Ceric and Ferrous Dosimeters Show Precision for 50–5000 Rad Range

The problem:
To develop ferrous and ceric dosimeters for use in mammalian radiobiology that would be sensitive in the biologically interesting 50 to 1500 rad range. The classical ferrous sulfate (Fricke) dosimeters have been used to date; however, they exhibit low sensitivity in the desired range, have a tendency to autoxidation, and require a temperature-controlled ultraviolet spectrophotometer. In developing the improved dosimeter, systems involving the addition of sensitizers (e.g., benzoic acid) before irradiation were avoided since this created a different radiochemical situation and required readetermination of radiochemical yields (G values) as functions of linear energy transfer (LET), dose rate, etc. Similarly, determinations of remaining ferrous ions were avoided, for they led to poor precision and reproducibility, especially if exposure lasted a period of several hours.

The solution:
Ammonium thiocyanate, added to the usual ferrous sulfate dosimeter solution, yielded a very stable, precise and temperature-independent system—eight times as sensitive as the classical Fricke system in the 50 to 5000 rad range.

Ceric dosimeters, which are promising for use in mixed radiation fields, possessed a response nearly independent of LET. Fluorometry of an irradiated ceric solution, to which fluorescein was then added, afforded a method several hundred times more sensitive than previous ceric dosimeters.

How it’s done:
The ferrous dosimeter was produced by adding ammonium thiocyanate to a ferrous sulfate dosimeter solution which had been irradiated and handled in the usual way. It was composed of 0.001 M in Fe(NH₄)₂(SO₄)₂, 0.001 M in NaCl, 0.4 M in H₂SO₄, and was aerated before use.

This combination produced a two-fold increase in sensitivity over previous thiocyanate methods because of the increased SCN concentration obtainable with the ammonium salt NH₄SCN, also diminished temperature sensitivity, and decreased the rate of ferrous ion autoxidation.

Two methods have been used successfully for the determination of the ferric thiocyanate complex. Both employ visible region colorimeters at 4750 and 4820 Å. In the first, one volume of a saturated aqueous solution of NH₄SCN was added to three volumes of irradiated dosimeter. In the second, dosimeter was added to a known mass of solid ammonium thiocyanate. This latter method was more sensitive because of the lack of dilution.

Absorbance was strictly proportional to ferric ion concentration, with ε₄₈₂₀ = 17,600 liter mole⁻¹ cm⁻¹, from A = 0 to A = 1.0. No temperature effect was found between 23.5° and 31° C and no time effect between 5 to 25 minutes after mixing. In a 10-cm cuvette, 50 rads gave A = 0.126 ± 0.001.

The ceric dosimeter system was based on the observation that the fluorescence of fluorescein is quantitatively destroyed by the ceric ion. Fluorescence of one volume of aqueous fluorescein, added to 12.5 volumes of irradiated ceric dosimeter, was found to be a quasi-linear function of dose over the 50–5000 rad range with a precision of ±1%, and no observable temperature or time dependence between 20° to 30° C and 5 to 100 minutes.

(continued overleaf)
Notes:

2. Inquiries concerning this innovation may be directed to:
   Office of Industrial Cooperation
   Argonne National Laboratory
   9700 South Cass Avenue
   Argonne, Illinois 60439
   Reference: B68-10426
   Source: N. A. Frigerio and V. D. Henry of the Biological and Medical Research Division
   (ARG-10173)

Patent status:
Inquiries about obtaining rights for commercial use of this innovation may be made to:
   Mr. George H. Lee, Chief
   Chicago Patent Group
   U.S. Atomic Energy Commission
   Chicago Operations Office
   9800 South Cass Avenue
   Argonne, Illinois 60439