Latent Image Fading in Nuclear Emulsions

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HE magnitude of the regression of the latent image produced by charged particles incident on a nuclear emulsion is, in general, a function of the emulsion characteristics, the flux, kind, and energy of the particles, and of the time and conditions of storage in the interval between irradiation and development. This effect is much more marked in nuclear emulsions than in optical-type emulsions because of the extremely small size of the silver speck development centers in the former, which is caused by inefficiency in the photolytic process when the time of exposure of the halide grains is very short. This occurs in traversal by a radiation particle, where it will be in the vicinity of a halide grain for a time of the order of 10^{-14} sec. Since latent image regression is a surface phenomenon of the specks, the smaller ones will be affected to a proportionally greater extent, accounting for the relative instability of nuclear emulsion latent images.

Investigations by Lauda¹ and Mather² demonstrate a reduction of about 90 percent in the fading of the latent image after predevelopment storage in an evacuated chamber. This indicates that the dominant agent involved is a constituent of the atmosphere, with the known susceptibility of the silver specks to oxidizing agents suggesting the oxygen and water vapor present as causative factors. This is corroborated by studies3 of the variation of fading with storage in atmospheres of oxygen, nitrogen, and air, and with water vapor added to the above. The fading rate was found to be greatest in oxygen and least in nitrogen, with the addition of water vapor greatly increasing the fading rate in all cases. The temperature of storage will influence the rate of latent image oxidation, and may also act in connection with a purely thermal regression process⁴ that could account for part of the 10 percent residual fading observed even after vacuum storage.

To obtain a relation between the magnitude of the regression and the time of storage, the fading rate under a constant set of conditions may be considered proportional to the number of development centers present at any time. In integrated form this gives the expression

$$(N_0 - N)/N_0 = 1 - \exp(-ct),$$

where N_0 is the initial number of centers present after irradiation, N the number after a time t, and c a constant dependent on the size of the silver specks and on the storage conditions. For nuclear emulsions the density D after development will, to a good approximation, be proportional to N. Thus, the fading coefficient will be given by

$$(D_0 - D)/D_0 = 1 - \exp(-ct),$$
 (1)

with D_0 being the density produced upon immediate development. $(D_0 - D)/D_0$ is plotted in Fig. 1 as a function of t for various values



FIG. 1. The variation of fading with time of storage for various values of c.

of c, in good agreement with the experimental values given by Yagoda and Kaplan⁵ for two storage conditions.

The variation of the fading index c with temperature and humidity may be determined from a consideration of the effect of temperature on the velocity of a gas-solid reaction. Combining the Arrhenius relation

$$dN/dt = A \exp(-E/RT)$$

where T is the absolute temperture and A, E, and R constants, and the empirical Freundlich adsorption isotherm

$$dN/dt = kC^n$$

where C is the concentration of the gas and n a fraction whose value lies between 0 and 1, gives for c in this case

$$=k_1H^n\exp(-k_2/T),$$

where H is the absolute humidity. For storage in air the oxygen concentration varies to a negligible extent and therefore can be included in k_1 . Since for reactions of this kind $n \sim \frac{1}{2}$,

$$c = k_1 H^{\frac{1}{2}} \exp(-k_2/T).$$
 (2)

Equations (1) and (2) are adequate to describe the major part of fading phenomena in nuclear emulsions.

¹ J. Lauda, Sitzber. Akad. Wiss. Wien, Abt. IIa 145, 707 (1936). ² K. B. Mather, Phys. Rev. 76, 486 (1949). ³ Albouy and Faraggi, Report of Conference on Fundamental Mecha-nisms of Photographic Sensitivity. Part III, ONRL-66-50, 4 (1950). ⁴ A. Beiser, Phys. Rev. **80**, 112 (1950). ⁵ H. Yagoda and N. Kaplan, Phys. Rev. **73**, 634 (1948).

Decay of the Hydrogen Discharge*

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HE decay of the electrodeless hydrogen discharge has been studied by the previously described techniques¹ of electron density, optical spectrographic, and optical light intensity measurements as a function of time after termination of the exciting fields. The electron density is measured by the shift of resonance of a microwave cavity due to the presence of free electrons, and this measurement has been improved by the technique of sweeping the exploring microwave signal through resonance in a few microseconds rather than depending upon the decay transient of the discharge to plot a resonance indication. Pressures studied varied from 1.0 to 19.8 mm Hg; times from 35 to 3500 µsec; and electron densities from 5×10^7 to 5×10^9 electrons/cm³.

The character of the decay curves for electron density indicates that two-body recombination of electrons occurs from about 7 mm Hg to 20 mm Hg, and that the recombination coefficient, α , defined by the relation $dn/dt = -\alpha n^2$, where n is the electron concentration, is constant within experimental error at 6.2×10^{-6} cm³/electron-sec. Measurements at the lower pressures indicate that ambipolar diffusion predominates. The decay curve is taken to be represented by Biondi's expression.²

$$n/(1+\alpha\tau n) = [n_0/(1+\alpha\tau n_0)] \exp[-(t-t\alpha)/\tau]$$

and by the use of three experimental points, τ is extracted without the necessity of assuming the behavior of α at low pressures. At the same time, α is determined, provided the distribution of electrons within the cavity can be assigned with certainty. The use of τ enables $D_a p = \Lambda^2 p / \tau$ to be calculated at the constant value 600 cm²-mm Hg/sec for pressures in the range 1 to 3 mm Hg, where D_a is the ambipolar diffusion coefficient, p the pressure, and Λ the characteristic diffusion length. In this range α is observed to rise from 2×10^{-6} cm³/electron-sec toward the above value at 7 mm Hg.

Spectrographic examination of the discharge under excitation shows most of the energy to be in the molecular spectrum of hydrogen, including the ultraviolet continuum associated with the

 $1s\sigma 2s\sigma^3\Sigma_{\sigma} \rightarrow 1s\sigma 2\rho\sigma^3\Sigma_{\mu}$ transition, with atomic lines also visible. Decay of the molecular spectrum is so rapid that it cannot be photographed within the 10-15-µsec resolution of our shutter. The light at times from 0-600 μ sec is continuous in character, having a maximum at about 4250A and cutting off at about 3400A; and is thus not identifiable with the molecular continuum. Changes in spectral distribution and absolute intensity are observed in passing from quartz to Pyrex containers.

Estimates to within a factor of 2 or 3 (by means of a calibrated 1P28 photo-tube) of the absolute energy lying in the band of the 1P28 per disappearing electron lead to results which (a) are less than 1/500 of the available energy (ionization potential) per recombining electron, and (b) vary widely with time instead of remaining constant. These and other results lead to the conclusion that the light associated with the hydrogen afterglow arises from the container walls, and that nonradiatiative volume recombination accounts for the removal of electrons. A possible mechanism is

$H_2^+ + e \rightarrow H + H + K.E.,$

with the $1s\sigma 2p\sigma^3\Sigma_u$ or similar state as the final state of the molecule.

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¹ AEC Predoctoral Fellow.
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² M. A. Biondi, Phys. Rev. 79, 733 (1950).

Californium Isotopes from Bombardment of Uranium with Carbon Ions*

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HE recent production and identification¹ of isotopes of elements with atomic numbers up to six higher than the target element through bombardment with approximately 120-Mev carbon (+6) ions made it seem worth while to apply this technique to the transuranium region.

Accordingly, small pieces of natural uranium metal (about 0.5 mil thick and 2.5 cm by 0.6 cm area) were irradiated in the internal carbon ion beam in the Berkeley 60-inch cyclotron. Following the irradiations, the uranium was dissolved in dilute hydrochloric acid containing hydrogen peroxide and a transplutonium fraction was isolated through the use of lanthanum fluoride and lanthanum hydroxide precipitation steps followed by the ion exchange adsorption column procedure, in which concentrated hydrochloric acid is used to separate the tripositive actinide elements from the rare earth elements.²

The transplutonium fractions in hydrochloric acid were evaporated as weightless films on platinum plates which were placed in the ionization chamber of the 48-channel pulse analyzer apparatus in order to measure the yield and energies of any alpha-particles which might be present. In the best experiment at about one hour after the end of the 90-min bombardment, some 50 disintegrations per minute of the distinctive 7.15±0.05-Mev alpha-particles3 of Cf²⁴⁴ were observed to be present and to decay with the 45-min half-life. The Cf244 was presumably formed by the reaction $U^{238}(C^{12}, 6n).$

After the decay of the alpha-particles due to Cf²⁴⁴, about five disintegrations per minute of alpha-particles with 6.75 ± 0.05 -MeV energy were observed and this alpha-radioactivity decayed with a half-life of about 35 hours. In subsequent experiments involving the use of an ion exchange method⁴ to separate the individual actinides from each other, both the \sim 45-min and \sim 35-hour activities were found together in the chemical fraction corresponding to the new element californium. A consideration of the systematics of alpha-radioactivity⁵ leads us to the view that this 35-hour period is due to the new isotope Cf²⁴⁶ formed by the reaction $U^{238}(C^{12}, 4n)$. The measured half-life agrees with the expected alpha half-life for the observed energy for an even-even isotope of the element with atomic number 98.

It is not possible at this time to obtain a good estimate of the intensity of the carbon ion beam. Therefore, the cross sections for the reactions given above cannot be calculated. However, it is interesting to note that the indicated yields for the (C, 6n) and (C, 4n) reactions are comparable.

If the mass assignment of the new 35-hour activity to Cf²⁴⁶ is correct, these new data give a better idea as to the slope of the alpha-energy vs mass number line for californium, which in turn makes it possible to make better predictions of the radioactive properties of the nuclides in this region.

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Thresholds of Photo-Neutron Reactions*

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EUTRON yields as functions of the maximum energy of the bremsstrahlung output of the University of Pennsylvania betatron have been measured for several substances in the energy region from threshold to 22 Mev. The samples, of the order of 50 grams of high purity materials, were inserted in a block of paraffin 9 in \times 9 in \times 20 in, in which was also placed a B¹⁰F₃ counter (Fig. 1). Shielding against background neutrons was obtained by surrounding the apparatus with sheet cadmium, then with about 4 inches of paraffin mixed with B₄C, then with about 1 foot of additional paraffin. The samples were $\frac{3}{4}$ -in diameter cylinders. The collimated x-ray beam was about $\frac{5}{8}$ in diameter and irradiated the samples axially. The distance from sample to betatron target was 8.5 feet. The counter was parallel to the beam and about one inch away.

In order to avoid counting pile-up due to the x-ray pulse, and also to avoid being bothered by transient effects on the counting system due to the x-ray burst and auxiliary betatron pulses, only those neutrons were counted which, having been delayed in the paraffin, entered the counter some time after each burst of x-rays. This was done by constructing an electronic gate signal which was delayed from each x-ray burst by about 30 µsec, and whose dura-



FIG. 1. Schematic diagram of the experimental arrangement (side view).