

INTERMETALLIC DIFFUSION IN GOLD-LEAD SYSTEMS

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Quelques résultats préliminaires sur la diffusion de l'or dans le plomb, obtenus par l'utilisation des traces radioactives, sont présentés.

One of the most typical applications of radioactive tracers in basic solid state physics and metallurgy is in the study of self-diffusion and inter-diffusion phenomena in solids and liquids.

Some results concerning the diffusion of gold in lead are briefly reported here. The general procedure involved is well known: a thin layer of a suitable radioactive isotope is plated onto the specimen to be investigated and, after a sufficiently long annealing interval, at high temperature, the distribution of activity within the specimen is studied by a sectioning and counting technique. It is found that the activity $A(x)$ at a distance x from the surface is given by

$$A(x) = \frac{c}{t^{\frac{1}{2}}} \exp\left(-\frac{x^2}{4Dt}\right)$$

where D is the diffusion coefficient at the temperature T , and c is a constant.

D depends upon temperature according to the law

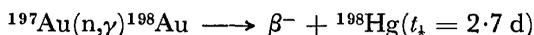
$$D = D_0 \exp\left(-\frac{Q}{kT}\right)$$

Q represents the activation energy for diffusion in the case in question. Previous research on the gold-lead system was done in 1933-1935, mainly by Seith, who used chemical methods in tracing gold atoms in the lead matrix. Rather large amounts of gold were needed. As D is expected to depend upon the mutual concentrations of the two elements it was thought convenient to repeat such measurements, using very small concentrations of radioactive gold.

PRESENT MEASUREMENTS

The use of radioactive tracers provides an extremely sensitive method of identifying very small amounts of gold within lead, and makes it possible to measure the diffusion coefficient of gold in pure lead. In a first run of measurements very pure lead (99.998 per cent) was used in the shape of cylindrical polycrystals 15 mm long and 1 cm in diameter, and of foils $\frac{1}{16}$ mm thick, 1 cm wide and 4 cm long.

Annealing was done in a furnace where the temperature was uniform and stabilized within $\pm 0.4^\circ\text{C}$, between 180°C and 300°C . Owing to the short half-life of ^{198}Au produced in the reaction



a slightly unconventional experimental procedure was used. It was made possible by the small activation cross-section of lead and the short half-lives of the lead isotopes, and was as follows:

- (1) Inactive gold was evaporated onto the specimen.
- (2) Diffusion annealings were done at the different temperatures.
- (3) Diffused samples were activated at Harwell.
- (4) Specimens were sectioned (50μ thick), and the distribution of gold was measured by counting the activity due to the 411 keV γ -rays from ^{198}Au .

The results suggested the following law

$$D = 2.8 \times 10^{-3} \exp\left(-\frac{0.39}{kT}\right) \text{ cm}^2 \text{ sec}^{-1} \quad (Q \text{ in eV})$$

Such results noticeably differ from those given by Seith:

$$\begin{aligned} D_0 &= 2.8 \times 10^{-3} \text{ instead of } 0.4 \times 10^{-3} \\ Q &= 0.39 \quad \text{instead of } 0.60 \end{aligned}$$

The disparity is not due to the effects of surface diffusion, as we found the same results with cylindrical probes and with thin foils. There are three further possible explanations:

(1) The irradiations after diffusion were performed at a low temperature (37°C), but it cannot be excluded that the temperature might have been somewhat higher so that some further diffusion might have taken place during irradiation.

(2) Diffusion at grain boundaries could have been considerable in the polycrystalline specimens. (Some of Seith's specimens were single crystals.)

(3) The fast neutron flux within the reactor might have had the effect of enhancing the diffusion process even at low temperatures. (This effect has been recently noted by Dienes and Damask in α -brass.)

Rough calculations were tried to test (1) and (3); it was concluded that their effect must have been rather small, and have tended to reduce the discrepancy. Further measurements were done by one of us (A.A.) with an entirely different procedure in order to compare diffusion coefficients in single crystals and in polycrystals, and to eliminate any possible spurious effects.

Single lead crystals were grown from high-purity lead as cylinders, 1 cm in diameter, 4 cm long. One of the ends was flattened by alternative sectioning and etching: this method was found to be effective in maintaining the single crystal structure of the specimens.

Carrier-free ^{198}Au ($t_{1/2} \simeq 200 \text{ d}$) was obtained from Philips Roxane after deuteron irradiation of platinum, and was electroplated in very thin films

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onto lead. After annealing, the specimens were sectioned (20μ thick) and the activity due to kx-rays from lead (^{195}Au decays by electron capture to Pb) measured by means of a scintillation spectrometer.

The investigated range of temperatures was widened between the melting point of lead (327°C) and 120°C . The results are not yet complete. The results obtained so far suggest that

$$D_0 \simeq 6.4 \times 10^{-3} \text{ cm}^2 \text{ sec}^{-1}, \quad Q \simeq 0.45 \text{ eV}.$$

It can therefore be deduced that none of the three effects listed changes the measured diffusion rate to any marked extent. Corrections to the previous results would tend to make the two curves coincide, but it is too early to form a definite opinion on this point.

If the two series of results are assumed to be equivalent, the parameters of gold-lead diffusion turn out to be

$$D_0 \simeq 1.2 \times 10^{-2} \text{ cm}^2 \text{ sec}^{-1}, \quad Q \simeq 0.46 \text{ eV}$$

which must probably be considered as the upper limit. We consequently conclude that the activation energy for the diffusion of gold in lead is smaller than previous results seemed to indicate. It is nevertheless remarkable that such reasonably accurate results were obtained even before radioactive tracers were available.