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Diffusion of copper in lead

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This paper presents a reanalysis of experimental data on the diffusion of Cu in Pb. The new values of the activation energy and volume are 8.17 ± 0.11 kcal/mole and 0.16 ± 0.02 atomic volumes, respectively.

In 1972, Candland, Decker, and Vanfleet[1] published the measurement of the effects of pressure on the diffusion of Cu in Pb. They reported an activation energy of $\Delta H = 5.6$ kcal/mole at atmospheric pressure and suggested that Cu diffuses in Pb by a pure interstitial mechanism. Their value of the activation energy, however, is not in good agreement with atmospheric-pressure measurements of Dyson, Anthony, and Turnbull[2] and Miller.[3] In attempting to correlate the measurement of the diffusion of the noble metals in Pb, using an extension of the theory of Miller,[4] we found[5] that the value of 5.6 kcal/mole for ΔH of Cu diffusing in Pb was inconsistent with the data for the other noble metals, while the value of 8 kcal/mole given by Dyson *et al.*[2] was consistent. It seemed that the analysis of the high-pressure data to obtain ΔH must be in error, so I reexamined the original data of Candland *et al.*[1] It was immediately apparent that Candland *et al.*'s earlier atmospheric-pressure measurements were taken prior to their preanneal technique and showed definite non-Gaussian penetration profiles. If I removed these points from their work, their remaining atmospheric-pressure points agreed well with Miller and Dyson *et al.* The lowest-temperature points along each of Candland *et al.*'s isobars had diffusion values higher than the higher-temperature measurements. The corresponding diffusion profiles had less than one decade of penetration and so these points were rejected. Their remaining data points were analyzed along with the atmospheric-pressure points of Miller and Dyson *et al.* to yield the following results[6]: $\Delta H = 8.17 \pm 0.11$ kcal/mole, $D_0 = 0.0086 \pm 0.0009$ cm^2/sec, $\Delta V/V_0 = 0.16 \pm 0.02$, $\delta(\Delta V/V_0)/\delta P = -(2.9 \pm 1.7) \times 10^{-3}$ kbar^{-1}, and $\delta(\Delta V/V_0)/\delta T = (0.8 \pm 0.4) \times 10^{-4}$ K^{-1}. A graph of the data is shown in Fig. 1. Another surprising change in this analysis is the considerably larger value for $\Delta V/V_0$ than previously reported. If Candland *et al.* had realized that this value was so large, they might have been more hesitant in claiming that the diffusion of Cu in Pb was pure interstitial.

After submitting this paper an article[7] was published which also considers Candland *et al.*'s original data. By rejecting data taken prior to the preanneal procedure, which was designed to avoid oxidation problems at the surface, Mundy *et al.*[7] found a value of 7.0 ± 0.5 kcal/mole for the activation energy from Candland *et al.*'s data. Using only the four atmospheric-pressure data points which we included in this reanalysis, we would obtain $\Delta H = 7.9 \pm 0.4$ kcal/mole, which agrees with the conclusions of Mundy *et al.* As they suggested, the earlier measurements of Candland *et al.* may have been distorted by solubility problems.

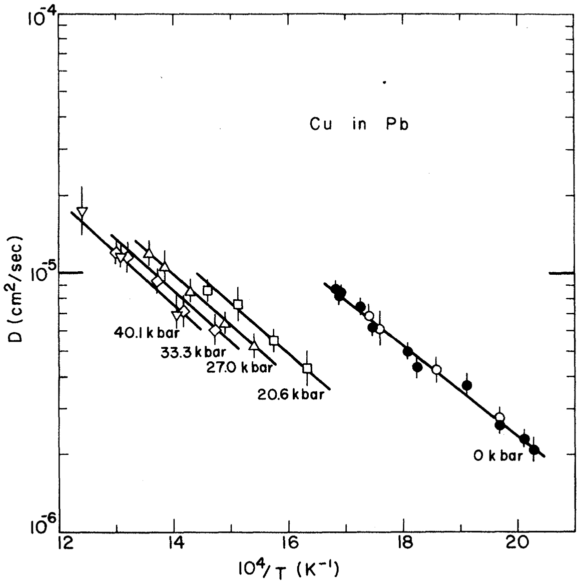

FIG. 1. Temperature dependence of the diffusion of Cu in Pb along several isobars. The solid lines are the computer fit to the data using Eq. (6) in Ref. 6. The open circles at atmospheric pressure are measurements of Ref. 1, while the solid data points come from Refs. 2 and 3. □, at 20.6 kbar; △, at 27.0 kbar; ◇, at 33.3 kbar; ▽, at 40.1 kbar.

[1] C. T. Candland, D. L. Decker, and H. B. Vanfleet, Phys. Rev. B 5, 2085 (1972).

[2] B. F. Dyson, T. R. Anthony, and D. Turnbull, J. Appl. Phys. 37, 2370 (1966).

[3] J. C. Miller, Argonne report (1972) (unpublished).

[4] J. W. Miller, Phys. Rev. 188, 1074 (1969).

[5] D. L. Decker and H. B. Vanfleet, Bul. Am. Phys. Soc. 18, 428 (1973).

[6] J. A. Weyland, D. L. Decker, and H. B. Vanfleet, Phys. Rev. B 4, 4225 (1971).

[7] J. N. Mundy, J. W. Miller, and S. J. Rothman, Phys. Rev. B 10, 2275 (1974). This is the same datum as in Ref. 3.