

Effect of Hydrostatic Pressure on the Self-Diffusion Rate in Single Crystals of Gold*†

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The rate of self-diffusion in gold was measured by the radioactive-tracer-lattice sectioning technique at 860, 910, and 960°C. and at hydrostatic pressures from 2.00 to 9.08 kbar. The diffusion coefficient was observed to decrease with increasing pressure, and the activation volume for self-diffusion was determined to be 7.2 ± 0.4 cm³/mole. This value is in good agreement with the sum of the experimental values of the activation volumes of formation and motion in gold, the experimental values of the activation volumes in other face-centered metals, and many of the theoretical values calculated for copper. The present value also is in fair-to-excellent agreement with the various expressions relating the activation volume to other experimentally derived quantities. The limiting source of error in pressure-diffusion experiments of the present type was found to be in controlling the measurement, reproducibility, and uniformity of the temperature.

I. INTRODUCTION

MANY experiments have been carried out in recent years in an effort to clarify the nature of atomic defects in metals. A series of such experiments by Simmons and Balluffi¹⁻⁴ has convincingly established that the predominant thermally-generated defects in the noble metals and aluminum are vacant sites, reinforcing the already general belief that diffusion-limited phenomena proceed by the vacancy mechanism in these metals. However, many of the details pertaining to the nature of these defects are still not well defined, and there are some important gaps in the experimental values of some of the basic properties of vacancies in metals.

Of particular use in clarifying the detailed nature of the defects in metals are the values of the total activation energy and the total activation volume of the defects, as well as the independently determined values of the energies and volumes of formation and motion. Self-diffusion experiments are appropriate for measuring the values of the total activation energy and volume. The radioactive tracer and sectioning technique may be used for most metals in such measurements, although other techniques are sometimes used, such as measurements of anelastic relaxation in certain alloys, or nuclear-magnetic-resonance techniques in certain metals. Diffusion rates have been measured for many of the pure metals at atmospheric pressure,

yielding values of the activation energy for these metals. Relatively few of these metals have been subjected to self-diffusion studies at elevated pressures because of the technical difficulties involved. For the lower melting-point metals, liquid pressure systems and external furnaces may be utilized. Nachtrieb and co-workers have thus determined the activation volumes for sodium,⁵ alpha-white phosphorus,⁷ and lead.⁸ Hudson and Hoffman⁹ used a "belt"-type pressure system¹⁰ in their determination of the activation volume in lead; they used silicone oil as the pressure fluid and an internal heater to maintain the diffusion temperature. However, self-diffusion measurements of the high-melting-point metals, such as the noble metals, require the use of a gas pressure system to insure both hydrostaticity, and chemical stability of the pressure medium at high temperatures. An internal heater also is required in order to make it possible to maintain the temperature of the pressure vessel sufficiently low by forced external cooling. Tomizuka¹¹ used such an arrangement to determine the activation volume of silver, and Albrecht and Tomizuka,¹² with the apparatus in this laboratory, have recently completed a determination of the activation volumes for diffusion of gold and silver tracers in a gold-34 atomic percent silver alloy.

Most of the theoretical calculations on point defects have been done for copper,¹³ which is representative of the face-centered cubic crystal structure, but for which

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