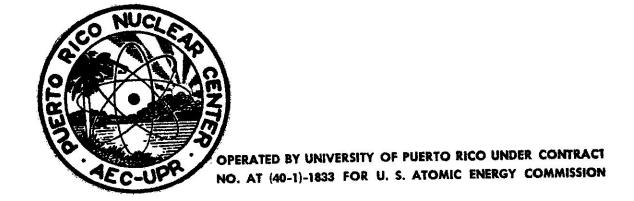
PUERTO RICO NUCLEAR CENTER

RESONANCE IN RADIATION EFFECTS

Progress Summary Report No. 1



RESONANCE IN RADIATION EFFECTS
Progress Summary Report #1
March 1963

Henry J. Gomberg - Principal Investigator Robert A. Luse Florencio Vázquez Martínez

Puerto Rico Nuclear Center
operated by the
University of Puerto Rico
for the
U. S. Atomic Energy Commission
under Contract No. AT-(40-1)-1833

INTRODUCTION.

To answer the question "What are the unique effects of ionizing radiation on matter?", our research program continues a study started at the University of Michigan under the principal investigator. The Michigan program is now under the direction of Prof. Hoyt Whipple, while in Puerto Rico a new project has been created.

The first objective of the Puerto Rico Program is to provide an independent test of the original results reported from Ann Arbor. The second is the extension of the investigation to new problems in biological, chemical and physical effects produced by monochromatic X-rays in the "less than 20 kilovolt" energy range.

Below is a summary of the results obtained during the first year of operation of the Puerto Rico project.

Section I. Resonance Radiation Effects of Low-Energy Monochromatic
X-rays on Catalase (by R. A. Luse)

Dilute solutions of the iron metalloenzyme catalase were irradiated with a beam of monochromatic X-rays having photon energies in the 6-9 Kev region and with a high purity (± 50 ev). Irradiation was carried out in a sample holder constructed of methacrylate plastic with a thin Mylar film window. Approximately 0.2 ml of sample could be irradiated in the chamber (6 x 9 x 4 mm HMD); a chamber of similar size, shielded from radiation, contained control solution. Masking of solution under the conditions of complete absorption was avoided by mechanical stirring with a fine glass rod. To minimize evaporation and inactivation of the catalase, solutions were covered and maintained at 5°C by passing water from a constant temperature, refrigerated bath through the sample holder.

In conjunction with this irradiation system, a ferrous-ferric micro-dosimetry technique was developed. The prime requirement specified for the dosimeter system was the ability of direct substitution for the sample, to ensure that values of radiation intensity measured with the dosimeter correspond directly to those absorbed by the biological sample. Other considerations were simplicity of use and reliability in the low dose ranges involved in this work. The Fricke ferrous-ferric dosimeter was chosen, since it is the best characterized secondary standard available. It relies on the oxidation, by ionizing radiation, of ferrous ion to ferric ion, and on determination of the concentration of ferric ion formed by its light absorption at 304 mu (cf. Schuler and Allen). Application of this dosimeter is primarily limited by the methods for ferric ion analysis.

A considerably more sensitive assay of ferric ion concentration is measurement of the absorption of the red-orange ferric-thiocyanate complex; the molar absorptivity of this complex is 10,000 - 14,000 liter mole-1 cm⁻¹ at 480 mu. Pribicevic, Gal, and Draganic have characterized this complex formation and proposed its use as a dosimetry system in the 300 - 100 rad dose range. Not all their results could be confirmed, so that further characterization and modification of the system were undertaken; these changes were incorporated in the procedure developed. To avoid dilution of the irradiated solution, optical measurements were taken with the Beckman DU spectrophotometer in cuvettes of 3 mm x 10 mm x 25 mm chamber dimensions; as little as 0.22 ml of solution may be assayed in such cuvettes. X-radiation intensities of 2 x 10¹¹ photons per hour in the sample holder have been determined by this method (6-hour irradiation periods were required). The results of this chemical dosimetry

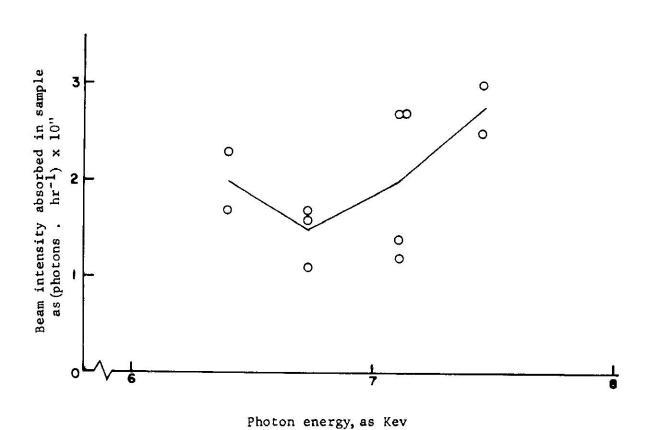


Figure 1. Monochromatic \mathbb{X} -ray beam intensity as a function of photon energy

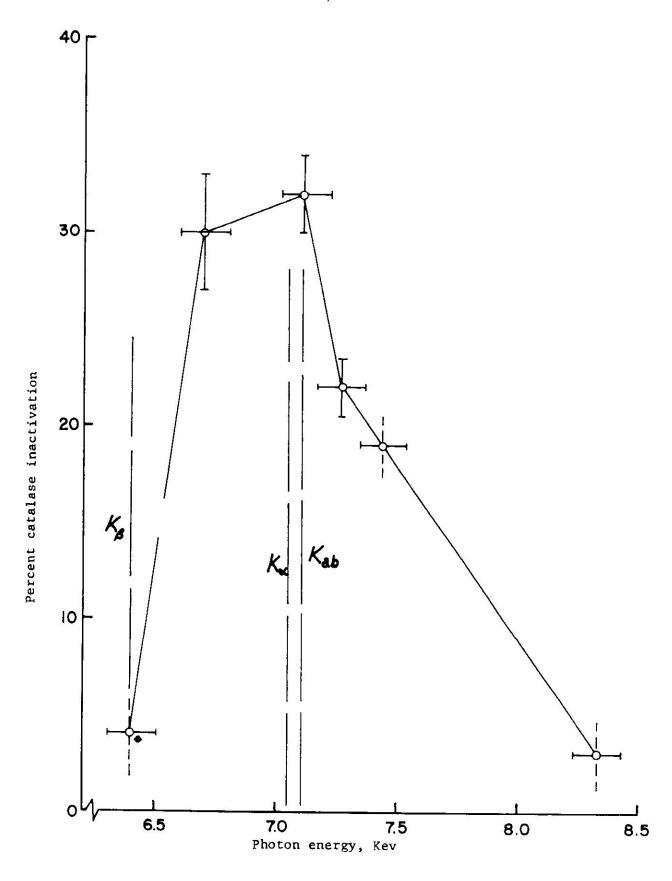


Figure 2. Resonance radiation effect of monochromatic X-rays on catalase

Total dose absorbed by sample = 1.4 x 10^{13} photons (except for starred point, where dose = 0.2 x 10^{13})

in the photon energy range of current interest are given in Figure 1.

There is no resonance radiation effect in the ferrous dosimeter at the K-absorption edge of iron; this is as expected, since the ferrous to ferric oxidation is due to "indirect" effects.

The extent of enzyme inactivation was determined using essentially the standard assay for catalase developed by Beers and Sizer in which the disappearance with time of substrate hydrogen peroxide is followed spectrophotometrically at 212 mu.

The results of such irradiation experiments using monochromatic x-rays in the energy range 6.4 - 8.3 Kev are given in Figure 2. Here increased inactivation of the iron enzyme at or near the K-absorption edge of iron (7.11 Kev) is obvious. This is taken to confirm the resonance radiation hypothesis of Gomberg and previous experimental work of Emmons and Paraskevoudakis.

Section II. Characterization of a High Intensity Monochromatic XIrradiation System in the 5-20 Kev. Region (by F. Vázquez
Martínez)

The x-ray emission system utilized for the present resonance radiation studies was characterized quantitatively as to intensity and photon energy distribution, and second harmonic contamination.

The monochromatic x-ray beam resulting from crystal diffraction and collimation was analyzed horizontally across its front for (a) intensity distribution, utilizing a special moving slit device; and (b) photon energy distribution, using double diffraction by a second analyzer crystal. The results of this work are shown as Figures 3 and 4.

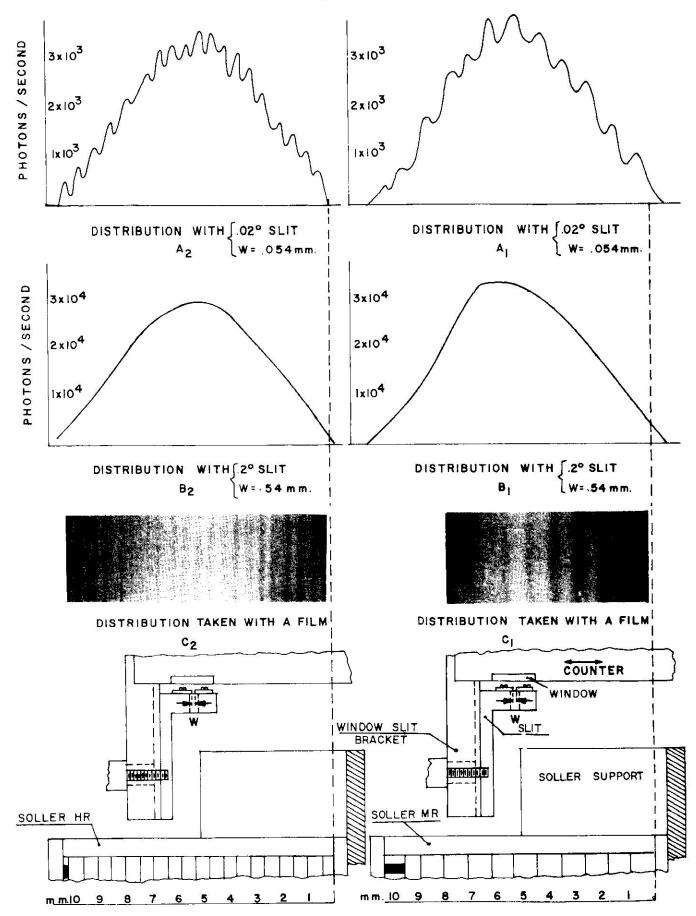


Figure 3. Intensity Distribution

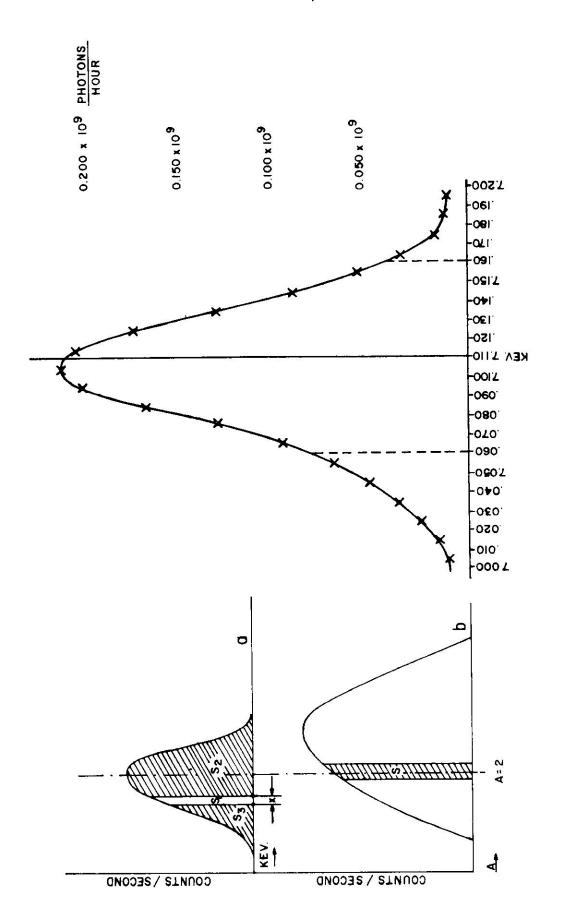


Figure 4. Intensity and Energy Distribution

across the beam and energy distribution at point A - 2

Total intensity distribution

Energy distribution of photons in the whole beam

Estimation of the extent of second harmonic energies was made on the basis of (a) absorption measurements relying on the different mass absorption coefficients at the first and second harmonic wavelengths and (b) double diffraction measurements in which photons with second harmonic energies were analyzed separately. Correction for percentage of reflection by second harmonic energies also was determined by the double diffractometer method. Contamination by higher harmonics was shown to be considerable at higher operating voltages (see Figure 5); monochromatic beams can be obtained only by proper selection of tube potential.

The effects on the beam of positioning the various components of the x-ray system (tube, diffraction crystal, two soller slits) were determined and the system was selected which provides high uniformity of photon energy distribution.

As a result, a diffraction system was developed which permits irradiation with photons of uniform energy distribution (± 50 ev in 6000-9000 ev), a selectivity of photon energies not possible with fluorescent emission systems.

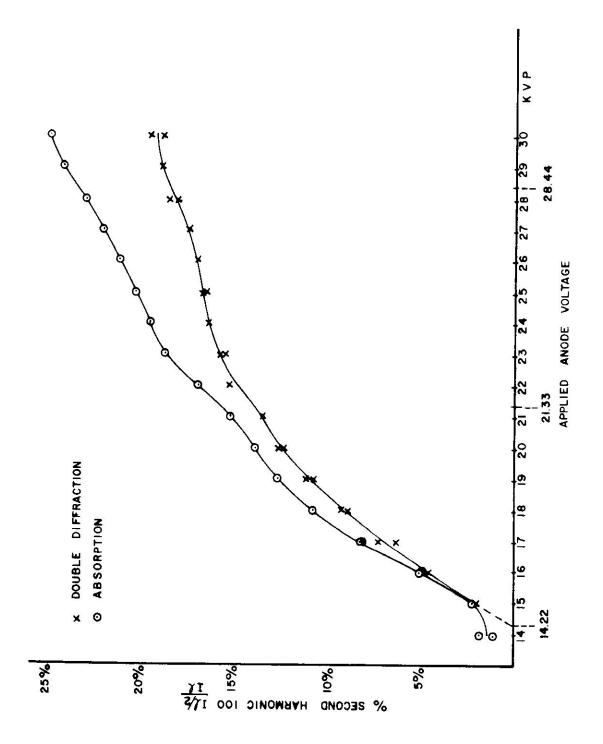


Figure 5. Percentage of the second harmonic contamination

REFERENCES

- 1. Schuler, R. H. and A. O. Allen, "Yield of the Ferrous Sulfate Dosimeter", J. Chem. Phys. 24, 56-9 (1956).
- Pribicevic, S. M., O. S. Gal, and I. G. Draganic, "Use of the Fricke Dosimeter for the Measurement of Doses Below 1000 Rads", Report NR-001-0022-1961 Inst. of Nuclear Sciences "Boris Kidrich".
- 3. Gomberg, H. J., "Resonance in Radiation Effects", Technical Report No. 1. University of Michigan 1960, Contract No. AT(11-1)-684.
- 4. Emmons, A. H., "Resonance Radiation Effects of Low Energy Mono-chromatic X-rays on Catalase". Doctoral Thesis University of Michigan 1959 (also Technical Report #2, Contract No. AT(11-1)-684
- 5. Paraskevoudakis, P., "Resonance in Radiation Effects" Technical Report #5 Contract No. AT(11-1)-684 November 1960.