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ENVIRONMENTAL STUDIES: RADIOLOGICAL SIGNIFICANCE
OF
NUCLEAR ROCKET DEBRIS

PROGRESS REPORT
FOR
PERIOD JULY 1, 1965 - JUNE 30, 1966

B.R. Fish
T.G. Clark
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HEALTH PHYSICS DIVISION

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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* For Space Nuclear Propulsion Office, United States Atomic Energy Commission,
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ENVIRONMENTAL STUDIES: RADIOLOGICAL SIGNIFICANCE OF NUCLEAR ROCKET DEBRIS

I INTRODUCTION

This report for the period July 1, 1965 to June 30, 1966 summarizes the radiological safety studies that are being conducted for the Space Nuclear Propulsion Office, Flight Safety Branch. A continuing assessment is being made on the significance of radioactive debris that may enter the biosphere following accidental, in-flight destruction of a nuclear propulsion system. This program emphasizes the production and dosimetry of micron-size particles containing either individual radionuclides or a spectrum of fission products and studies of the interaction of such particles with man and his environment.

Two dosimetric techniques are used to extrapolate to minute volumes of tissue the dose absorbed from small spheres of reactor fuel material. The first of these utilizes an air ionization chamber. The second employs a novel technique in which progressively smaller discs of an organic scintillator (anthracene or NE-102-A plastic) yield pulse height spectra. These are converted to energy spectra using a computer program to apply appropriate corrections, and the absorbed dose is calculated. Individual dose measurements are then assembled into extrapolation curves which relate dose to tissue volumes at specified absorber thicknesses. This technique is useful for measurements with sources of activity in the range of 0.05 to 500 μCi , which is below the range for which an ionization extrapolation chamber can be used. The two techniques are complementary, each having inherent advantages; generally the air ionization chamber is required for those radiation fields in excess of 50 rads per hour.

Spherical particles from 1-mm to 1-cm diameter have been made in thermal gradient columns or in split molds of aluminum or teflon. "Tagging" of these sources has been accomplished by addition of radioisotopes to a uranium-carbon mix prior to insertion in the mold or by neutron irradiation following particle fabrication.

A complete analysis of the hazards arising from the accidental fragmentation of nuclear reactors or isotopic power sources requires a knowledge of the deposition and adhesion of radioactive debris on the skin. In addition to information on radiobiological effects and on the dosimetry of small particles some estimate of source-skin contact probability is needed. A study has been initiated to determine the degree of initial retention on the arm of particles falling vertically in a uniform fallout field. Coated UC_2 fuel beads have been used in tests to determine the initial deposition and the retention time on the skin of this type material. A wide variation was noted between individuals in the average length of time these particles remained on the arm under identical environmental conditions. Skin decontamination experiments showed that simple sanitary habits suffice to remove particles ranging from 1 to 1000 microns in diameter.

Preliminary determination of gastrointestinal tract transit time in man, using insoluble particles, showed a significant fraction to be retained for periods longer than the 31-hour "standard" value.

2.0 DOSIMETRY

2.1 Ionization Extrapolation

2.1.1 Irradiated Fuel Beads, Coated

Dose rate measurements have been made on several coated fuel beads whose diameters ranged from 253 to 286 microns. Figure 2.1 shows a typical particle. Irradiation of the beads was performed in a pneumatic tube facility at the Oak Ridge Research Reactor (ORR) at a thermal flux level of 5.0×10^{13} neutrons/cm²-sec, the resonance flux level being 2.3×10^{12} neutrons/cm²-sec. Flux levels were determined by activation analysis of Au-Al and Au-Mn monitors irradiated concurrently with the fuel material.

Two methods were employed to estimate the fissions per particle. In the preferred method, the area under the ¹⁴⁰La photopeak, found by gamma spectroscopy, was used as an index of the total number of fissions occurring.⁽¹⁾ For a second estimate, the total fissions were calculated by the $n\sigma\phi t$ formula based on particle weight and neutron flux values. The small difference in experimental and calculated values can be explained by neutron shadowing effects. For a sphere the reduction in flux is given by:⁽²⁾

$$f = 1 - 3/4 n \sigma r$$

where

σ = activation cross-section in barns

n = ²³⁵U density in atoms per cm³

r = particle radius in cm

f = fractional reduction in flux level

Dose rate values for five different particles at a depth of 7.62 mgm/cm² in tissue equivalent material (polystyrene) are listed in Table 2.1. Each of these beads was irradiated for 10 minutes. Dose measurements were made with the ionization extrapolation instrument using a 3-mm diameter collecting electrode. For ease of interpretation, all data have been converted to rads per hour per fission occurring in the particle. Variation in dose rates between similar particles may be

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Fig. 2.1 Pyrolytic Carbon Coated UC_2 Particle. ($\sim 275\mu$ Diameter.)

Table 2.1

Ionization Extrapolation Dosimeter Measurements

Particle Number	Diameter (Microns)	Weight (μgms)	Fissions per Particle ($\times 10^{12}$)		Time after irradiation (minutes)	Dose Rate *	
			from ^{140}La photopeak	from weight, and flux **		rads/hr	rads/hr -- fission
66-2	256	93.4	2.53	2.96	330	11,800	4.68×10^{-9}
66-4	286	126.1	3.68	4.68	266	6,900	1.88×10^{-9}
66-5	283	126.5	3.71	4.89	231	8,100	2.18×10^{-9}
66-6	273	112.0	4.09	4.36	356	29,100	7.11×10^{-9}
66-7	253	92.0	3.04	3.47	326	22,400	7.38×10^{-9}

* At depth of 7.62 mg/cm^2 .

** Does not include correction for neutron shadowing.

due to an inhomogeneous distribution of fissionable material within the source volume with a resulting inhomogeneity of fission product activity. Dose rates given in Table 2.2 for a 100- μ particle were calculated by ZAP31, a computer program that uses an iterative procedure to follow the various interactions of β and γ rays as they traverse matter.⁽³⁾ These theoretical dose rates are averaged over one square centimeter area and are based on a reactor operating time of nine minutes at a power level of 1120 MW.⁽⁴⁾

Table 2.2

ZAP31 Computer Program Calculations for a 100- μ Reactor
Debris Particle

Time after reactor operation	Dose Rate (depth of 7 mg/cm ²)	
	rad/hr	rad/hr-fission
360 minutes	320	5.5×10^{-10}
12 hours	160	2.7×10^{-10}
24 hours	75	1.3×10^{-10}
84 hours	23	3.9×10^{-11}
168 hours	6.1	1.0×10^{-11}

2.1.2 Irradiated Fuel Beads, Uncoated

Dose rate data have been obtained on uncoated UC₂ fuel beads using the ionization extrapolation instrument. These beads were obtained from the U.S. Naval Radiological Defense Laboratory and are approximately 125- μ diameter. X-ray photographs, shown in Fig. 2.2, indicated the absence of any significant coating on these spheres.

Irradiation of this material was performed at the ORR facility at flux levels of greater than 10^{13} neutrons/cm²-sec for periods up to 20 minutes. Measurements have been taken at times out to 25 days after irradiation. Comparative data from two such fuel beads are given in Fig. 2.3. There is an apparent decrease in

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Fig. 2.2 X-ray Photograph of Typical Uncoated UC_2 Particle
($\sim 125 \mu$ diameter).

Fig. 2.3 Tissue Dose Rates of Irradiated Uncoated UC₂ Fuel Beads
(Extrapolation Chamber - 3mm Electrode)

the slope of the dose rate curves with increasing absorber thickness. This seeming anomaly is probably due to inherent variability in dose rate and fissions in the two particles. The activity in particle N-23 as a function of time is shown in Fig. 2.4. This is an uncoated UC_2 sphere having a diameter of 129 microns. A 1-mm diameter collecting electrode was used to measure the dose rates; absorber thickness was 2.54 mg/cm^2 . Dose rate measurements were made at intervals from 2.5 to 840 hours after irradiation. The solid line can be expressed by the equation

$$D = 5.6 \times 10^5 t^{-1.17}$$

where

D = dose rate in rads per hour

t = decay time in hours.

With a constant absorber thickness the ionization current is a function of the electrode diameter as shown in Fig. 2.5. These measurements were made at a depth of 2.54 mg/cm^2 on a 129- μ diameter uncoated fuel bead, 48 hours after irradiation. At this time the dose rate was changing at a rate less than 10% per hour, Fig. 2.6.

A curve similar to that shown in Fig. 2.5 was obtained using a 1-cm diameter sphere containing ^{45}Ca . However, the differences in dose rate values using various size electrodes was not as great as those seen for the smaller fuel beads, indicating that chamber response is also a function of source dimensions.

2.1.3 Irradiated 1-mm Diameter Fuel Spheres

It has been assumed that there is a finite probability for the ingestion of particles as large as 1-mm diameter ($\sim 1/32''$) and for their retention on the skin over a significant period of time. The dose rate from a 1-mm simulated reactor debris particle is given in Fig. 2.7 as a function of time after activation. This spherical source, containing ^{235}U , was irradiated for 20 minutes in the Low Intensity Test Reactor (LITR) at a flux level of 1.36×10^{12} neutrons/ cm^2 -second, and the radioactive decay followed out to 12.5 days. The dose rate was measured over an area of one square centimeter at a depth of 7.62 mg/cm^2 in tissue equivalent material.

Fig. 2.4 Tissue Dose Rates of Irradiated Uncoated UC_2 Fuel Bead
(Extrapolation Chamber - Imm Electrode).

Fig. 2.5 Tissue Dose Rate as a Function of Electrode Diameter
 $129 \pm 1 \mu$ Particle (N-23)(45 hours after irradiation).

Fig. 2.6 UC_2 Particle Data Showing Time at which there is <10%
Change in Dose per Hour.

Fig. 2.7 Tissue Dose Rates of Irradiated 1-mm UC_2 Source
(Extrapolation Chamber - 11-mm diameter electrode)

2.2 Empirical Dose Model

A previous report⁽⁵⁾ describes an empirical dose model in which the radioactive source may differ in composition from the dose receptor (e.g., a reactor debris particle in contact with tissue). In this model a library of dose rates and spectra is accumulated from sources of known elemental composition and size. Linear combinations of these spectra can be made to approximate the spectrum from a source of unknown or complex isotopic composition such as the fission product inventory in reactor fuel material. Dose rates from the standard sources are then added in this same proportion to obtain an estimate of the dose rate from the sample.

The beta ray energy spectrum from a 1-cm diameter sphere of a graphite-UC₂ bead mixture simulating a reactor fuel particle is shown in Fig. 2.8. This particle was irradiated at the LITR for 10 minutes and the spectrum analyzed 2 1/2 hours later using the anthracene spectrometer described in section 2.3.

Graphite spheres tagged with $^{45}\text{Ca} + ^{89}\text{Sr}$ have been used to simulate this spectrum quite well up to ~ 1 Mev. The spectrum of the reactor particle superimposed on one composed of $.46\ ^{89}\text{Sr} + .43\ ^{45}\text{Ca} + .10\ ^{204}\text{Tl}$ is shown in Fig. 2.9. Experimental measurements of the dose rate from the irradiated particle are a factor of five higher than that predicted by the simulation. This discrepancy is due to the large contribution to dose rate by the high energy components (> 2 Mev) in the irradiated source. Plans are underway for simulating these higher energies with $^{106}\text{(Ru-Rh)}$ labeled particles.

Extrapolation chamber data for the 1-cm irradiated sphere is given in Table 2.3.

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Fig. 2.8 Beta Spectrum from 1 cm Diameter Irradiated Source
(512 Channels - 10 min. Count).

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Fig. 2.9 Simulation of 1 cm Diameter Source Spectrum using ^{45}Ca , ^{89}Sr , and ^{204}Tl (256 Channels).

Table 2.3

Dose Rate from 1-cm Diameter Irradiated Fuel Particle

Tissue Depth (mg/cm ²)	Decay Time (min.)	Dose Rate (rad/hour)
2.54	70	519
8.52	85	358
17.20	100	263
32.86	130	114

(Particle was irradiated 10 minutes at a flux level of
 5.68×10^9 neutrons/cm² - second)

2.3 Scintillation Extrapolation Dosimetry

2.3.1 Design and Calibration

The continuing development of theoretical models and experimental techniques for tissue-equivalent dosimetry is important in medicine and biology. The field of beta dosimetry has received considerable attention particularly since 1950, and much progress has been made in the basic understanding of both the physical processes involved in electron slowing-down and in the biological damage resulting to tissue from energetic electrons.

L. V. Spencer⁽⁶⁾ has discussed calculation of the dose due to negatons, incorporating the Bethe-Bloch stopping power formula for the determination of the slowing-down flux and using the Mott theory of nuclear scattering for describing the electron scattering. This method has a sound theoretical basis for dose calculations. Another approach has been proposed by Loevinger.⁽⁷⁾⁽⁸⁾ This method, based on experimental measurements, describes a point-source function which, with the proper parameters, may be integrated over a distributed source area or volume to arrive at estimates of absorbed energy at points inside a medium. Both of these approaches require the medium to be of infinite extent and homogeneous in composition, although the beta sources need not be uniformly distributed. The Loevinger method is applicable in a strict sense only to media of low atomic number.

There remain physically important areas of investigation for which there are no applicable theories and for which adequate experimental techniques have not been developed. This is the case regarding dosimetry of beta particulates in inhomogeneous and nonuniform systems. Scintillation-extrapolation dosimetry is a technique that can be used in determining the absorbed dose to vanishingly small volumes of tissue. This technique may be applied to physical situations as for example, a radioactive particle on the surface of the skin.

The scintillation method has been applied to determining beta dose by several authors. Brannen and Olde⁽⁹⁾⁽¹⁰⁾ have used an optical lens and a photo-

multiplier tube to view a small volume of plastic scintillator sheet imbedded in the center of a 2 in x 2 in x 2 in lucite phantom to obtain depth-dose estimates relative to the surface dose delivered by a 1.5 Mev beam of electrons. The effective volume from which scintillations were observed was 1.1 mm³. Other authors⁽¹¹⁾⁽¹²⁾⁽¹³⁾ have used thin anthracene and plastic scintillators mounted on lucite light pipes to measure depth-dose relative to surface dose from plane sources of beta emitters. Nentwig⁽¹⁴⁾ has used thin plastic scintillators for the development of a portable scintillation detector for the purposes of radiation protection. In all of these investigations the dose measurements were relative, and the output incorporated current measuring devices. The currents observed were assumed directly proportional to dose, and this assumption can result in serious error. More accurate dosimetry is possible using an ionization extrapolation chamber such as described by Loevinger.⁽¹⁵⁾

In order to use scintillators for the determination of the energy deposited by the absorption of beta particles, it is necessary to know the fluorescence response of each detector in terms of light output per unit energy absorbed. It is well known that organic scintillators exhibit a characteristic nonlinear response to surface-incident negatons of low energy. Birks⁽¹⁶⁾ has discussed the scintillation process in detail and has proposed a theoretical relation describing the response function of scintillators which adequately follows experimental data. In a previous work Wilkie and Birkhoff⁽¹⁷⁾ explain its use with reference to the response of anthracene to surface-incident beta particles.

Thin scintillators of anthracene and NE-102 (Nuclear Enterprises, Winnipeg) were used in the present work. These scintillators were joined to polished lucite light pipes 1-inch diameter by 1/2-inch thick. Scintillator-light pipe assemblies were optically coupled with clear silicone oil to an end-window photomultiplier tube (a selected, low noise Amperex, Type XP-1010). Pulse output from the photomultiplier was recorded using a multichannel analyzer (Nuclear Data Model ND-110) the linearity of which was adjusted using a precision pulse generator (ORNL Model Q1212-C). Response characteristics of these scintillator assemblies were studied

with the aid of a linear accelerator designed and built for this purpose. The accelerator provided a beam of monoenergetic electrons with energies variable between zero and 60 kev. A schematic of this apparatus is shown in Fig. 2.10. An alpha source of ^{241}Am provided a reference peak which was used to normalize the data from different scintillation assemblies. A point calibration of beta particles was not practical since the scintillator thicknesses (3 to 50 mg cm^{-2}) were not sufficient to provide a total absorption peak whereas the alpha particles were completely absorbed by 3 mg cm^{-2} (16) giving a relatively sharp peak equivalent to 473 kev betas absorbed in NE-102 or 520 to 560 kev betas absorbed in anthracene depending on the condition of the surface of the particular crystal. Figure 2.11 shows a typical alpha spectrum for anthracene.

Figure 2.12 shows a comparison of the response of plastic and anthracene to surface-incident negatons. The response curve of the plastic exhibited considerably less curvature at the low energies, and the straight line fitted to the data points below 60 kev extrapolated to the response at 624 kev within experimental error. All of the assemblies exhibited essentially the same response curve except in the case of several obviously inferior anthracene crystals. The pulse-height distributions for the mono-energetic negatons were distinguishable from the electronic noise down to 5 kev for both types of scintillators. No corrections were made for the difference between the mean and the mode of the distribution caused by the asymmetry due to statistical fluctuations in the number of photoelectrons emitted by the photocathode when low-energy negatons were absorbed by the scintillators. (18)

Figure 2.13 shows the response curve with additional points obtained using internal conversion electrons from ^{137}Ba , ^{203}Tl and ^{114}Cd . Figures 2.14 and 2.15 show the pulse-height distributions of 30 and 60 kev negatons on the two types of scintillators.

A schematic of the apparatus used to obtain the data for determining the absorbed dose is shown in Fig. 2.16. The instrumentation is calibrated prior to each dose measurement by positioning an alpha source of ^{241}Am 15-cm from the

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Fig. 2.10 Schematic of Linear Accelerator

Fig. 2.11 Typical Alpha Spectrum for Anthracene.

Fig. 2.12 Comparison of the Response of Plastic and Anthracene to Surface-incident Negatons.

Fig. 2.13 Response Curve using Internal Conversion Electrons.

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Fig. 2.14 Pulse-height Distributions for Anthracene.

Fig. 2.15 Pulse-height distributions for NE-102.

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Fig. 2.16 Schematic of Apparatus used to Obtain Data for Determining Absorbed Dose.

scintillator face, evacuating the chamber to a pressure of less than 100 μ Hg, and adjusting the amplifier gain so that the alpha peak appears in the proper channel of the analyzer. Whenever an absorber is to be used to obtain a depth-dose measurement it is necessary to place a similar absorber having a 1-mm aperture in the center over the scintillator face in order to approximate the light collection geometry for the dose measurement. Failure to use this technique incurs a calibration error of approximately 10%. After calibration, air is readmitted to the chamber, and the radioactive particle for which dose information is to be obtained is placed on the scintillator axis with the appropriate absorber of mylar (polyethylene terephthalate) or polystyrene. The gross pulse-height spectrum is then recorded at a total counting rate of less than 10^4 per second in order to minimize difficulties due to accidental coincidences, phototube drift, and errors in the determination of total live time of the count. After the gross pulse-height spectrum is recorded the scintillator assembly is removed, and a lucite cylinder with the same dimensions as the light pipe of the scintillator assembly is coupled to the phototube. The source and absorbers are placed on the lucite cylinder in the same configuration as before, and a background is subtracted from the previously recorded gross spectrum. The background spectrum is quite significant for the scintillators with thicknesses below 75 microns. It includes contributions from the fluorescence of the lucite, Cerenkov radiation, and electronic noise from the phototube and the analyzer. Of these the least significant is the electronic noise. Failure to subtract the composite background from the gross spectrum can result in errors of as much as 50% in the dose estimates for the thinnest scintillators.

2.3.2 Dosimetry Calculations

After a net pulse-height spectrum is obtained the absorbed dose rate in rads per hour per millicurie is calculated using the relation

$$D = \frac{B}{P \exp[-\lambda t]} \sum_c^C \left[\frac{N_c E_c}{F_c} \left(\frac{S_T}{S_s} \right)_c \right]$$

where

B = conversion factor

P = count period

$\exp[-\lambda t]$ = decay correction

N_c = number of counts in channel c ,

E_c = energy associated with channel c ,

F_c = nonlinear correction factor for channel c ,

$\left(\frac{S_T}{S_s} \right)_c$ = ratio of stopping-power of tissue to scintillator.

The nonlinear correction factor, F_c , should be used whenever a nonlinearity is observed in the pulse-height versus energy response which results in a nonuniform energy band width per channel. A computer program was written to perform the required mathematical manipulations for the dose calculation.

2.3.3 Data from Fuel Beads and ^{32}P -Sulfur Spheres

Much information may be obtained from the scintillation method described. Every dose estimate is derived from an energy spectrum which is plotted in graphical form by the computer program. Typical spectra are shown in Fig. 2.17 for a sulfur sphere using three 11-mm diameter scintillators of NE-102 with thicknesses of 405, 93, and 32 microns. If dose estimates for a radioactive particulate are obtained using a series of scintillators of the same diameter but different thicknesses an extrapolation interpretation may be applied which yields a dose estimate for that diameter and zero thickness. In addition, if the dose estimates for zero thickness are plotted as a function of diameter the resulting curve may be extrapolated to zero diameter to give an estimate of the zero volume dose. Figures 2.18 and 2.19 illustrate this procedure for two sulfur spheres of different diameters.

Depth dose curves and data for the determination of dose rate as a function of time for decaying sources may be obtained as previously described. The dose values if not extrapolated by the use of additional scintillation assemblies may be interpreted directly as the absorbed dose (or dose rate) averaged over the volume of the scintillator used for the measurement. A typical depth dose curve for a sulfur sphere obtained using a plastic scintillator 11-mm diameter and 93 microns thick is illustrated in Fig. 2.20. Dose data obtained for a neutron-activated UC_2 sphere using an anthracene dosimeter 11-mm diameter by 105 microns thick is shown in Fig. 2.21. The dose measurements were corrected to zero thickness by extrapolation using a series of 11-mm diameter scintillators with thickness ranging from 405 microns to 32 microns. The magnitude of this correction was +24%. The UC_2 bead was uncoated, had a diameter of approximately 145 microns, and had been irradiated in the Low Intensity Test Reactor for 10 minutes. Monitors showed the thermal flux to be 5.8×10^8 neutrons/cm²-second.

Fig. 2.17 Sulfur sphere using three 11-mm diameter scintillators of NE-102 with thicknesses of 405, 93, and 32 microns.

Fig. 2.18 Extrapolation curves for sulfur sphere, 620 μ diameter.

Fig. 2.19 Extrapolation curves for sulfur sphere, 1.56-mm diameter.

Fig. 2.20 Depth Dose Rate for 1.71-mm Diameter Sulfur Sphere

Fig. 2.21 Dose data obtained for a neutron-activated UC₂ sphere

2.3.4 Discussion

The two major advantages of the scintillation method are its great sensitivity and its yield of information concerning the absorbed energy spectrum. Failure to use the energy spectrum data to correct for nonlinear response can lead to errors on the order of 15 per cent for a typical beta energy spectrum and an anthracene scintillator.

It is important to make an accurate determination of the sensitive volume of each scintillator because this figure enters directly into the dose calculation. In the work reported here the scintillator thicknesses were measured with a Leitz microscope having a fine focus adjustment calibrated in microns and the diameters were determined with an eyepiece micrometer.

There are limitations in this approach to experimental dosimetry. The primary disadvantage involved with the use of NE-l02 is the lesser amount of light collected by the photocathode. Besides a lower inherent fluorescent efficiency than anthracene more light is lost at the interface of the scintillator and the lucite pipe. This is due to critical angle considerations resulting when the refractive index of the scintillator is greater than that of the light pipe. The refractive indices for NE-l02 and lucite are 1.58 and 1.50, respectively. Polystyrene, $\mu = 1.59$, would be a better choice of light pipe were it not for the much higher fluorescence response of polystyrene to ionizing particles. Anthracene has a higher specific fluorescence to ionizing particles and hence better energy resolution, but the disadvantages outweigh the advantages for this type of dosimetry. The quality of the crystals is variable, and the edges tend to crumble upon machining. Furthermore, the crystals must be joined to the lucite light pipes by an optical cement such as Canada balsam dissolved in a solvent. Both of these factors tend to make the accurate determination of the sensitive volume of the scintillator quite difficult.

In addition, the surface is subject to deterioration in an oxygen atmosphere, and sublimation in a vacuum changes the response characteristics and reduces the volume of the scintillator. Plastic scintillators do not have these disadvantages, and may be successfully used in a dosimetric experiment of this type provided care is taken in the construction of the assemblies, and a high quality phototube is used.

There is room for improvement in the scintillation-extrapolation technique and efforts are being directed toward this end. Closer tolerance is needed for machining the diameter of the scintillators, and more thicknesses are needed for each diameter in order to increase the reliability of the extrapolation curves and remove subjective judgments. We are investigating the difficulties involved in reducing the dimensions of the scintillators. These include calibration for thicknesses less than the range of the alpha particle and assessing the significance of the fraction of events resulting in an energy absorption of less than 1000 ev per incident-negaton. This is the approximate amount of energy which on the average results in the emission of one photoelectron from the photocathode of the phototube.⁽¹⁶⁾ The determination of this fraction of incident-negatons whose energy loss within the scintillator is not detected is a function of the emission spectrum, the dimensions of the scintillator and the source and the composition of the surrounding media. If no applicable theory is found, perhaps the problem is amenable to a computer solution using Monte Carlo calculations.

It is clear that the scintillation dosimetric method described is applicable to a wide range of problems. It has high sensitivity and is useful for estimating the absorbed dose to small volumes of tissue using sources with activities in the range of 0.05 to 500 μCi . The larger activities, however, are restricted to geometries which result in counting rates of less than 10^4 per second.

2.4 Comparison of Scintillation and Ionization Extrapolation Dosimetry.

Application of the ionization extrapolation chamber used in this laboratory is limited (for a 3-mm diameter collecting electrode) to radiation fields in excess of 50 rads/hr. Scintillation extrapolation is a factor of 10^3 more sensitive. Careful measurements with the conventional extrapolation chamber may be more precise, but electrical field distortion limits the electrode spacing to about 500 μ when one of the electrodes is a flexible mylar film. The scintillation method may be used with thicknesses down to 30 μ when using the total energy peak of a 5 Mev alpha source and as thin as 5 μ if the alpha peak observed is recognized as corresponding to a fraction of the energy absorbed. Limitations due to difficulties in measuring scintillator volumes and in calibration of response have been mentioned in section 2.3. In addition to greater sensitivity the chief assets of scintillation dosimetry are the information this technique provides on the spectral distribution of absorbed energy and better approximation by the organic scintillator to the dose delivered to biological material.

The relative agreement between the two methods is shown in Fig. 2.22. The solid circles are air ionization extrapolation measurements on a UC_2 particle 120 μ in diameter that had been irradiated for 10 minutes at a flux level of 5.8×10^{11} neutrons/cm²-sec. Electrode diameter was 11-mm so the dose rate shown is that delivered to a one square centimeter area. An absorbing layer of 7 mg/cm² mylar was placed between the particle and the sensitive volume. The open circles are anthracene measurements on a similar source 145 μ in diameter that had been irradiated also for 10 minutes but at a flux level three orders of magnitude lower. This was necessary to obtain an activity level suitable for use with the scintillation method. The data shown were made with an anthracene scintillator crystal 11-mm in diameter and 105 μ thick. A correction factor determined by the method described in section 2.3.3 was applied in order to correct to zero thickness. Data were normalized to a per fission basis for comparison.

Fig. 2.22 Comparison of the two dosimetric methods.

3.0 Source and Particle Fabrication

3.1 Production of Simulated Debris Particles

Although the nonuniform character of the core debris resulting from nuclear or ordnance type destruct tests is well documented,⁽¹⁹⁾⁽²⁰⁾⁽²¹⁾ a more orderly geometry is required for dosimetric studies. Small spheres of homogeneous composition, uniformly labeled throughout with radioactivity, have been fabricated in a size range 1-mm to 1-cm in diameter.

The reference fuel material to be simulated consists of pyrolytic carbon coated UC_2 beads distributed in a graphite matrix. This composition is maintained in the sources produced in the laboratory; the binding material has been changed however, to a thermosetting epoxy or plastic for ease of fabrication. Before addition of the binder the radioactivity is added, usually as the chloride solution, and thoroughly mixed in the dry formulation of powdered graphite and fuel beads. The epoxy (Ciba Araldite 502) or plastic (Turtox Embedding Plastic) is added in an amount sufficient to wet the mixture which is then placed in a mold and allowed to polymerize over a period of 24 hours. Upon removal from the mold the sources are given a thin coating of acrylic lacquer to prevent contamination of equipment and instruments.

One-half of the aluminum mold used to manufacture sources 1-cm in diameter and larger is shown in Fig. 3.1. A 1-cm particle can be seen in the lower left hand corner. Figure 3.2 shows a similar mold made of Teflon which is used to produce particles either 1- or 3-mm in diameter. Filling of the mold through sprue holes resulted in void spaces in the particles due to the high viscosity of the mix. To avoid this a technique was developed in which the hemispherical cavities on both mold sections were filled with the mixture and then the two parts brought together quickly and clamped using small bolts. Also shown with the Teflon mold are particles 1- and 3-mm in diameter.

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Fig. 3.1 Aluminum mold

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Fig. 3.2 Teflon Mold for Making 3mm and 1mm Particles

3.2 Sulfur - ^{32}P Sources

A thermal gradient column has been used to produce spherical particles used in the development of the plastic and anthracene extrapolation dosimeters. Sulfur spheres were formed by dropping molten sulfur into a column of glycerol 1-1/2 inches diameter by 36 inches long. The upper 12 inches of the glycerol was maintained at a temperature above 120° C by the use of resistance heaters. The drops of sulfur formed spheres ranging in size from .1 to 3- or 4-mm in diameter which solidified as they slowly sank into the cooler portion of the column. The feed material for this process was a high purity sulfur produced by four distillations in a nitrogen atmosphere of C.P. grade sulfur.

The resulting spheres were activated at the Oak Ridge Research Reactor in a neutron flux to produce ^{35}S and ^{32}P . Figure 3.3 shows the relative abundance of these two radionuclides after 20 minutes irradiation and the subsequent change in fractional activities with decay time. Sulfur-35 has a half life of 87.1 days with a maximum beta energy of 167 kev while Phosphorus-32 has a half-life of 14.3 days with a maximum beta energy of 1.7 Mev. Consequently, the shift to lower energy of the emission spectrum will be reflected in dose measurements.

Fig. 3.3 Fractional Activities in Neutron-Activated Sulfur.

3.3 Other

Work is continuing on the production of spherical particles using the air-driven grinder described in a previous report.⁽⁵⁾ Efforts are underway to modify this device (Fig. 3.4) so that it might be easily disassembled with remote manipulators. This would allow its use in hot-cells in the fabrication of small, highly radioactive spherical sources that are needed for dosimetric and radiobiological studies.

In one other development spheres of iron measuring tenths of millimeters in diameter have been coated with a mixture of ZnS-CdS and carnauba wax. The wax provides protection against the corrosive action of digestive juices when these particles are used in gastrointestinal transit time and wall-proximity studies. The phosphor fluoresces brightly under ultra-violet illumination allowing the rapid detection of these small spheres in excreta.

Fig. 3.4 Air Grinder

4.0 PARTICLES ON SKIN

4.1 Deposition of Particles

A complete analysis of the hazards arising from the accidental fragmentation of nuclear reactors or isotopic power sources requires a knowledge of the deposition and adhesion of radioactive debris on the skin. In addition to information on radiobiological effects and on the dosimetry of small particles some estimate of source-skin contact probability is needed. A study has been initiated to determine the degree of initial retention on the arm of particles falling vertically in a uniform fallout field.

Figure 4.1 shows the device designed and constructed to give such a distribution pattern. The closed copper chamber is cylindrical measuring 6.5 centimeters in diameter and 4.5 centimeters high with two 1-centimeter ports. Perforations in the base-plate are made with a #52 twist drill (.0635" dia.) and are in a hexagonal array with 1-centimeter spacing. A double-faced sticky tape is applied to this plate, small holes punched in the tape at the 36 orifices and Eaton-Dikeman #953 sharkskin filter paper placed over the tape. Suction is applied to the top port providing an air flow of 66 linear feet per minute. This assures a face velocity sufficient to retain a wax particle up to 500 microns in diameter at each orifice.

After loading, the chamber is positioned above the forearm with the aid of small plumb-bobs in a draft-free area. The particles are released by simultaneously interrupting the vacuum, instituting a momentary back-flow in the side port, and mechanically vibrating the chamber. Particle deposition is observed to be uniform for a drop of several centimeters under normal room conditions.

In the experiments the device is held at a height that will assure the particles have attained their terminal velocity before impaction on the arm. For unit density particles of 140 micron diameter, this is on the order of a few centimeters.

Preliminary results show that initial deposition is dependent upon particle size and skin surface moisture. In the size range of 297-500 μ from 6.2 to 44 per cent of the particles impacting on arms of average dryness were retained long enough to observe them (≥ 1 min). In one trial sweating was induced before exposure by

Fig. 4.1 Particle Dispersion Device

enclosing the arm in a polyethylene envelope. Under these conditions, simulating natural occurrence of perspiration, retention values approached 100% for 104-125 spheres. Only 55% were retained by this individual under normal conditions using this size particle.

4.2 Retention of Particles

Hazards arising from the deposition of micron-size particles on the skin are proportional to the residence time of these sources. Efforts are continuing to determine the variation of this time as a function of particle size and density.

4.2.1 Wax Spheres

Earlier studies using spherical particles of carnauba wax showed an inverse relationship between particle diameter and retention time. These data are shown in Fig. 4.2 where the vertical lines are standard deviations of the retention times and the horizontal lines indicate the size range of the particles used. These points have been fitted (at the U.S. Naval Radiological Defense Laboratory) with the empirical function

$$T = 6.0 D^{-0.16} - 1.1 \times 10^5 D^{-2.65}$$

where

T = retention time (hours)

D = particle diameter (microns)

Although some particles are entrapped by hairs, adhesion is due primarily to capillary forces acting between particles and the surface layer of skin oils and moisture. The nature of this layer varies widely because of the environmental and physiological differences between individuals.

4.2.2 Depleted Fuel Beads

Trial runs were made with simulated reactor debris on the forearms of three subjects. Depleted uranium-dicarbide spheres, carbon coated were dropped onto the arm from a height of a few centimeters, the subjects engaged in routine laboratory or clerical activity and periodic counts were made of the rate of loss of

Fig. 4.2 Particle Retention on Skin

particles. Low magnification was required to observe these 230 micron diameter spheres among the hairs of the arm. Kraft paper was taped onto the floor and lower edge of walls of the room in which these experiments took place to assure recovery of all the test material even though the radiotoxicity of the depleted uranium is negligible. Figure 4.3 shows the observed wide range of retention times which was not unexpected. Subjects TGC and BRF are moderately hirsute with hair of the arm measuring from 2.5 to 3.0 cm long and quite dense (~ 20 hairs/cm²). Under low magnification the surface of the skin appears moist. Subject JLT's arm is almost devoid of hairs (12 per cm²) which are fine and short (.68 cm): the surface appears somewhat dryer. Numerical integration of the areas under the curves give a range of average particle retention from 0.73 to 6.9 hours.

4.3 Skin Decontamination

A series of tests was carried out on one individual to determine the effectiveness of simple countermeasures in removing small spherical particles. Experiments were conducted in an airconditioned laboratory where the temperature varied from 68° to 70° F and the relative humidity was 60%. Test material consisted of fluorescent wax spheres from 100 to 1,000 microns in diameter. From 50 to 170 of these were dropped onto the surface of the left arm and counts made under ultraviolet light before and after the decontamination process. These trials were carried out "blind," that is, under standard laboratory illumination. During brushing, etc., the particles were not visible to the experimenter and no conscious effort was made to restrict the removal forces to the test contaminants.

Three techniques were used: (1) Brushing with the right hand for a short time (less than 5 seconds); (2) Rinsing under running tap water for a few seconds; and (3) Washing with soap and water with the right hand and rinsing off under running water. Results are shown in Table 4.1. Brushing would not be quite so effective were the skin moist or oily. It can be seen that decontamination of the arm is a natural consequence of simple sanitary habits.

Fig. 4.3 Retention of 230-Micron Diameter UC₂ Beads on the Arm.

Table 4.1

Decontamination of Particulate Contamination* on Skin

Particle Diameter (microns)	Per Cent Removed		
	Brushing	Rinsing	Washing
500 - 1000	100	100	100
297 - 500	100	100	100
250 - 297	97	100	100
210 - 250	95	100	100
179 - 210	98	100	100
125 - 149	92	100	100
104 - 125	89	98	100
88 - 104	86	97	97
(1 - 2)**	.1	.1	99

* Fluorescent - carnauba wax spheres

** Zinc sulfide powder.

5.0 GASTROINTESTINAL PASSAGE TIME

Recent studies on humans of passage time through the gastrointestinal tract (GIT) of small insoluble particles indicated a narrow range of values (.5 to 4 days) when the ingested tracer material consisted of only three particles.⁽²²⁾ Other investigators, however, report that although the average time of passage falls in a fairly predictable time interval that the residence time in the GIT of any one particle cannot be foretold and may range from a few hours to several weeks.⁽²³⁾⁽²⁴⁾⁽²⁵⁾⁽²⁶⁾ An analysis of the hazards arising from the accidental ingestion of small, highly radioactive particles must take into consideration such deviations from the normal. Damage to the stomach wall or intestinal mucosa depends on the total radiation dose received by a unit of tissue, which in turn is a function of particle location within the tract and the total residence time of this source. Age, diet, and stress are factors affecting the rate of passage while particle density has been reported to be either of little consequence⁽²²⁾ or of major importance.⁽²³⁾

Some studies have been made during this report period on the passage times in rats and man of small, insoluble particles. Rats were dosed by gavage with fluorescent wax particles 200 μ and 300 μ in diameter and with 300 μ iron spheres. In the dosing procedure from 45-100 spherical particles were placed in ~.5 ml of water or corn syrup and the slurry drawn up into the lumen of a stomach tube. A filter was used between the tube and syringe to prevent the particles from entering the latter and a plug made of a food-water mash was inserted in the end of the stomach tube to avoid premature ejection of the test material. After insertion into the stomach of the rat the tube was flushed out with 0.5 ml of water. Feces were collected on a modified chart recorder moving at 2-inches per hour. No particles were recovered after the fifth day, however, collections continued for as long as two weeks since a quantitative recovery of all beads was desired.

Fluorescent particles were recovered from the feces by sieving the excreta under ultraviolet illumination. Iron spheres were separated from the bulk of the

material in two stages, first gravimetrically and second magnetically. They were counted under stereo-magnification in order to differentiate them from metallic fragments of food and cage material.

Results of the experiments are shown in Figs. 5.1 and 5.2. Additional data will be required to determine if the small differences seen are statistically significant.

Two gelatin capsules were each loaded with 200 inert fluorescent wax particles and ingested by two human volunteers. The particles were spheres in the size range 500 to 595 micron diameter and with a density of 1.22 grams per cm³.

Excreta collections were made over a period of eight days. Recovery and counting of the beads was made by sieving under ultraviolet light. This technique is somewhat tedious and a quantitative recovery of the test material was not made. Figure 5.3 illustrates the data obtained and indicates that a holdup of several days in the GIT, even for low specific gravity material should be expected.

In addition, the passage time of highly radioactive particles may differ from that of test materials used in humans up to this time. Entrapment of particles in radiation-induced ulcers has been observed in the pig⁽²⁷⁾ and the change in intestinal motility due to the radiation insult must be considered.

One brief study using rats indicated no correlation between the radioactivity levels of ingested particles and retention time.⁽²⁸⁾ A collection of animal data should be made to clarify this aspect of the "hot particle" problem.

Fig. 5.1 Excretion Patterns of Insoluble Particles in Rat.

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Fig. 5.2 Passage Time of Insoluble Particles through GIT of Rat.

Fig. 5.3 Differential and Cumulative Excretion Patterns in Humans
using 500 - 595 μ Wax Spheres.

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7.0 TRAVEL*

<u>Date (1965)</u>	<u>Location and Purpose</u>	<u>Traveler</u>
Sept 13-17	Fiuggi, Italy. Meeting of International Commission on Radiological Protection.	KZM
Sept 22-28	Rome, Italy. International Congress of Radiology (11th meeting)	KZM
Sept 24	Rome, Italy. Symposium on Radiation Exposure of Human Population	KZM
Sept 28-Oct 1	Cambridge, England. Second International Symposium on Inhaled Particles and Vapors.	BRF
Sept 30 -Oct 1	Gatlinburg, Tennessee. Tennessee Valley Industrial Health Conference.	TGC WSS
Oct 7-8	Albuquerque, N.M. 11th Annual Bioassay and Analytical Chemistry Meeting	WSS
Nov 1 - 3	Charlottesville, Va. Southeastern Section Meeting, American Physical Society.	KZM
Nov 3-5	Chicago, Ill. First AEC Symposium on Accelerator Radiation Dosimetry Experience (Argonne National Laboratory)	WSS
Nov 15-16	Washington, D. C. American Nuclear Society	TGC
Dec 13-15	Paris, France. International Conference on Radiological Protection in Industrial Uses of Radioisotopes.	KZM WSS
Dec 16-17	Paris, France. IRPA Provisional Executive Council Meeting.	KZM
(1966)		
Feb 7- 8	Berkeley, Calif. USAEC BioMedical Directors Meeting (University of California)	KZM
Feb 9-10	Cleveland, O. Space Review Committee Meeting for NERVA2 Program(NASA)Lewis Space Flight Center.	BRF TGC
Feb 16-18	Cleveland,O. Wm. Voelker, Western Reserve (Discussions concerning dosimetry equipment)	WHW
	Winnipeg, Canada. Nuclear Enterprises, Ltd. (Discussions concerning dosimetry equipment)	WHW

7.0 TRAVEL* (continued)

1966

Mar 17	Washington, D. C. NCRP Annual Meeting	KZM
Mar 18	Washington, D. C. NCRP Subcommittee 23	WSS
June 24	Strontium and Cesium Dosimetry.	
Mar 25	Washington, D. C. Advisory Committee on Civil Defense.	KZM
Mar 29-Apr 5	Houston, Texas. 5th Annual Technical Meeting of American Association for Contamination Control. Albuquerque, N.M. M.J. Fulwyler, Sandia Corporation (Discussions concerning dosimetry problems) Menlo, Calif. R. E. Yoder, Lawrence Radiation Laboratory (Discussions concerning dosimetry problems.	WHW
April 6	Nashville, Tennessee. Meetings with dermatologist	TGC
May 26	and Tennessee Department of Corrections on	BRF
June 21	Biological Effects of β -rays on Skin.	
April 19-20	Germantown, Md. Meeting on Aerospace Safety of SNPO Devices Washington, D.C. M. Goldman, Nuclear Utilities Services, Inc. (Discussions concerning dose rate in tissue from reactor debris particles.	BRF RHB
April 20	Germantown, Md. Strontium-90 Space Study.	WSS
May 16-20	Pittsburgh, Pa. Annual Conference of American Industrial Hygiene Association.	WSS
June 18-19	Sterling Forest (New York). IRPA Provisional Executive Council.	WSS KZM
June 27-30	Houston, Texas. 11th Annual Health Physics Society Meeting.	KZM WSS

 *KZM, K.Z. Morgan; WSS, W.S. Snyder; TGC, T. G. Clark; RHB, R. H. Boyett; WHW, W. H. Wilkie; BRF, B. R. Fish.

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