

following way. The sample was first brought to its exhausted state by irradiating with 970-m μ light for four minutes. The light was removed and the paramagnetic spectrum of Eu²⁺ in the exhausted sample was plotted. The sample was then irradiated with the particular wavelength of exciting light for four minutes. The light was removed. The Eu²⁺ paramagnetic spectrum was replotted, the diminution in its intensity being proportional to the stored energy in the phosphor. The sample was then once again exhausted with 970-m μ light before a second exciting wavelength was used. The results are given in Fig. 1. The heights have been corrected for constant number of exciting photons. The graph of Fig. 1 agrees favorably with that found by Keller, Mapes, and Cheroff³ who used as a measure of the stored energy the intensity of the light emitted by the

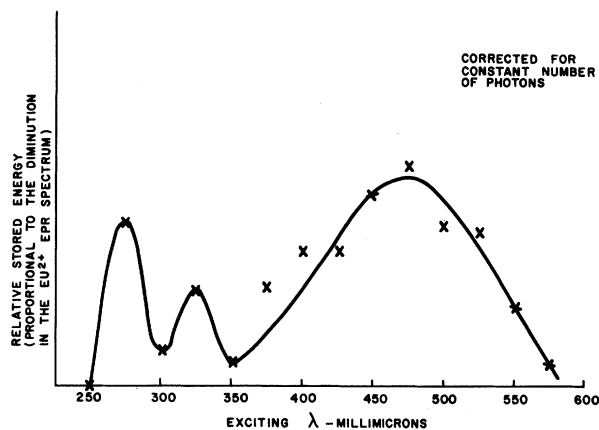


FIG. 1. Relative stored energy as measured by the diminution in the Eu²⁺ paramagnetic spectrum versus exciting wavelength.

sample when 970-m μ light was incident on it.

As a measure of the efficiency of optical excitation it is noted that at the point of maximum optical excitation, 475 m μ , the Eu²⁺ paramagnetic spectrum decreased by 13% in magnitude. This should be taken as a lower limit of the efficiency as the light absorption is not uniform throughout the sample, decreasing exponentially from the surface inward. We plan to measure the variation of efficiency as a function of the relative europium and samarium concentrations. The experiment will also be repeated at liquid helium temperatures looking for a change in the Sm paramagnetic spectrum.

It may be added that 590-m μ light was found to be equally as effective as 970-m μ light in exhausting the phosphor, which agrees with the results of Keller and Pettit.³

In conclusion the use of paramagnetic resonance absorption to detect the optical excitation provides an independent verification of the simplified band-theory model proposed by Keller³ for this phosphor.

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¹W. D. Hershberger, *J. Chem. Phys.* **24**, 168 (1956); W. D. Hershberger and H. N. Liefer, *Phys. Rev.* **88**, 714 (1952).

²B. Bleaney and W. Low, *Proc. Phys. Soc. (London)* **A68**, 55 (1955); W. Low, *Phys. Rev.* **98**, 426 (1955).

³Keller, Mapes, and Cheroff, *Phys. Rev.* **108**, 663 (1957); S. P. Keller and G. D. Pettit, *Phys. Rev.* **111**, 1533 (1958); S. P. Keller, *Phys. Rev.* **113**, 1415 (1959).

TRITIUM AS A PRODUCT OF FISSION*

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Evidence is described that indicates that the triton is a previously unrecognized fission product. Tritium was shown to be present in a ratio of one triton per $(1-2) \times 10^4$ fissions in samples of irradiated natural and enriched uranium and in an irradiated mixture of transuranium isotopes. The triton-to-fission ratio was within a factor of two for these samples even though the concentrations of tritium per gram of irradiated material ranged over a factor of one thousand.

The lithium content of the irradiated uranium was proved not to be the source of the tritium; neither was the tritium present as the result of diffusion from the tritiated heavy water moderator of the Savannah River piles. Confirmation of the triton as a product of fission may contribute to the understanding of ternary fission and may have a practical application to burnup analysis.

A search of the literature of ternary fission showed that the formation of the triton in fission

had not been observed or postulated, although ejection of an alpha particle for approximately each 300 fissions had been well established.^{1,2} Proton emission had also been suspected although this appeared unlikely from a theoretical standpoint.^{1,3}

Recent data collected at the Savannah River Laboratory suggest that a direct correlation exists between triton formation and the number of fissions. These data were taken on irradiated samples of natural and enriched uranium and on an irradiated sample of fissionable transuranium nuclides. In each case, aliquots of nitric acid solutions of the irradiated material were made alkaline with sodium hydroxide, distilled, diluted, and analyzed for tritium with a Packard "Tri-Carb" liquid scintillation spectrometer. For these three cases, the observed frequency of formation was one triton per $(1-2) \times 10^4$ fissions.

Three experiments were carried out to provide proof that the radioactive species being measured was tritium, that the presence of tritium was not due to irradiation of a lithium impurity in the uranium fuel, and that the tritium did not come from the heavy water moderator system of the piles:

(1) A sample of water from the acid solution in which irradiated natural uranium had been dissolved was converted to hydrogen and the gas was diffused through a palladium barrier. The diffusate was reoxidized to water and the expected activity was verified by means of liquid scintillation counting.

(2) The lithium content of the uranium fuel was determined. A 100-gram sample of natural uranium from a typical fuel element was dissolved in nitric acid and the uranium was precipitated by means of pyridine as $UO_4 \cdot 2H_2O$. The supernate, which contained any possible lithium impurity in solution, was evaporated to dryness and the residue analyzed by emission spectrography. The lithium content of the uranium was found to be 0.01 ppm, less than 6% of the amount required to produce the levels of tritium observed in irradiated natural uranium.

(3) The third experiment was more qualitative.

A sample of enriched uranium as U_3O_8 , with a lithium content of < 0.2 ppm, was irradiated in a graphite-moderated experimental reactor which contained no heavy water. The sample was dissolved in nitric acid and the water was separated by distillation from alkaline solution. The water was then reacted with hot zinc to produce hydrogen that was diffused through a palladium barrier. The diffusate was reoxidized to water and counted in the liquid scintillation counter. An estimated 10^{15} fissions in the original sample were found to produce about 2×10^{-3} μC of tritium, a ratio of about one triton per 3×10^4 fissions. Less than 4% of the observed activity could be attributed to the irradiation of lithium-6.

It is postulated on the basis of these findings that like the alpha particle, the triton is produced by direct emission in ternary fission.

Further experiments are planned to establish the frequency of formation with a greater degree of accuracy over a wide range of exposure for a variety of fissionable nuclides and to investigate, as a possible application, tritium analysis as an index of fuel "burnup."

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¹D. L. Hill, Phys. Rev. 87, 1049 (1952).

²C. B. Fulmer and B. L. Cohen, Phys. Rev. 108, 370 (1957).

³D. L. Hill and J. A. Wheeler, Phys. Rev. 89, 1102 (1953).