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DOSIMETRY IN THE ARGONNE HIGH-LEVEL
GAMMA-IRRADIATION FACILITY

by

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At the present time chemical dosimetry utilizing the oxidation of ferrous sulfate is used as the method of measurement of radiation intensity of the Argonne National Laboratory's High-Level Gamma-Irradiation Facility.

Ferrous sulfate (0.001 N) and sodium chloride (0.001 N) are dissolved in 0.8 N sulfuric acid. The solution is placed in an appropriate container and irradiated in the source for a predetermined period of time, depending on the approximate intensity of the source.

As most food samples are irradiated in No. 2 cans ($3\frac{7}{16}$ in x $4\frac{9}{16}$ in), the ferrous sulfate solution is placed in either a glass cell surrounded by water inside of the can or in a polystyrene cell in a polystyrene holder inside the can (see Figures 1, 2, and 3).

Five No. 2 cans containing ferrous sulfate solutions in the cells are placed one on top of the other inside the irradiation urn (see Figure 4). The urn is lowered by cable through a chute into the irradiation rack. The time of irradiation is determined by a stop watch or an electric timer. The use of the hand cable and timing by a stop watch is shown in Figure 5; use of an electrically timed cable is shown in Figure 6. A schematic diagram of the irradiation urn and rack is shown in Figure 7.

The so-called "hot" spots, i.e., those having four fuel rods surrounding the sample, are measured by allowing the ferrous sulfate to remain in the source for only 15 to 60 seconds, as 0.001 N ferrous sulfate solution can only be utilized for intensity measurements as high as 50,000 r.

Preparation of Ferrous Sulfate Solution

Reagents:

FeSO₄ (0.001 N)
NaCl (0.001 N)
H₂SO₄ (0.8 N)

Method

Prepare an approximately 0.8 N sulfuric acid solution (22.5 ml concentrated reagent grade H₂SO₄ made up to one liter

with very pure distilled water* in a volumetric flask). The acidity is critical, so that the acid should be standardized against sodium carbonate, using methyl orange as the end-point or a pH of 4.3 on a pH meter. The normality of the acid should lie between 0.79 and 0.81.

Make up a 1.0 N sodium chloride solution (58.46 gm reagent grade NaCl per liter).

Weigh out exactly 0.2780 gram ferrous sulfate (reagent grade $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) and place this in a clean, dry, one liter volumetric flask. Add about 500 ml 0.8 N H_2SO_4 to dissolve the ferrous sulfate. Add 1 ml of the 1 N NaCl solution, then make up to volume with 0.8 N H_2SO_4 . The resulting solution will be 0.001 N NaCl and 0.8 N H_2SO_4 . The optical density of this solution, when read at 3020 Å, should be less than 0.010.

Irradiation

The ferrous sulfate solution is placed in clean irradiation vials, preferably made of thin-walled Pyrex glass, and then in the irradiation rack. The irradiation period should be so chosen that the difference in optical density between the irradiated and unirradiated ferrous sulfate solution is about 0.5. After irradiation the solution is transferred to quartz Beckman cells and its optical density determined in a Beckman DU spectrophotometer in the ultraviolet using a wave length of 3020 Å. The Beckman cells must be very clean. These are cleaned by rinsing in weak detergent solution, followed by thorough rinsing with distilled water. Four Beckman cells are placed in the cell holder: the first cell is filled with distilled water, the second with the unirradiated ferrous sulfate solution, and the third and fourth with the irradiated ferrous sulfate solutions. The unirradiated ferrous sulfate solution should have an optical density less than 0.010. Whenever the optical density of the unirradiated solution (or blank) becomes greater than 0.030 it should be discarded. The temperature is critical, so that a constant temperature must be maintained or a temperature correction factor used. All of our readings are made at 25° C.

*Very pure distilled water, free of organic matter, is required. If there is any question about the purity of the water, double or triple distillation is recommended. At Argonne a Barnstead still is used and it is piped through aluminum lines. It is freshly distilled daily, thus reducing the possibility of impurities.

FERROUS DOSIMETRY

Calculations

$$\frac{dN}{dt} = G \frac{dE}{dt} \quad (1)$$

where:

N = number of molecules or number of ions
 E = energy absorbed
 E₁₀₀ = energy absorbed per 100 ev.
 G = "G" value = number of molecules reacted per 100 ev.
 t = time

If N is in molecules/liter, then E is in units of energy/liter.

$$\frac{d \mu N}{dt} = G \frac{dE}{dt} \times \frac{1}{6.02 \times 10^{17}} \quad (2)$$

$$\mu N = \frac{G E_{100}}{6.02 \times 10^{17}} \quad (3)$$

$$E_{100} = \frac{\mu N \times 6.02 \times 10^{17}}{G} \quad (4)$$

$$E_{ev/1} = \frac{\mu N \times 6.02 \times 10^{17} \times 10^2}{G} \quad (5)$$

$$E_{erg/1} = \frac{\mu N \times 6.02 \times 10^{19}}{G} \times 1.602 \times 10^{-12} = \frac{0.9644 \times 10^8 \mu N}{G} \quad (6)$$

$$E_{ergs/ml} = \frac{0.9644 \times 10^5 \mu N}{G} \quad (7)$$

$$E_{rad}(Soln.) = \frac{E_{ergs/ml}}{100} = \frac{0.9644 \times 10^5}{15.6 \times 100} \mu N = 0.6182 \times 10^2 \mu N \quad (8)$$

$$E_{rad} = 61.82 \times 449 \times O.D. = 2.78 \times 10^4 O.D. \quad (9)$$

$$E_{rad} (FeSO_4 \text{ in } 0.8 \text{ N } H_2SO_4) = \frac{2.78 \times 10^4 O.D.}{1.024} = 2.71 \times 10^4 O.D.$$

$$E_{rad} (H_2O) = 2.71 \times 10^4 O.D. \times \frac{97.54}{97.23} \quad (10)$$

$$= 2.71 \times 10^4 \times 1.003 O.D. = 2.72 \times 10^4 O.D.$$

$$1 \text{ r of } \gamma \text{ radiation in H}_2\text{O} = 97.5_4 \text{ ergs/gm}$$

$$1 \text{ r of } \gamma \text{ radiation in } 0.8 \text{ N H}_2\text{SO}_4 = 97.2_3 \text{ ergs/gm}$$

$$1 \text{ r of } \gamma \text{ radiation in air} = 87.7 \text{ ergs/gm}$$

$$1 \text{ erg} = 6.24 \times 10^{11} \text{ ev}$$

$$1 \text{ ev} = 1.602 \times 10^{-12} \text{ erg}$$

$$G = 15.6 \text{ for Fe}^{+++}$$

$$\mu\text{N (Fe}^{+++})_{25\text{C}} = 449 \times \text{O.D.}$$

$$\text{O.D.} = \text{optical density}$$

$$\text{rad} = 100 \text{ ergs/gm}$$

Glass Dosimetry Cell is Being Placed in Polystyrene Holder.
Cell is 2 in. long and $\frac{3}{4}$ in. in diameter. The base is a 1-in. long rod.
The cell is made of thin-walled Pyrex glass
and has a capacity of 13 ml.

Figure 1

Glass Dosimeter Cell shown in the Center of a Polystyrene Holder
Inside a No. 2 Can.

Figure 2

Polystyrene Dosimeter Cell and Holder

The upper portion of the photograph shows a No. 2 can, a solid polystyrene insert, the polystyrene cell and the top of the insert. The lower portion of the photograph shows the dosimeter cell in place and the polystyrene cap being placed on the insert.

Figure 3

Loading Cans Containing Dosimeters into the Irradiation Urn.

Figure 4

Hand Cable Being Used for Immersing Urn in Irradiation Rack.
Timing Being Done With a Stop-Watch.

Figure 5

An Electrically Operated Cable, Electrically Timed to 0.001 Min.
Timers are Shown on Wall in Background.

Figure 6

Gamma Irradiation Rack

F = fuel elements

Location of Sample in Urns

A Schematic Diagram, Top View, of the Gamma-Irradiation Rack.

Figure 7