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ENVIRONMENTAL STUDIES: RADIOLOGICAL SIGNIFICANCE OF NUCLEAR ROCKET DEBRIS

SEMI-ANNUAL PROGRESS REPORT
FOR
PERIOD JANUARY 1 - JUNE 30, 1965

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HEALTH PHYSICS DIVISION

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JANUARY 1966

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For Space Nuclear Propulsion Office, United States Atomic
Energy Commission, National Aeronautics and Space Administration.

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
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ENVIRONMENTAL STUDIES: RADIOLOGICAL SIGNIFICANCE
OF NUCLEAR ROCKET DEBRIS

I INTRODUCTION

Radiological safety studies are being conducted for the Space Nuclear Propulsion Office, Flight Safety Branch. This report for the period January 1 through June 30, 1965, outlines the status of our work related to the significance of radioactive debris which may be deposited on the earth following an accidental or intentional fragmentation of a nuclear rocket in space. An assessment of the radiological hazards arising from the exposure of skin or gastrointestinal tract to reactor debris particles is being continued. Emphasis of this program is on the production, and behavior of micron size inert particles, and on the dosimetry of similar particles that have been made radioactive by spiking with radioisotopes or by reactor irradiation.

A computer program has been used to estimate the dose rates from 100 μ and 1000 μ diameter reactor core fragments as a function of time following a typical operational sequence of a NERVA engine. Dose rates averaged over 1 cm² at 7 mg/cm² depth, and surface dose rates were computed for times from 6 hours to 10 years after shutdown.

Tissue depth dose rates from activated fuel particles were measured with an extrapolation chamber as a function of decay times from 6 to 28 days after neutron irradiation. As a further aid in the determination of the spatial distribution of β dose in tissue, an empirical dose model has been formulated to approximate any arbitrary beta spectrum by a linear combination of the dose data of a few known beta-active isotopes for which depth doses and emission spectra have been cataloged for several standard source geometries. Preliminary work has been completed on the approximation of the spectra of a fission product mixture by the use of ⁴⁵Ca and ⁸⁹Sr.

A novel technique employing small radioactive particles adhered to an intragastric balloon was developed for the in vivo irradiation of small areas of the gastric mucosa. Simulated reactor fuel particles, spiked with ^{89}Sr , have been fabricated, monitored to determine tissue depth dose and attached to a balloon. This apparatus was used by Battelle Northwest in effects studies using miniature swine.

Further developments were made on the production of small spherical particles to be used in gastrointestinal transit time and skin retention studies. Inactive spherical particles, tagged with ultraviolet phosphor, were fabricated in the 50 to 1000 μ size range using a thermal gradient technique and air driven grinders. Particle density was varied from 1.2 to 5 g/cm³ by the addition of metallic powders to carnauba wax which is spheroidized in a thermal column.

2.0 COMPUTER PROGRAM

Pending the availability of computer programs being developed at other establishments, we are continuing to use the ZAP code to estimate tissue depth doses from reactor debris. Calculated dose rates are shown in Fig. 2.1 for 100 μ and 1000 μ diameter fragments of a NERVA reactor core operated nine minutes at a power level of 1120 MW. Information on spectra and activity levels were obtained from Westinghouse Astronuclear Laboratories.¹

Neither contained activity, surface dose rate nor a combination of these two parameters sufficiently characterize the biological hazards of this material. A thousand-fold difference in volume of the two particles is offset somewhat by the greater concentration of uranium in the 100 μ bead so that the larger particle contains approximately thirty times the activity of the smaller. Still, because of differences in geometry and self-absorption within the source matrix, the surface dose rate from the 100 μ bead is a factor of three or four times greater than that of the 1000 μ particle. However, tissue dose rates averaged over 1 cm² area at a depth of 7.0 mg/cm² are proportional to contained activity, the 1000 μ particle delivering an order of magnitude greater dose at this depth.

Fig. 2.1 Computer estimates of dose rates from reactor debris.

ZAP has been used also to estimate the degradation of beta spectra and the production of an electromagnetic spectrum (Bremsstrahlung) from various isotopes contained in high-Z spherical sources. The derived spectra for a 1 cm diameter reactor fuel source containing ^{89}Sr are shown in Fig. 2.2. Self-absorption within the source was estimated by ZAP to be 71.7%. ORNL TM-1160³ gives FORTRAN listings and flow charts for ZAP31 as written for the CDC-1604A computer system.

3.0 DOSIMETRY TECHNIQUES

3.1 Introduction

The significance of beta radiation dosimetry of nuclear rocket debris particles has been discussed in previous reports.^{4,5,6} Two considerations are important: First, the spatial distribution of absorbed energy and its relation to available radiobiological data; and the comparison of absorbed doses with radiological safety control criteria. This section summarizes our current efforts to provide adequate experimental dose data for these purposes using simulated and real reactor fuel particles.

3.2 Empirical Dose Model for Reactor Debris Particles

Determination of radiation dose to tissue from a discrete β -emitting source is a notoriously difficult problem. The most successful and widely applicable approach to date is an empirical treatment due to Loevinger.⁷ Loevinger dealt with the homogeneous case in which the radioactive material is uniformly distributed in a volume of tissue and where the activity, absorption and backscatter media are assumed to be of unit density. This case is applicable to a wide variety of problems, but differs significantly from the situation in which a high-Z radioactive source is in contact with tissue (e.g., a neutron irradiated UC_2 particle on the skin). These two cases are contrasted in Fig. 3.1.

This section describes preliminary results of spectrum and radiation dose measurements which we have initiated to provide a basis for an empirical

Fig. 2.2 Degradation of β energy spectrum and production of Bremsstrahlung in a high-Z source.

DOSE ? → ← SENSITIVE VOLUME

HOMOGENEOUS CASE

DOSE ? → ← SENSITIVE VOLUME

INHOMOGENEOUS CASE

Fig. 3.1 Comparison of homogeneous and inhomogeneous source-tissue geometries.

treatment suggested by us for application to the inhomogeneous case.⁸

These data are specific for the case of 1 cm diameter sources composed of graphite containing 17 wt % of uranium; however, additional measurements will be made using UC_2 sources of various sizes. The principal is that of approximating the spectrum for which a dose estimate is desired by superposition of dose and spectra data from a library of known measurements made using the desired geometry and source composition. Obviously, application of the technique is not limited to UC_2 and, in principle, could be used for other compositions including particles of activated structural materials.

The spectra shown in Fig. 3.2 were obtained from two 1 cm diameter sources of UC_2 plus graphite, one of which contained ^{45}Ca and the other contained ^{89}Sr (plus 3% $^{90}Sr + ^{90}Y$ contaminants). These emission spectra were measured with a 1/4" thick x 1/2" diameter anthracene crystal, under 100 μ Hg vacuum. Surface and depth dose measurements were made on the 1 cm diameter particles with an extrapolation chamber using a 3 mm diameter collecting electrode.

An example of this approach is shown in Fig. 3.3. A linear combination of the spectra from two known sources is compared with that of a reactor fuel particle. Obviously, a closer simulation could be made by the use of spectra from additional radionuclides having intermediate energies or energies higher than that of ^{89}Sr .

For the example of Fig. 3.3 it was necessary to add 27% of the standard ^{45}Ca spectrum and 3% of the standard ^{89}Sr spectrum to approximate the spectrum from the reactor fuel particle. Thus an estimate of the dose rate from the reactor particle can be obtained by adding the same fractions of the measured dose rates obtained from the two standard sources. Dose rate estimates as a function of depth in tissue are shown in Fig. 3.4. This approach is adaptable to automatic curve fitting on a computer or to simplified techniques of hand calculation, e.g., using energy bands and solving a set of simultaneous linear equations.

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Fig. 3.2 Spectra of ^{89}Sr and ^{45}Ca as emitted from a high-Z source.

ORNL-DWG. 65-5730R

Fig. 3.3 Simulation of a reactor particle spectrum using ^{89}Sr and ^{45}Ca .

Fig. 3.4 Use of beta dosimetry model to estimate depth dose rates from a mixed fission products source.

4.0 SIMULATED REACTOR DEBRIS SPIKED WITH ^{89}Sr .

4.1 Introduction

A cooperative program with Battelle Northwest Laboratories on the radiobiological effects of simulated reactor debris was initiated during this report period.

Ideally, particles used in the initial phase of these studies would be a maximum of 1.0 mm in diameter, and contain a single high energy beta emitter of relatively long half-life and high specific activity, in a matrix similar to NERVA reactor debris. The radioactivity should be homogeneously distributed throughout the source material and remain immune to dissolution by body fluids or abrading by normal handling. Most of these conditions were met using the fabrication techniques outlined below.

ORNL assumed the responsibility for the production and dosimetry of a group of 1.0 mm diameter particles containing ^{89}Sr , and in addition, designed and prepared a source applicator to be used by Battelle Northwest in the irradiation of the gastric mucosa of miniature swine. These sources also were used in skin irradiation experiments with swine and were given by gastric intubation to rats, although the leaching out of the ^{89}Sr by intestinal fluids in the latter case made an estimation of mucosal dose quite difficult.⁹ Results of the radiobiological studies will be reported by Battelle Northwest.

4.2 Production and Particle Description

Fabrication techniques and source production were carried out by Union Carbide Nuclear Division, Development Engineering Group, Y-12 Plant, Oak Ridge.

4.2.1 Process Description - The one millimeter spherical sources were prepared in the following steps:

(1) A fine graphite flour containing 85wt% graphite and 15wt% carbon black was blended with UO_2 powder so that the blend contained 45wt% UO_2 . A small amount of this blend (Table 4.1) was placed on a watch glass and

Table 4.1

Data for 1. mm Diameter Sources

Desired mc/source	0.7	0.35	0.07
Wt graphite- UO_2 (g)	0.0375	0.0440	0.0440
^{89}Sr added (mc)	51.	30.	6.
Volume of ^{89}Sr (ml)	2.4	1.4	0.3
Varcum added (g)	0.0179	0.0215	0.0210
Wt of sources before curing (g)	0.0515	0.0596	0.0688
Wt of sources after curing (g)	0.0465	0.0535	0.0622
Wt of round sources (g)	0.0301	0.0317	0.0398
Number of round sources	28	30	33
^{89}Sr as of March 16, 1965 (mc/source)	1.04	0.496	0.104
^{90}Sr (mc/source)	.042	.021	.0042

^{89}Sr as an aqueous chloride solution (Table 4.2) was added to the dry mixed powders and the water was evaporated.

(2) The dried graphite- UO_2 powder mixture, containing the ^{89}Sr isotope, was placed in the head of an 18 gauge, blunt end hypodermic needle which had been shielded with lead foil (Fig. 4.1). Furfural alcohol binder (Varcum 8251), polymerized with maleic anhydride, was added so that the mixture contained 33wt% binder. The formulation was mixed using the shielded stirring rod shown in Fig. 4.1 and extruded using the same rod. To form each source, a small amount of material was extruded past the end of the needle, cut off with a razor blade, and allowed to fall into a pan containing dry UO_2 -graphite powder. This powder served to coat the particles and thereby prevented sticking.

After extrusion of a group of beads, the dry powder was removed by screening, and the particles were rounded by gently rolling them with a circular motion between a strip of fine emery cloth and a soft rubber pad. The particles were then placed in a glass dish, covered with aluminum foil, and cured in a muffle furnace which attained a maximum temperature of 260°C gradually in a 26-hour heating period. Cured sources were spray coated with a thin acrylic resin to prevent contamination of shipping container and handling equipment.

Thirty preliminary batches of sources, in which ^{89}Sr was omitted, were prepared in order to develop the techniques of producing small spherical sources of the proper uranium content and of the proper ^{89}Sr content in each source. In addition, considerable effort was expended in finding methods of reducing process losses so that a minimum quantity of radioactivity would be required. At the beginning of the work 228 mc of ^{89}Sr would have been necessary to fabricate the sources using existing techniques. After the development work, 96 mc ^{89}Sr were required.

Table 4.2

Description of Isotope
 SrCl_2 in HCl Solution.

Concentration	21 mc/ml
Normality	0.94 HCl
Radiochemical Purity	> 99 % (exclusive of ^{90}Sr and ^{90}Y)
^{90}Sr Content	~0.86 mc/ml
^{141}Ce , ^{144}Ce	$< 2.3 \times 10^{-4}$ mc/ml
^{140}Ba	$< 5.4 \times 10^{-5}$ mc/ml
Heavy Metals	< 10 ppm
Total Solids	2.0 mg/ml

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Fig. 4.1 Apparatus for extrusion of graphite-uranium - ^{89}Sr , one millimeter sources. Lead shielded plunger is shown at left, shielded hypodermic needle at top.

Data pertinent to the production of the three activity levels of sources are given in Table 4.1. Figure 4.2 shows several 1 mm spheres from preliminary batches, while Fig. 4.3 shows a cross-section of one of these. The small white flecks visible in Fig. 4.4 are UO_2 particles which illustrate the uniformity of distribution in the graphite. Chemical analysis of sources made without ^{89}Sr indicate a uranium content of 0.1712 grams U per gram of cured source. Using a value of 1.7 g/cm^3 as the average density of the graphite (as determined by microscope measurements and weighing of a large number of sources), an approximate value of 290 mg U per cm^3 was obtained.

One-centimeter diameter sources for corollary dosimetric studies were prepared using split hemispherical plaster molds, sized so that the green graphite compact would shrink to 1 cm diameter after curing. The ^{89}Sr was dried on a layer of the graphite- UO_2 blend, the binder was added, and a predetermined weight of this mixture was pressed in the mold. The spheres were cured at 260°C in the mold. After curing, the equatorial flash was filed away and the sphere was spray coated in a manner similar to the 1 mm sources. Table 4.3 lists the data pertinent to the preparation of these sources.

4.2.2 Health Physics Experience

Hand exposure to the operator during preparation of these sources was approximately 50 rem/hand. This exposure exceeded the AEC Radiation Protection Standard of 25 rem/calendar quarter, but was within the allowable 75 rem/year. The exposure was considerably more than had been expected from the extrapolation of radiation exposures experienced in previous preparations of this type. In addition, it was found that the activity levels of dry material were much greater than the wet formulation activity, which resulted in unexpectedly high exposure levels during the dry steps of processing. Should further work be carried out, maintenance of constant beta shielding of the hands would be well advised. The shielding provided by various materials is listed in Table 4.4.

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Fig. 4.2 Uranium-graphite-binder after extrusion, rounding and polymerizing.

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Fig. 4.3 Polished cross-section of 1 mm diameter simulated reactor debris particle (100X).

Fig. 4.4 Polished cross-section of particle showing UO_2 dispersion in graphite matrix (X250).

Table 4.3

Data for 1. cm Diameter Sources

Source Number	1	2
Desired mc/source	3.0	3.0
UO ₂ (wt% of graphite)	42.0	42.0
Dry UO ₂ -Graphite (g)	1.3198	1.3198
Varcum added (g)	0.3828	0.3828
Wt finished sources	1.3878	1.3428
Polar diameter (cm)	1.049	1.053
Equatorial diameter (cm)	1.041	1.035
⁸⁹ Sr content (Mar. 25, 1965) (mc/source)	3.52	3.48
β-γ Activity at 3" in air March 25, 1965 (mr/hr)	2000	2000

Table 4.4

Shielding Provided by Various Materials

Activity of 23, 1. mm diameter sources containing 1.04 mc ^{89}Sr /source		
Distance from Source (inches)	Shield Material	(1) mr/hr
3	None (air)	280
3	3 mils paper	250
3	1/8 inch lead	1
3	20 mils lead	120
3	1/2 inch plastic	10

(1) Measurements made with a paper shell Cutie Pie.

4.2.3 Variability of Activity

To check the homogeneity of ^{89}Sr distribution both among and between batches, 26 of the sources were counted using an 8" x 4" NaI crystal and a Nuclear Data 512 channel analyzer. Individual sources were counted in glass vials, 10, 20 and 30 inches from the face of the crystal with corrections made for the different geometries. Although a slight variation between vials was seen, it was decided, in the interest of radiological safety, to forego the multiple transfers that would have been required for a common counting vial. The results are given in Table 4.5 expressed as counts per minute per microcurie of activity, corrected to a 10-inch source-to-detector geometry. Analysis of all values gives

$$n = 26 \text{ sources}$$

$$\text{avg. activity} = 1274 \text{ counts/min per } \mu\text{c}$$

$$\text{standard deviation} \pm 342 \text{ counts/min}$$

Rejecting the obvious extraneous values, marked with an asterisk, gives:

$$n = 21$$

$$\text{avg. activity} = 1237 \text{ counts/min per } \mu\text{c}$$

$$\text{standard deviation} \pm 133 \text{ counts/min}$$

A typical spectrum is shown in Fig. 4.5.

4.3 Dose Rate Determinations

An extrapolation chamber was used to estimate dose rates from the 1. mm diameter ^{89}Sr -spiked sources. The sources contained 868 μc ^{89}Sr on 1 April 1965, and 736 μc ^{89}Sr on 12 April 1965, plus 41.2 μc ^{90}Sr + ^{90}Y (total of 82.4 μc) indicating about 5% each of ^{90}Sr + ^{90}Y contaminants in the original isotope. Table 4.6 shows the source number and dose data which have been corrected for the stopping power ratio.¹⁰ All the dose figures were calculated from the slope of the linear portion of the average ionization curve and are believed to be no greater than 8 - 10% in error including corrections for standard temperature and pressure. A 3 mm diameter collecting electrode was used,

Table 4.5

Check of Source Uniformity by γ -Counting

Activity (μc)	Source No.	counts/min per μc
52.9	.07-1-1	1417
	-2	1404
	-3	1414
	-4	1088
	-5	1341
	-6	1257
52.9	.07-2-1	1436
	-2	1210
	-3	1076
	-4	929*
	-5	1289
242.	.35-1-1	1466
	-2	1080
	-3	1284
	-4	872*
	-5	1242
	-6	1076
242.	.35-2-1	1176
	-2	922*
	-3	2656*
	-4	1191
	-5	1144
529.	.7-1-2	1128
	-3	1760*
	-4	1176
	-5	1086

*

See text page 22 (4.2.3).

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Fig. 4.5 Typical spectrum from high -Z source containing $^{89}\text{Sr} + ^{90}\text{(Sr+Y)}$.

Table 4.6

Dosimetry of 1. mm, ^{89}Sr -Spiked, Reactor Fuel Sources

Extrapolation Chamber Measurements*

Source Number	Tissue Depth (Mg/cm ²)	Dose Rate (Rad/hr)**	Date of Readings
0.7 - 1 - 1	2.54	13,800	3/26/65
	7.62	13,600	3/26/65
0.7 - 1 - 2	2.54	13,400	4/6/65
	7.62	12,300	
	12.7	10,700	4/1/65
	23.3	7,900	
	52.07	4,400	
	300.0	380	
0.7 - 1 - 3	2.54	14,200	4/6/65
	7.62	12,800	
0.7 - 1 - 4	2.54	13,000	4/8/65
0.7 - 1 - 5	2.54	12,700	4/8/65
	7.62	12,500	

* Dose rate measurements taken with a 3 mm diameter collecting electrode.

** To $\pm 10\%$ and corrected for stopping power ratio.

thus the data do not represent center line dose rates from the particles. One would expect the center line dose rate to be much higher than indicated by these data, especially at the surface of the particle. Center line dose and the 3 mm diameter average dose should approach the same value at greater depths (on the order of 300 mg/cm^2). Particle No. 2 was the only particle for which depth dose measurements were carried beyond 7.62 mg/cm^2 . As shown in Table 4.7, it is difficult to normalize the dose data to dates different from the time of measurement because of the $^{90}\text{Sr} + ^{90}\text{Y}$ contaminants. Six particles were exposed to duPont Type 834 film in order to get an estimate of the uniformity of particle surface dose. These films indicated a symmetrical distribution of activity within each particle and fair uniformity of surface dose between particles.

4.4 Beta and Gamma Spectra

Beta and gamma spectra were obtained using solid-crystal scintillation techniques. Gamma spectra were detected with a sodium iodide crystal 8" dia x 4" thick, and a Nuclear Data, Model ND-130, 512 channel analyzer. Data in the form of counts per channel were recorded on punched paper tape and typewriter.

Beta spectra were analyzed using an anthracene crystal $1/2$ " dia x $1/4$ " thick mounted on a low-noise phototube having a photocathode one inch in diameter. The crystal assembly was enclosed in a vacuum chamber, and the particles were placed 10 to 35 cm from the crystal depending on their activity. Pressure during the measurements was maintained at less than 0.1 torr to eliminate scatter by the air, and lucite collimators also were used to help prevent spectral distortion due to scattering. The pulse height spectra were analyzed with a 512 channel analyzer.

4.5 Source Applicator Preparation

By using radioactive sources secured to the outside of intracavitary latex bags, small areas of the mucosal lining of esophagus, stomach or distal end of

Table 4.7

Particle Dosimetry - Extrapolation Chamber Measurements

Date	Particle Number	Dose Rate (rad/hr)	Depth * (mg/cm ²)	Normalized to 8 Apr 1965	Average (rad/hr) 8 Apr 1965
3/26/65	0.7-1-1	13,800	2.54	11,600**	
4/6/65	0.7-1-2	13,400	"	13,100	
4/6/65	0.7-1-3	14,200	"	13,500	13,100 ± 330
4/8/65	0.7-1-4	13,000	"	13,000	
4/8/65	0.7-1-5	12,700	"	12,700	

* A similar table can be constructed for the 7.62 mg/cm² depth.

** Not included in average.

gastrointestinal tract can be irradiated without surgery. A pattern of placement of the particles should aid in the correlation of irradiated area and biological damage. This laboratory has devised a technique for securing sources containing millicurie amounts of a beta emitter to gastric balloons (Supplied by Pioneer Rubber Company, Willard, Ohio). The procedure involved a minimum of exposure to personnel.

A thin sheet (.025") of latex rubber is cemented over the sources to hold them in position. Some compromise was made between security of attachment and the thickness of the absorbing layer thus interposed between tissue and particle.

Initially, the balloon was partially inflated and six equally spaced marks were made around its greatest equatorial diameter, Fig. 4.6. Cold patch cement (Western Auto No. A-4523) was spread over a 1.5 x 1.5 cm area at each site and a source placed on each mark. A pair of modified sponge forceps was then used to stretch the latex rubber cover over the source, Fig. 4.7. After the cement had dried the forceps were removed and excess covering material trimmed.

All operations were carried out in a standard chemical hood equipped with filtered exhaust. Smears of the work area and source handling instruments showed no detectable contamination. Dose rates of 10 rad/hr two inches from the balloon were measured with a paper shell Cutie Pie when all six sources were in place. Film finger badges were worn during source manipulation; readings from these indicated that the exposures to the hands were less than 200 mrad.

Fig. 4.6 Modified sponge forceps (top) used for attachment of beta sources to intragastric balloon (bottom).

Fig. 4.7 Method of attaching ^{89}Sr sources to intragastric balloon.

5.0 PRODUCTION OF SPHERICAL PARTICLES

5.1 Thermal Gradient Columns

A previous report⁶ describes the techniques by which nonradioactive spherical particles of carnauba wax tagged with fluorescent chemicals were produced by dropping the mixture into a 72-inch long water column with a thermal gradient of 87°C to 25°C.

By the addition of WS_2 to a wax-ZnCdS(Ag) mixture, spherical particles of 177 to 1000 microns diameter and having a bulk density of 5. gms/cm³ were produced which fluoresced brightly under ultraviolet light. The surface of these particles had a "whiskered" appearance, Fig. 5.1, but could be polished in the air grinder described in Section 5.2.

Spheres of high purity sulfur ranging from 200 to 2000 microns in diameter were produced by a similar technique in a 24-inch column of glycerin.

5.2 Air Powered Grinder

An air driven grinder was built as a modification of a design described by Bond.¹¹ Grinding and polishing of irregular shaped particles into spheres requires from a few minutes for graphite or amorphous carbon to several hours for metallic zinc or $CuSO_4 \cdot 5H_2O$ crystals. Finished sizes range from 20 to 1000 microns in diameter. Figure 5.2 shows an exploded view of one of the grinders designed to spheroidize and classify radioactive particles. Air under 40 lbs/in² pressure is introduced tangentially into the sample chamber which is lined with an abrasive of the desired coarseness. The rough particles spin rapidly about their axes of greatest moment of inertia abrading off the material most distant from the centroids. When the resulting spheres become smaller than the classifying screen they drop into the smooth surfaced collection chamber. Dust and fines are blown out through the outlet pipe and trapped on an absolute filter. A screen prevents the loss of large particles through this route. This grinder is designed to be operated remotely in a hot cell and is disposable since the costs of materials and fabrication are quite low.

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Fig. 5.1 Carnauba wax spheroids containing ultraviolet phosphors and WS_2 .

Fig. 5.2 Exploded view of air-driven grinder.

Figures 5.3 and 5.4 show examples of small spheres that have been produced by the air driven grinders.

6.0 IRRADIATED REACTOR PARTICLES

6.1 Description

Two hundred nineteen particles containing enriched uranium and simulating reactor fuel material were obtained from the Development Engineering Division, Y-12 Plant. These are spherical particles 1.0 mm in diameter and were fabricated by extrusion of a carbon-Varcum-uranium mixture similar to the process described in Section 4.2. X-ray plates were made of all sources showing the distribution of the uranium. Average ^{235}U loading per source is about 0.16 milligrams. Helium density measurements on the sources gave a value of 2.40 gms/cm^2 .

Neutron irradiation was carried out at the ORNL, Low Intensity Test Reactor (LITR), a water cooled, water moderated 3 MW reactor, fueled with a fully enriched U-Al alloy. Flux levels from 5×10^9 to 2×10^{13} neutrons/sec/cm² are available at this facility; samples are inserted into and removed from beam holes via a pneumatic system.

6.2 β - γ Spectra

Equipment and techniques used in obtaining the β - γ spectra of these sources are described in Section 4.4. Figure 6.1 shows the change of the gamma spectrum of a small pyrolytic carbon coated uranium sample as a function of time after irradiation; Figure 6.2 shows the β spectrum for like times. This particle was irradiated for 10 minutes at a neutron flux density of $2.0 \times 10^{10} \text{ n/sec/cm}^2$, determined by activation of cobalt wire.

6.3 Dose Estimates

Dose rate measurements have been made on the 1 mm reactor irradiated fuel particles with an extrapolation ionization chamber having

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Fig. 5.3 Metallic zinc sphere produced in air-driven grinder.

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Fig. 5.4 Graphite spheroids produced in air-driven grinder.

Fig. 6.1 Gamma spectra of a UC_2 particle after a 10 minute irradiation.

Fig. 6.2 Beta spectra of a UC_2 particle after a 10 minute irradiation.

a 3 mm diameter electrode. Table 6.1 shows the results which were obtained. Because of differences in the exact amount of uranium in each particular 1 mm particle, the total number of fissions varied somewhat between particles, compare particles 109-IC and 109-IF in Table 6.1. The apparent discrepancy in which the particle undergoing the smallest number of fissions (IC) resulted in the highest dose rate at 307 hours, may be explained by a nonuniform distribution of fuel beads in the 1 mm sources. Such variations can be expected in a real exposure situation and a realistic assessment of dose should include this consideration.

Table 6.1

Dose Rate Measurements for Irradiated 1 mm Diameter Simulated Reactor
Fuel Particles

Particle No.	Irradiation Time (min)	Flux Level $n/cm^2/sec$	Fissions	Decay Time (hrs)	Dose Rate (Rad/hr)*	Tissue Absorber (mg/cm ²)
109-1C	100	1.5×10^{13}	1.75×10^{13}	307.3	538.2	2.54
109-1F	100	1.5×10^{13}	1.93×10^{13}	136.0	1116.7	2.54
"	"	"	"	308.2	455.4	"
"	"	"	"	668.0	153.8	"
"	"	"	"	136.5	483.0	7.62
"	"	"	"	668.5	121.7	"
109-2H	10	8.0×10^{11}	6.52×10^{10}	46.0	64.2	2.54

* Corrected for STP and Stopping Power Ratio, and averaged over a 3 mm diameter collecting electrode.

7.0 TRAVEL

<u>Date (1965)</u>	<u>Location and Purpose</u>	<u>Traveler*</u>
Jan. 27-28	Washington, D.C. Discussions with Dr. S. Allan Lough and Ralph S. Decker.	BRF
March 1-2	San Francisco, Calif. SNPO Radiation Effects Working Group at USNRDL.	BRF
March 1-2	Washington, D.C. Federal Radiation Council, meeting of Ad Hoc Group on Sr and Cs Dosimetry.	WSS
March 18	Washington, D.C. NCRP (Executive Committee meeting)	KZM
March 25-26	Washington, D.C. ASA Committee Z-54, Subcommittee on Radioactive Surface Contamination and Panel Discussion on Decontamination Levels, USPHS.	BRF
April 20-30	Stockholm, Sweden. Meeting of ICRP Committee 2.	WSS
April 20-May 1	Stockholm, Sweden. Meeting of ICRP Main Commission and Committees.	KZM
May 21	Washington, D.C. Discussions with Dr. S. Allan Lough.	WSS BRF
June 10-11	Salt Lake City, Utah. Meeting of NCRP Committee 2.	WSS
June 14-17	Los Angeles, Calif. Health Physics Society meeting and discussions with R. S. Decker, P. E. Zigman and W. H. Ellett.	RHB BRF
June 25	Toronto, Canada. ASA N-7, Working Group on Sampling Air and Water for Radioactive Materials.	BRF

* BRF, B. R. Fish; WSS, W. S. Snyder; KZM, K. Z. Morgan; RHB, R. H. Boyett

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