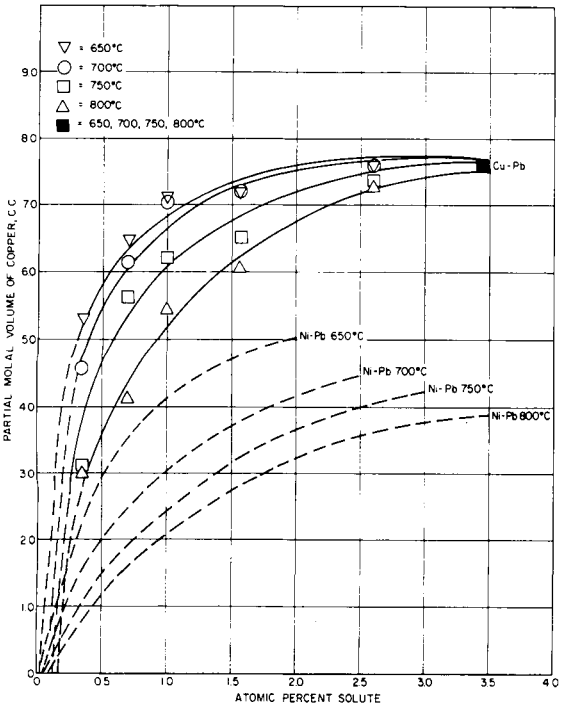

FIG. 1. Partial molal volume-composition isotherms for copper in liquid lead-copper and for nickel in liquid lead-nickel solutions.

ess than those calculated from the rule of mixtures even though less dense copper was added to the liquid lead.[2]

Partial molal volumes for lead and copper were estimated by the method of intercepts (*1*, *4*). It was found that, as in the case of the lead-nickel solutions, the partial molal volume of lead in the lead-copper solutions did not differ by more than about 0.2% from the molal volume of lead. Figure 1 gives partial molal volume-composition isotherms for copper in the lead-copper solutions, and similar curves for nickel in lead-nickel solutions are included for comparative purposes. The data indicate that the same general trend is observed for both systems; the partial molal volume of the solute extrapolates to a value of zero at high dilution, decreases with increase in temperature at constant concentration of solute, and increases with increase in solute content. These observations, in addition to the fact that the metallic radii of the solutes, their electronegativities, as well as their temperature coefficients of solubility in liquid lead (*2*) are comparable, suggests that the model used as a basis to explain the composition dependence of the partial molal volume of nickel (*1*) should be equally applicable to explain that of copper. Briefly, in each of these binary systems, the radius ratio of solute to solvent (*5*) is not far from Hägg's atomic radius ratio of 0.59 for pairs of atoms the smaller of which should fit into interstitial positions in the lattice of the larger. Furthermore, since liquid metal solutions may display a distribution in the sizes of interstitial holes (*6*), one can visualize that at high dilution the relatively small solute atoms (copper or nickel) enter interstitial sites in the quasi-lattice of the molten lead with the result that the partial molal volume of the solute approaches a value of zero. As more solute is dissolved, the partial molal volume increases with increase in composition because some of the solute atoms must enter substitutional sites (*1*). It is to be noted in Fig. 1 that the partial molal volume of copper increases more rapidly with composition than that of nickel. This is still consistent with the proposed model for the solution process (since the atomic volume of copper is about 5% larger than that of nickel) and further emphasizes the importance of the atomic size effect in liquid alloys.

[2] In the absence of experimental data on the densities of liquid copper undercooled to temperatures as low as 650°C, the densities used in the rule of mixtures calculations of the molal volumes of copper were obtained by extrapolation on a density-temperature plot using Lang's (*3*) density data.

REFERENCES

1. S. W. STRAUSS, L. E. RICHARDS, AND B. F. BROWN, *Nuclear Sci. and Eng.* **7**, 442–447 (1960).
2. S. W. STRAUSS, J. L. WHITE, AND B. F. BROWN, *Acta Met.* **6**, 604–606 (1958).
3. J. I. LANG, Measurement of the heat capacity and density of liquid copper. WADC (Wright Air Develop. Center) Tech. Rept. 57-488 (1957).
4. G. N. LEWIS AND M. RANDALL, "Thermodynamics and The Free Energy of Chemical Substances," p. 38, McGraw-Hill, New York, 1923.
5. L. S. DARKEN AND R. W. GURRY, "Physical Chemistry of Metals," p. 61, McGraw-Hill, New York, 1953.
6. F. SAUERWALD, *Z. Metallk.* **41**, 97–104 (1950).

S. W. STRAUSS
L. E. RICHARDS

Physical Metallurgy Branch
Metallurgy Division
U. S. Naval Research Laboratory
Washington, D. C.

Received August 17, 1960

Reactivity Effect of (*n*, 2*n*) Reactions in D_2O*

Recent measurements (*1*) of the (*n*, 2*n*) cross section of deuterium by Hill, Goldberg, LeBlanc, and Taylor at the Lawrence Radiation Laboratory give results in good agreement with theory (*2*) and permit reasonably accurate calculations to be made of the effect of this reaction on the multiplication factor of heavy water moderated reactors. Multigroup calculations were made for an infinite sea of D_2O containing a uniformly distributed source of fission neutrons to determine not only the magnitude of the (*n*, 2*n*) effect, but also the effect of (*n*, α) and (*n*, *p*) reactions with oxygen. The effects were found approximately to cancel with a net loss of neutrons per fission neutron of 0.06%.

In the multigroup calculation, a group width (common to all groups) of 0.1 lethargy unit was employed where the

* The information contained in this article was developed during the course of work under contract AT(07-2)-1 with the U. S. Atomic Energy Commission.

lethargy, u, is given by $u = \log 10/E$ with E the energy in Mev. The highest energy group employed had an upper bound of 14.9 Mev. The tabulation of the fission spectrum (*3*) given in reference *4* was employed. Oxygen cross sections and anisotropic scattering data were taken from reference *5*. Legendre polynomial fits to the available data for deuterium (*6–10*) were employed to express the anisotropic scattering data in the same form as that for oxygen in reference *5*, with weight given to what appeared to be the best data (*6*, *7*, *10*). The calculation gave the results that, per fission neutron, there are 0.00414 $(n, 2n)$ reactions with deuterium, 0.00472 (n, α) and (n, p) reactions [mainly (n, α)] with oxygen, and 0.00097 inelastic collisions with oxygen. The net loss of neutrons per fission neutron is thus 0.00058. The $(n, 2n)$ effect is somewhat larger than stated by Weinberg and Wigner (*11*), but much smaller than would be calculated from the cross sections assumed by Howerton (*12*) or Emmerich (*13*).

REFERENCES

1. J. A. Harvey, Reports to the AEC Nuclear Cross Sections Advisory Group. WASH 1028, p. 63 (April, 1960).
2. R. M. Frank and J L. Gammel, *Phys. Rev.* **93,** 463–70 (1954).
3. L. Cranberg, G. Frye, N. Nereson, and L. Rosen, *Phys. Rev.* **103,** 662–70 (1956).
4. R. Avery, ed., Reactor physics constants. ANL-5800, p. 18 (1958).
5. H. Lustig, H. Goldstein, and M. H. Kalos, An interim report on the neutron cross sections of oxygen. NDA-086-2 (January, 1958).
6. J. C. Allred, A. H. Armstrong, and L. Rosen, *Phys. Rev.* **91,** 90–99 (1953).
7. J. D. Seagrave, *Phys. Rev.* **97,** 757–65 (1955).
8. R. K. Adair, A. Okazake, and M. Walt, *Phys. Rev.* **89,** 1165–1170 (1953).
9. E. Wantauch, *Phys. Rev.* **84,** 169–172 (1951).
10. J. D. Seagrave and L. Cranberg, *Phys. Rev.* **105,** 1816–1820 (1957).
11. A. M. Weinberg and E. P. Wigner, "The Physical Theory of Neutron Chain Reactors," p. 449. Univ. Chicago Press, Chicago, Illinois, 1958.
12. R. J. Howerton, Semi-empirical neutron cross sections. UCRL-5351 (1958).
13. W. S. Emmerich, Multigroup neutron data for deuterium. NP-8363, Research Report 410 FF 323-R1, Westinghouse Research Laboratories (September, 1958).

H. K. Clark

Savannah River Laboratory
E. I. duPont de Nemours & Company
Aiken, South Carolina

Received August 22, 1960

Formulas for Thermal Reactors with Some Resonance Fission

The contribution of fissions induced by epithermal neutrons to the reactivity of primarily thermal reactors is of interest in reactors having low moderator to fuel ratios. In treating this effect it has been common to use the following definitions:

$$(k_{\text{thermal}}) \qquad k_t = \epsilon\eta_t f p_{28} p_{25}$$
$$(k_{\text{resonance}}) \qquad k_r = \epsilon\eta_r p_{28}(1 - p_{25})$$

where p_{28} and p_{25} are the resonance escape probabilities for the fertile and fissile materials, respectively; the η's are for average thermal and resonance fissions, respectively; and the other symbols have their usual meanings.

Commonly the total k is obtained by adding k_t and k_r, but in the following it is shown that this is correct only in a very restricted sense and that a more general formulation is of the form

$$k_{\text{eff}} = \frac{k_t}{1 - k_r}$$

in those cases where $k_r < 1$, and where the average lifetime of neutrons captured at resonance energies is short compared to the thermal neutron lifetime.

This is most easily shown by adding a term for resonance fission to the usual two-group formulation:

(Fast)

$$-D_f\nabla^2\phi_f + \Sigma_f\phi_f = \epsilon\eta_t f\Sigma_t\phi_t + \epsilon\eta_r\Sigma_f p_{28}(1 - p_{25})\phi_f \qquad (1a)$$

(Thermal)

$$-D_t\nabla^2\phi_t + \Sigma_t\phi_t = p_{28}p_{25}\Sigma_f\phi_f \qquad (1b)$$

Introducing the buckling, B^2, in the usual way and solving the simultaneous equations leads to a criticality condition:

$$[1 + \tau B^2 - \epsilon\eta_r p_{28}(1 - p_{25})][1 + L^2B^2] - \epsilon\eta_t f p_{28}p_{25} = 0 \qquad (2)$$

and to terms of first order,

$$B^2 = \frac{k_t + k_r - 1}{(1 - k_r)L^2 + \tau} \qquad (3a)$$

In reactors where $k_r \ll 1$, the correction term to L^2 may be ignored, and in this sense then the k_t and k_r are simply additive. However, a more general interpretation of Eq. (3) may be obtained by solving (1a) for the ratio of the fluxes

$$\phi_f/\phi_t = \frac{k_t/p_{28}p_{25}}{(1 - k_r + \tau B^2)} \cdot \frac{\Sigma_t}{\Sigma_f} \qquad (4)$$

When leakage may be neglected, this ratio is larger by the factor $1/(1 - k_r)$ than in the corresponding case without resonance fission and suggests the interpretation of the effect as a convergent, iterative process with the multiplication at each cycle being k_r. With leakage included, it is just $k_r - \tau B^2$. This conclusion is reinforced by the observation that, for zero leakage, the critical condition can be written in the form

$$k_t/(1 - k_r) = 1$$