EMP

702256

ANL-7360

ANL-7360

Argonne National Laboratory

RADIOLOGICAL PHYSICS DIVISION

ANNUAL REPORT

JULY 1966 THROUGH JUNE 1967

TABLE OF CONTENTS

THERMOLUMINESCENT MICRODOSIMETRY IN DENTAL RADIOLOGY. I. TECHNIQUE*

C. E. Rehfeld,† J. Kastner, B. G. Oltman, H. M. Rosenberg,‡ and D. E. Doyle†

Irradiation of lithium fluoride (LiF) crystals leads to an excited energy state due to the entrapment of freed electrons in the LiF crystal defects. The electrons remain trapped at room temperatures, but are released at higher temperatures, e.g. 200°C. When the irradiated LiF crystals are raised to a high temperature in a short time (15–30 seconds), de-excitation occurs with an emission of visible light. This emitted light is registered by an electronic phototube and recorded in the form of a glow curve on a strip chart. The integral of the curve is directly proportional to the ionizing radiation exposure.

Nine milligrams of annealed LiF powdered crystals are loaded into each section of a plastic tube measuring 1 × 10 mm in outside dimensions. Each section of loaded tube is heat-sealed to form one microdosimeter; it is small enough to be placed in a large dental cavity or in the dental supporting tissues. After x-ray exposure, the powder is emptied from the tube onto a small pan, heated to obtain a glow curve, and weighed as a final step. The integral of the glow curve is divided by the weight of LiF in the pan to determine the response per mg of LiF.

Calibration, in roentgens, is accomplished by exposing a number of microdosimeters to radioactive Ra or Co standards. The irradiation response per mg of the LiF exposed to Ra or Co provides the factor for conversion of response per mg of x-ray exposure to roentgens. Ionizing effects of beta, gamma, and x-ray radiation can all be measured equally well. The variation in 0.1 and 1.0 R readings is ±5% and ±2%, respectively.

* Abstract of a paper presented at the International Association of Dental Research (Radiology Section) held in Washington, D. C., March, 1967. Publication is planned for the *Journal of Oral Surgery, Oral Medicine and Oral Pathology.*

† Biological and Medical Research Division.

‡ University of Illinois College of Dentistry, Chicago.

THERMOLUMINESCENT MICRODOSIMETRY IN DENTAL RADIOLOGY. II. CLINICAL APPLICATION*

H. M. Rosenberg,† C. E. Rehfeld,‡ J. Kastner, B. G. Oltman, D. E. Doyle,† and D. J. Hauptfuehrer†

Lithium fluoride microdosimeters are being used to measure relative and absolute x-ray doses to skin, teeth, and other oral structures in dental patients. This pilot study was designed to develop a technique for the employment of these dosimeters in radiology.

Each microdosimeter is placed in one of three anatomical locations to be aligned in an x-ray beam. The dental films are processed to assure adequate exposure to produce routine x-ray images. The microdosimeters are read in the manner described by Kastner.[1]

The initial group consisted of 50 randomly selected patients. Eight primary dental x-ray examination areas on the right side of the face were selected, and two or three patients were employed for each area. Fixed wet specimens of maxillae and mandibles, from which no tissue had been removed, provided teeth for intradental measurements. Microdosimeters were placed in the pulp chambers and in the adjacent gum tissue of 20 teeth. All x-ray exposures were made at 90 kVp, 15 Ma at $\frac{1}{10}$ second, using a standardized reproducible technique.

This study demonstrates that lithium fluoride microdosimeters can provide total dose and dose ratio data. Approximately $\frac{1}{3}$ roentgen is required to produce an intraoral x-ray film. The average intradental dose was 142 millirads with a range of 126 to 153, and the gum level dose averaged 207 millirads with a range of 159 to 223.

* Abstract of a paper presented at the International Association of Dental Research (Radiology Section) held in Washington, D. C., March, 1967. Publication is planned for the *Journal of Oral Surgery, Oral Medicine and Oral Pathology.*

† University of Illinois College of Dentistry, Chicago.

‡ Biological and Medical Research Division.

REFERENCE

1. J. Kastner, R. Hukkoo, and B. G. Oltman. Argonne National Laboratory Radiological Physics Division Annual Report, July 1964 through June 1965. ANL-7060, p. 15.

THOROTRAST-EXPOSED PERSONS IN THE CHICAGO AREA

*Harold May and Edwin Kingsley**

INTRODUCTION

Examination in the whole-body counter of four individuals containing colloidal Th^{232} and its daughter elements in the liver and spleen, accompanied by skeletal deposits resulting from translocation and redeposition, has been previously reported.[1] These individuals were referred to us from a large medical center in a neighboring state as part of a study to demonstrate the feasibility of a large-scale study of many persons known to have been injected with Thorotrast at this hospital. In the course of evaluating the significance of these measurements and the means for obtaining more detailed information from whole-body counting, certain conclusions have been reached. These are:

(a) The principal gamma rays observed originate in the decay of the daughter elements actinium-228, lead-212, bismuth-212, and thallium-208. Calculation of the burden of Bi^{212} based on the 1.62 and 2.2 MeV photons emitted is always in agreement with the Tl^{208} as calculated from the 2.62 MeV gamma ray. While a certain amount of simplification in the observed spectra results from coincidence or summing-coincidence modes of operation, these seem to offer no compelling advantage for the purposes of estimating total body burden.

(b) Accuracy in the quantitative measurement of Ac^{228} from gamma emission at 907 and 964 keV is significantly affected by the amount of potassium-40 present, since degraded photons resulting from Compton scattering of the 1.46 MeV radiation of K^{40} form an underlying continuum upon which the thorium contribution in the 900 to 960 keV region must be "stripped." The K^{40} content is, in turn, somewhat difficult to determine because of interfering radiation at 1.62 MeV from Bi^{212}. Normal metabolic processes, principally excretion of radium-224 (half-life of 3.6 days) and exhalation of radon-228 (thoron, half-life of 54 seconds), result in approximately 80% retention of the late daughter elements relative to the Ac^{228}. The relative amounts of each isotope are expected to differ with deposition site, being most nearly in radioactive equilibrium in the primary deposit. If the crystal detector is positioned some distance from the body so as to afford constant response to all locations, accuracy is impaired by the reduced number of events which may be detected in a time period limited by patient discomfort.

(c) Some success in estimation of skeletal burden from measurement over the tibiae and fibulae has been achieved. Information about the amounts of activity translocated to other organs such as the lungs and kidneys is desired. The close proximity of these organs to the primary deposits, as well as their much lower expected total burden, make this very difficult, and it remains to be seen whether direct, *in vivo* detection of these secondary sites is indeed possible.

Thus, the further application and improvement of procedures briefly outlined above call for improved spatial resolution and sensitivity, accompanied by freedom from, or at least some systematic and rational compensation for, normal variability in patient size, physique, and habitus, with the concomitant fluctuations in potassium-40 (and, to a constantly diminishing extent, of cesium-137) content, organ size and location, etc. Further development of measurement techniques and the application of sophisticated computer manipulation of data are desirable.

But in addition to these technical problems, there are logistic ones. We refer to the desirability, if not necessity, of having available for study some Thorotrast-exposed individuals who are accessible and cooperative. The first requirement may best be realized by residence within 40 or 50 miles of the Laboratory. It also requires that the persons be reasonably free to be called in at our convenience; i.e., that they not be employed full time. The cooperativeness of these individuals involves personality factors as well as age and physical condition.

The results of direct, *in vivo* measurements of activity translocated to secondary sites will be, at very best, of low accuracy. Therefore, these should eventually be correlated with measurements of individual tissue samples obtained at time of death and autopsy. The applicability of the above qualifications for this further phase of the investigation is obvious.

CHICAGO AREA SEARCH

Since Thorotrast has been used extensively as an x-ray contrast media in cerebral angiography, a search of medical records for the names of individuals undergoing such examination was undertaken. It was ascertained that of the many neurosurgeons in practice in the Chicago area during the period when use of Thorotrast was common, only two individuals em-

* Summer employee, 1966.

ployed it extensively. One of these is still active, and was able to furnish us with a log-book listing all angiographic examinations performed between 1942 and 1956, listed consecutively and summarized yearly. The other individual had retired from practice and left the state; however, it was known that most of his practice was done at one hospital. Since the hospital records of this institution were found to be exceptionally well organized, all of the pertinent records were located and examined with no difficulty.

The task of record search was carried out by the junior author, a medical school student of the University of Cincinnati employed in the Radiological Physics Division during the summer of 1966. It was found that the type of records kept, and their accuracy and completeness, differed greatly among the various institutions. The operative record sheet normally would contain spaces to record site of injection, type of contrast medium used, and amount injected. Frequently, these spaces were found blank, supplemented only by the cryptic remark "Usual dye injection made."

The results of this record search are summarized in Tables 20 and 21.

As the names of verified Thorotrast cases were obtained, search procedures were instituted, usually by reference to telephone and address listings, contact with the listed family physician or place of employment, or next of kin. In some cases, a personal visit to the last known address and interview with former neighbors was necessary. In view of the great mobility of most city-dwellers, and the rapid changes that have taken place in most of the city neighborhoods during the past two decades, it is a matter of great satisfaction that 90% of the patients on record have been either located or their death authenticated.

TABLE 20. SUMMARY OF RECORD SEARCH

Source of records	Records examined	Use of Thorotrast verified
Hospital W	850	31
Hospital X	300	57
Hospital Y	40	4
Neurosurgeon Z	115	1
Miscellaneous	4	4

TABLE 21. BREAKDOWN OF RECORDS EXAMINED, HOSPITAL W

Type of dye not specified	208
Diodrast employed	421
Thorotrast employed	31
Other contrast media used	63
Patient record not found—missing	53
Patient record not found—error in admission number versus patient's name, etc.	62
Arteriogram not successful	12
	850

TABLE 22. TIME DISTRIBUTION, KNOWN THOROTRAST CASES

Year	All cases	Known deceased	Known living	Not located
1934	1		1	
1945	2			2
1946	3	2	1	
1947	3	2	1	
1948	6	5	1	
1949	3	1	2	
1950	4	4		
1951	8	4	3	1
1952	15	8	5	2
1953	18	12	6	
1954	10	5	4	1
1955	8	4	3	1
1956	5	3	1	1
1957	3	3		
1958	3	3		
1959	1	1		
Total	93	57	28	8
Date unknown	4	2	2	
Grand total	97	59	30	8

We had expected to find that Thorotrast was most frequently used around 1938–1941, but analysis of all verified cases reveals a much later peak date. A partial explanation for this lies in the statement of the neurosurgeon associated with hospital W, to the effect that the radioactivity of Thorotrast was known, and its possible hazards recognized; hence, it was used only after careful consideration of the patient's age and probable prognosis, with due consideration being given to the possible long-term radiation effects versus the immediate need for radiographs having optimum information content. The time distribution of injection for all records examined is summarized in Table 22.

A brief analysis of the ages of these patients reveals the following:

Age at injection, located (mean value)	47.9 years
Age at injection, known deceased (mean value)	51.8 years
Age at death (mean value)	55.0 years
Present age, located (mean value)	62.5 years

For individuals known to be dead, the average time elapsed between injection and death is 3.2 years, indicating that Thorotrast was the preferred contrast medium in cases of short life expectancy. By contrast, those located and hence still known to be living

have survived for an average period of 14.6 years. Many of these are approaching or have exceeded their normal life expectancy—four are in their 70's and nine more are between 65 and 70. If this small sampling can be considered representative of the situation in the entire United States, it is obvious that the majority of recorded Thorotrast-exposed individuals have already expired, and that prompt measures should be taken to examine those remaining.

It is impossible to mention all the persons that have been contacted and have made suggestions or have been of help in the patient search. Our special thanks go to Miss Rita Finnegan and Mrs. Yamaguchi, medical record librarians at the two hospitals where most of this work was done. Mr. Robert Xenos of the Security Division, ANL, was able to locate several of these persons when there were seemingly no leads at all. The unfailing interest and encouragement of Mr. L. D. Marinelli is also deeply appreciated.

REFERENCE

1. H. A. May and L. D. Marinelli. A Preliminary Study of Gamma Rays Emitted by Thorotrast Patients. Argonne National Laboratory Radiological Physics Division Annual Report, July 1964–June 1965. ANL-7060, pp. 93–99.

PHOTOIONIZATION STUDIES IN THE VACUUM ULTRAVIOLET*

James C. Person

Recent results on the effects of deuterium substitution on the photoionization of gaseous molecules [1, 2] by photons with energies up to 11.7 eV are presented. The results in benzene and acetone indicate that preionization of superexcited states is an important ionization mechanism for these molecules, whereas the results in ammonia, *n*-butane, and ethylene indicate that direct ejection of an electron during the excitation act is the dominant ionization mechanism for these molecules. Ethylene is an especially interesting case, since the increase in the photoionization yield upon deuteration is caused by an isotope shift in the production of neutral excited states rather than by preionization. Another interesting result is the observation of chemi-ionization in acetone just below the ionization threshold.

* Abstract of an invited paper presented before the Lake Superior Section of the American Chemical Society, Duluth, Minnesota, April 27, 1967.

REFERENCES

1. James C. Person. *J. Chem. Phys.* **43**, 2553 (1965).
2. James C. Person and Paul P. Nicole. This report.

ISOTOPE EFFECTS IN THE PHOTOIONIZATION OF ACETONE, BUTANE, ETHYLENE, AND AMMONIA

James C. Person and Paul P. Nicole

INTRODUCTION

As a continuation of a program of studies of isotope effects on the photoionization of gaseous molecules,[1] we have studied the photoionization of acetone, *n*-butane, ethylene, and ammonia, together with their fully deuterated analogues. Measurements were made of the total absorption cross section, $\sigma(E)$, and the photoionization yields, $\eta(E)$, (this quantity is also called the photoionization efficiency and it is defined as the probability that photon absorption will produce an ion pair) as a function of photon energy, E, for $E \leqq 11.7$ eV.

From observations of an increase in $\eta(E)$ upon deuteration, one can determine some of the cases where the preionization of superexcited states makes a large contribution to the ionization, since the preionization processes, presumably invariant under isotopic substitution, compete with atomic rearrangement processes, such as dissociation, which are slower in heavier isotopic molecules.[2] Such an effect was observed in the photoionization of benzene,[1] and similar isotope effects had been observed earlier in the cases where ionization was produced by collision with excited atoms,[3, 4] by β particles,[5] and by 70-eV electrons.[6]