For fast electrons internal conversion of energy by electronically excited molecules is almost certainly the predominant quenching process. It is consistent with the efficiency being independent of electron energy, and Birks[1] has shown that it can quantitatively explain the low values found. For more heavily ionizing particles the scintillation efficiency may become very small, yet the accompanying decrease in decay time is relatively slight.[2] This indicates that the additional quenching is due to bimolecular processes, e.g., neighboring ionized and electronically excited molecules may interact to form new chemical entities (such as dianthracene in anthracene), excited molecules may be affected adversely by the coulomb fields of neighboring ions and return non-radiatively to the ground state, and interaction between excited molecules may occur leading to non-radiative dissipation of their electronic energy. This latter process is discussed by Black[3] who shows that it can provide an interpretation of the experimental data. This author also considers the effects of migration of energy out of the excitation column. However, due to the rapidity with which the bimolecular processes occur, expansion of the excitation column may be neglected whether taking place by resonance exchange of energy between molecules[4] or by photon emission and reabsorption.[1] Considering then that quenching of fluorescence is due to the combined effect of monomolecular and bimolecular processes in the excitation column, one may readily calculate the scintillation efficiency.

In penetrating a distance Δx the particle loses energy $(dE/dx)\Delta x = \epsilon_0 \Delta x$ to the molecules along or near its path. At a time t after absorption the excess electronic energy of the molecules in this length of the excitation column is ϵ, and we have

$$d\epsilon = -(p+k+a\epsilon)\epsilon dt, \qquad (1)$$

where $a\epsilon$ = rate of energy dissipation by bimolecular quenching processes, k = rate of energy dissipation by monomolecular quenching processes, and p = rate of energy dissipation by fluorescence emission. The total fluorescence emission from these molecules is

$$dL = p\Delta x \int_0^\infty \epsilon dL = \frac{p}{a} \ln\left(1 + \frac{a}{p+k} \cdot \frac{dE}{dx}\right)\Delta x. \qquad (2)$$

When dE/dx is small this may be written in the approximate form

$$\frac{dL}{dx} = \frac{p}{p+k} \cdot \frac{dE}{dx} \Big/ \left(1 + \frac{1}{2} \cdot \frac{a}{p+k} \cdot \frac{dE}{dx}\right), \qquad (3)$$

in agreement with the semi-empirical expression proposed by Birks.[5] The total scintillation emission excited by the particle is

$$L = \int_0^x dL = \int_E^0 \frac{dL}{dE} \cdot dE. \qquad (4)$$

When dE/dx is small, Eq. (4) reduces to $L = pE/(p+k)$; thus the constant $p/(p+k)$ denotes the phosphor efficiency for fast electrons. For the particular case of anthracene the complete integration of Eq. (4) has been performed using a graphical method. The differential energy loss dE/dx was calculated from the theoretical stopping formula, except for alpha particles with energies below 10 Mev for which a semi-empirical method based on the range-energy curves for air was used;[6] the constant $p/(p+k)$ is 0.05 for anthracene, and the ratio a/p was determined from the data for 5-Mev protons to be 0.23. The curves obtained are illustrated in Fig. 1 and compared with the experimental data.[7,8] The agreement is very satisfactory, and since the response of all organic phosphors is very similar we may expect this in other cases.

A point of interest concerns the decay time of fluorescence, which is longer when excited with particles than with ultraviolet quanta. This has been interpreted as the result of additional absorption and emission processes undergone by fluorescence photons emitted from initially ionized molecules;[1] an alternative explanation is that particle excitation involves an effective delay in the emission of fluorescence by ionized molecules due to the finite time of ion recombination. It is hoped that measurements of decay times and efficiencies of fluorescence excited by radiations in the
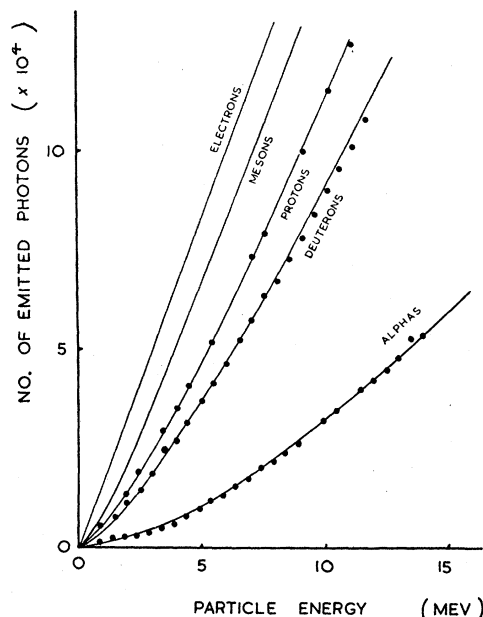
wavelength range 3000A to 100A will resolve this question; such measurements are in progress in this laboratory.

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Variable Activation Energy and the Motion of Lattice Defects*

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IN a recent paper, Overhauser[1] successfully analyzed isothermal annealing curves of irradiated copper on the basis of a variable activation energy, i.e., an activation energy which depends on the number of disturbances left in the lattice. The writer was concerned with the analysis of this type of annealing kinetics in another connection[2] and arrived at some general conclusions on the basis of a simple example. It is the purpose of this note to summarize the results briefly.

Let n denote the number of lattice disturbances present at any time t, and assume that over the range of interest the activation energy, E, decreases linearly with increasing n. This simple case can be handled analytically. The source of this functional dependence of E on n is most likely the strains introduced in the lattice by the disturbances, particularly intersitial atoms.[1,3] A linear dependence is a reasonable approximation to the relation derived by Overhauser[1] on the basis of lattice strains. E is expressed, therefore, as

$$E/k = E_0/k - \alpha n/k, \qquad (1)$$

and the general rate equation for the disappearance of n is

$$dn/dt = -\nu n^\gamma e^{-E_0/kT} e^{\alpha n/kT}. \qquad (2)$$

Integrating formally gives

$$\int_{n_0}^{n} \frac{e^{-bn}}{n^\gamma}dn = -\nu t e^{-E_0/kT}, \tag{3}$$

where $b=\alpha/kT$. The integrals of (3) are expressible in terms of the exponential integral for any integer γ. At a fixed value of n one may write

$$t = K'e^{E_0/kT}e^{-\alpha'/kT}, \tag{4}$$

where K' is a constant and $\alpha'=\alpha n$. Thus, a plot of $\log t$ versus $1/T$ obtained at a fixed n is linear just as in the simple case of constant activation energy but the effective activation energy, $E_{\text{eff}}=E_0-\alpha'$, depends on the value chosen for n; i.e., the phenomenologically determined activation energy[4] varies systematically with n.

The influence of a variable activation energy on the nature of the annealing curve is best discussed on the basis of an actual example. n versus t curves were calculated at a fixed temperature (250°C) for $n_0=0.10$; $E_0/k=15\,000$ with $\alpha/k=25\,000$; $E_0/k=12\,500$ with $\alpha=0$; $\nu=10^{13}$, for $\gamma=2$ and 3. For $\gamma=2$, Eq. (3) integrates to

$$\frac{e^{-bn_0}}{n_0}-\frac{e^{-bn}}{n}+b[\text{Ei}(-bn_0)-\text{Ei}(-bn)] = -\nu t e^{-E_0/kT} \tag{5}$$

(using the notation of Jahnke and Emde). By simple integration by parts the integral for any γ may be obtained. The results obtained are shown in Fig. 1. (The semi-log plot is used for convenience only.) From the curves of Fig. 1 it is clear that a variable
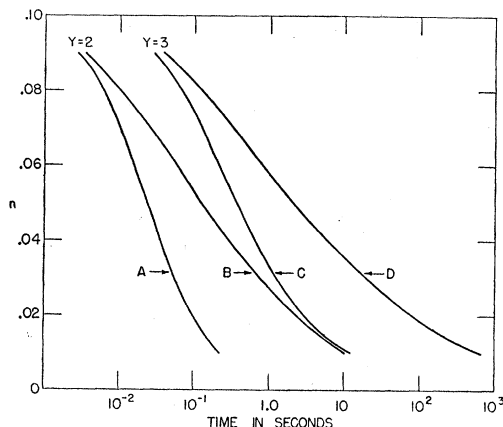

Fig. 1. Relaxation of n at 250°C. Comparison of constant and variable activation energy. A and C: $E_0/k=12\,500$, $\alpha/k=0$. B and D: $E_0/k=15\,000$, $\alpha/k=25\,000$.

activation energy which increases with decreasing n has the effect of lengthening the time scale for any given isothermal annealing curve. A rather small variation of E with n leads to a very large change in the time scale. Figure 1 also shows that an increase in the order of reaction (from $\gamma=2$ to $\gamma=3$ in Fig. 1) has a qualitatively similar effect to that of an E increasing with decreasing n. Computations showed that an annealing curve characterized by a given γ and a variable E can often be fitted by a constant E but a higher and not necessarily integer value of γ. This conclusion is in accord with Overhauser's experimental results.[1]

The results of this study may be summarized as follows:

1. A variable activation energy leaves the nature of the plot of $\log t$ vs $1/T$, taken at any fixed n, unchanged but manifests itself in a systematic change of the slopes of these lines as a function of the amount of anneal at which E is being determined.

2. A rather small variation of E with n leads to a very large change in the time scale for the relaxation of n.

3. An annealing curve of low γ and variable E is very similar in character to a reaction of higher order (not necessarily integral) and of constant E.

4. More complex expressions of $E=f(n)$ may be required in specific cases. As long as E is a decreasing function of n the above conclusions are qualitatively valid.

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Determination of the Neutron-Proton Capture Cross Section

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A DIRECT determination of the neutron-proton capture cross section has been made by a measurement of the mean life of neutrons in water, as first done by Manley, Haworth, and Luebke.[1] In the present method, which is capable of high precision, the diffusion correction which was the main source of uncertainty in their investigation has been avoided by making the measurements in a large tank, of dimensions $97\times97\times107$ cm, with the modulated neutron source, a stainless steel target bombarded by deuterons from a 150-kv accelerator, in the center. The decay of the neutron intensity at a number of positions was analyzed by a multichannel time analyzer, the detectors being cylindrical brass counters, 1 cm in diameter and filled to 600 mm of BF_3, which extended through the entire depth of the tank and thus automatically performed an integration of the neutron intensity in the vertical direction. By subsequent integration of the data in the radial direction the decay of the total number of neutrons in the tank was obtained and found to be accurately exponential apart from a small constant background.

The period of the exponential decay of the neutrons was found from 3 complete runs to be 206.6 ± 1.5 μsec, the estimated uncertainty being due mostly to the uncertainty of the correction for the constant background, since the statistical accuracy and consistency of the measurements was much better. A small correction of -0.4 μsec for counting losses in the time analyzer has been included in this result. The result has to be corrected for the influence of the counters on the neutron distribution. Since the brass counters used in the experiment have a larger capture cross section than the corresponding amount of water, the decay in the counters will be more rapid than in the water. This effect was investigated by comparing the decay curves obtained at three positions in the tank with the brass counters and with aluminum counters filled to such pressure of BF_3 that they have the same absorption cross section per volume as water. This measurement yielded a correction of $+5.5$ μsec with an uncertainty of 2 μsec. Because of the absence of moderating nuclei in the counter, a depression in neutron density will develop during the first intervals of time when the neutrons are being slowed down. As soon as the neutrons have become thermal this hole will, however, quickly fill up by diffusion. The effect of the absence of scattering in the counter once the neutrons have become thermal should be proportional to the Laplacian of the neutron density and will disappear in the integration over the whole tank. These considerations were verified by comparing the decay curves measured with two aluminum counters of different volume, both having the same absorption as water. The presence of a depression in the larger counter during the first intervals of time after the neutron burst from the source was clearly visible, but after about 50 μsec the decay curves became identical. In calculating the period, the initial part of the decay curve is therefore discarded.

For the corrected mean life in distilled water at 22.6°C, we thus obtain 212 ± 3 μsec, and the corresponding neutron-proton capture cross section for a neutron velocity of 2200 m/sec is 0.321 ± 0.005 barns, in satisfactory agreement with the most recent determinations by quite different methods of Hamermesh, Ringo, and Wexler,[2] 0.329 ± 0.004 barn, and of Muehlhause et al.,[3] 0.332