , plays a very substantial role. The phase diagram of the Ag–W system has not yet been sufficiently studied; moreover, in Hansen's monograph ⁽¹⁾ it is stated that silver and tungsten form no solutions either in the solid or in the liquid state. It should be borne in mind, however, that the works ⁽²⁾ and ⁽³⁾, on the basis of which Hansen's above-mentioned assertion was formulated, were carried out by methods of comparatively low sensitivity.

At the same time, some processes are now known, in particular the coarsening of particles of solid tungsten present in molten silver, which make it possible to suppose that the solubility of tungsten in silver may occur.

In connection with this, using radioactive isotopes we undertook an attempt to establish whether the process of penetration of silver into tungsten can take place. For this purpose a tungsten plate was held in liquid radioactive silver at an elevated temperature for a certain time. Then, on the basis of measuring the activity of the specimen, the total amount of silver that had penetrated into the tungsten was determined.

The experiment was carried out as follows. A plate of rolled technically pure W, measuring $0.015 \times 0.4 \times 1.2 \text{ cm}^3$, was fused into silver enriched with the radioactive isotope Ag^{110} , containing 0.01% Si; 0.01% Fe; 0.01% Cu; 0.001% Al. The block thus obtained was placed in a quartz crucible, which was then introduced into the experimental tube (see Fig. 1). After evacuation to a pressure of the order of 10^{-4} mm Hg, the tube was filled with helium to a pressure somewhat

Fig. 1. Schematic of the experimental tube

Figure 1: Fig. 1. Schematic of the experimental tube

above atmospheric. A tubular furnace, in which the operating temperature had been established in advance, was slid over the prepared system. The tungsten immersed in the liquid radioactive silver was held in it for 4-24 hours. At the end of the holding period, the tungsten plate, by means of a thin tungsten thread that had previously been welded to each specimen, was withdrawn from the silver into the cold zone of the tube.

The temperature of the furnace during the experiment was monitored with a chromel-alumel thermocouple connected to a PPTV-1 potentiometer. The constancy of the working temperature, with an accuracy of $\pm 1.5^\circ$, was maintained manually by means of a LATR-1 autotransformer. The low inertia of the furnace and careful observation of the temperature regime made it possible to ensure the constancy of the working temperature with the accuracy indicated above.

After cooling, the specimen was removed from the experimental tube, and the excess silver covering the tungsten surface was removed by etching the specimen in dilute HNO_3 at a temperature of 40° (in a TS-15 m thermostat). After etching and repeated washing in dilute HNO_3 and distilled water, the specimen was dried and its activity was counted on a VSP-type counting installation. The completeness of removal of the fused-on silver was determined by preliminary experiments, for which a tungsten specimen was fused into silver enriched with the radioactive isotope and was removed from it without holding. Owing to this, penetration of silver into tungsten was practically absent. After the chemical treatment described above, such specimens gave no excess count above background. In addition, we carried out a number of preliminary experiments to estimate the effect of extraneous factors that could have an additional influence on the increase in activity of the specimens under study during preliminary treatment. In the experiment, annealing was carried out at a temperature of 1000° for 8, 16, and 24 h, and at 1080° for 4, 8, 12, and 16 h.

Fig. 1. Schematic of the experimental tube:

1 –tap for raising the specimen; 2 –suspension thread for the plate; 3 –thermocouple; 4 –experimental tube; 5 –crucible; 6 –liquid silver; 7 –tungsten plate; 8 –tubular furnace.

In determining the activity of the specimens we took into account the change in background, the decrease of specimen activity with time, the presence of self-absorption, β -radiation, and the occurrence of secondary β -electrons produced by γ -radiation. The data obtained made it possible to calculate the number of pulses emitted per unit surface area of the specimen per unit time. These data are presented in Fig. 2. Knowledge of the specific activity of silver made it possible, taking into account the counting coefficient, to determine the total amount of silver that had “penetrated” through the surface of the specimen into

Fig. 2. Dependence of specimen activity on annealing time in liquid silver

Figure 2: Fig. 2. Dependence of specimen activity on annealing time in liquid silver

tungsten at a given temperature.

Fig. 2. Dependence of specimen activity on annealing time in liquid silver.

Since even after 24-hour annealing the specimen was still far from saturated with silver, we were unable to relate the silver contained in tungsten to the entire mass of the specimen. For an approximate estimate of the depth of penetration of silver into tungsten, we removed a layer of tungsten 20μ thick from both sides of the plate. It then turned out that the specimen gave no excess count above background. As is seen from Fig. 2, for both temperatures the dependence of the number of pulses counted per unit time from unit surface area on annealing time is linear. This allows one to assert that in the present case diffusion does not take place. Apparently, one may speak

one can speak only of some other penetration process. This process, in contrast to diffusion, is characterized by the constancy of the penetration-rate value at a given temperature; its value reaches $7.62 \cdot 10^{-8} \text{ g/cm}^2 \cdot \text{s}$ at a temperature of 1080° . The activation energy of this process, E , calculated from the angles of inclination of the experimental straight lines, proved to be

$$E = 825000 \text{ cal/g-at.}$$

An additional fact that may help clarify the process of silver penetration into tungsten is the sharp decrease in the strength of tungsten during prolonged annealing in liquid silver, which in a number of cases leads to the destruction of the tungsten plate. Taking this into account, the penetration of Ag into W can be explained by a process of etching of the tungsten grain boundaries by liquid silver. Silver atoms penetrate into the "pores" thus formed. This process should depend linearly on time, which is indeed reflected in the experimental graphs. Similar manifestations of metal destruction in the process of annealing in a foreign liquid metallic phase are also observed in a number of other cases.

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CITED LITERATURE

¹ M. Hansen, *Structures of Binary Alloys*, Moscow–Leningrad, 1941. ² F. A. Bernoulli, *Pogg. Ann.*, **111**, 587 (1860). ³ M. V. Schwarz, *Metall- und Legierungskunde*, Stuttgart, 1929, p. 73.

Note: Figure translations are in progress. See original paper for figures.

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