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S-611 - Rpt #4(Final)
Contract: DA19-129-qm-1900
Stanford Research Institute

A Review of Dosimetry Field

Period: 14 September 1961 - 13 September 1962

ARMED FORCES FOOD AND CONTAINER INSTITUTE
U. S. Army Research and Engineering Command
Chicago 9, Illinois

AD Accession No.
STANFORD RESEARCH INSTITUTE, Menlo Park, California

REVIEW OF DOSIMETRY FIELD—S. I. Taimuty
Report No. 1, 1 Sept. 1962, pp. 24
(Contract DA 19-129-QM-1100)
Project No. 7-84-01-002, UNCLASSIFIED REPORT

This report presents a review and critical evaluation of the literature on high-level radiation dosimetry published since 1958. The purpose of the review was to establish whether recent dosimetry research has developed any new dosimeters which are suitable for use in the Quartermaster Radiation Laboratory and for inclusion in a dosimetry handbook to be prepared as part of this contract work. Of the 13 new dosimeters discussed in the report, only three, oxalic acid, polyisobutylene, and Mylar film, seem sufficiently promising to warrant further development. Their current states of development are not sufficiently far advanced, however, to warrant their inclusion in a definitive dosimetry handbook. It is recommended that only the Fricke and ceric sulfate

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September 10, 1962

CONTRACT RESEARCH PROJECT REPORT

QUARTERMASTER FOOD AND CONTAINER INSTITUTE FOR THE ARMED FORCES, CHICAGO
HQ., Research and Engineering Command, QM Research and Engineering Center,
Natick, Massachusetts

Stanford Research Institute
Menlo Park, California

Project No: 7-84-01-002
Contract: DA 19-129-QM-1900
(OI 6004-62)

Official Investigator: S.I.Taimuty

Report No. 4 (Final)

Period: 14 Sept 61 - 13 Sept 62
Initiation Date: 14 Sept 61

Title of Contract: Review of Dosimetry Field

SUMMARY

This report presents a review and critical evaluation of the literature on high-level radiation dosimetry published since 1958. The purpose of the review was to establish whether recent dosimetery research has developed any new dosimeters which are suitable for use in the Quartermaster Radiation Laboratory and for inclusion in a dosimetry handbook to be prepared as part of this contract work. Of the 13 new dosimeters discussed in the report, only three, oxalic acid, polyisobutylene, and Mylar film, seem sufficiently promising to warrant further development. Their current states of development are not sufficiently far advanced, however, to warrant their inclusion in a definitive dosimetry handbook. It is recommended that only the Fricke and ceric sulfate dosimeters be included in the dosimetry handbook, but that additional work should be done on oxalic acid, polyisobutylene, and Mylar as dosimetry materials.

REVIEW OF DOSIMETRY FIELD

I INTRODUCTION

In September 1956, SRI began research directed toward the development of dosimetry systems applicable to the requirements of the QMC Radiation Preservation of Foods Project. That research program, carried out under Contract No. DA 19-129-QM-766, was completed in January of 1959, and the results were reported in the final project report dated March 16, 1959.[1]* The program included:

1. A critical review of the dosimetry literature published prior to January 1958 and surveillance of the literature published during 1958,
2. An experimental evaluation of the promising dosimetry systems uncovered by the literature review,
3. Selection (based on the literature review and the experimental evaluation) of three integrating dosimeters: the Fricke ferrous sulfate dosimeter for use as a working reference standard, the cobalt glass dosimeter (melt F-0621) for use in the range 10^4 to 10^6 rads, and the ceric sulfate dosimeter for use in the range 10^5 to 10^7 rads,
4. Development of a graphite cavity ionization chamber for use as a continuous production monitor at dose rates up to 10^7 rads/hr,
5. Development of beam current and beam energy monitors for use with electron beam sources.

A detailed discussion of these items can be found in Reference 1.

The scope of the foregoing program was determined by the characteristics and production requirements of the irradiation facilities that were to be included in the U.S. Army Ionizing Radiation Center. Plans for the Center, which was to serve as a pilot plant for the production of irradiated foods for the Armed Forces, were deferred indefinitely in October 1959 and the QM food irradiation research program was reoriented. The reoriented program included a continuing and extended basic research program and the establishment of an experimental irradiation facility at

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* Superscript numbers refer to references collected at the end of the report.

the Headquarters of the QM R&E Command at Natick, Mass. The QM Radiation Laboratory (QMRL), which is currently being established, includes a 1-megacurie Co^{60} source and a 24-Mev high-current linear electron accelerator. It is expected that this facility will serve as a focus for the QMC's continuing research program on the radiation preservation of foods.

The reorientation of the Army food irradiation program did not diminish the need for precise and reliable dosimetry systems. However, it did result in a time lapse of approximately 3 years between completion of the earlier program and establishment of QMRL, during which developments in the dosimetry field continued. Because of this time lapse, the QMC decided during 1961 that the earlier study should be extended to insure that the best available dosimetry systems were placed at the disposal of QMRL. Work on the new study, of which this report is a part, began at SRI in September 1961. The scope of the program includes

1. An intensive review of the dosimetry literature published since 1958. The review is to include (a) a critical evaluation of each system of dosimetry studied, (b) a recommendation for selection of the systems most suitable for the needs of the QMRL.
2. After consolidation of all information obtained in the literature review and coordination of these findings with the Project Officer, detailed dosimetry handbooks are to be prepared for each system agreed upon by SRI project personnel and the QMC Project Officer. These handbooks are intended to be complete working laboratory manuals for use by personnel making routine dosimetry measurements.

The purpose of this report is to present the results of the literature review discussed in Item 1 above. The report is organized as follows: in Section II, the desirable performance characteristics of the dosimeters required by QMRL are presented. The results of the literature review are discussed in Section III in light of these performance characteristics. These results are summarized and evaluated in Section IV, and Section V contains resulting conclusions and recommendations.

II DESIRABLE PERFORMANCE CHARACTERISTICS

Two types of dosimeters are required to meet the needs of QMRL. The first is a standard dosimeter for source calibration and the second is a go-no-go dosimeter for production control. Both are integrating dosimeters in that they are intended to measure the total absorbed dose during a given exposure period. However, there are basic differences between the two types of dosimeter. The go-no-go dosimeter is a threshold device. It should give an unequivocal indication that the threshold dose has been achieved but it should give no indication of exposure below the threshold dose. Thus, it is a discontinuous device capable of indicating only whether or not a threshold dose has been received. The standard dosimeter, on the other hand, should give a response which is either proportional to absorbed dose or which can, at least, be quantitatively related to absorbed dose. Thus it should be able to measure with some specified precision any absorbed dose falling within the region of its sensitivity. Because of the basic difference between the two types of dosimeter, and the different applications for which they are intended, the desirable performance characteristics of the two differ in some respects. The performance characteristics desired of these two dosimeter by the Quartermaster Corps are compared in Table I.

The characteristics listed in Table I were determined both by the requirements of the QMC Radiation Preservation of Foods Program and by the types of radiation sources specified for QMRL. The requirements of type of radiation, energy range, dose rate, and beam power are determined by the characteristics of the gamma and electron beam sources installed at QMRL. The remaining requirements are determined by the needs of the QMC program. Among these the precision, dose level, stability, and temperature dependence are the most important. The precision requirement for the standard dosimeter is more stringent than that for the go-no-go dosimeter because the standard dosimeter is expected to provide the source calibrations on which all experimental work conducted at QMRL will be based. The dose levels covered by the two dosimeters extend over approximately the same range since both will be used in the experimental program. The stability requirement for the standard dosimeter is based on the

Table I DOSIMETRY SCHEDULE

Criteria	Standard Dosimeter for Source Calibration	Production Control (Go-No-Go) Dosimeter
Type of Radiation	e^-, γ, or x-ray	e^-, γ, or x-ray
Accuracy	±8% or better	±15%
Energy Range		
Gamma	0.6 to 1.5 Mev	0.6 to 1.5 Mev
X-Ray	0.6 to 24 Mev	1 to 12 Mev
Electron Beam	1 to 32 Mev	1 to 12 Mev
Dose Rate		
Gamma	10^5 to $5\text{x}10^7$ rad/hr	10^5 to $5\text{x}10^7$ rad/hr
X-ray	10^4 to $3\text{x}10^8$ rad/min	10^4 to $3\text{x}10^8$ rad/min
Electron Beam	10^5 to 10^9 rad/min	10^5 to $4\text{x}10^8$ rad/min
Power of Electron Beam	10 to 20 Kw	10 to 20 Kw
Power of Gamma Beam	~50 Kw	~50 Kw
Dose Levels Required to be Measured (rad)	50,000 $5\text{x}10^5$ 10^7	Variety of doses from 10,000 to 10^7. Primary emphasis on sterilizing doses(4-6 M rad).
Stability	Change in dose indicated not to exceed 5% after storage at room temperature over a 72-hr period	Change in dose indicated not to exceed 15% after storage at room temperature over a 2-month period
Temperature Dependence[x]	Response independent of processing area temperatures in the range of 10-40°C	Response independent of processing area temperatures in the range of 2-75°C
Cost of System		As low as possible consistent with required precision
Reading	Permit use by trained technical personnel	Permit use by semi-skilled personnel. Due consideration will be given to permit record of obtaining dose
Size		
Overall	Refer to Project Officer	Very small
Sensitive Volume	0.1cc < V > 10 cc	Refer to Project Officer

[x] In conduct of review, Contractor to make special note of dosimeter devices with possible application to extreme temperature conditions such as -196, -76, -29, 80, and 90°C.

assumption that these dosimeters will be read within 72 hours after exposure. A longer storage period is desired for the go-no-go dosimeters, because these are intended to accompany the packages being irradiated and it is desirable to have as permanent a record as possible of the fact that the packages have been irradiated. Ideally, the indication given by the go-no-go dosimeters should be permanent. The wider operating temperature range desired for the go-no-go dosimeters reflects the fact that wider temperature variations in the processing area may be expected during their use than during use of the standard dosimeters.

It should be noted that the characteristics set forth in Table I are desirable characteristics rather than mandatory requirements. It is intended that the dosimeters selected for use in the QMRL approach these characteristics as closely as is presently possible. That criterion was used as a guide in preparing this report.

III LITERATURE SURVEY

The reference sources used in the literature survey were the major abstracting journals, Physics Abstracts, Chemical Abstracts, and Nuclear Science Abstracts. These journals were searched for references to publications which might be of interest to the present work, and the complete original papers or reports were then examined.

A. Fricke and Ceric Sulfate Dosimeters

The most significant development with the Fricke ferrous sulfate dosimeter during the past four years is its adoption by the American Society for Testing Materials as a tentative method of test for absorbed gamma radiation dose. The method, given ASTM designation D 1671-59T, has been published in the ASTM Standards.[2]

Three papers of interest in ferrous and ceric dosimetry have been published during this period. The first of these[3] describes a precise measurement of the molar extinction coefficients of ferric and ceric ions. The ferric ion extinction coefficient in 0.8N sulfuric acid at 24.9°C and a wavelength of 304 millimicrons was $\epsilon = 2199 \pm 4$ liters mol^{-1} cm^{-1}. Reference 3 also reports measurements of the extinction coefficient in 0.1N and 0.01N sulfuric acid and measurement of the temperature dependence of the extinction coefficient at these acid concentrations in the range 20-30°C. In agreement with earlier work, the extinction coefficient in 0.8N sulfuric acid increases by 0.7%/°C in this range. The extinction coefficient of ceric ion in 0.8N sulfuric acid at 320 millimicrons was found to be 5565 ± 15 liters mol^{-1} cm^{-1}.

The second paper[4] also reports a precise measurement of the extinction coefficient of ferric ion and, in addition, reports a calorimetric calibration of the Fricke dosimeter with Co^{60} gamma radiation. In this paper the extinction coefficient is given as 2187 ± 5 liters mol^{-1} cm^{-1} at 305 millimicrons and 25°C. The calorimetric measurement was based on ingenious use of the fact that the density of an ice-water mixture increases as the ice melts. The calorimeter used for source intensity measurements consisted of a dewar containing an ice-water mixture connected to a calibrated water-filled capillary. Energy absorbed from the radiation field by the ice-water mixture converted some of the ice to water so that the total volume of the mixture decreased. The volume change was measured by measuring the displacement of the water column in the calibrated capillary. Following the calorimetric measurement of source intensity, a quantity of ferrous sulfate solution was irradiated in the same flask under identical conditions. The G-value then calculated from the measured amount of ferrous ion oxidized as a result of the known energy absorption was 15.68±0.07. This compares favorably with the values of 15.6±0.3 quoted in Reference 5 and 15.8±0.3 quoted in Reference 6, but is a little higher than the value of 15.45±0.11 quoted in Reference 7. The weighted mean of these four measurements is 15.63±0.04, leading to the conclusion that the best value for use in routine experimental work is G = 15.6 molecules of iron oxidized per 100 ev of energy absorbed.

The effects of high dose rates on the Fricke and ceric sulfate dosimeters have been measured with an electron linear accelerator that provided doses as high as 10,000 rads in a 1.3-microsecond pulse, corresponding to instantaneous rates of $8x10^9$ rads/sec.[8] For both ferrous and ceric the yield remained constant within experimental error (±3%) up to a rate of about 100 rads/pulse, corresponding to an instantaneous rate of about 10^8 rads/sec. At $8x10^8$ rads/sec the Fricke dosimeter yield was 6% lower and the ceric dosimeter yield 13% higher than the respective low dose rate values. At $8x10^9$ rads/sec the Fricke dosimeter yield was 15% lower and the ceric dosimeter yield 36% higher than the respective low dose rate values. These results indicate that both the Fricke and ceric sulfate dosimeters will be satisfactory at the maximum rates provided by the Quartermaster electron linear accelerator.

B. Liquids

Sodium Formate. An interesting new dosimeter[9] is based on the radiolysis of aqueous solutions of sodium formate. Hardwick[10] showed that the reaction products are hydrogen and sodium oxalate plus small amounts of formaldehyde and glyoxalic acid; however, for dosimetric purposes the exact identity of the reaction products is unimportant except for the fact that they can be quantitatively titrated with acid permanganate. The analytical technique consists merely of a titration with potassium permanganate, and the dose is calculated from the quantity of permanganate used in the titration.

This system has the following advantages:

1. It is non-corrosive to normal structural metals and hence can be used for dosimetry in metal containers.
2. Hardwick[11] reports that a precision of 2 to 3% has been obtained in his laboratory.
3. The yield of the dosimeter is the same when either ordinary distilled water or water redistilled from alkaline permanganate are used, indicating that the reaction is less sensitive to impurities than is the ceric sulfate dosimeter.
4. The products of radiolysis are stable for at least one week.

The yield of reductant is 3.40 molecular equivalents per 100 ev and the usable dose range extends from 1 to 80 megarads. The yield is independent of temperature in the range 0 to 40°C and independent of sodium formate concentration in the range 0.05 to 0.3M. The major disadvantage of the system is that it is not usable below one megarad.

Oxalic Acid. The oxalic acid dosimetry system has received a good deal of attention in Europe but has been neglected in this country until recently. The most recent publications describing the dosimeter are those of Draganic[12,13] and Matsui.[14] The dosimeter can be used in the dose range 1.6 to 160 megarads when spectrophotometric analysis is used. Doses as low as 2.2×10^5 rads can be measured if chemical analysis is used. Thus the dosimeter covers approximately the same range as that covered by the ceric sulfate dosimeter. Recently, a dosimetry subcommittee of ASTM Committees D-9 and D-20 has begun to consider use of the oxalic acid dosimeter as a tentative method of test for gamma radiation by chemical dosimetry.

The dosimeter is a solution of oxalic acid in distilled water. The radiation effect is decomposition of the oxalic acid into CO_2 and trace amounts of aldehydes. Absorbed dose is measured by measuring the amount of decomposed oxalic acid. One analytical technique[13] is a spectrophotometric method based on measurement of the optical density of an oxalic acid/cupric-benzidine complex. The advantage of this method is that the decomposition products do not interfere with the determination of the oxalic acid concentration. Another analytical technique[14] to measure doses smaller than 1.6×10^6 rads is titration with a 0.1N caustic soda solution. Because the radiolysis product CO_2 can interfere with this method of analysis, it is necessary to heat the irradiated samples for about 30 minutes to remove CO_2. A third analytical method, which has been suggested but not yet tried, is oxidation with an excess of ceric ion and subsequent analysis for residual ceric ion either by titration with ferrous ion or by spectrophotometric analysis.[15] This method has advantages over both the cupric-benzidine spectrophotometric method and the caustic soda titration method. It is likely to be at least twice as sensitive as the first and is not affected by CO_2 as is the second.

The dosimeter has a number of desirable features. Draganic[13] quotes a precision of better than 10% and Matsui[14] demonstrates a precision of better than 5%. The dosimeter solutions are stable for several months against the influence of light and air. Irradiation temperature has no effect on yield between room temperature and 70°C. The system has two drawbacks which are not serious: first the G-value is concentration-dependent. For initial concentrations higher than 25 millimolar and a decomposition of less than 30% of the initial concentration, the G-value is constant. If the decomposition exceeds 30%, a break in the yield curve is observed, indicating a decrease in yield at that point. This drawback can be overcome by adjusting the initial concentration so that the maximum decomposition during exposure is less than 30%. Unfortunately, this technique limits the usefulness of the dosimeter to sources about which some dose rate information is available. The second defect is that the yield is affected by the composition of the gas phase above the solution. If either nitrogen or oxygen gas is continuously blown over the solution during irradiation, the yields are lower than the yield obtained with free air.[14] This drawback can be eliminated by irradiating the dosimeter solutions in closed containers.

Benzene-Water. The use of aqueous solutions of benzene as a dosimeter system was first suggested by Day and Stein[16] more than 12 years ago, but received little acceptance. The system can be used in the dose range 10,000-70,000 rads and is thus a competitor of the Fricke dosimeter. A recent reevaluation[17] suggests that the system is much less sensitive to impurities than is the Fricke dosimeter and is much less corrosive than the acid solution required for the Fricke dosimeter so that it can be used safely in metal, glass, or plastic containers. The main product of radiolysis is phenol but lesser amounts of diphenyl and various oxygenated products are also found. The yield in aerated solutions is 2.14 molecules/100 ev up to 70,000 rads and decreases sharply above that because the oxygen in the solution is consumed. Analysis for phenol requires measurement of the optical density of neutral and alkaline samples at 2900A. Some care must be exercised in sample preparation to insure that the solutions are saturated with benzene. There is a small post-irradiation effect evidenced by an increase in optical density of the irradiated solution that reaches a maximum at about 10 minutes after irradiation. Dose rate independence has been demonstrated only up to a rate of about 100 rads/sec. The precision is about ±10% and the solutions are stable for long periods both before and after irradiation.

Polyisobutylene. The use of polymer degradation for dosimetry purposes has not received much attention during recent years. In 1958, Feng[18] described the use of solutions of polystyrene in carbon tetrachloride as dosimeters in the range 100 to 10^5 rads. The radiation effect (cleavage of polymer bonds) was observed as a decrease in the viscosity of the polymer solution measured by a simple viscometer. More recently solutions of polyisobutylene in heptane or carbon tetrachloride have been used for dosimetry purposes.[19] Both techniques involved measurement of the decrease in viscosity of the solutions produced by irradiation. However, by proper selection of the polymer molecular weight and the use of appropriate solvents, the range of polyisobutylene can extend from 1000 rads to as high as 10^8 rads.[19] The useful dose range of any given solution depends on the polymer molecular weight and on its concentration in the solution. If polyisobutylene with a viscosity average molecular weight of about 200,000 is used, a concentration of 20 gm/liter in heptane can be used to measure doses in the range 10^3 to $3\text{x}10^6$ rads. Material with a viscosity average molecular weight of about 5000 and a concentration of 480 grams/liter heptane can be used in the range $4\text{x}10^5$ to at least $1.5\text{x}10^7$ rads and perhaps to doses as high as 10^9 rads.

The desirable features of the system include the following:

1. Commercially available polyisobutylene and heptane can be used without further purification or fractionation.

2. The solutions are stable for at least 2 months if exposure to sunlight is avoided.

3. The precision of the system can be as good as ±1%.[19]

4. The system is independent of temperature in the range 20-80°C and it is quite possible that it can be used at liquid nitrogen temperatures although with some loss in sensitivity.

5. Irradiation in an atmosphere of oxygen or nitrogen shows no different in viscosity decrease when compared with irradiation under identical conditions in air.

6. The analytical method is quite simple and the apparatus (a viscometer) is quite inexpensive. Analysis requires only measurement of the flow time of a measured quantity of solution through a calibrated capillary and, if an Ostwald viscometer is used, only 2 ml of solution are required.

7. The influence of dose rate is not known but it should be small.

The only drawback to the system in its present state is that additional work is required with starting materials of other molecular weights than those already used[19] to establish which molecular weights of starting material will be optimum for the requirements of QMRL.

C. Glass

Glass dosimeters have continued to receive attention since 1958. Studies have been made of an experimental high-antimony glass and of Corning Code 8392 glass[20] and of combinations of Mn-Fe, Mn-V, and

Mn-V-Fe.[21] These glasses do not represent a very significant advance over the Corning F-0621 cobalt silicate glass. In some cases the upper limit of a dose range has been extended measurably, but post-irradiation fading is still a problem. A more promising bismuth lead borate glass has been reported to be usable in the range 10^5 to 10^8 rads.[22] It exhibits no fading during a 1000-hour period of room temperature storage after receiving doses of 10^6 rads or greater. Fading is observed, however, after doses between 10^5 and 10^6 rads. Since it is an experimental glass, it is not available in quantity.

D. Plastic Films

The development of plastic film dosimeters is still in an unsatisfactory state. Like all of the film dosimeters considered heretofore, neither of the plastic dosimeters discussed below is completely acceptable.

Saran[*] *(polyvinyl vinylidene)*. Saran No. 7 in a thickness of 0.001-in. has been used in the dose range from 5×10^4 to 10^7 rads with a precision of approximately 8%.[23] Optical density is measured at 2600A.

Saran has two major drawbacks. First, to insure reproducible stability is is necessary before irradiation to place the Saran materials in a vacuum chamber at about 20μ Hg pressure for 24 hours. The treated samples must be stored in a dessicator until used. Second, there is a post-irradiation effect that causes the samples to continue to darken. The time required for this process to reach saturation was about 100 hours for doses of 10^7 rads and correspondingly less for smaller doses. No method was found to shorten saturation time.[23]

Mylar. Mylar which was first investigated by Boag, Dolphin, and Rotblat[24] has been re-investigated by Ritz.[25] Boag, *et al*, who studied films of 55μ thickness and greater found a rapid initial fading of the UV absorption during the first 24 hours after irradiation. The optical absorption was stable thereafter.

Ritz studied the thickest (0.001 inch or 178μ) and the thinnest (0.00025 inch or 6μ) Mylar films available in this country. In the thick

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[*] Dow Chemical Company

material he found the same rapid initial fading followed by the same stability after 24 hours that was observed by Boag. Ritz was able to trace this phenomenon to oxygen diffusing into the film. In the thin film no fading occurred over periods of 30 minutes to 6 months after the end of irradiation, apparently because oxygen diffuses easily into the thin material. The 0.001-inch material covers the dose range from $5\text{x}10^{6}$ to 10^{8} rads and the 0.00025-inch film covers the range $5\text{x}10^{7}$ to 10^{9} rads. Lower doses could probably be measured by using multiple thicknesses of the 0.001-inch film.

In contrast to the results of Boag, Ritz found a dose rate dependence in both thicknesses of film. For example, when samples of the 0.001-inch film were irradiated to total doses of 10^{7} rads of Co^{60} radiation at rates of $4.5\text{x}10^{5}$ rads/hr and $2.2\text{x}10^{6}$ rads/hr, optical densities differing by almost a factor of two were obtained. A rate dependence was also observed at rates of $5.6\text{x}10^{7}$ and $5.6\text{x}10^{8}$ rads/hr obtained with a 1-Mev Van de Graaff electron accelerator. Ritz' results indicate that the rate dependence is much more severe at lower than at higher dose rates. It is possible that this phenomenon can explain the discrepancy between the results of the two studies. Boag performed irradiations with an electron linear accelerator at instantaneous rates of approximately $4\text{x}10^{8}$ rads/sec, and it is reasonble to infer from the trend of Ritz' data, that the rate dependence disappears at high dose rates. In any case, the rate dependence at lower dose rates limits the use of the material to sources in which the dose rate does not vary too much during the exposure. If such is the case a correction can be made for the rate dependence if the exposure time is known.

E. Photographic Film

An intriguing application of photographic film to high level dosimetry is based on the fact that sufficiently high exposures to radiation form a print-out image that is observable without photographic development.[26] The density of the print-out image, which is a function of radiation exposure, can then be measured with a densitometer using red light. Eight industrial x-ray films were studied and by suitable choice of film type the exposure range from 2×10^4 to 10^8 rads could be covered with a precision of ±5%.

Additional advantages are:

1. The stability is such that the density of the print-out image did not vary during a 4-month storage period after irradiation.
2. No rate dependence was observed over the exposure range of 2×10^2 to 5×10^5 rads/hr.
3. The material is energy-independent at photon energies above 0.3 Mev.

Balanced against these advantages are the following limitations:

1. A dark room with proper safety lights is necessary for densitometry since the film cannot be exposed to normal room light after exposure to radiation.
2. The print-out response is temperature-dependent so that the temperature during exposure must be controlled to within ±10°C to avoid errors in exposure interpretation greater than ±10%.

F. Go-no-go Dosimeters

Further development of the radiation polymerization dosimeter proceeded since 1958.[27] The precision, dose rate and temperature dependence, and stability have been improved to the point where this system now seems moderately satisfactory. No new concepts usable for go-no-go dosimetry were recorded in the literature during this period.

IV EVALUATION AND SUMMARY

The characteristics of the dosimeters discussed in the preceding section are summarized in Tables II, III, and IV. Energy, temperature, and dose rate dependence data are omitted except where noted in the comments or footnotes. Omission of these data for a given dosimeter indicates that the dosimeter will satisfactorily meet the QMC requirements.

Convenience rating is based on four requirements: ready availability of materials used to prepare the dosimeter, ready availability of instrumentation to measure the radiation effect, simplicity of preparation of the dosimeter, and simplicity of measurement and interpretation. If the dosimeter meets all four of these requirements, a rating of A is given. Ratings of B, C, D, indicate that only 3, 2, or 1, respectively, of the requirements are met. As noted, the entries under stability refer to the percentage change in radiation effect during the first 72 hours after completion of the exposure. If the change is much smaller than 1% the dosimeter is considered stable. Pre-irradiation stability (shelf life) is satisfactory for all of the dosimeters.

Table II summarizes the characteristics of the liquid dosimeters considered. The Fricke and ceric dosimeters are included in this list for purposes of comparison. Of the remaining four, the benzene-water dosimeter has only limited usefulness. It can serve as an alternate to the Fricke dosimeter when a non-corrosive liquid dosimeter covering the low dose range is desired. However, such occasions are not frequent and other more satisfactory alternates may be available. Both the sodium formate and the oxalic acid dosimeters show some promise as potential alternates to the ceric sulfate dosimeter. Both are considerably less sensitive to impurities than is the ceric dosimeter, but they require considerable further development before either can be recommended for use. Of the two, the sodium formate dosimeter is apparently more precise in its present stage of development. However, it has been used only in one laboratory so this conclusion is subject to modification. On the other hand, the oxalic acid dosimeter is potentially capable of measuring lower doses if a reliable method of chemical analysis can be developed.

Table II COMPARISON OF LIQUID DOSIMETERS

Dosimeter	Range (rads x 10^{-6})	Precision (%)	Stability	Convenience[a]	Comments
Fricke	0.004-0.04	±1	Stable	B	Widely used as standard
Ceric	0.1-140[b]	±2	Stable	B	High purity necessary for precision
Sodium formate	1-80	±3	Stable	B	Untried, but potentially good; non-corrosive
Oxalic acid	0.22-160[c]	5-10	Stable	B	Needs development; potential alternate to ceric sulfate
Benzene-water	0.01-0.07	±10	Stable	B	Useful only as non-corrosive alternate to Fricke
Polyisobutylene	0.001-10^3	±1	Stable	A	Needs development, but shows great promise

[a] See text for explanation of rating

[b] Nonlinear above 1.[illegible]x10^7 rads

[c] Two analytical methods needed to cover dose range; method used below 1.6x10^6 rads sensitive to CO_2.

Table III COMPARISON OF GLASS DOSIMETERS

Dosimeter	Range (rads x 10^{-6})	Precision (%)	Stability[a]	Convenience[b]	Comments
Cobalt F-0621[c]	0.01-4	±2	10%[d]	A	Readily available; well tried; best range: $2x10^4$-10^6 rads
High-Sb	0.1-600	Note e	26%	B	Special glass; needs annealing for stability
Corning 8392	0.1-300	"	40%	B	Needs annealing for stability
Mn-Fe	0.1-3	"	15%	B	Special glass; no advantage over F-0621
Mn-V	0.1-4	"	12%	B	Special glass; not readily available
Mn-V-Fe	0.1-20	"	13%	B	Special glass; not readily available
Bi-Pb	0.1-10^3	"	Note f	B	Special glass; not readily available

[a] Change during 72-hr period after irradiation

[b] See text for explanation of rating

[c] Bausch and Lomb Optical Co.

[d] Dependent on dose and wavelength

[e] No precision data available

[f] 27% fading at $4x10^5$ rads; stable above 10^6 rads

Table IV COMPARISON OF FILM DOSIMETERS

Dosimeter	Range (rads x 10^{-6})	Precision (%)	Stability[a]	Convenience[b]	Comments
Saran No. 7	0.05-10	±8	Note c	C	Needs vacuum pre-treatment for precision
Mylar	0.1-[illegible]00[d]	±5	Note e	AB	Rate dependent; potentially good for electron depth-dose measurements
Photographic film	0.02-[illegible]00	±5	Stable	B	Dark room required; response is temperature dependent

[a] Change during 72-hr period after irradiation

[b] See text for explanation of rating

[c] [illegible]0% at 10[illegible] rads; 220% at $5 \times 10^{[illegible]}$ rads

[d] 0.01-inch thickness needed for 10^5-5×10^6 rads

[e] 0.00025-inch film is stable; thicker films stable after 24 hrs

Consequently it may be more suitable for the purposes of QMRL. In sum then, neither of the dosimeters in their present stages of development is a satisfactory alternate for the ceric sulfate dosimeter. More experience has been accumulated with the oxalic dosimeter than with the sodium formate dosimeter and, in addition, the oxalic acid dosimeter is currently being tested in a number of laboratories as a part of the ASTM program. For these reasons, further development of the oxalic acid dosimeter should be watched carefully and it is advisable, when facilities are available, for the QM Radiation Laboratory to participate in the ASTM program. Because of the attention that is being given to the oxalic acid dosimeter and because of the great promise shown by the polyisobutylene dosimeter (see below), it does not seem worthwhile at this time to consider further development of the sodium formate dosimeter.

Of all the new dosimeters listed in Table II, the polyisobutylene dosimeter shows the greatest promise. It covers the widest dose range, exhibits the greatest potential precision, is stable, is non-corrosive, and is the only dosimeter that may be usable at liquid nitrogen temperature. In addition, it is not sensitive to impurities (commercially available materials may be used). Finally, the analytical method is simple and the apparatus required for analysis is quite inexpensive. Additional development of the dosimeter is required, but in view of the foregoing advantages, such a development effort seems to be worthwhile.

The characteristics of the glass dosimeters that have been studied recently are summarized in Table III. The characteristics of cobalt glass (Bausch and Lomb Melt F-0621) are included for comparison. The drawback that all of the remaining glasses share in common is that they are not readily available. Four of the new dosimeter glasses can be used at significantly higher doses than the cobalt glass. The extension of range is obtained, however, at the cost of sensitivity in the dose range of interest to the QM Radiation Laboratory. The two glasses (Mn-Fe, Mn-V), which cover approximately the same range as the cobalt glass, offer no advantages over that glass. The bismouth-lead-borate is unique in exhibiting no fading at doses greater than 10^6 rads. If this glass were readily available at a reasonable cost, further study of its characteristics would be worthwhile. The need for such a dosimeter is not sufficiently pressing, however, to warrant the expense of special procurement for purposes of additional study.

Table IV contains data on three film dosimeters studied recently. The Saran No. 7 film does not warrant further consideration because of the need for vacuum pre-treatment and the lack of stability. Mylar film has a number of attractive features, among which are its wide dose range and relatively good precision. However, it is necessary to use multiple thicknesses of film to measure doses smaller than $5\text{x}10^6$ rads. Its chief drawback is its dose rate dependence in the range of dose rates primarily encountered with gamma radiation sources. It is possible that the rate dependence vanishes at the higher rates characteristic of electron linear accelerator sources. If this proves true (and this point should be investigated), Mylar film may prove to be a very satisfactory material for measuring depth dose distributions produced by electron beams. Aside from this possible application, its usefulness as a dosimetry material is probably limited. Industrial x-ray film does not seem to have any outstanding features that warrant ranking it above the other dosimeters considered here. Furthermore, it has two disadvantages that weigh against its selection for QMRL. First, a dark room is required for post-irradiation analysis. (A dark room has not been provided at QMRL.) Second, although the film can be used at temperatures other than room temperature, it must, for precision, be calibrated at each temperature at which it is to be used, and the temperature during exposure must be controlled to $\pm 10^\circ$C.

V CONCLUSIONS AND RECOMMENDATIONS

Two general conclusions may be drawn from the literature survey. First, only a few of the new dosimetry systems studied during the last several years are promising enough to warrant further consideration in their present stage of development. Second, none of the promising new systems is sufficiently well-developed, or supported with a sufficient background of use in different laboratories, to warrant recommendations for routine use in QMRL.

More specifically, the following conclusions emerge from the survey:

1. The Fricke and ceric sulfate systems are still the most acceptable dosimeters for source calibration, despite the impurity sensitivity of the ceric sulfate dosimeter.

2. The oxalic acid dosimeter clearly warrants further study and development as a possible alternate to the ceric sulfate dosimeter.

3. The polyisobutylene system shows great promise for use as a routine, general purpose dosimeter and further development of the system should be worthwhile.

4. The Mylar film dosimeter may be satisfactory for measuring depth-dose distributions produced by the QMRL electron linear accelerator, if its dose rate dependence at high rates is sufficiently small.

In view of these conclusions, the following steps are recommended:

1. A dosimetry handbook based on the present state of the art should be prepared under this contract only for the Fricke and ceric sulfate systems. Appendixes on the oxalic acid and polyisobutylene dosimeters, as described in the following paragraphs, should also be included.

2. QMRL should participate in the evaluation of the oxalic acid dosimeter now being undertaken by the Task Group on Gamma Dosimetry, Section B, Joint Subcommittee II of ASTM Committees D-9 and D-20. In addition, the possible advantages to be gained by using the ceric sulfate method of analysis for oxalic acid should be investigated. The ASTM "Proposed Tentative Method of Test for Gamma Radiation by Chemical Dosimetry" using oxalic acid should be included as an addendum to the dosimetry handbook along with a proposed procedure for investigating the ceric sulfate analytical method.

3. The polyisobutylene system should be investigated further. In particular, starting materials with molecular weights higher than 5000 and lower than 2×10^5 (the materials used by Wiesner[19]) should be investigated to find a material that better spans the dose range of interest to QMRL. Procedures to be used in conducting this investigation should be included as an addendum to the dosimetry handbook, along with pertinent details of the work done by Wiesner.

4. The dose rate dependence of Mylar film at the rates available from the QMRL linear accelerator should be investigated to determine whether Mylar will be useful as a dosimeter for electron depth-dose measurements.

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October 1, 1962

CONTRACT RESEARCH PROJECT REPORT

QUARTERMASTER FOOD AND CONTAINER INSTITUTE FOR THE ARMED FORCES, CHICAGO
HQ., Research and Engineering Command, QM Research and Engineering Center,
Natick, Massachusetts

Stanford Research Institute
Menlo Park, California

Project No: 7-48-01-002
Contract: DA 19-129-QM-1900
S-611 - [illegible]

Official Investigator: S.I.Taimuty

Report No. 4 (Addendum to Final Rpt)

Period: 14 Sept 61 - 13 Sept 62
Initiation Date: 14 Sept. 61

Title of Contract: Use of the Fricke and Ceric Sulfate Dosimeters

SUMMARY

This is a handbook containing procedures for the laboratory use of the Fricke and ceric sulfate dosimeters in the measurement of gamma ray and electron radiation dose. For each dosimeter, background information is included on reaction mechanisms, effect of impurities, yield, precision, usable dose range, dose rate and energy dependence, stability and temperature dependence. In addition, specific directions for preparation of solutions, for calibration of instrumentation, and for use of the solutions in dosimetry measurements are included. Appendices on the oxalic acid and polyisobutylene dosimeters are also included. These appendices contain information and suggested procedures for use by the Quartermaster Radiation Laboratory (QMRL) in further evaluation and investigation of these dosimeters.

FOREWORD

In September 1961 Stanford Research Institute (SRI) undertook a study of radiation dosimetry systems for the Quartermaster Research and Engineering Command under Contract No. DA 19-129-QM-1900. Under an earlier contract (DA 19-129-QM-766) an extensive literature survey and experimental evaluation of dosimetry systems were carried out. As a result of this earlier work and a further extensive literature survey under the present contract (see Technical Report No. 1[1]*), two dosimeters were selected which are, in my opinion, the best systems currently available for source standardization measurements in the dose range 10^4 to 10^7 rads. These are the Fricke and ceric sulfate dosimeters.

This is a laboratory manual containing suggested procedures for use of the Fricke and ceric sulfate dosimeters in measurement of absorbed gamma ray and electron radiation dose. In preparing the procedure for the Fricke dosimeter, I have borrowed heavily from the "Tentative Method of Test for Absorbed Gamma Radiation Dose in the Fricke Dosimeter", adopted by the American Society for Testing Materials.[2] The procedure for the ceric sulfate dosimeter is based primarily on work performed by Stanford Research Institute under the earlier contract referred to above [3,4]

Two additional conclusions, contained in Technical Report No. 1, were that the oxalic acid and the polyisobutylene dosimeters are sufficiently promising to warrant further study and development. Consequently, in addition to the handbook, I have included appendices containing suggested procedures for further study and development of these two dosimeters.

S. I. Taimuty
Project Leader

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* Numbered references are listed at the end of this report.

CONTENTS

I THE FRICKE (FERROUS SULFATE) DOSIMETER

A. General Description

1. Reaction Mechanism

The Fricke dosimeter is an aqueous solution dosimeter that functions by virtue of the radiation-induced oxidation of ferrous ion (Fe^{++}) in air-saturated 0.4M sulfuric acid solution. The reaction mechanism is well established. Starting with the well-founded assumption that water is dissociated by radiation into hydrogen (H) and hydroxyl (OH) radicals, the reactions which result in the oxidation of ferrous ion can be written as follows:

$$Fe^{++} + OH = Fe^{+++} + OH^- \quad (1)$$

$$H + O_2 = HO_2 \quad (2)$$

$$Fe^{++} + HO_2 = Fe^{+++} + HO_2^- \quad (3)$$

$$HO_2^- + H^+ = H_2O_2 \quad (4)$$

$$Fe^{++} + H_2O_2 = Fe^{+++} + OH + OH^- \quad (5)$$

To this reaction sequence, which results from the presence of H and OH radicals and oxygen, must be added the effect of hydrogen peroxide formed by the reaction,

$$OH + OH = H_2O_2 \quad (6)$$

followed by Reactions 5 and 1, in that order.

2. Effect of Impurities

The above reaction scheme is affected by organic impurities. Formic acid, for example, produces the following sequence

$$OH + HCOOH = H_2O + HCOO \quad (7)$$

$$HCOO + O_2 = HO_2 + CO_2 \quad (8)$$

These reactions have the net effect of changing an OH radical into an

HO_2 radical which can oxidize two ferrous ions by Reactions 3, 4, and 5, whereas an OH radical can oxidize only one ferrous ion by Reaction 1. In addition, the OH radical resulting from Reaction 5 can initiate the sequence of Reactions 7 and 8. The result is a chain reaction that can lead to a substantial increase in the yield of ferric ion (Fe^{+++}). Fortunately, the effect of organic impurities can be inhibited by chloride ion as follows:[5]

$$OH + C\ell^- = OH^- + C\ell$$

Thus, the chlorine atom replaces the OH radical and Reaction 1 becomes Reaction 1'.

$$Fe^{++} + C\ell = Fe^{+++} + C\ell^-$$

The inhibiting action is effective because organic impurities are less reactive with chlorine atoms than with hydroxyl radicals.

3. Yield

The yield of ferric ion has been measured by absolute methods.[6-9] The four most reliable values of G (number of ferrous ions oxidized per 100 ev of energy absorbed) are 15.6 ± 0.3, 15.8 ± 0.3, 15.45 ± 0.11, and 15.68 ± 0.07. The weighted mean of these four measurements is 15.63 ± 0.04, leading to the conclusion that the best value for use in routine experimental work is $G = 15.6$.

4. Precision

Only one controlled study of precision has been reported.[10] The results indicate that a precision of better than ±1% can be obtained even by relatively untrained operators given brief instruction and some supervision. This conclusion has been verified in numerous other laboratories, where a precision of ±2% is routinely obtained and a precision of ±1% is obtainable with only moderate care.

5. Dose Range

The dose range in which air-saturated ferrous sulfate solutions can be used with precision extends from 4×10^3 to 4×10^4 rads. The lower limit is set by the precision with which analysis of small concentrations of ferric ion can be made routinely. Using spectrophotometric analysis the ferric ion concentration produced by 4000 rads of absorbed dose can be measured with a precision of 1%. Smaller concentrations and, hence, lower doses can be measured with some loss of precision. The upper limit is set by oxygen depletion in the solution. Although the upper limit can be extended to 2×10^5 rads by saturating the solution with oxygen rather than air, this procedure is not recommended.

6. Dose Rate

The yield of the dosimeter is independent of dose rate up to average rates of at least 2×10^6 rads/sec[8] and peak rates of about 10^8 rads/sec.[11] At such high rates special precautions must be taken to avoid depletion of oxygen and ferrous ion in the reaction zone. In practice such precautions are difficult and time consuming, so that the maximum practically usable dose rate is much lower. The maximum is, in fact, set by the shortest exposure that can be timed accurately, and that will simultaneously limit the total dose to 4×10^4 rads. To a considerable extent, the timing accuracy is determined by the speed with which the dosimetric solution can be introduced into and removed from the radiation field or, if an electron accelerator is used, by the ease with which the dose can be kept below 4×10^4 rads. If manual manipulation is used, timing errors of the order of seconds will be involved, and the maximum usable dose rate at 1% timing accuracy will be less than 10^6 rads/hour. Mechanical manipulation can reduce the timing errors and increase the usable dose rate at the cost of some increase in complexity.

7. Energy Range

The lower and upper limits of energy independence have not been completely established. However, it may be concluded from published reports[12-15] that the system is independent of energy at least in the range 0.1 to 16 Mev. The range may extend to lower energies and almost certainly extends to higher energies.

8. Stability and Temperature Dependence

No data on stability were found in the literature. Experience in many laboratories has shown that unirradiated solutions can be satisfactorily stored for months if protected from light. Post-irradiation stability is such that delays in reading of a week or even a month have a negligible effect on the measurement if the solution is protected from light and temperature extremes. The yield has been found to be independent of the irradiation temperatures in the range 4°C to 54°C.[16]

B. Use of the Dosimeter

1. Reagents

The reagents required in the Fricke dosimeter are ferrous sulfate (or ferrous ammonium sulfate), sulfuric acid, sodium chloride, and distilled water. Reagents should be American Chemical Society analytical reagent grade. Ordinary commercial distilled water is adequate for routine work.

2. Preparation of Solutions

Pyrex glass should be used for preparation of the solutions. Before use all glassware should be washed thoroughly with detergent and rinsed thoroughly with distilled water. Precautions should be taken to avoid contact between the solutions or any of the reagents and organic materials.

The concentrations used in the dosimetric solution are 0.001M ferrous sulfate or ferrous ammonium sulfate, 0.001M sodium chloride, and 0.4M sulfuric acid. The most convenient way to prepare the dosimetric solution is to make up a stock solution 0.5M in ferrous sulfate or ferrous ammonium sulfate, and 0.5M in sodium chloride. To prepare the stock solution dissolve 200 g of $Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$, or 140 g of $FeSO_4 \cdot 7H_2O$, and 30 g of $NaC\ell$ in sufficient distilled water to make 1 liter of solution. This stock solution may be stored in amber bottles for up to three months.

The stock solution may be tested occasionally for depletion of ferrous ion by the following procedure. Dilute 2 ml of the stock solution to 1 liter with 0.4M sulfuric acid solution. The acid solution is made up by dissolving 22 ml of concentrated (95 to 98%) sulfuric acid in sufficient distilled water to make up 1 liter of solution. The optical density of the dilute ferrous solution is then measured with a spectrophotometer at a wavelength of 305 mμ (slit width, 0.5 mm, using quartz cells) with distilled water as a blank. If the optical density of the dilute solution is greater than 0.4, the solution must be discarded.

3. Spectrophotometer Calibration

Analysis of the amount of ferric ion produced in the irradiated dosimeter solution is performed by measuring the optical density of the solution with a spectrophotometer at a wavelength of 305 mμ. For this measurement a Beckman Model DU spectrophotometer with hydrogen lamp attachment, and 1 cm matched quartz cells or equivalent apparatus should be used.

To calculate the concentration of ferric ion from the measured optical density, it is necessary to know the extinction coefficient of ferric ion at the measuring wavelength. For simplicity, we define the extinction coefficient, k, as the optical density observed in a 1 cm cell containing a solution concentration of 1 mole/liter. The concentration C of an unknown solution in moles per liter is then $C = \frac{OD}{k}$ where OD is the optical density of the unknown solution.

The appropriate value of the extinction coefficient may be derived in two ways: (1) the values quoted in the literature may be adopted or (2) the spectrophotometer may be calibrated with ferric solutions of known concentration. The first method is by far the simpler and, for most work, gives adequate accuracy despite the fact that there is some variation in the value of the extinction coefficient measured on different spectrophotometers by different investigators. The validity of this approach for ferric ion determinations can be determined by examination of 12 values of the ferric ion extinction coefficient published by different investigators during the last 10 years (see References 6-9, 12, 15, 17-21). These range from a minimum of 2167 to a maximum of 2240. The mean and standard deviation of the mean of these 12 values is k = 2199 ± 7 mole^{-1} cm^{-1}. The standard deviation of the mean is 0.32% and the deviation of the extreme value from the mean is 1.9%. Thus, if an accuracy of 2 to 3% in routine measurements is satisfactory, as it is for most applications, use of the mean value of the extinction coefficient quoted above is acceptable.*

If greater accuracy is desired the spectrophotometer must be calibrated with solutions of known ferric ion concentration. The procedure for performing this calibration is as follows. Add a weighed quantity of ferric sulfate to the calculated amount of 0.4M sulfuric acid required to make up a solution approximately 0.1M in ferric ion. Place this mixture (with cap loose on the bottle) in an oven at 90 to 95°C overnight to obtain a solution. Analyze a sample of the cooled ferric solution for iron using the following procedure. Reduce the ferric iron to ferrous, either with a Jones reductor or with test lead, and analyze the reduced solution for iron by potentiometric titration with ceric sulfate. Dilute 1, 2, 3, 4, 5, 6, and 7 portions of the stock solution to 1 liter with 0.4M sulfuric acid solution. Read the optical densities of the diluted solutions in the spectrophotometer, set at a wavelength of 305 mμ, (slit

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* A distinction needs to be made between the precision (reproducibility) and the absolute accuracy with which dose measurements can be made when the mean value is used. The precision of the measurements is unaffected by the value of k adopted. It is determined by the care with which the measurements are performed, by the precision of the apparatus used in performing the measurements, and by random errors, as it the case in all experimental measurements. However, the absolute accuracy (the discrepancy between the measured value and the true value of dose) may be affected because spectrophotometers differ slightly, as is evidenced by the spread in the measurements quoted above. Adoption of the above value rather than a value determined by calibration of the instrument to be used in the measurements may introduce a systematic error. The effect of a systematic error is to shift all of the measured values in the same direction away from the true values by the amount of the systematic error. In the case considered here, this shift, if it exists, will be small and, in fact, will be comparable to the errors normally associated with dosimetric measurements.

width, 0.5 mm, using quartz cells) and using a portion of 0.4M sulfuric acid solution as the blank in the spectrophotometer. From these measurements prepare a curve of optical density versus concentration of ferric ion. Calculate the extinction coefficient (optical density units per mole per liter) from the slope of this curve.

The extinction coefficient at 305 mμ wavelength (the absorption maximum) increases by 0.7%/°C in the range from 15 to 35°C.[20] Consequently, the spectrophotometer should have provisions for maintaining solutions at constant temperature (25°C is commonly used), or alternatively the solution temperatures should be measured and the extinction coefficient corrected accordingly.

4. Irradiation Cells

Cells used for dosimetry measurements should be Pyrex glass or equivalent. Lucite and polystyrene may be used if the solution is irradiated immediately after filling the container and measured immediately after irradiation.[13] However, such containers are not recommended for precise work. Before use, the cells should be cleansed thoroughly with detergent and rinsed with distilled water.

To avoid wall effects, no internal dimension of glass containers should be less than 8 mm. Wall effects are not important in lucite or polystyrene vessels because the radiation absorption characteristics of these materials are close to those of water.

5. Dosimetric Procedure

Expose a quantity of the dosimetric solution in the radiation field for a carefully measured time. Measure the optical density of the irradiated solution in the spectrophotometer using a portion of the unirradiated solution as a blank, keeping temperature effects in mind as noted above. The dose (D) is calculated as follows:

$$D(\text{rads}) = 9.65\text{x}10^{8}\,\frac{\text{OD}}{G\rho k}$$

where

OD = optical density of irradiated solution,

G = yield (molecules/100 ev),

ρ = solution density, (g/ml)

k = extinction coefficient.

For the Fricke dosimeter, $\rho = 1.024$ and $G = 15.6$; consequently

$$\text{Dose(rads)} = 6.04\text{x}10^{7}\ \frac{\text{OD}}{k}$$

If the mean of the published values of k is used ($k = 2199$), then

$$\text{Dose(rads)} = 2.75\text{x}10^{4}\ \text{OD}$$

II CERIC SULFATE DOSIMETER[3,4]

A. General Description

1. Reaction Mechanism

Like the Fricke dosimeter, the ceric sulfate dosimeter is an aqueous solution dosimeter. The over-all radiation effect is the reduction of ceric ion to cerous in solutions of ceric sulfate in 0.4M sulfuric acid. In contrast to the Fricke dosimeter the reduction of ceric ion is independent of oxygen concentration. The reaction mechanism is

$$H_2O = H_2,\ H_2O_2,\ H,\ OH \qquad (1)$$

$$Ce^{4+} + H = Ce^{3+} + H^+ \qquad (2)$$

$$H + O_2 = HO_2 \qquad (3)$$

$$Ce^{4+} + HO_2 = Ce^{3+} + H^+ + O_2 \qquad (4)$$

$$HO_2 + HO_2 = H_2O_2 \qquad (5)$$

$$Ce^{4+} + H_2O_2 = Ce^{3+} + H^+ + HO_2 \qquad (6)$$

$$Ce^{3+} + OH = Ce^{4+} + OH^- \qquad (7)$$

The over-all yield is G = 2.50 ceric ions reduced per 100 ev of energy absorbed in dilute solutions (less than or equal to 0.05M in ceric ion) and is linear with decrease in ceric ion concentration.

2. Effect of Impurities

The ceric sulfate dosimeter, particularly in the more dilute solutions, is sensitive to impurities. As yet, no additive has been found that will inhibit impurity effects. Consequently, satisfactory performance can be obtained only through the use of pure reagents and scrupulously clean experimental techniques.

3. Precision

The dosimeter is capable of high precision, although it is not as precise as the Fricke dosimeter. A precision of ±2% can be obtained in routine use by skilled personnel. However, precision and accuracy are possible only if the ceric sulfate solutions are checked periodically against the Fricke dosimeter.

4. Dose Range

The dosimeter is usable with linear response (G = 2.50) from about 10^5 rads up to 1.5×10^7 rads. The lower limit is set by impurity effects in dilute solutions and by the difficulty in measuring small changes in ceric concentrations. The dosimeter can be used for measurement of doses up to 1.4×10^8 rads if more concentrated (up to 0.4M) solutions are used. In such solutions, the yield is nonlinear but predictable. The technique of high dose measurements is described in Reference 22.

5. Dose Rate

No dose rate dependence has been found at average rates up to 2×10^6 rads/sec. A rate dependence has been observed with pulsed electron irradiation at peak rates above 10^8 rads/sec.[11]

6. Energy Range

The yield is independent of electron energy from 45 kev up to a least 24 Mev. The only data on photon energy dependence indicate that the lower limit extends to an x-ray energy of 200 kvp (mean energy $\sim$ 100 kv). From the results of electron irradiation, it may be deduced that the upper limit for photons extends well above a mean energy of 24 Mev.

7. Stability

Ceric sulfate solutions are very stable when stored in light-tight bottles. Over a period of two months, unirradiated 0.1M solutions are stable to within 0.7%. Over a similar period, ceric ion depletion of approximately 5% is observed in 10^{-4}M solutions, as is expected. Irradiated 10^{-3}M solutions are stable to within 1 to 3% during a one month period after irradiation.

The solutions are, however, sensitive to visible and ultraviolet light.[23] The effect is most pronounced at low concentrations. Because of this effect, solution concentrations giving optical densities within the instrumental range (0.4 to 0.2 millimolar) should be measured immediately after preparation.

8. Temperature Dependence

Between 7°C and 35°C the yield is constant within experimental error. At higher temperatures a steady decrease in yield to 2.23 at 61°C is observed. Thus, a temperature correction is necessary if the dosimeter is irradiated at elevated temperatures.

B. Use of the Dosimeter

Reliable results are not as easy to obtain with the ceric dosimeter as with the Fricke dosimeter. If the procedure outlined below is followed carefully, however, good precision can be achieved.

1. Reagents

The purity of reagents used in preparation of solutions is a critical factor in successful use of the ceric dosimeter. The following reagents have been found satisfactory:

(a) G. F. Smith reagent grade ceric sulfate (solid) or Baker and Adamson reagent grade ceric ammonium sulfate.

(b) 95 to 98 wt % Baker and Adamson or duPont reagent grade sulfuric acid.

(c) Triply distilled water. Barnstead distilled water redistilled twice from potassium permanganate/calcium oxide is acceptable.

2. Preparation of Solutions

The solutions should never contact any surface other than a clean glass surface and all glassware should be Pyrex or equivalent. Plastic containers or glass containers with plastic caps are not satisfactory. Prior to use, all glassware should be rinsed with singly distilled water, cleaned with hot concentrated nitric acid, and rinsed thoroughly with triply distilled water. The glassware may be stored under triply distilled water until used.

Prepare a stock solution, 0.1M in ceric sulfate and 0.4M in sulfuric acid, as follows. Dissolve 53 g of $Ce(HSO_4)_4$ (G. F. Smith ceric sulfate) or 63 g $(NH_4)_4Ce(SO_4)_4 \cdot 2H_2O$ (Baker and Adamson ceric ammonium sulfate) and 22 ml concentrated (95 to 98%) H_2SO_4 (Baker and Adamson or duPont) in sufficient triply distilled water to make one liter of solution. Dosimetric solutions are prepared by dilution of the stock solution with 0.4M sulfuric acid. Solutions should be stored in light-tight bottles to avoid possible photochemical effects, and must not be allowed to contact any organic material. Whenever fresh ceric stock solutions are prepared they should be checked against the Fricke dosimeter. If the stock solutions are used over a long period of time, they should be checked monthly. Stock solutions should be discarded if their G values deviate from the expected value of 2.50 (ceric ions reduced per 100 ev of energy absorbed) by more than ±4%.

3. Analytical Methods

Absorbed dose is determined by measuring the depletion of ceric ion in the ceric sulfate solutions. This may be done either by potentiometric titration or by measurement of the optical absorption at 320 mμ wavelength in a spectrophotometer as described below.

a. Potentiometric Titration. The titration is performed with sulfuric acid solutions of ferrous ammonium sulfate. Reagent grade ferrous ammonium sulfate is a satisfactory secondary standard if precautions are taken to prevent efflorescence and oxidation of the ferrous ion. This can most safely be done by purchasing the reagent in small bottles which may be discarded after preparation of one solution. Alternatively (and less acceptably), the reagent bottle should be kept tightly sealed, and in any case should not be used over a long period of time.

The molar concentration of the ferrous solution should be approximately equal to the molar concentration of ceric sulfate in the solution to be titrated. This concentration can usually be estimated closely enough. The solutions are prepared by dissolving the appropriate amount of ferrous ammonium sulfate in 0.4M sulfuric acid to make a solution of the desired molar concentration.

The apparatus required for titration includes: a vacuum tube voltmeter with a full-scale sensitivity of 1.5 volts or better, a titrating burette, platinum and calomel electrodes, a beaker of the appropriate volume, and a magnetic stirrer. First 25 ml of the ceric sulfate solution to be analyzed are poured into the beaker (dilute to 25 ml with 0.4M sulfuric acid, if necessary). The solution should be stirred continuously during titration. Measured quantities of the ferrous ammonium sulfate solution are added by means of the burette. After each addition, the quantity added and the voltmeter reading are recorded after the reading becomes steady. The titration curve is then plotted, as shown in Fig. 1. Figure 1 illustrates titration curves for two concentrations of ceric sulfate. Note that the end point is less sharp in the dilute solution (curve B) than in the concentrated solution (curve A). The end point can be estimated from the direct titration curve. The proper point to be chosen is the point of maximum slope; this point can be determined more precisely by plotting the slope of the direct titration curve against the quantity of ferrous ion added.

b. Spectrophotometric Method. The spectrophotometric method is more convenient and simpler than the titration method of analysis. When the spectrophotometric method is used the apparatus should be equivalent to that used with the Fricke dosimeter (see Section I-B-3). The spectrophotometer may be calibrated by the following method. Prepare a 0.1M solution of ceric sulfate as outlined in Section II-B-2 above; determine the ceric ion concentration by titration with standard ferrous ammonium sulfate, as outlined in Section II-B-3 above. Prepare seven samples spanning the concentration range 0.04 to 0.2 millimolar by diluting aliquots of the analyzed solution with 0.4M sulfuric acid. Measure the optical densities of these samples (0.5 mm slit width; 1 cm matched quartz cells) at a wavelength of 320 mμ against 0.4M sulfuric acid as a blank. Prepare a curve of optical density versus concentration and calculate the spectrophotometer extinction coefficient, k, from the slope of this curve. The value of k should be near 5600 optical density units per mole per liter.

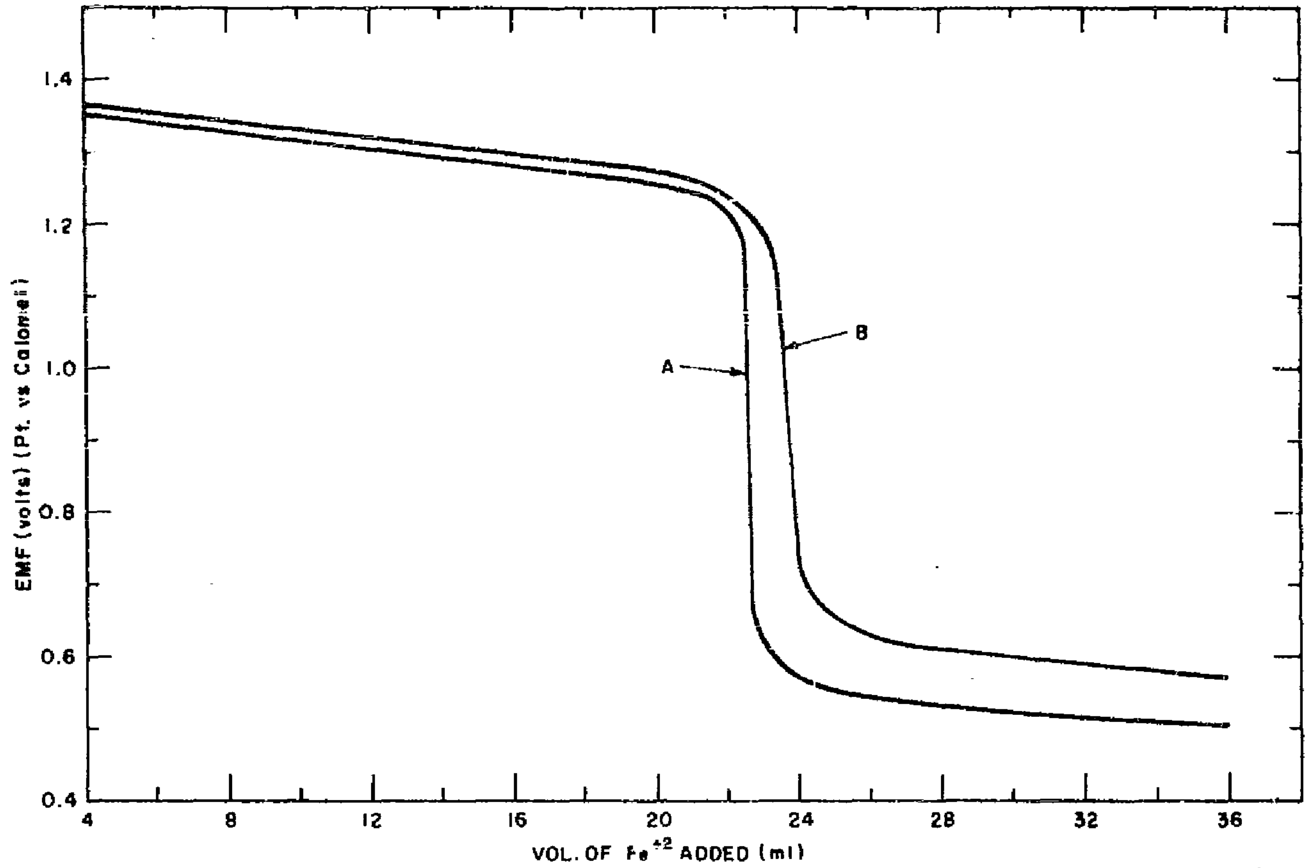

FIG. 1 POTENTIOMETRIC TITRATION OF Ce^{+4} WITH Fe^{+2}
CURVE A – 0.05 M Fe^{+2} IN 25 ml OF ~0.05 M Ce^{+4}
CURVE B – 0.001 M Fe^{+2} IN 25 ml OF ~0.001 M Ce^{+4}

4. Irradiation Cells

Irradiation cells should be Pyrex glass or equivalent. As noted above, plastic containers or plastic closures are not satisfactory. The cells should be cleaned before use by the procedure outlined in Section II-B-2 above. To avoid wall effects the minimum internal dimension of the container should be 8 mm.

5. Dosimetric Procedure

Since the absorbed dose is determined from the change in ceric concentration induced by radiation, a suitable initial concentration should be chosen for the dose region to be studied. The following ceric concentrations should be used in the dose ranges indicated:

Initial Ceric Ion Concentration	Dose Range (rads)
10^{-3}M	10^5 to $3x10^5$
$4x10^{-3}$M	$3x10^5$ to $1.2x10^6$
0.02M	$1.2x10^6$ to $6x10^6$
0.05M	$6x10^6$ to $1.5x10^7$

These solutions are prepared from a 0.1M stock solution by dilution with 0.4M sulfuric acid. It is advisable to maintain a stock of 0.4M sulfuric acid solution for use in the dilution.

To measure absorbed dose, expose a quantity of the dosimetry solution in the radiation field for a carefully measured time. After irradiation measure the residual ceric concentration either by the spectrophotometric method or by titration as outlined in Section II-B-3 above. If the spectrophotometric method is used, dilute the irradiated solution with 0.4M sulfuric acid solution so that the resulting concentration is within the instrumental range (0.04 to 0.2 millimolar). Pipettes and volumetric flasks used in performing the dilution should be calibrated since a small error may be considerably magnified by the large dilutions that are required.

If the spectrophotometric method is used, the dose is calculated from the following expression:

$$D(\text{rads}) = 9.65\times10^{8} \times f \times OD/G\rho k$$

where f = dilution factor (final volume/initial volume) of the measured solution,

OD = change in optical density,

ρ = density of undiluted solution (grams/ml),

k = extinction coefficient,

G = 2.50.

The following densities should be used:

Ceric Ion Concentration	Density, ρ
0.001M	1.024
0.004M	1.025
0.02M	1.030
0.05M	1.040

If potentiometric titration is used the dose is calculated from the following expression:

$$\text{dose (rads)} = 9.65\times10^{8}\ \Delta C/G\rho$$

where ΔC = change in concentration of ceric ion (moles/liter),

G = 2.50,

ρ = density (grams/ml) as given above.

6. Variations of Procedure

It is not intended to imply that other procedures or deviations from the procedure recommended here will not yield equally reliable results with the ceric sulfate dosimeter. However, the procedure outlined above has proved to be satisfactory, and it is strongly recommended that acceptability of any deviation be established by comparison with the Fricke dosimeter. In fact, acceptability of these techniques is based on routine periodic checking of the ceric sulfate solution against the

Fricke dosimeter. Routine use of the Fricke dosimeter as a standard is a powerful technique for revealing defects in the procedure and for tracing sources of error.

Appendix A

THE OXALIC ACID DOSIMETRY SYSTEM

This appendix consists of the complete text of "Proposed Tentative Method of Test for Gamma Radiation by Chemical Dosimetry" prepared by Joint Subcommittee II of Committees D-9 and D-20 of the American Society for Testing Materials (ASTM). This text contains the procedure now being used in the Round Robin evaluation of the oxalic acid dosimeter by the ASTM. It is included here for information and for possible use by personnel of the Quartermaster Radiation Laboratory (QMRL) in their evaluation of the oxalic acid dosimeter. In addition to the proposed ASTM method, a brief suggested procedure for investigating the analysis of oxalic acid with ceric sulfate is included.

1. ASTM Proposed Tentative Method for Oxalic Acid Dosimetry

PROPOSED TENTATIVE METHOD OF TEST FOR GAMMA RADIATION BY CHEMICAL DOSIMETER

SCOPE:

1. This method describes a procedure for measurement of gamma radiation in the range of absorbed dose of 1.6×10^6 - 5×10^7 rads, through the decomposition of oxalic acid in aqueous solution. The absorbed dose is derived from the difference in concentration of oxalic acid before and after irradiation.

SIGNIFICANCE:

2. (a) Dose Rate - No influence of dose rate in range of 720 rads per hour to 3.96×10^5 rads per hour.

(b) Temperature - The irradiation temperature should be between 15-85°C. The readings of the samples with the spectrophotometer may be carried out at room temperature.

(c) Energy Dependence - For Round Robin.

DEFINITIONS:

3. Rad - The recommended unit for reporting absorbed radiation dose, defined as 100 ergs of energy absorbed per gram of specimen.

APPARATUS:

4. (a) Spectrophotometer - Beckman Model DU hydrogen lamp attachment, and 1 cm matched quartz cell, or equivalent apparatus.

(b) Containers

(1) Chemically resistant glass(1) shall be used to hold the reagents.

(2) Containers should be of approximately the same dimensions, geometry and materials as those employed to hold specimens for irradiation. A thin-walled glass liner, 1.0 to 1.2 mm maximum thickness may be employed inside the specimen container where necessary. No internal dimensions of the containers should be less than 8 mm in diameter. The containers shall contain ground glass stoppers or be flame sealed.

(3) All containers should be cleaned with chromic-sulfuric acid cleaning solution, then rinsed thoroughly with distilled or deionized water. Care should be taken that the last three rinsings are water clear.

PURITY OF WATER AND REAGENTS:

5. (a) All reagents shall conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society.(2)

(b) Distilled or deionized water used in this method shall be free of organic impurities.

METHOD:

6. (a) Reagents

(1) Dosimetric Solution - Prepare a solution of oxalic acid using the initial concentrations shown in Table 1. A suitable initial concentration should be chosen for the dose region to be studied.(3) The dosimeter solution is stable against the influence of light and air for several months.

(2) Cupric benzidine reagent - Prepare two solutions, A and B.

Notes: (1) - Pyrex glass has been found to be satisfactory for this purpose.

(2) - "Reagent Chemicals, Am. Chem. Soc. Specifications," Am. Chem. Soc., Washington, D.C.

(3) - A decomposition less than 30% of the initial concentration of oxalic acid is necessary to maintain a constant radiation yield.

Solution A: Dissolve 32.5 mg recrystallized benzidine(4) and (5) in 2 ml 30 vol-% acetic acid and dilute with distilled water to 250 ml in a volumetric flask.

Solution B: Dissolve 125.0 mg cupric acetate in 250 ml distilled water in a volumetric flask.

(b) Exposure and Reading in the Spectrophotometer

(1) Place the dosimetric solution in the radiation field for a carefully measured length of time (see Table 1).

(2) After removal from the radiation field, withdraw an accurately measured aliquot, as indicated in Table 1, from this solution and dilute this aliquot in a volumetric flask to the volume given in Table 1, adding the cupric-benzidine reagent also indicated in Table 1 before diluting to the mark on the flask. Obtain the complexing reagent (A + B) by mixing equal volumes of A and B. The above reaction is instantaneous at room temperature and the complex formed is stable. As reference for the optical density measurements, a solution of 200 ml of the cupric benzidine reagent and 800 ml of distilled water is used.

Absorbed Dose, Megarads	Initial Conc. of $H_2C_2O_4$, m Mol/L	Aliquot of Exposed Dosimeter Solution ml	Volume of Cupric-Benzidine Reagent, ml	Total Dilution, Volume, ml	Calculation Factor Megarads
1.6 - 6	100	2	200	1000	43.55
6 - 18	300	1	400	2000	174.2
18 - 50	750	1	0	1000	---
		Then.			
		25 (from 1000)	20	100	348.4

Table 1

Notes: (4) The molar extinction coefficient of benzidine should be checked for each recrystallized batch. The usual molar extinction coefficient is, k = 2260 l mol^{-1} cm^{-1} at room temperature. The k value is independent of the temperature.

(5) Considerable caution should be exercised in using benzidine as it is considered to possess carcinogenic properties. References: Carcinogens in the Human Environment, W. C. Heuper, M.D., Archives of Pathology, March 1961, Vol. 17, (p.19) pp. 237-367 and W. C. Hueper, Cancer and the Law, Medicolegal Considerations of Occupational and Non-Occupational Environmental Cancers, par. 38, p. 564, from the Lawyers Medical Cyclopedia, C. J. Frankel, Editor, published by Allen Smith Company, Indianapolis, Indiana.

(3) The measurements in the spectrophotometer are performed in 1 cm matched quartz cells at 248 mμ and a slit width of 1 mm., with a hydrogen lamp attachment. Both irradiated and unirradiated oxalic acid dosimetric solutions (of the same initial concentration) must be measured in order to obtain the change in optical density during irradiation.

(c) Dose Calculation

The absorbed dose D can be calculated from the equation(6);

$$D = K \cdot \Delta OD \text{ Megarads}$$

where K = calculation factor from Table 1

ΔOD = change in optical density measured at 248 mμ

DEVELOPMENT OF EQUATION FOR CONVERTING CHANGE IN OXALIC ACID CONCENTRATION TO RADIATION EXPOSURE UNITS(a)

$$D = \frac{N \cdot d \cdot \Delta OD \cdot 100}{k \cdot Q \cdot 10^3 \cdot G} \text{ eV/g}$$

Where: N = Avogodro's number ($=6.020 \times 10^{23}$ molecules/mol)

ΔOD = the change in optical density measured at 248 mμ

k = the molar extinction coefficient ($=2260$ 1 · mol cm^{-1})

Q = the density of the dosimeter solution (=1.0 g/ml)

G = The G-value (=4.9 molecules decomposed per 100 eV)

d = the dilution factor (final volume/initial volume)

$$D = \frac{6.02 \cdot 10^{23} \cdot d \cdot \Delta OD \cdot 100}{2260 \cdot 1.0 \cdot 10^3 \cdot 4.9} \text{ eV/g}$$

$$D = \frac{6.02 \cdot 10^{23} \cdot d \cdot \Delta OD \cdot 100}{2260 \cdot 1.0 \cdot 10^3 \cdot 4.9} \cdot 1.602 \cdot 10^{-14} \text{ rads}$$

$$D = 8.71 \cdot 10^4 \cdot d \cdot \Delta OD \text{ rads}$$

Note: (6) The above equation and the calculation factors of Table 1 were derived from the equation of I.G. Gragonic, as shown in the Appendix, by combining 8.71×10^4 with the dilution factors (d) and then dividing by 1×10^6 to convert rads to megarads.

Note: (a) The information for the dosimetry procedure reported in this document and the development of the equation as shown above were taken from the work of I. G. Draganic as reported in the J. Chim. Phys. 56, 9-20 (1959)

2. Analysis of Oxalic Acid with Ceric Sulfate

As pointed out in Technical Report 1 under Contract DA 19-129-QM-1900,[1] it seems possible that the range of the oxalic acid dosimeter may be extended below 1.6×10^6 rads by selecting a more sensitive method of analysis than that proposed in the preceding section. Matsui[24] was able to measure doses as small as 2.23×10^5 rads in solutions containing 0.016M oxalic acid by titration with 0.1N caustic soda solution. Because the radiolysis product, CO_2, can interfere with this method of analysis, it is necessary to heat the irradiated samples for about 30 minutes to remove CO_2. A third possible analytical method, as yet untried for dosimetry, is oxidation with an excess of ceric ion and subsequent analysis for residual ceric ion either by titration with ferrous ion or by spectrophotometric analysis. The ceric sulfate method of analysis is discussed below.

The basis of the method is complete oxidation of the oxalic acid by an excess of ceric sulfate. The oxidation is performed by adding ceric sulfate in acid solution to the oxalic acid and refluxing for 15 to 30 minutes until the oxidation reaction is complete. Analysis of the residual ceric ion in the cooled solution may be performed either spectrophotometrically or by potentiometric titration with ferrous ion.

Measurements of doses smaller than 1.6 megarads will probably require more dilute solutions of oxalic acid than those recommended in the ASTM procedure. This modification is necessary so that the oxalic acid destruction produced by radiation causes a measurable change in concentration. Since $G = 4.9$, a dose of 2×10^5 rads will destroy 1 millimole of oxalic acid per liter of solution. Thus, to measure a dose of 2×10^5 rads the initial and final concentration of oxalic acid should be such that the errors in their measurement are small (within the desired precision) compared to 1 millimole/liter. For such doses it is likely that a concentration of the order of 20 millimolar or less will be required.

Appendix B

THE POLYISOBUTYLENE DOSIMETER

The following discussion of a polyisobutylene dosimeter is based on the work by Wiesner. Wiesner[25] studied the use of radiation-induced depolymerization of polyisobutylene solutions in organic solvents to measure gamma radiation doses and found that the system met many of the requirements for an ideal dosimeter. Solutions of polyisobutylene (viscosity average molecular weights of 200,000 to 300,000) in heptane were used to measure doses in the range 1×10^3 to 3×10^6 rads. Solutions of polyisobutylene (viscosity average molecular weight of 5000) in heptane were used to measure doses from 4×10^5 to 1.5×10^7 rads. It seems likely that polyisobutylene with a viscosity average molecular weight intermediate between these two values can better cover the range from 10^4 to 10^7 rads that will be encountered in the QMRL. The following is a brief outline of Wiesner's technique for use in examining commercial materials for their potential applicability.

The following table contains trade names and suppliers of semi-solid and liquid isobutylene polymers that may be used in this study.

Trade Name	Supplier
Vistac	Advance Solvents and Chemical Corp.
Vistanex	Enjay (Standard Oil Co. of New Jersey)
Indopol	Standard Oil Co. of Indiana
Polybutenes	Oronite Chemical Co.

Viscosity measurements can be made simply with a capillary viscometer. Any of the viscometers and methods described in the ASTM Standard Method of Test for Kinematic Viscosity (ASTM Designation D445-61)[26] may be used.

Following Wiesner, a sufficient quantity of polyisobutylene of the appropriate viscosity average molecular weight is dissolved in heptane to yield a solution with a viscosity of 312 centistokes (cS). The viscosity of the solution, η_o, and the viscosity of the solvent, η_s, are then measured. The viscosity measured after irradiation is η. On log-log paper the relative viscosity $\eta - \eta_s/\eta_o - \eta_s$, is plotted against radiation dose. The linear portion of the curve on the double logarithmic plot is the usable dose range of the materials studied. The linear portion of the curve can be extended to lower doses by plotting the dose on a logarithmic scale versus $[1 - (\eta-\eta_s)/(\eta_o-\eta_s)]^{\frac{1}{2}}$ on a linear scale.

Different batches of a commercial polymer of the same nominal molecular weight exhibit differences in the chain length. Consequently the molecular weights of different batches will differ with the result that the calibration curves of different batches will also differ. Wiesner found, however, that the different calibration curves can be made to coincide by displacement along the dose scale. The reason for this is as follows. Assume two batches, A and B, in which the molecular weight of A is higher than that of B. During irradiation Batch A will be depolymerized to the point where eventually it will have the same molecular weight and hence the same viscosity as Batch B, after which it will behave like Batch B under further irradiation. Thus, subtraction of the dose required to give Batch A the same viscosity as that initially possessed by Batch B, when the two solutions have the same initial polymer concentration, will produce coincidence of the two calibration curves. This method may be used to correct for batch differences. However, it is probably advisable, for maximum precision, to calibrate each batch against the Fricke dosimeter.

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DEPARTMENT OF THE ARMY
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REPLY TO
ATTENTION OF

November 1, 2001

Dr. R. William Field
University of Iowa
Department of Epidemiology
College of Public Health
Iowa City, IA 52242-5000

SUBJECT: Freedom of Information Act Request (FOIA)

Dear Dr. Field:

This is in reply to your Freedom of Information Act request, dated April 16, 2001 originally sent to the Defense Technical Information Center requesting the following document, "AD296591 Review of Dosimetry Field". When this document was originally published it was categorized as, "unclassified/limited" distribution. A current review of this document determined that is should be "delimited" and available to the public.

There are no search, review, or reproduction costs associated with this request. Please feel free to contact me if I can be of further assistance. The telephone number is (508) 233-4513.

Cordially,

Teresa Crouse
Paralegal Specialist

Cc:
Rose Marie Christensen
Kelly D. Akers

NATIONAL PERFORMANCE REVIEW

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