UN in UO_2 is not stable in vacuum) a reasonable assumption is that nitrogen ions become free and occupy anionic and interstitial positions, whence they migrate through the intersticialcy mechanism, involving interchanges with the oxygen ions, the self-diffusion of which becomes the controlling factor. (Since nitrogen ions are somewhat bigger than oxygen ions, they are supposed to move faster, because they tend to make the lattice unstable[18]). Otherwise, the nitrogen ions might occupy only the interstitial positions $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ and $<\frac{1}{2},0,0>$ and migrate jumping from one to a next neighbor in the [1,1,0] directions.

Returning to xenon, one may remark that the Pauling univalent positive ion has the radius of 0.75 Å, while the univalent negative ion has the radius of 1.90 Å (comparable to that of a hypothetical di-valent nitrogen anion). Obviously, polyvalent ions would be smaller. It is very unfortunate that we cannot have any ideas of their compressibilities (except perhaps that they are, like any ions, very rigid), because of the inexistence of ionic xenon compounds.

However, one may attempt to draw the following conclusions:

—if xenon is in the neutral atomic state, then probably it occupies interstitial sites and moves by jumping directly from one to another, its diffusion energy being in this case of the order of 30,000 cal/mol;

—if xenon is a positive ion, it occupies cationic sites, whence it diffuses like a uranium ion, being smaller and having a somewhat lower activation energy (though higher than the energy of formation of a cation vacancy);

—if xenon is a negative ion, either it occupies interstitial sites or both anionic and interstitial sites, moving in one of the ways proposed previously for nitrogen, with activation energies in the range of 30,000 cal/mol.

Diffusion experiments carried out up to date do not allow conclusions. The B.M.I. Group[19] found 30,200 cal/mol for xenon diffusion from a fused UO_2 single crystal; though the experiment was very accurate, fused UO_2 always contains carbides, nitrides and other impurities, which make its use questionable. Almost all of the other researchers measured higher energies (sometimes in excess of 100,000 cal/mol) using UO_2 powders and polycrystalline sinters; if one is tempted to reject their results, because too many factors (including surface conditions, grain boundaries and inhomogeneities) might have interfered, he must very well consider also that the experiments on uranium, oxygen and nitrogen diffusion have been carried out on these "undesirable" materials.

Activation energies for krypton diffusion in UO_2 have been measured more seldom than for xenon, but the results lie well in the same range established for the latter.

This paper ends with a remark. In xenon and krypton diffusion experiments with UO_2 and UC, it has always been found that a sudden gas release (burst), much larger than would be expected from the solution of the diffusion equations for the transient period, takes place any time the temperature is stabilized at a certain level. Similar features are reported by Kelly[20] for the release of argon and xenon radiatively absorbed on Nb_2O_5, SiO_2 and TiO_2, by the Hahn-Meiter Institut Group for argon diffusion from fluorides,[6] by Bauer for xenon diffusion from stainless steel[21] and by Ferrari for nitrogen diffusion from UO_2.[9]

Thus, the so-called "burst-effect" is turning out to be not typical of any particular kind of solids (having been reported for ionic and covalent compounds and metals as well), nor of inert gases alone (since nitrogen is not), nor of any way of introducing the gas into the solid (fission, neutron capture, knocking-in, chemical decomposition and fission-product bombardment having been used).

The only common feature to all cases is the little chemical affinity, at the experimental conditions, of the diffusing gas to the lattice atoms; one may be left with the impression that the "burst-effect" is a much more general phenomenon in gas diffusion through solids than supposed.

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[19] R. H. BARNES *et al.*, Battelle Memorial Institute Report BMI-1533 (1961).

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Measurement of Doppler Coefficient by Heating a Small Region of a Fast Reactor Critical Assembly*

Several of the methods proposed for measuring the Doppler coefficient in a fast critical facility involve heating only a very small region of the reactor. (The oscillating-rod Doppler experiment is one example.) It has been recognized that there is a difficulty in interpreting the results of such

*Research performed under contract to the USAEC.

an experiment since the change of resonance-neutron absorption in the heated region is, in general, considerably different from the Doppler effect obtained when the entire reactor is heated. Recently Khairallah and Ozeroff[1] showed that for a small temperature increment the total change in neutron absorption, taking into account both the small heated region and the unheated portion of the reactor near the heated region, was virtually the same as the normal Doppler effect. (The term "normal Doppler effect" here refers to the change in absorption that would have occurred in the heated region alone if the remainder of the reactor were also heated to the same temperature.) In this note a general expression is derived for the total change in neutron resonance absorption when only a portion of the reactor, Region 1, is heated and the remainder, Region 2, has the same composition as Region 1. The conclusion of Khairallah and Ozeroff that a very small heated sample gives the normal Doppler effect is shown to be in error by about 25 per cent in the limit of an infinitesimally small heated region. This result is obtained for a representative temperature rise in a rod-oscillator experiment and for a uranium-238 resonance with parameters pertinent to the interesting resonance region for the Doppler effect in a large fast oxide reactor.

Consider a single isolated resonance with a uniform neutron source $\sigma_p\phi$ (narrow resonance approximation), where σ_p is the potential scattering cross section per absorber isotope and ϕ is the unperturbed neutron flux at energies just above the resonance. The total neutron absorption in the reactor per unit unperturbed flux per absorber atom in the heated region is given by

$$A = (1+V)\sigma_p \int_{res} \frac{\sigma_a}{\sigma_t} dE \quad \text{for Region 1 unheated, and} \tag{1a}$$

$$A' = \sigma_p \int \left[\left(\frac{\sigma'_a}{\sigma'_t}\right)(P_{1,1} + V\,P_{2,1}) + \left(\frac{\sigma_a}{\sigma_t}\right)(P_{1,2} + V\,P_{2,2})\right] dE \quad \text{for Region 1 heated} \tag{1b}$$

where V is the volume of Region 2 divided by the volume of Region 1, (σ_a, σ_t) are (absorption, total) cross sections per absorber atom in the unheated condition, (σ'_a, σ'_t) are the same cross sections for the heated condition and E is neutron energy. (Note that the E^{-1} multiplicative factor usually contained in the expression for a resonance integral is omitted here since the flux in a fast reactor does not exhibit an E^{-1} variation.) $P_{i,i}$ is the probability that a source neutron in region i $(i = 1,2)$ suffers its next collision in that region, $P_{i,j} = 1 - P_{i,i}$ is the probability that a source neutron in Region i suffers its next collision in the other region (escape probability for Region i).

The reciprocity relationship[2]

$$P_{2,1} = \frac{1}{V}\left(\frac{\sigma'_t}{\sigma_t}\right) P_{1,2} \tag{2}$$

permits the collision probabilities $P_{i,i}$ and escape probabilities $P_{i,j}$ to be expressed in terms of the single escape probability $P_{1,2}$. The change of absorption per unit unperturbed flux per absorber atom in the heated region, $A' - A$, may be considered as the change of effective resonance integral due to the Doppler broadening in the heated region; it is given by

$$\Delta I = A' - A = \sigma_p \int_{res} \left(\frac{\sigma'_a}{\sigma'_t} - \frac{\sigma_a}{\sigma_t}\right)\left[1 + P_{1,2}\left(\frac{\sigma'_t - \sigma_t}{\sigma_t}\right)\right] dE\,. \tag{3}$$

The integral on the right side of Equation (3) may be divided into two terms:

$$\Delta I = (\Delta I)_v + (\Delta I)_s \tag{3a}$$

where

$$(\Delta I)_v = \sigma_p \int_{res} \left(\frac{\sigma'_a}{\sigma'_t} - \frac{\sigma_a}{\sigma_t}\right) dE, \text{ and} \tag{3b}$$

$$(\Delta I)_s = \sigma_p \int_{res} P_{1,2}\left(\frac{\sigma'_t - \sigma_t}{\sigma_t}\right)\left(\frac{\sigma'_a}{\sigma'_t} - \frac{\sigma_a}{\sigma_t}\right) dE. \tag{3c}$$

The term $(\Delta I)_v$ gives the normal Doppler effect (change of effective resonance integral) and the term $(\Delta I)_s$ gives the excess Doppler effect due to the finite volume of the heated region (the surface effect). Exact expressions for $P_{1,2}$ may be obtained for simple geometries;[3] for the general case, the escape probability may be approximated by

$$P_{1,2} = \frac{1 - \exp(-N\sigma'_t\bar{\ell})}{N\sigma'_t\bar{\ell}}\,, \tag{4}$$

[1] A. KHAIRALLAH and W. J. OZEROFF, "The Calculation of the Doppler Effect in the Case of Spatial Temperature Variation," presented at *E.A.E.S. Symposium on Advances in Reactor Theory*, Karlsruhe, West Germany, (April 23-24, 1963).

[2] B. DAVISON, *Neutron Transport Theory*, Oxford University Press, London (1957).

[3] K. M. CASE, F. DeHOFFMAN, and G. G. PLACZEK, "Introduction to the Theory of Neutron Diffusion," Los Alamos Scientific Laboratory, 1, 21 (June 1953).

where $\bar{\ell} = \frac{4V_1}{S_1}$, with V_1 and S_1 being respectively the volume and bounding surface area of Region 1, and N is the concentration of absorber atoms in this region. Equation (4) is adequate for most applications and approaches the same limits as the exact expressions for specific geometries when the heated region is made very large or very small.

The following observations are made concerning the excess Doppler effect:

1. As the heated region becomes very large ($N\sigma_p \bar{\ell} \gg 1$),

$$(\Delta I)_s \rightarrow 0 \text{ since } P_{1,2} \rightarrow 0 .$$

2. As this region becomes very small

$$\left[\left(\frac{\sigma_0 \Gamma}{\Delta} + \sigma_p\right)\right] N\bar{\ell} \ll 1,$$

where σ_0 is the peak total cross section for the natural resonance, Γ is the total natural resonance width, and Δ is the Doppler width for the temperature of the heated region, $P_{1,2} \rightarrow 1$ over the entire resonance and $(\Delta I)_s$ assumes its maximum value.

3. For a weak resonance ($|\sigma_t' - \sigma_t| \ll \sigma_p$) or for a very small temperature change ($|\sigma_t' - \sigma_t| \ll \sigma_t$ at all energies), $(\Delta I)_s \rightarrow 0$ and $[(\Delta I)_s/(\Delta I)_v] \rightarrow 0$ regardless of the size of the heated region.

4. $(\Delta I)_s$ is always positive. This can be seen by dividing the integral of Equation (3c) into two components. One of these integrates over the center resonance region where the conditions $\sigma_t' < \sigma_t$ and $\sigma_a'/\sigma_t' < \sigma_a/\sigma_t$ are satisfied. (Note that the second inequality follows from the first for a Breit-Wigner resonance for $\sigma_p > 0$.) The second integral component covers the outer resonance regions defined by $\sigma_t' > \sigma_t$ and $\sigma_a'/\sigma_t' > \sigma_a/\sigma_t$. Since the two terms in parenthesis in Equation (3c) are either both negative or both positive and $P_{1,2}$ is positive at all energies, it follows that $(\Delta I)_s$ is always positive.

The conclusion by Khairallah and Ozeroff that the normal Doppler effect is obtained even for very small samples is based on the assumption that $|\sigma_t' - \sigma_t| \ll \sigma_t$ is generally valid (see observation 3 above). For a strong resonance (such as those responsible for the large negative Doppler coefficient in a UO_2/PuO_2-fueled fast reactor) and for a large enough temperature rise in the very small heated sample to give a measurable reactivity signal, this assumption is not correct. To evaluate the departure from the normal Doppler effect ΔI_s, ΔI_v, and the ratio $\Delta I_s/\Delta I_v$ are listed in Table I for a uranium-238 resonance at 1000 eV, assuming a temperature rise from 300 to 600 K for several diameters of a cylindrical heated region. The uranium-238 resonance parameters used are $\Gamma_\gamma = .0246$, $\Gamma_n = .0965$ (which is $\sqrt{\langle \Gamma_n^2 \rangle}$ at 1000 eV); and $\sigma_p = 60$ barns corresponding to the following reactor volumetric composition: 52% sodium, 32% PuO_2-UO_2, and 16% steel; U^{238} is 77% of the total U + Pu (see Table 18 of Reference 4 for more details on the composition).

TABLE I

Diameter (cm) of Heated Region	$(\Delta I)_s$	$(\Delta I)_v$	$(\Delta I)_s/(\Delta I)_v$
0	1.18	4.22	0.28
1	0.86	4.22	0.20
5	0.36	4.22	0.085
∞	0	4.22	0

It is seen that the departure from the normal Doppler effect is significant in the limit of an infinitesimally small heated region. However, the large - region approximation, which gives the normal Doppler effect, appears adequate for regions of diameter greater than five centimeters, which probably covers most practical applications for small-sample reactivity-change measurements.

The most likely Doppler experiments with heated samples so small as to involve the above considerations are differential activation measurements using, for example, heated and unheated uranium or plutonium foils. For such experiments, the increased absorption per unit volume of heated sample reduces to

$$\Delta I = \sigma_p \int_{res} \left(\frac{\sigma_a' - \sigma_a}{\sigma_t}\right) dE \qquad (5)$$

in the small sample limit. This expression differs from the normal Doppler effect since it does not involve a temperature dependence of the flux spectrum.

P. Greebler

[4] P. GREEBLER and E. GOLDMAN, "Doppler Calculations for Large Fast Ceramic Reactors - Effects of Improved Methods and Recent Cross-Section Information," GEAP-4092 (December, 1962).

Nuclear-Reactor Contribution to the Van Allen Belt

There is considerable development effort towards producing nuclear reactors for SNAP applications. This program now consists of four re-