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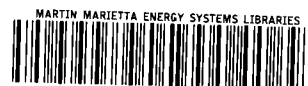
For Period Ending July 31, 1958

K. Z. Morgan, Director

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HEALTH PHYSICS DIVISION ANNUAL PROGRESS REPORT

WASTE DISPOSAL RESEARCH AND ENGINEERING

E. G. Struxness

R. J. Morton

LOW-LEVEL WASTE WATER TREATMENT

K. E. Cowser

H. J. Wyrick

Process wastes at ORNL come from equipment cooling water, floor drains, decontamination pad drains, storage canals, laboratory sinks, and discharges from low-activity operations. The waste volumes range from 200 to 300 million gal/year, and the radioactivity varies from 50 to several hundred beta counts per minute per milliliter (counts/min/ml). Occasionally, transient levels of radioactivity above 1000 counts/min/ml are observed in these wastes.

Expanded research and development programs at ORNL have increased gradually the amount of activity released to White Oak Creek, White Oak Lake, and the Clinch River. The yearly volumes and activity levels during 1950 to 1957 are summarized in Table 1. The total gross beta activity of discharges from the process-waste system and from the retention pond¹ has ranged from 172 to 498 curies per year. Strontium-90 is the most hazardous radioisotope in the waste.

To minimize the danger of accidental or emergency discharges and to increase the factor of safety for normal operations, a process-waste-treatment plant was deemed essential. Laboratory and pilot-plant studies of water decontamination processes and the anticipated future needs of the system served as the basis for determining the waste treatment requirements and establishing basic criteria for the design of the plant.

A lime-soda softening plant² of a straight-line type was adopted, and plant construction was completed in August 1957, at a cost of \$265,000. As shown in Fig. 1, the plant utilizes gravimetric chemical feeding, flash mixing, coagulation, and sedimentation, and has a capacity of 500,000

¹Former monitoring point for the detection and measurement of any leakage in the ORNL waste storage tank facilities.

²K. E. Cowser, R. J. Morton, and E. J. Witkowski, "The Treatment of Large-Volume, Low-Level Waste by the Lime-Soda Softening Process," paper 2354 in *Proc. Intern. Conf. Peaceful Uses Atomic Energy, Geneva, 1958* (to be published).

Table 1. Yearly Volumes and Radioactivity of Discharges of Liquid Wastes to White Oak Creek at ORNL, 1950-57*

Year	Total Volume (gal)	Gross Beta Activity (curies)			Gross Beta Activity Identified with Specific Radionuclides									
		Settling Basin	Retention Pond	Total	TRE**	Ce	Ru	Zr	Cs	I	Sr	Nb	Ba	Co
	($\times 10^6$)													
1950	226.4	172	15	187										
1951	297.6	169	3	172										
1952	268.2	411	87	498										
1953	239.4	289	140	429	53.1	2.6	0.8	2.6	8.5	0.2	27.6	1.0	0.8	
1954	164.3	237	17	254	34.6	19.1	0.5	1.0	20.3	0.5	23.3	0.4	0.3	
1955	210.6	213	54	267	30.3	14.7	3.1	0.6	31.6	0.2	18.7	0.6	0.2	
1956	260.7	253	20	273	24.4	12.3	2.0	0.5	42.1	0.1	15.1	1.0	0.2	2.2
1957	272.3				25.8	4.4	1.0	10.2	36.5	0.0	18.0	0.7	0.0	1.8

*Volume of settling basin effluent is measured in weir box with liquid-level float recorder. For determinations of radioactivity, continuous proportional samples of the effluent are composited for daily gross beta measurements and monthly radiochemical separations and analysis.

**Trivalent rare earths exclusive of cerium.

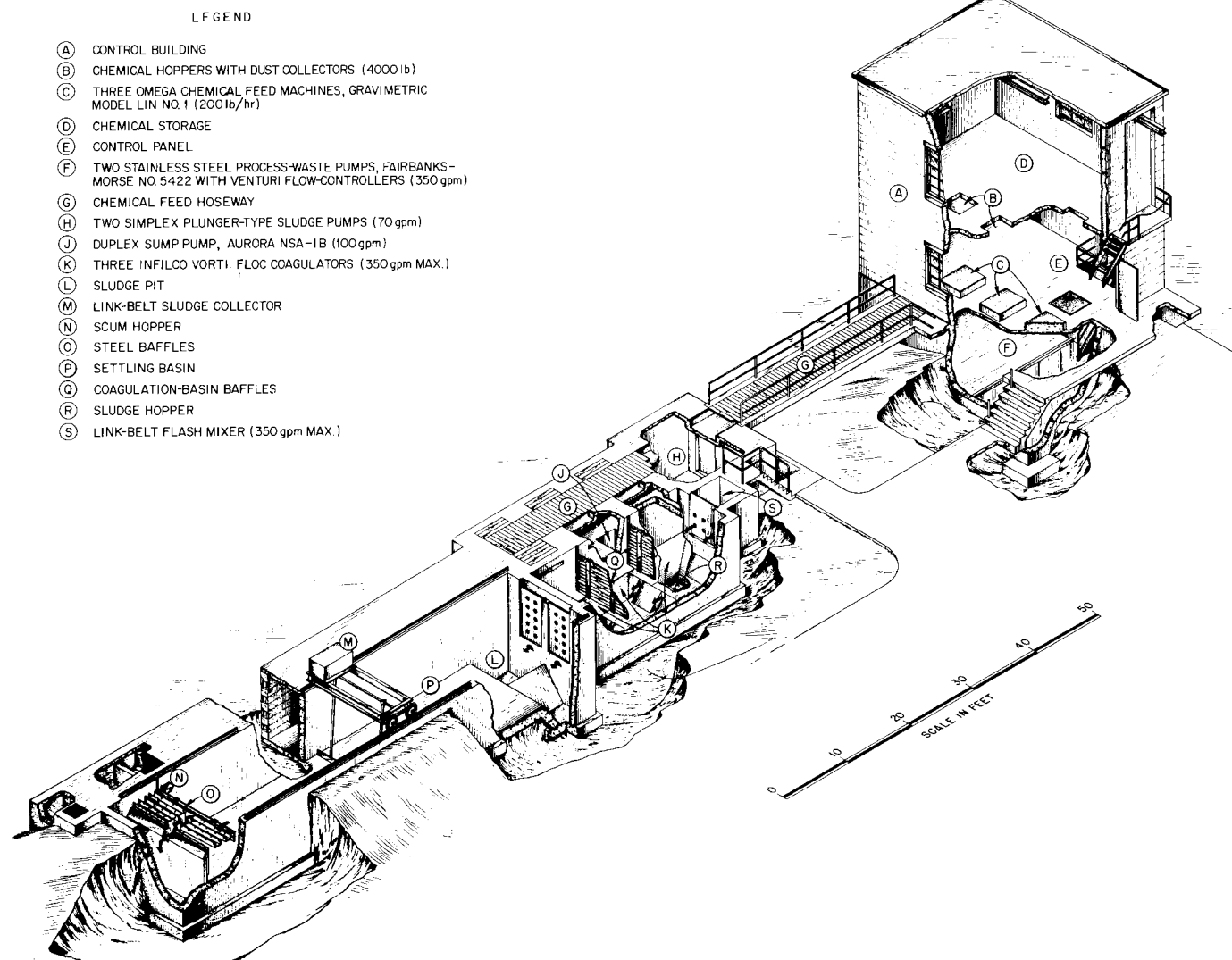


Fig. 1. Process-Waste-Treatment Plant.

gal/day. Flexibility in design provides for recirculation of the plant effluent, the return of preformed crystals (sludge), precipitation, and the addition of clay when necessary. An equalization basin with an operating capacity of 700,000 gal is provided to minimize fluctuations in chemical composition of the influent waste.

During the first six months of operation, 58.55 million gal of waste was treated. This amounted to 43% of the process waste, 57% having been bypassed and discharged to White Oak Creek without treatment, because, at times, the level of activity is so low that treatment is not necessary. Diversion of waste to the equalization basin for treatment or bypassing through a 1.5-million-gal settling basin to the creek is now accomplished automatically at a central monitoring station.³ A flow diagram of the waste handling and treatment system is shown in Fig. 2.

Tests of Plant Performance

The efficiency of treatment is determined by radiochemical analyses of composite samples of both the plant influent and the effluent, as listed in Table 2. The amounts of individual radio-nuclides removed each month are also shown. A total of 16.93 curies of strontium and 14.73 curies of total rare earths (TRE) was removed during the first six months of operations. This activity is associated with the calcium carbonate sludge that results from the treatment. For the first six months, 208,000 gal of sludge containing an average of 49% solids by volume was transferred by trailer truck to the waste disposal pit system.

With the use of the data in part A of Table 2, the percentage removals of the important radio-nuclides by the treatment plant for monthly periods (part B) were calculated. From an average of the percentage removals of strontium and total rare earths, computed from weekly composite samples, the reliability of the monthly composite samples

was confirmed. Strontium removals ranged from 66.4 to 87.4% and were as high as 91.6% for individual weekly periods. The removal of total gross beta activity ranged from 52.7 to 87.3% and was dependent upon the composition of the waste and the type and amount of chemical added. The increased reduction of gross activity during November 1957 was due to the efficient removal of Pa^{233} , which was present in the waste in unusual quantities during that month. No Pa^{233} was detected by analysis of the composite sample of the plant effluent. Since clay was not added continuously, cesium removals were low, as was anticipated. During the week of December 9, 1957, 30 ppm of a local clay was added to the waste, and cesium removals increased from about 16% to 55%. A source of Grundite, a clay similar to that found in the local shale, has been located, and continuous addition of this material will be initiated.

Weekly composite samples were analyzed for calcium hardness, total hardness, strontium, and total rare earths. The percentage removals of strontium and of total rare earths as a function of the residual calcium in the plant effluent are shown in Figs. 3 and 4. In these figures it is apparent that increased softening (low calcium concentration) results in increased removal of strontium and total rare earths. With the use of an excess of soda ash above that required for the reaction with the noncarbonate hardness and the excess lime, increased removals of calcium are obtained, as would be expected from the common-ion effect. At present, no excess lime is used; however, 200 ppm of excess soda ash is fed. There is no apparent relationship between the amount of calcium carbonate precipitated and the removal of strontium.

A relationship exists between the percentage removal of strontium and the total rare earths and is expressed as an index of correlation of 0.87. With percentage removal of total rare earths plotted as the ordinate Y and percentage removal of strontium as the abscissa X , the relationship is given by the equation

$$Y = 15.4 + 0.825X$$

³F. E. Gillespie, *Continuous Water Monitor for Radioactive Waste*, paper to be presented at the Instrument Society of America's 13th Instrument-Automation Conference-Exhibit, Philadelphia, Sept. 15-19, 1958.

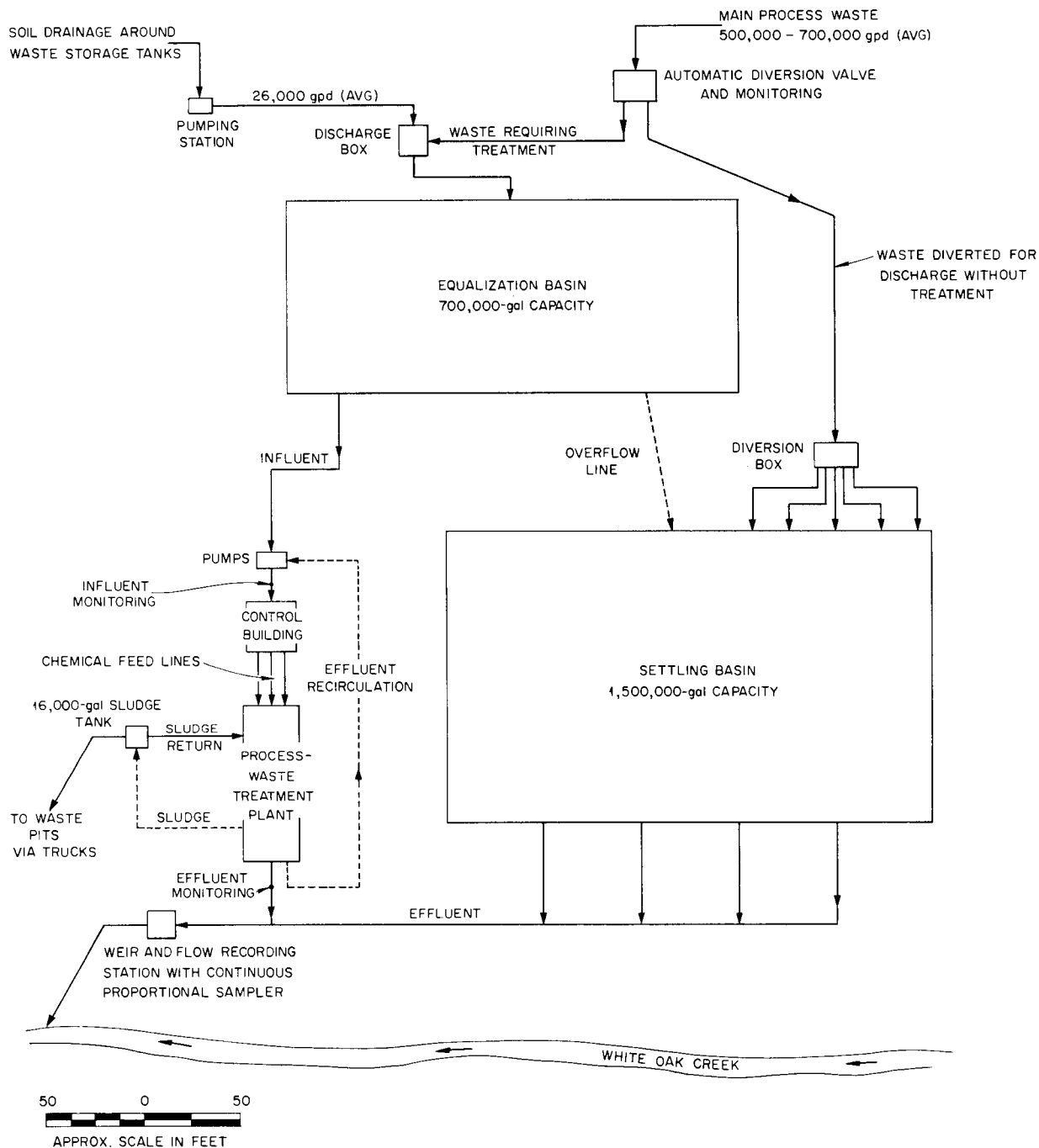


Fig. 2. Flow Diagram of Process-Waste System.

Table 2. Summary of Operating Results for Process-Waste-Treatment Plant

Monthly composite samples

A. Concentration of Radionuclides in Influent and Effluent

Month	Influent Concentration (counts/min/ml)								Effluent Concentration (counts/min/ml)							
	Gross Beta	Sr ⁸⁹	Sr ⁹⁰	Total Sr	TRE	Cs	Co	Ru	Gross Beta	Sr ⁸⁹	Sr ⁹⁰	Total Sr	TRE	Cs	Co	Ru
Sept. 1957	107.3	4.10	13.74	17.84	21.63	17.61	1.15	8.59	35.6	0.52	1.73	2.25	2.34	15.94	0.56	7.94
Oct. 1957	63.8	1.34	10.03	11.37	20.48	14.52	0.97	3.38	20.7	0.37	2.76	3.13	4.75	11.80	0.75	
Nov. 1957	155.3	1.59	10.66	12.25	18.01	11.18	0.54	6.93	19.7	0.32	2.16	2.48	3.60	8.85	0.15	1.20
Dec. 1957	70.4	1.17	9.60	10.77	21.00	9.77	0.75	3.36	14.5	0.19	1.59	1.78	2.67	5.89	0.13	1.11
Jan. 1958	77.0	1.58	15.60	17.18	24.96	13.42	0.71	4.24	36.4	0.41	4.08	4.49	6.61		0.17	1.48
Feb. 1958	45.8	1.59	7.18	8.77	17.16	11.27	0.59	2.23	19.7	0.66	2.29	2.95	5.68	6.57	0.26	1.29

B. Removal of Radionuclides by Treatment

Month	Percentage Removals							Curies Removed						
	Gross Beta	Total Sr	TRE	Cs	Co	Ru		Sr ⁸⁹	Sr ⁹⁰	Total Sr	TRE	Cs	Co	Ru
Sept. 1957	66.8	87.4 (87.9)*	89.2 (88.9)*	9.5	51.3	7.6		0.48	3.21	3.69	2.58	0.62	0.02	0.19
Oct. 1957	67.6	72.5 (72.6)*	76.8 (73.0)*	18.7	22.7			0.16	2.39	2.55	2.57	0.84	0.12	
Nov. 1957	87.3	79.8 (74.8)*	80.0 (82.5)*	20.8	72.2	82.7		0.18	2.24	2.42	1.91	0.23	0.12	0.74
Dec. 1957	79.4	83.5 (83.3)*	87.3 (84.9)*	39.7	82.7	67.0		0.13	2.26	2.39	2.58	0.74	0.21	0.31
Jan. 1958	52.7	73.9 (71.3)*	73.5 (75.4)*		76.1	65.1		0.17	3.38	3.55	2.65		0.19	0.38
Feb. 1958	57.0	66.4 (65.2)*	66.9 (64.9)*	41.7	55.9	42.2		0.23	2.10	2.33	2.44	1.36	0.17	0.18

*Average of weekly composite samples.

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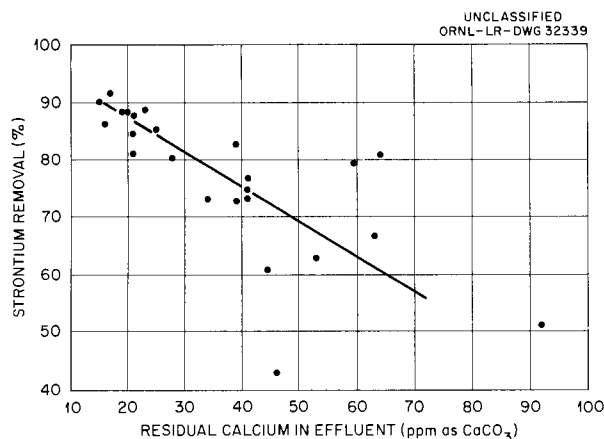


Fig. 3. Relation of Residual Calcium to the Removal of Strontium.

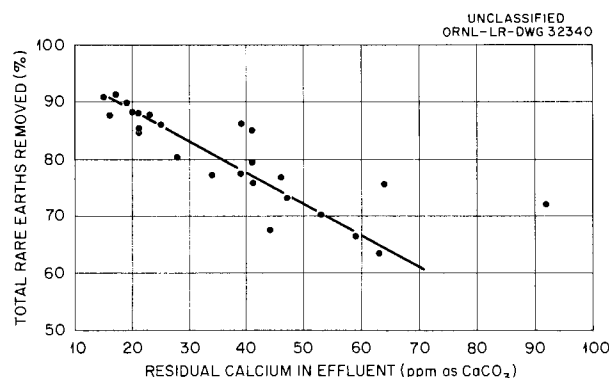


Fig. 4. Relation of Residual Calcium to the Removal of Total Rare Earths.

SOIL DISPOSAL OF INTERMEDIATE-LEVEL WASTES

K. E. Cowser	R. L. Bradshaw
D. G. Jacobs	F. L. Parker
W. de Laguna	H. J. Wyrick
E. R. Eastwood	

Inventory of ORNL Liquid Wastes

The liquid waste pumped to the pits comes from different sources in the Laboratory and varies in composition. From June 1952 through December 1957, 8.6 million gal of waste, containing about 112,000 curies of beta activity, was released to the pits. The principal contributors to radioactivity in the waste were Cs^{137} , Ru^{106} , and the rare earths. Lesser amounts of Sr^{89} , Sr^{90} , Co^{60} , and Sb^{125} were present. The quantity of the materials released is summarized in Table 3.

About 800 tons of sodium and 1100 tons of nitrate, the principal stable ions in the waste, have gone into the three pits. The concentration of fission products in the waste is given in Table 4. The concentration of fission products in the waste is reduced as the wastes are passed from Pit 3 through Pit 2 into Pit 4. Cesium and ruthenium are reduced by about 75 to 80%, strontium is reduced by 94%, and essentially all the rare earths are removed. These reductions are the result of precipitation in the first two pits, sorption on the side walls of the pits, dilution by rainfall, and radioactive decay.

The use of these pits has reduced the potential hazard of contamination of the Clinch River by the intermediate-level wastes. If in the past three years all the intermediate-level waste from ORNL had found its way into the river at the rate at which it was produced, and if complete mixing had occurred, a dilution of about 530,000 to 1 would have resulted. This gives a concentration value below the maximum permissible concentration for drinking water $(\text{MPC})_w$ for occupational use but exceeds the $(\text{MPC})_w$ for use by neighboring populations.⁴ While these $(\text{MPC})_w$'s are based on the exponential model, which assumes that biological elimination from the critical organ follows a single exponential law, there is some evidence that the elimination rate varies inversely with time and, consequently, can best be represented by a power-function model. This question is presently under consideration by the International Commission on Radiological Protection and the National Committee on Radiation Protection and Measurement. If the power-function model is selected as being more appropriate in the case of Sr^{90} - Y^{90} , the $(\text{MPC})_w$ values are increased by a factor of 10-15, depending on the particular form chosen.⁵⁻⁷ Because of the sorption and filtration properties of the soils, a great majority of

⁴Calculations of the $(\text{MPC})_w$ values were based on the *Report of Committees on Permissible Dose for Internal Radiation—1958 Revision* (Joint report of the International Commission on Radiological Protection and the National Committee on Radiation Protection; data unofficial and subject to change) (to be published).

⁵J. W. Healy, *Radiation Research* 4, 367-72 (1956).

⁶L. D. Marinelli, *Estimates of Bone Pathology To Be Expected from Sr^{90}* , ANL-5716 (April 22, 1957) (preliminary unpublished report).

⁷M. A. Van Dilla, S. Wallach, and J. S. Arnold, *Semi-annual Progress Report of the Radiobiological Laboratory, Salt Lake City, Utah, University of Utah College of Medicine*, Sept. 30, 1956, p 48-58.

Table 3. Waste Disposal System Inventory^a

	Pit 2	Pit 3	Pit 4	Total
Fission products released to pits, curies				
Gross beta emitters	26,100	80,900	5,600	112,600
Cs ¹³⁷	26,800	33,700	7,400	67,900
Ru ¹⁰⁶	8,500	9,500	1,400	19,400
St ⁹⁰	(260) ^b	2,580	90	
St ⁸⁹	(50) ^b	950	10	
TRE	(<100) ^b	11,000	<100	
Volume, thousands of gallons	3,897	658	3,990	8,545
Chemicals released to pits, thousands of pounds				
Na	983	216	360	1,559
NO ₃	1,320	304	600	2,224
Volume, thousands of gallons	3,987 ^c	658	3,990	8,635

^aAmounts are cumulative through December 1957.

^bCuries in waste overflowed to Pit 2 from Pit 3 from June 1955 through December 1957.

^cIncludes 90,000 gal of nonradioactive chemical waste.

the radioisotopes has been retained in the pit system, and the concentration of fission products reaching the Clinch River is thereby greatly reduced. The factors responsible for the reduction are discussed later.

Geology of Waste Pit Area

The waste pits are excavations in weathered Conasauga shale, a light-brown, layered and banded weak rock which is broken by many little joints into small prisms.

The depth of weathered shale extends 30–40 ft below the low ridges and about 10 ft under the valleys; in many places the bottom of the weathered zone is roughly at the water table. The underlying shale is composed of thin alternate layers of well-consolidated dark-gray calcareous shale and light-gray shaly limestone. Much of the carbonate has been leached out by the weathering; in general the limestone layers are weathered to silty clay and the shale layers to a fine silty sand.

The shale beds dip to the southeast at about 35 deg, but there are several types of structure which complicate this pattern. At the north end

of Pit 3 (Fig. 5) a reversal of dip has produced a small anticline and syncline; the width of the structure is about 150 ft. The dips are gentle and there is no evidence of crumpling or faulting. In the area occupied by Pit 4, the beds are intensely and irregularly folded and crumpled (Fig. 6). Exposures suggest that this belt of crumpled beds is roughly 200 ft wide and that it extends east and west along the strike for at least 1000 ft. There is some hydrologic evidence which suggests that this belt of crumpled rock is bordered on the south by a fault, possibly a thrust fault related to the Copper Creek fault. South of Pit 4, and south of the presumed fault, there are a number of small folds. Exposures in and near Pit 2 are poor, but probably there are no folds or faults in this area.

Hydrology of Waste Pit Area

Movement of Water in the Conasauga Shale. – The weathered shale is much more permeable than the unweathered portion. Water moves through the weathered rock along the many joints; since these are more closely spaced and more regular along the bedding planes, the water moves much more

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Table 4. Concentration of Radionuclides in Liquid Waste

Pit No.	Type of Waste	Concentration ($\mu\text{c/ml}$)					
		Gross Beta Emitters*	Cs ¹³⁷	Ru ¹⁰⁶	Sr ⁹⁰	Sr ⁸⁹	TRE**
3	Wastes pumped to Pit 3						
	Minimum	0.086	0.0081	0.0081	0.0009	0.0005	0.0005
	Mean	4.0	3.3	0.65	0.13	0.051	0.45
	Median	1.3	0.62	0.26	0.021	0.063	0.06
	Weighted average	3.5	2.0	0.47	0.10	0.038	0.40
	Maximum	45	61	6.9	1.6	0.72	5.6
2	Overflow from Pit 3 into Pit 2						
	Minimum	0.23	0.24	0.018	0.0005	0.0003	
	Mean	0.62	0.79	0.13	0.0090	0.0050	
	Median	0.57	0.68	0.10	0.0045	0.0027	
	Weighted average	0.59	0.77	0.13	0.010	0.0046	
	Maximum	1.3	1.7	0.37	0.056	0.023	
4	Overflow from Pit 2 into Pit 4						
	Minimum	0.14	0.18	0.014	0.0005		
	Mean	0.37	0.48	0.088	0.0077		
	Median	0.33	0.45	0.089	0.0036		
	Weighted average	0.37	0.49	0.090	0.0059		
	Maximum	0.64	0.88	0.24	0.069		

*Based on a counting efficiency of 12.6%.

**Exclusive of Y⁹⁰.

easily parallel to the strike than at an angle to it. There is some movement of water through the fresh shale, not through the rock itself, but along joints and fractures. Figure 7 shows the results of pressure tests of some of the observation wells near Pit 4. The method consisted in introducing water under pressure into short vertical sections of the well. The most permeable section of the fresh shale is near the top; there are some fractures down to 100 ft, but apparently very few below 200 ft. This method does not test the permeability of the weathered shale, because the test wells are cased down to fresh rock. The results of pumping tests and tracer tests have been reported previously.⁸

⁸K. E. Cowser *et al.*, *H-P Semiann. Prog. Rep.* Jan. 31, 1956, ORNL-2049, p 33-34.

The conclusion reached from pumping, pressure, and tracer tests is that it is virtually impossible to predict, from any known method of pretesting, how a waste disposal pit will operate in a proposed site in the Oak Ridge area. In a pumping test, the response of the water levels in the observation wells may be due to pressure effects or to actual removal of water; the magnitude of the response depends on the hydraulic connection with the pumped well, on the hydraulic connection with outlying areas which may supply water to replace that removed, and on the porosity of the soil. These factors cannot always be separated. The only reliable test would consist in filling the pit with water that contains an appropriate tracer and studying the underground movement of the water. However, this test will not furnish

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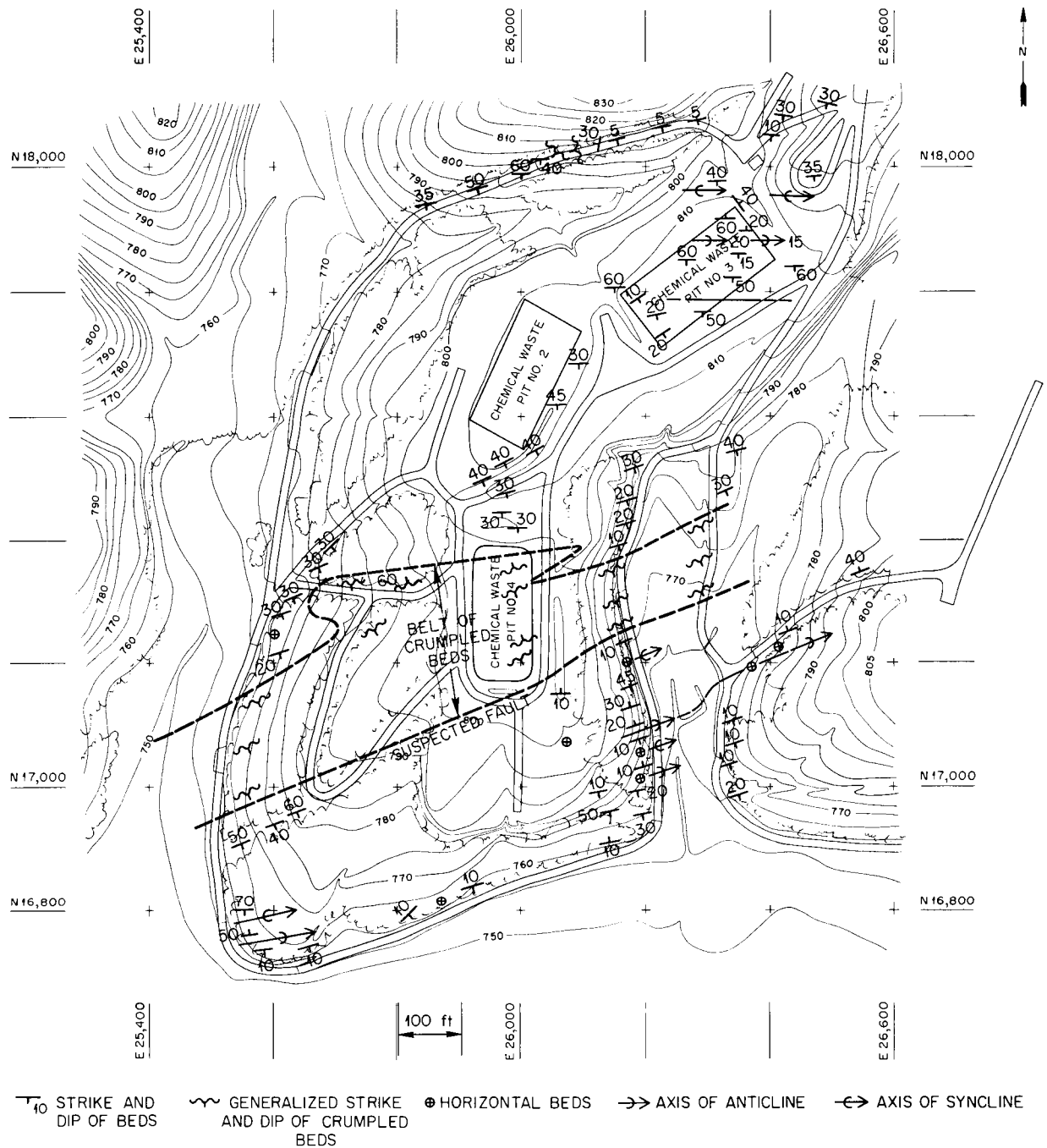


Fig. 5. Geologic Map of Pit Area.

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Fig. 6. South End of Pit 4.

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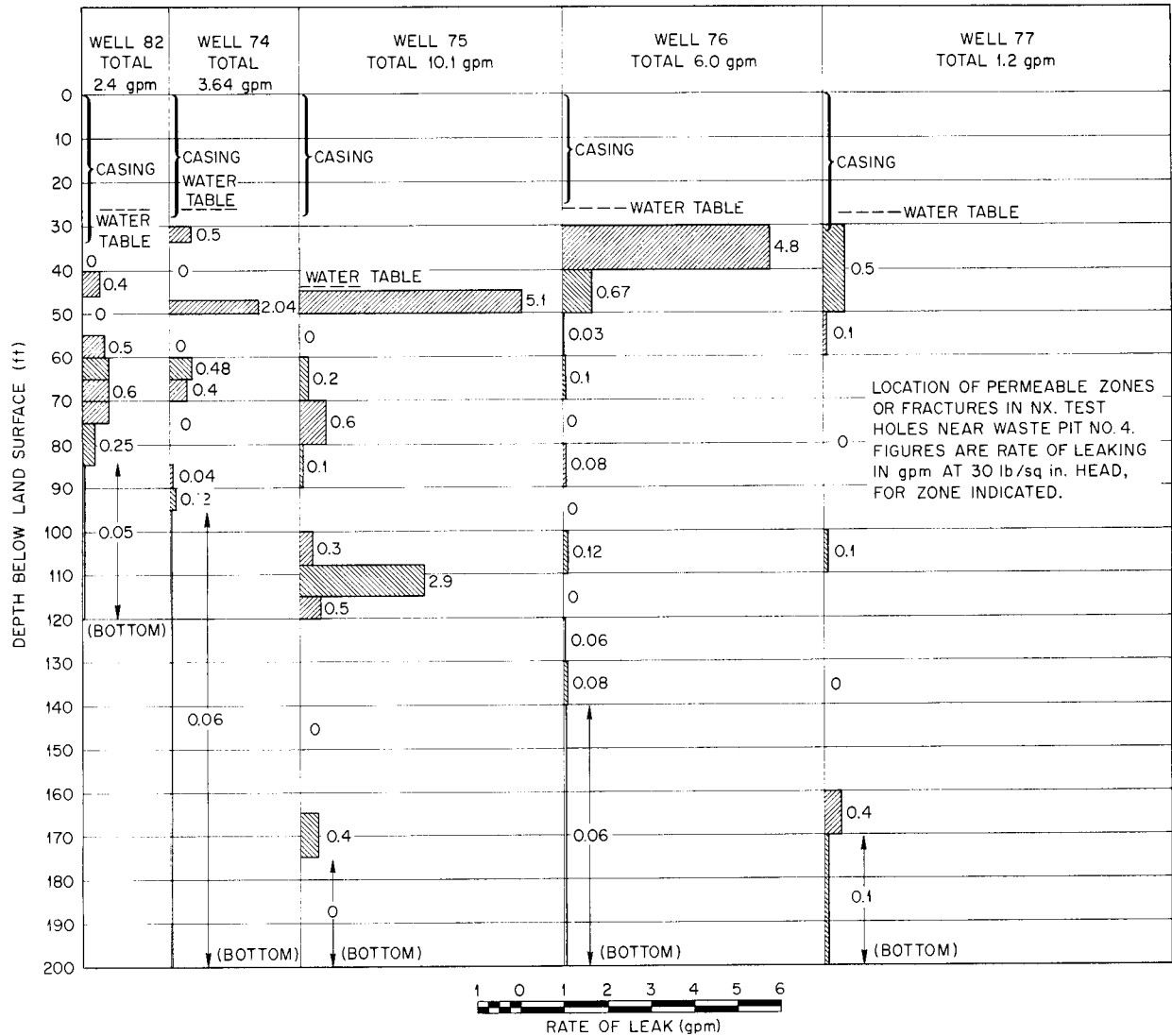


Fig. 7. Pressure Testing of Wells in Pit 4 Area.

complete information on the movement of all radioactive materials.

Hydrologic Characteristics of the Operating Pits. — The record of waste transfer and overflow is shown graphically in Fig. 8. In summer, when evaporation and seepage increase and rainfall decreases, Pits 3 and 2 can dispose of most of the waste. In winter, waste is frequently overflowed to Pit 4.

Figure 9 shows the level of the water table on January 10, 1958. For over a year prior to that time, the waste levels in Pits 2 and 3 had been

maintained at close to the 13-ft stage, and the water table had stabilized around the pits. Pit 4 was put into service in April 1956, and by the end of 1957 it had received about 4 million gal of waste. The water table around Pit 4 is never stable. Waste is run into the pit in batches, usually during the winter months, and it is impossible to maintain any fixed liquid level in the pit. When the level is fairly high, the waste leaks out rapidly (20,000 gal/day at the 8-ft stage; when the level falls, the rate decreases). At the same time, the water table rises under and

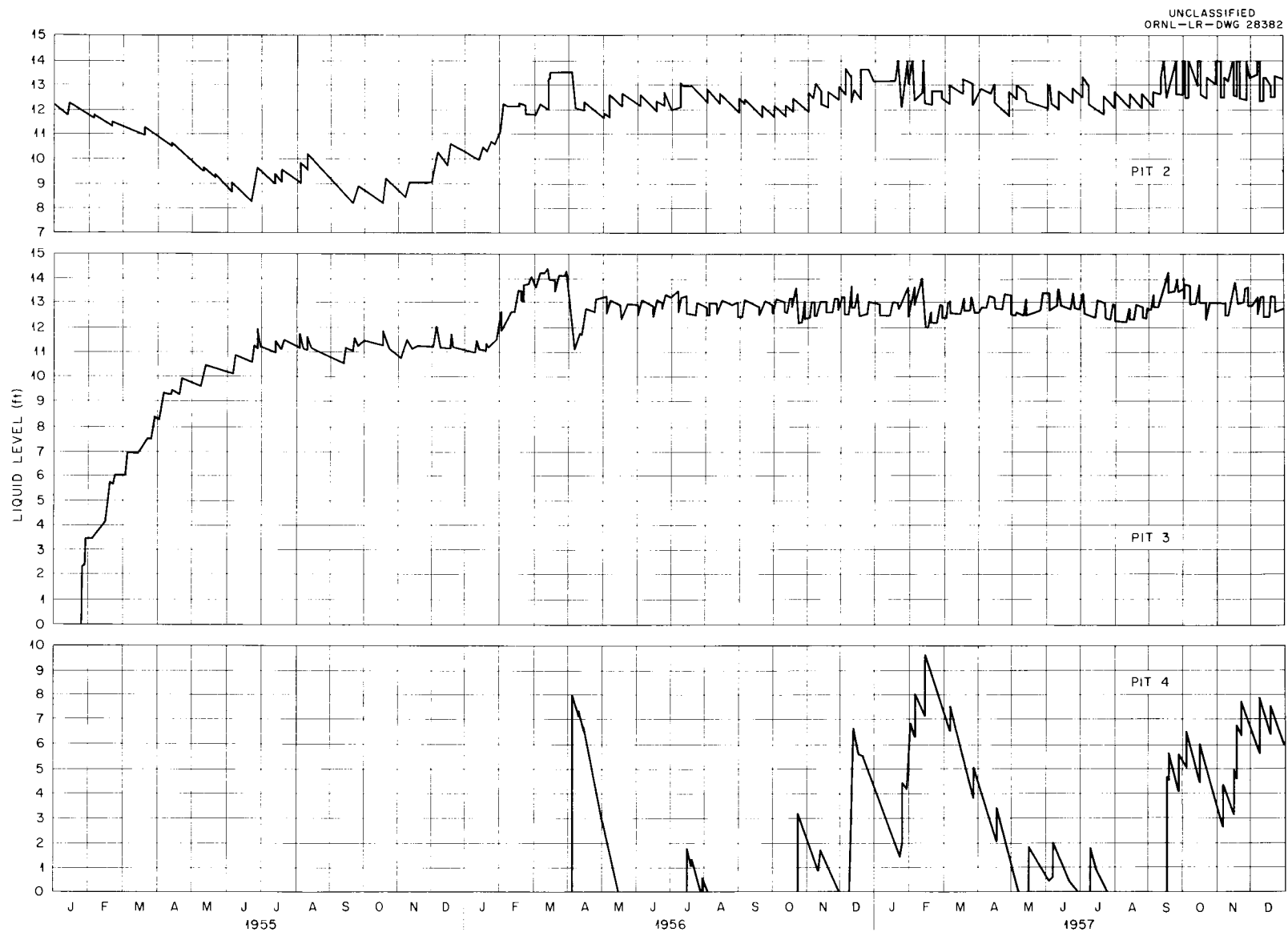


Fig. 8. Record of Liquid Levels of Operating Waste Pits.

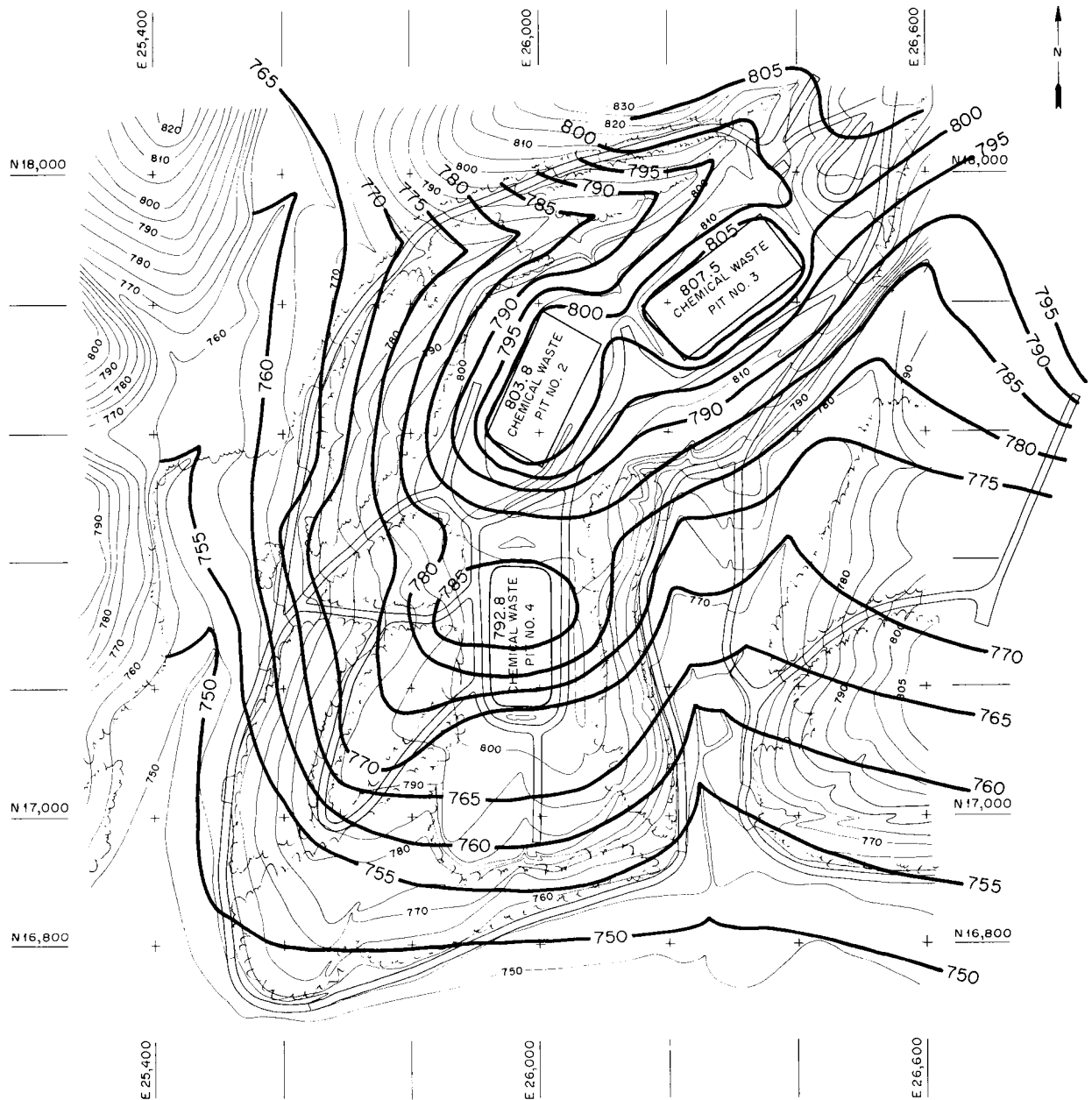


Fig. 9. Water Table Contour Map, January 10, 1958.

around the pit, the water-table gradients are steepened, and the rate of movement of liquid out from the pit area increases (Fig. 10). When the pit level falls and the rate of leaking from the pit decreases, the water table rises less rapidly, until eventually an equilibrium is reached. When the pit is empty, the water table falls. The water-table map of January 10, 1958, was made at a time when there had been liquid in Pit 4 for some time, and the water-table mound under the pit was approximately at a maximum.

Evaporation and Seepage from Waste Pits

The number of pits required is a function of the volume of input and the loss of liquid from the system. The rate of seepage from the pits cannot be measured directly and must be determined as the net loss after accounting for waste pumpage, rainfall, and evaporation.

The volume of waste pumped to the pits can be determined with sufficient accuracy by measuring the changes in levels that occur in the storage tanks as a result of the transfer. The volumes transferred between pits cannot be determined as accurately as desired. The amount of rainfall entering a pit can be estimated within 3% by simply considering the rain falling within the catchment area of the pit. Pit evaporation was estimated from the observed losses of several evaporation pans and was also calculated from meteorological observations by use of Kohler's technique.⁹ While the different methods for estimating evaporation gave results that differed by

about 10%, the effect of these differences on seepage calculations becomes negligible when seepage accounts for more than 70% of the liquid loss. Such is the case for Pit 2 and Pit 4.

The rate of seepage from a pit is influenced by the liquid stage in the pit, the depth to the groundwater table around the pit, the viscosity of the liquid waste, and the permeability of the soil. The estimated rates of seepage from Pits 2 and 3 are shown in Table 5. Until 1957, the rate of seepage from Pit 2 increased each year. The apparent reduction in the rate of seepage from Pit 2 in 1957 cannot be considered significant,

⁹M. A. Kohler, T. J. Nordenson, and W. E. Fox, "Evaporation from Pans and Lakes," U.S. Weather Bureau Research Paper No. 38, GPO, Washington, May 1955.

Table 5. Estimated Seepage from Pit 2 and Pit 3

Year	Pit 2		Pit 3	
	Seepage (gal/day)	Pit Stage (ft)	Seepage (gal/day)	Pit Stage (ft)
1953	1900	3.7		
1954	2700	8.3		
1955	3400	9.9	1650	9.2
1956	3850	12.2	950	12.7
1957	3700	12.5	450	13.0

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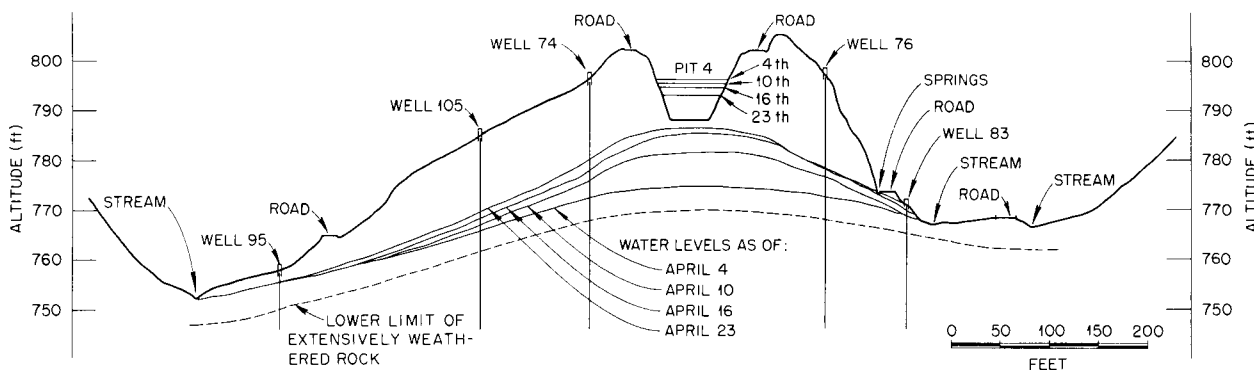


Fig. 10. East-West Section Through Pit 4 Showing Changes in the Water Table Following First Discharge of Waste on April 4, 1956.

since the inaccuracy of the data used in the calculations may give rise to errors of about 10% in seepage. The reduced rate of seepage from Pit 3 in 1956 was thought at that time to be due to the general rising of the water table around the pit, resulting in lower water-table gradients. Although the water table around the pit became essentially stable, a significant reduction in rate of seepage occurred in 1957. It is believed that the ability of the shale around Pit 3 to transmit liquid has been reduced. The reduction may be due to a mechanical plugging of the passageways by silt or precipitates formed in the pits, or it may be the result of a gradual swelling of the clay by the substitution of sodium for calcium in the clay mineral structure. If a similar reduction in seepage should occur in Pits 2 and 4, additional pits will be necessary. The seepage rate of both pits is about 15–20% higher in summer than in winter, probably due to temperature-induced changes in the viscosity of the waste.

The entrance of surface runoff to Pit 4 has prevented an accurate liquid balance for this pit. Nevertheless, records of liquid stage and evaporation show that seepage accounts for more than 95% of the gross liquid loss from the pit, even during periods of maximum evaporation.

Extent of Dispersion from Waste Pits

Local Dispersion. — Liquid samples collected from observation wells provide the data for constructing a map of the concentration of radioactivity in the pit area, as shown in Fig. 11. The activity has spread principally east and west from the pits in a direction parallel to the bedding. Very little activity has moved south from Pit 4, that is, across the bedding. The pattern of waste movement is largely as anticipated. The concentration of radioactivity in wells around Pits 2 and 3 has been relatively constant for some time, while around Pit 4 the concentration of radioactivity in the several wells fluctuates in response to the amount of waste in the pit and the rate at which the pit is leaking.¹⁰

To date, the two most hazardous isotopes, Sr^{90} and Cs^{137} , have not been detected in the waste escaping from the pits. Ruthenium-106 is the only isotope that has been identified in important

amounts in any of the observation wells, seeps, and streams. Laboratory and field results indicate that a continuing removal of ruthenium occurs as long as the waste remains in contact with the shale.¹¹

In Fig. 12 it is shown that the concentration of activity in the streams east and west of the pits fluctuates rapidly and depends on stream discharge. The total transport of activity per unit time did not fluctuate rapidly; it increased in 1957 presumably because of the rapid seepage from Pit 4. An estimated 200 curies of Ru^{106} escaped from the pits into the bed of former White Oak Lake during 1957, while about 60 curies was released from White Oak Creek to the Clinch River. Apparently, additional amounts of ruthenium are removed as the liquid moves across the lake bottom. The operation of the waste-pit system from June 1952 through December 1957 has been responsible for the release of about 150 curies of Ru^{106} from White Oak Lake to the Clinch River. With dilution in the Clinch River, the average concentration of ruthenium would be about 10^{-8} $\mu\text{c}/\text{ml}$.

Although Cs^{137} has not reached the test wells in detectable quantities, it has been detected in trees and their litter in the immediate environs of the pits. The vehicle for transport cesium is uncertain. It may be the ground water, or the cesium may be air-borne. Air monitoring stations were installed around the pits, and the gross activity collected by standard gum paper for 25 weekly periods was statistically analyzed. These analyses showed that the amount of air-borne activity in the pit area was significantly greater than at similar off-site stations, the average concentration being about twice that at the off-site stations. Work is under way to determine the specific radionuclides involved.

Although the general pattern of waste movement from the pits has been established, the movement of individual radionuclides has not been defined in as much detail as desired. The underground movement of water and of any mobile waste materials is restricted to the north, and cannot extend beyond the streams east and west of the pits. To the south, the streams drain into the bed

¹⁰R. L. Blanchard *et al.*, *H-P Semiann. Prog. Rep.* July 31, 1957, ORNL-2384, p 79–81.

¹¹R. L. Blanchard, B. Kahn, and G. G. Robeck, *Laboratory Studies on the Ground Disposal of ORNL Intermediate-Level Liquid Radioactive Wastes*, ORNL-2475 (March 14, 1958).

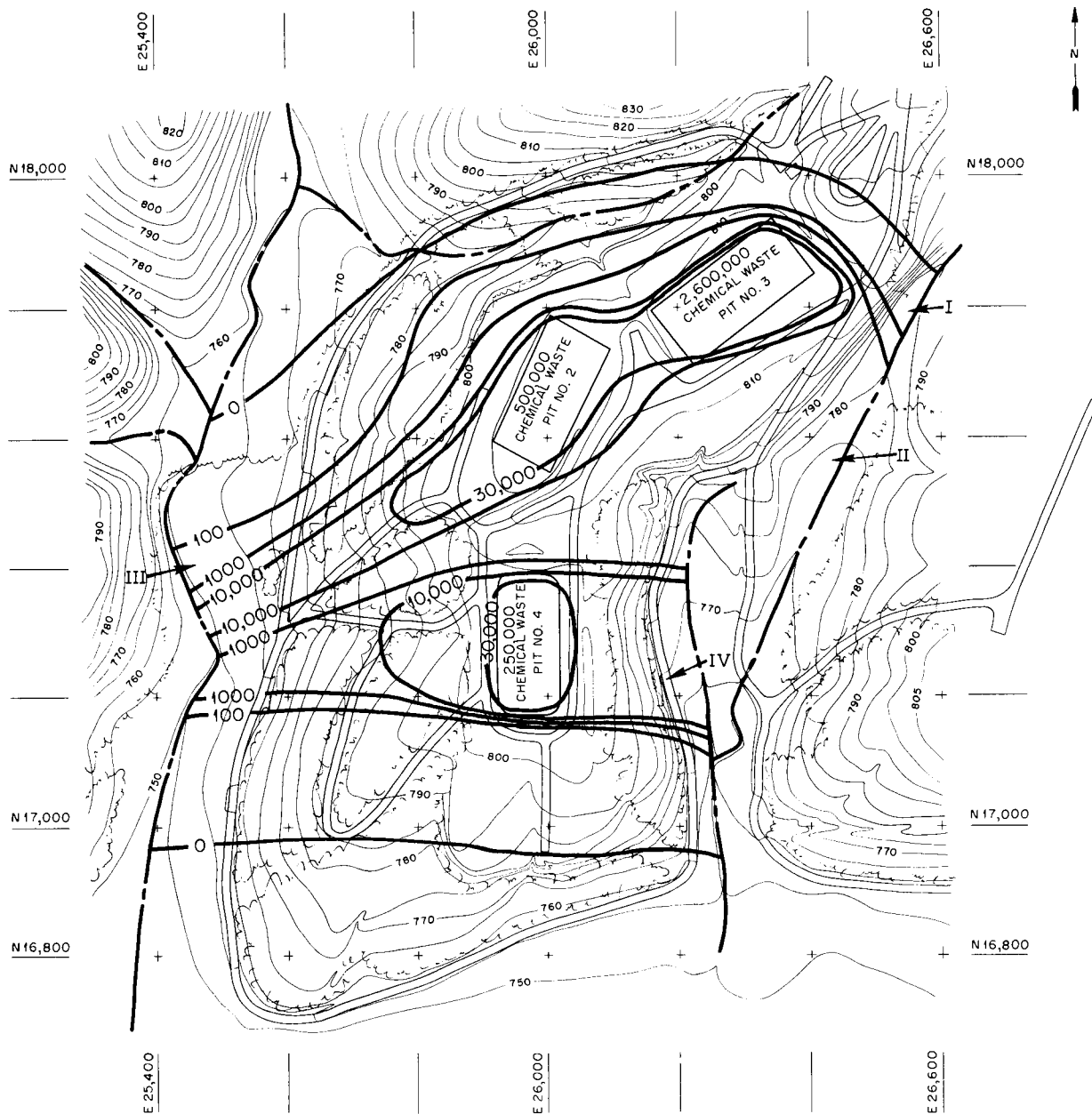
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Fig. 11. Dispersion of Radioactive Wastes - February 1958. I, II, III, and IV represent radioactive seeps and springs; unit of concentration is $\mu\text{mc}/\text{ml}$.

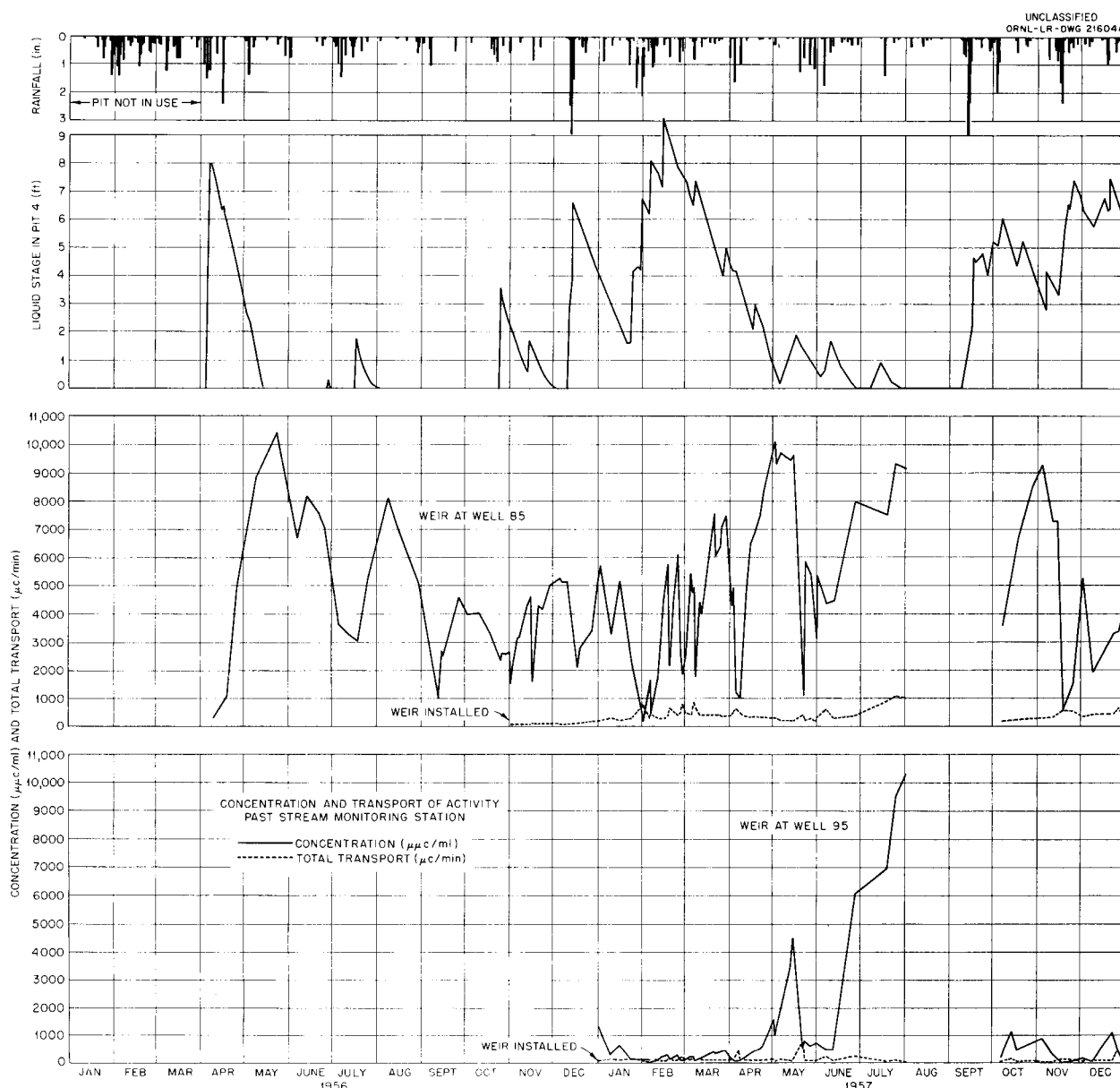


Fig. 12. Transport of Activity Past the Stream-Monitoring Stations at Wells 95 and 85.

of former White Oak Lake. The liquid escaping from the pits is monitored at numerous wells at weirs which are located on the streams, and before discharge from White Oak Creek to the Clinch River. Therefore, the material escaping from the pits cannot inadvertently become a hazard to the public even if the details of its movement are unknown.

Dispersion in the Clinch River. — The fate of fission products discharged from White Oak Dam is of great importance, because the concentrations in the Clinch River must be below the maximum permissible concentrations for drinking water. White Oak Dam lies 0.6 mile above the confluence of White Oak Creek and the Clinch River at Clinch River mile 20.8. Measurements on the amounts of

activity entering the Clinch River system have been made,¹² and the bottom muds of the Clinch River have been sampled in order to measure the amount of fission products in the mud.^{13,14} The role of the river system in reducing the concentration of the radionuclides had not been previously studied; therefore, when TVA decided to determine the re-aeration of streams below the dams, the opportunity was taken to determine the effectiveness of the White Oak Creek–Clinch River System in diluting and dispersing the wastes from ORNL.

After leaving the immediate environs of the Laboratory and the waste pits, the wastes flow into White Oak Creek and then into the Clinch River. White Oak Creek can be dammed at the White Wing Road, and the wastes can be treated or diluted before release to the Clinch River. However, the watershed of White Oak Creek above the White Wing Road is 6.0 square miles, and the capacity of the lake bed at the overflow elevation of 750 ft above mean sea level is only 15 million cubic feet. A 1-in. runoff from the drainage area is equivalent to 14.1 million cubic feet. Therefore, the capacity of the lake would be almost exceeded by a runoff of 1 in. over the drainage area, and a runoff of this magnitude is not unusual. Even at the yearly average flow of 7.5 cfs in White Oak Creek, the wastes would be detained only 23 days behind the dam at elevation 750 ft. Therefore, the White Oak Dam System would not be able to detain the wastes for any appreciable length of time before they would be discharged to the river.

Tracer-level tests were run on July 4–7 and July 9–13. The TVA maintained a steady flow in the Clinch River of 3000 cfs for the first test and 6000 cfs for the second test. On each test, 5 curies of I^{131} was injected into White Oak Creek at its confluence with the Clinch River. Liquid samples were taken periodically during the course of the test at Clinch River miles 19.6, 17.8, 15.5, 14.0, and 13.1 (stations A, B, C, D, and E, respectively) (Fig. 13). Scintillation counters were used for measuring the concentration of radioiodine flowing past the sampling stations. Through

a cooperative arrangement with the U.S. Geological Survey, the flow through this reach of the river was measured during the test. The velocity and temperature profiles at the sampling stations were also determined by the U.S. Geological Survey.

Additional liquid samples were collected downstream at Centers Ferry (Clinch River mile 4.4) and at the Kingston Steam Plant (Emory River intake). While the results have not been completely evaluated, preliminary studies at Clinch River mile 19.6 (station A) show clearly that complete dilution was not attained, under the river conditions existing during the time of the test. A definite temperature gradient exists (Fig. 14), causing the concentration profile shown in Fig. 15. The contaminated White Oak Creek water flows below the surface waters of the Clinch River and remains close to the right bank, only gradually spreading across the river. The study of patterns of dispersion and dilution further downstream and a quantitative evaluation of the phenomena are under way.

Cesium Retention by Conasauga Shale

Equilibrium Studies. – For further understanding of the problems of waste-pit management, a study of the mechanism of fission product retention by Conasauga shale was desirable. A knowledge of the sorptive behavior of cesium in the presence of various complementary cations affords a guide to understanding the mechanism of cesium sorption. Conasauga shale from the waste-pit disposal area was used in all studies.

Two hundred grams of shale was leached with 3 liters of 1 *N* chloride salt solution in order to obtain an essentially homionic shale. Six grams of this shale, having 1 milliequivalent (meq) of cation exchange capacity (CEC), was added to a bottle containing 100 ml of tagged $CsNO_3$ solution of the proper concentration. After sufficient time for completion of the exchange reaction, 1-ml aliquots were removed for counting, and a known amount of the complementary cation (Na^+ , K^+ , Rb^+ , NH_4^+ , Ca^{++} , or Al^{+++}) was added to each bottle. This was repeated for each complementary cation concentration desired. The amount of cesium sorbed by the shale was computed by comparing the initial and final counting rates of the supernatant.

Conasauga shale is very effective in the removal of cesium from solution. The effectiveness is more pronounced as the cesium concentration is decreased to tracer quantities. The ability of the

¹²The Effect of Discharge of Radioactive Substances from White Oak Lake into Clinch River (undocumented report).

¹³J. M. Garner, Jr., and O. W. Kochtitzky, *Proc. Am. Soc. Civil Engrs.* 82(SA4), paper 1051 (1956).

¹⁴W. D. Cottrell, *Preliminary Report of 1954 Survey of the Clinch and Tennessee Rivers*, ORNL CF-55-10-125.

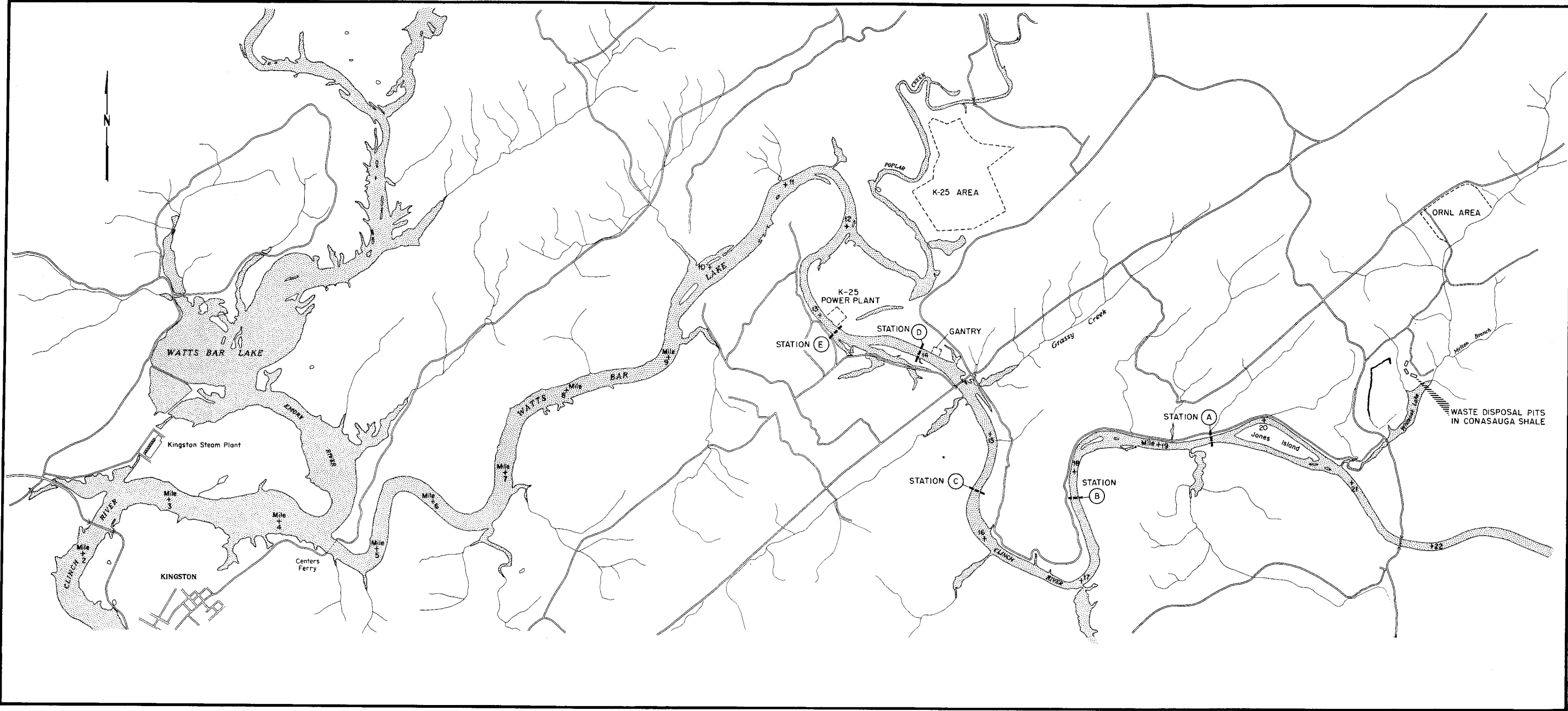


Fig. 13. Location of Sampling Stations A, B, C, D, and E.

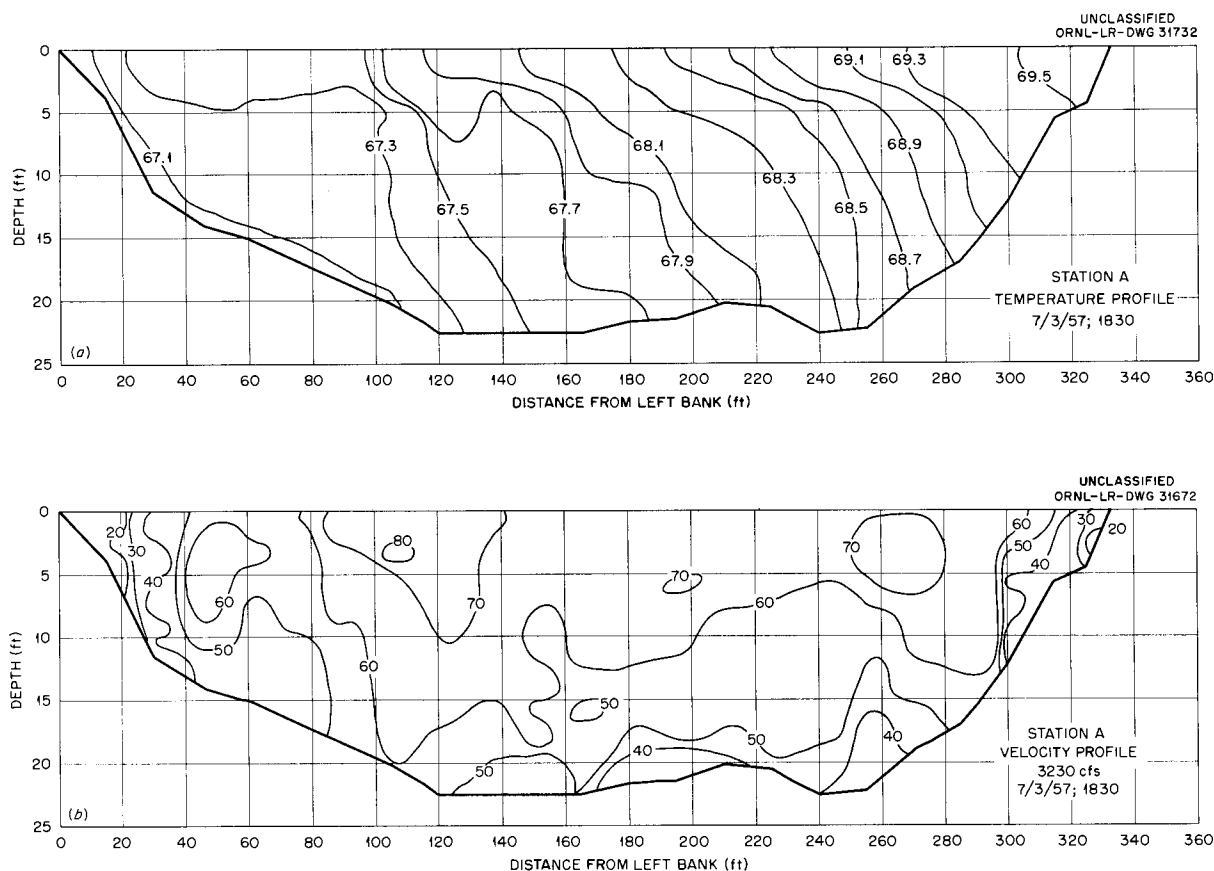


Fig. 14. Temperature (a) and Velocity (b) Profiles at Clinch River Mile 19.6, Test 1.

shale to retain cesium is enhanced by the presence of fixation sites which have a high affinity for cations of the size of cesium.

As the valence of the complementary cation is increased, the sorption of cesium is decreased. However, the complementary cation is not detrimental to the sorption of tracer quantities of cesium by Conasauga shale. It is believed that in this case the specific bonding energy for cesium greatly overshadows the increased affinity of the charged surface for counterions of higher valence.

Increased concentrations of the complementary cation result in decreased sorption of cesium. The effect is very small for tracer quantities of cesium, except when the complementary cation has an ionic radius similar to that of cesium. Large amounts of K^+ , Rb^+ , or NH_4^+ markedly decrease the efficiency of the shale to retain tracer quantities of cesium.

Column Desorption Studies. — Sodium-saturated shale was leached with a tagged $CsNO_3$ solution, and the amount of sorbed cesium was calculated. This shale was then used in a series of desorption experiments.

Six-gram portions of the Na-Cs shale (1 meq CEC) were placed on Buechner funnels and leached with appropriate leaching solutions. Aliquots of the effluent were counted for Cs^{137} activity.

The first series of leachings were made by using NaCl solutions of different concentrations. The amount of cesium leached from the shale is related only to the total amount of NaCl passed through the shale and appears to be independent of the concentration of the leaching solution. Table 6 gives a résumé of these data.

Decinormal solutions of various sodium salts were then used as leaching solutions; 10 meq of $NaHSO_3$ removed as much Cs^{137} from the shale

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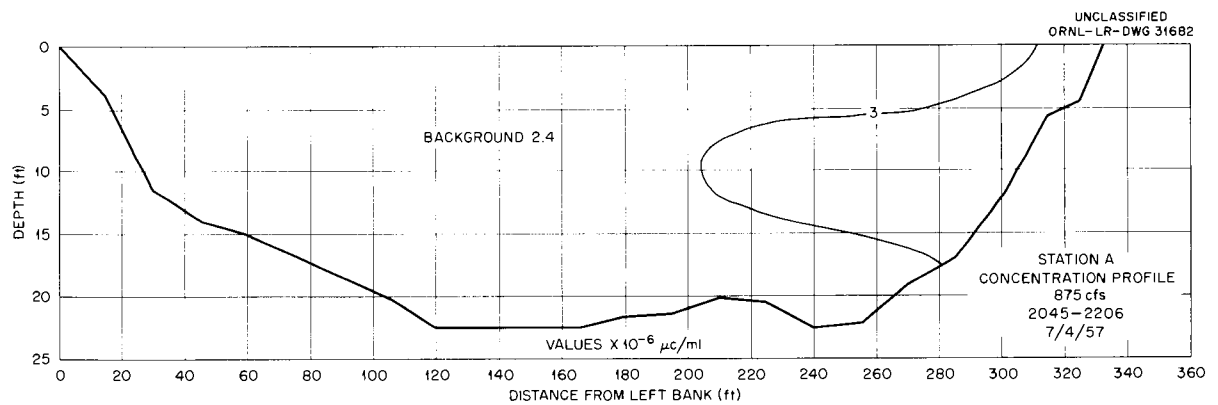
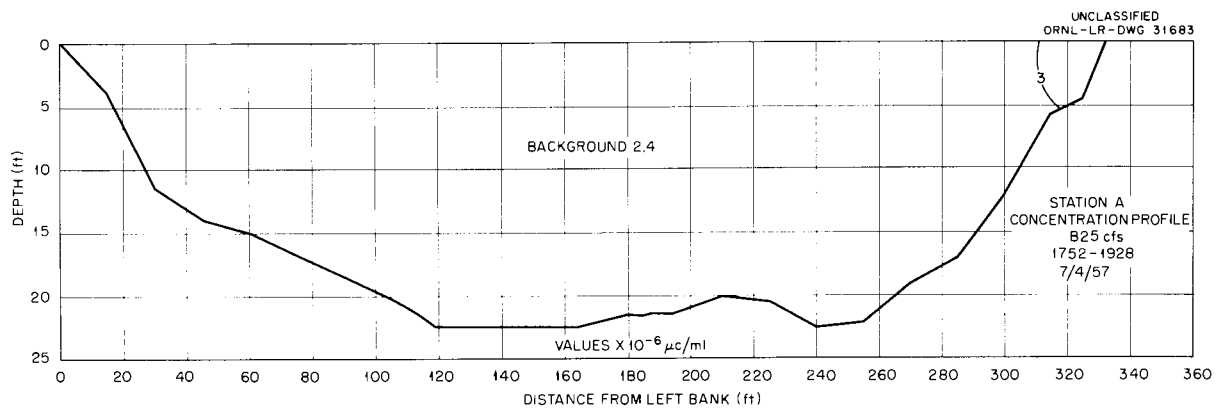
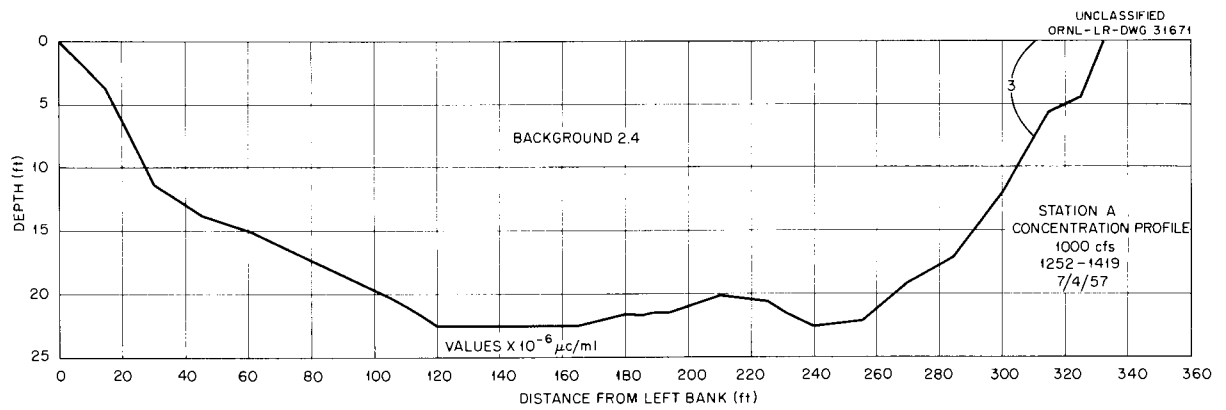


Fig. 15. Concentration Profiles at Clinch River Station A.

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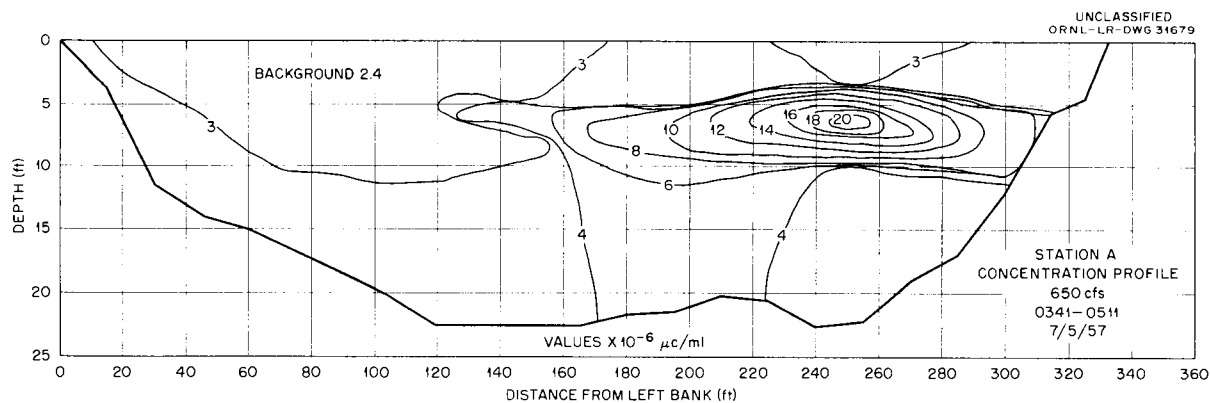
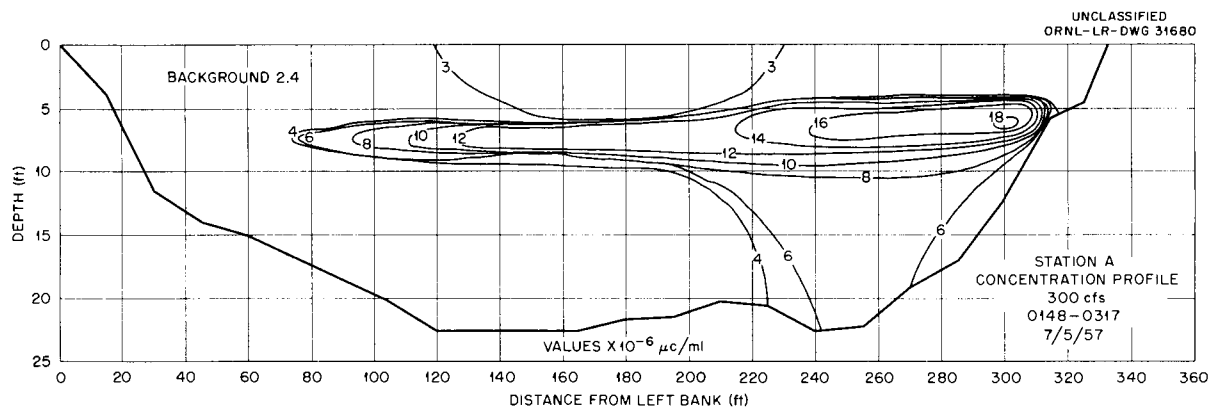
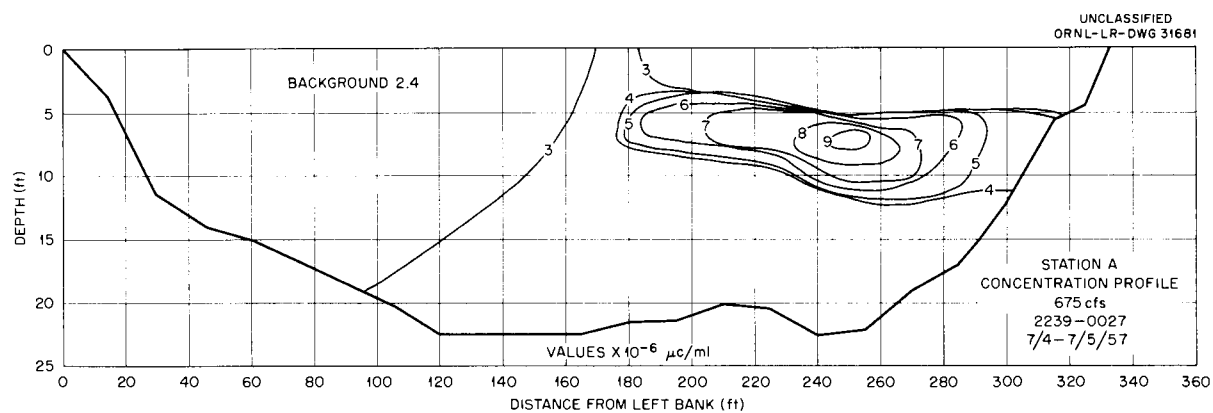


Fig. 15 (continued). Concentration Profiles at Clinch River Station A.

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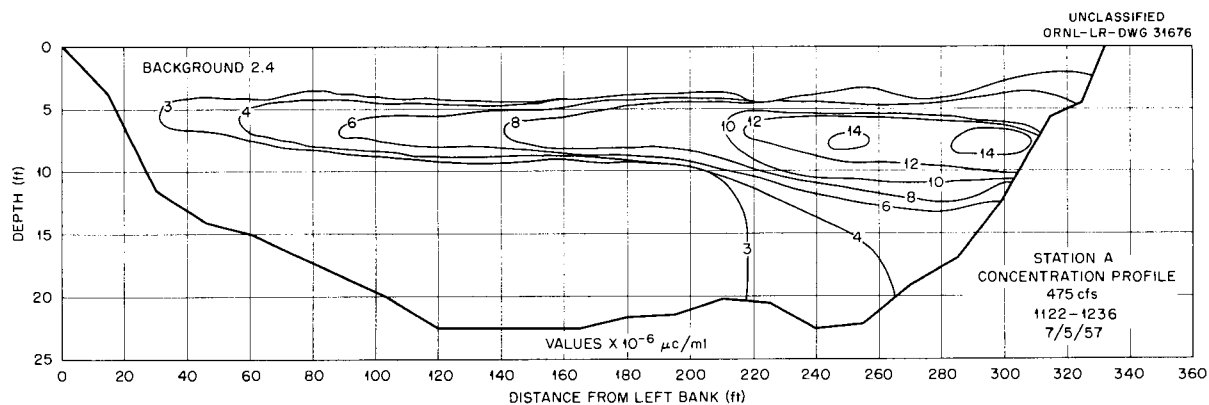
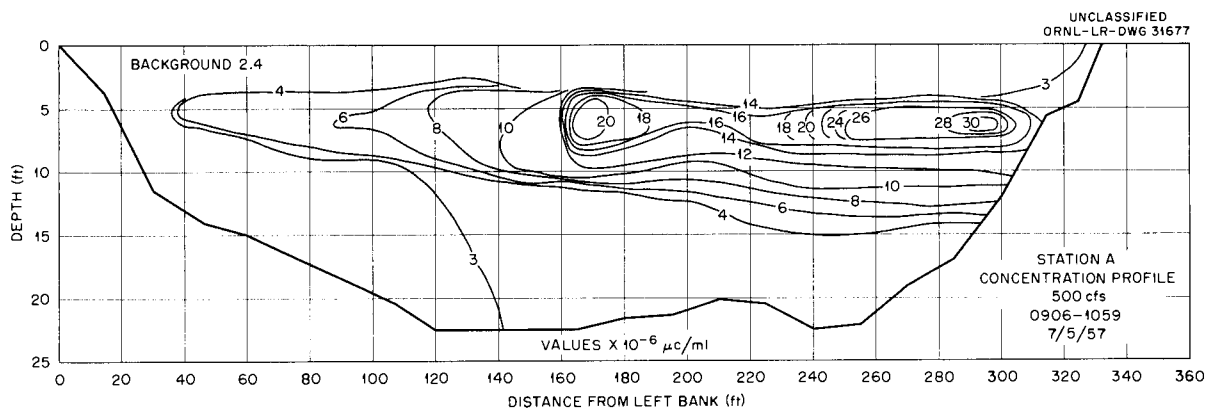
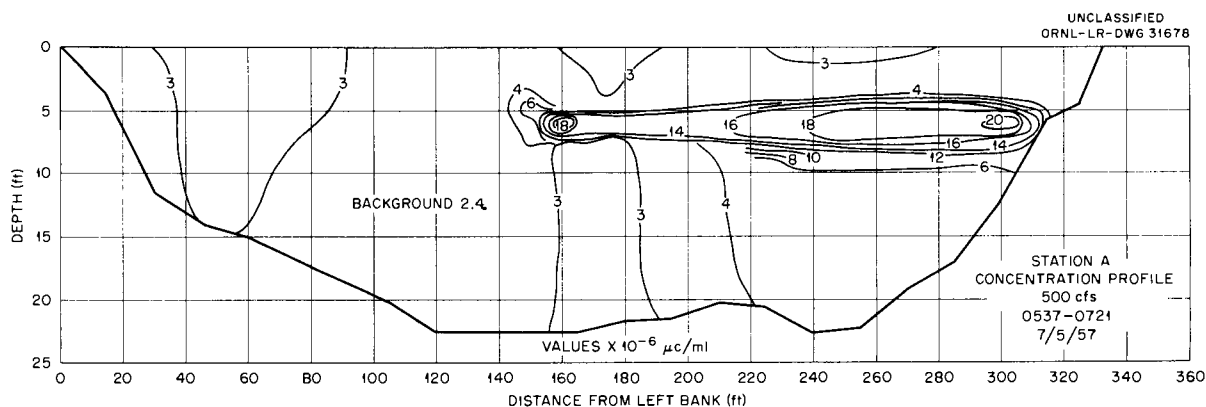


Fig. 15 (continued). Concentration Profiles at Clinch River Station A.

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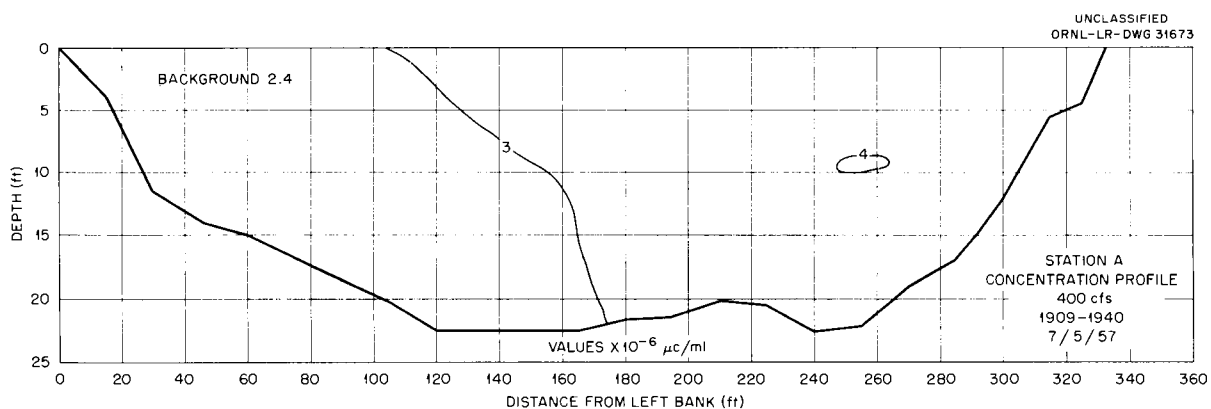
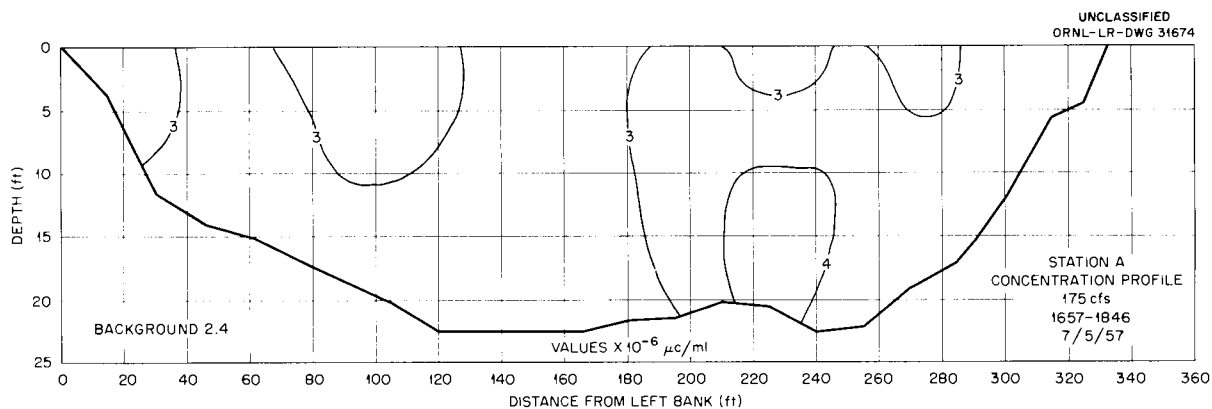
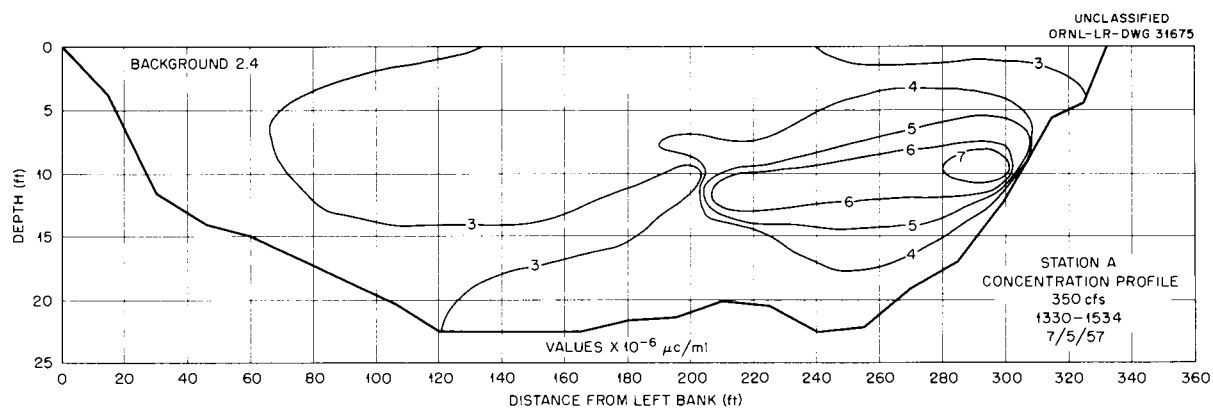


Fig. 15 (continued). Concentration Profiles at Clinch River Station A.

Table 6. The Removal of Cesium from Conasauga Shale by NaCl Solutions of Various Concentrations

Amount of NaCl (meq)	Cesium Removed (μ eq)*				
	NaCl				
	0.1 N	0.2 N	0.3 N	0.5 N	1.0 N
60	69.8	72.1	77.0	(59)	(62)
100	93.5	101.4	(96)	82.6	85.6
120	102.2	111.9	106.1	(93)	(97)
150	112.9	(126)**	117.0	105.0	(112)
180	123.9	135.8	126.2	(117)	(122)
200	128.4	141.2	(131)	123.1	126.3
300	144.8		150.5	145.2	146.0
400	153.3			159.3	157.6

*From 6 g of shale originally containing 225 μ eq of cesium.

**Values interpolated, not experimental.

as did 13.6 meq of NaCl. Thus NaHSO_3 was given an effectiveness rating of 1.36. The rank of the anions in order of decreasing effectiveness was HSO_3^- (1.36 ± 0.01) > NO_3^- (1.25 ± 0.10) > HCO_3^- (1.18 ± 0.02) > EDTA^{4-} (1.16 ± 0.02) = OAc^- (1.16 ± 0.02).

Synthetic waste solution of the same stable-element composition as that found in the waste pits was prepared and used as a leaching solution.¹⁵ The pH of the solution (originally 10.9) was adjusted by adding HCl. Leaching solutions of pH 10.9, 9.8, 6.9, 4.1, and 1.8 were used. The pH of the leaching solution had no effect on the amount of cesium leached until the pH was reduced to 1.8. Table 7 shows the influence of pH on the amount of cesium leached. The increased removal of cesium probably was due to an increase in the total ionic activity of the leaching solution rather than to a simple pH effect.

The fourth series of leachings involved the use of leaching solutions of various cations. These cations were compared with Na^+ (NaCl) for effectiveness in removing successive portions of the

Table 7. The Effect of the pH of the Leaching Solution on the Removal of Cesium from Conasauga Shale*

Leach No.	Cesium Removed (μ eq)				
	pH 10.9	pH 9.8	pH 6.9	pH 4.1	pH 1.8
1	37.6	33.9	36.0	35.8	51.5
2	65.2	58.3	64.3	62.8	93.3
3	85.0	76.8	84.1	81.5	120.6
4	99.6	92.7	98.8	96.0	137.6
5	112.0	106.3	110.6	107.9	149.2
6	121.1	117.3	119.9	118.1	157.5
7	129.3	126.3	127.7	126.2	164.2
8	136.2	131.9	134.3	133.6	169.7
9	142.8	137.3	140.0	139.7	174.1
10	149.2	142.2	145.1	145.3	177.8

*225 μ eq. per 6 g of shale (original).

sorbed cesium. The rank of the cations in order of decreasing effectiveness was Cs^+ (20.6 ± 4.3) > Rb^+ (15.3 ± 1.5) > H^+ (13.2 ± 1.6) > NH_4^+ (11.3 ± 3.5) > K^+ (9.2 ± 1.9) > Al^{+++} (7.5 ± 2.6) > Ca^{++} (6.2 ± 2.9) > methylene blue⁺ (1.3 ± 0.2). The relative effectiveness of Al^{+++} and Ca^{++} decreased markedly as the amount of cesium remaining on the shale decreased. The relative effectiveness of H^+ increased as the amount of cesium decreased, probably because of destruction of the clay mineral lattice when leached with 1 N HCl. As was expected, the nature of the cation of the leaching solution was of more importance in influencing cesium removal than was the anion.

Mineralogical analysis of the Conasauga shale from the waste-pit disposal area by the U.S. Geological Survey indicates that the shale is essentially illitic in nature. The illite group of clay minerals has a lattice consisting of an aluminum octahedral sheet sandwiched between two silicon tetrahedral sheets. The arrangement of the silicon tetrahedral sheet is such that there is an open hexagonal network of oxygen atoms on the exposed surface. Successive layers are stacked with the voids in the hexagonal networks superimposed. The void can accommodate spheres with a radius of $(\sqrt{5} - 1)$ times the radius of the surrounding atoms, or 1.64 Å. If cations of this approximate size, such as cesium (ionic radius of

¹⁵The composition of the synthetic waste solution was the same as that of a sample from the overflow of Waste Pit 3 to Pit 2 on Oct. 19, 1956. See W. J. Lacy, *Radioactive Waste Disposal Report on Seepage Pit Liquid Waste-Shale Column Experiment*, ORNL-2415 (Nov. 12, 1957).

1.65 Å), move into these exchange positions they become tightly bonded.¹⁶ In addition to the Coulombic forces of attraction resulting from the charge deficiency in the crystal lattice, the cations will be held by the formation of covalent bonds with the surrounding 12 oxygen atoms. The results of these experiments confirm this mechanism of cesium sorption.

Cesium-137, in detectable quantities, has not reached the test wells surrounding the ORNL waste pits.¹⁷ The fixation of cesium by Conasauga shale is responsible for the deterred lateral movement from the waste pits. The high affinity of the shale for cesium will also minimize the danger of Cs¹³⁷ reaching natural drainageways in appreciable concentrations after the abandonment of the waste pits. However, as a consequence of the restricted movement of Cs¹³⁷, the immediate vicinity of the waste pits has become highly contaminated. Though fixed Cs¹³⁷ is less available for utilization by biological systems than is exchangeable Cs¹³⁷, it is conceivable that, over an extended period of time, dispersal by biological systems may be the limiting factor in the safe disposal of intermediate-level radioactive wastes.

Development of Improved Well Probe

Existing radiologging equipment at ORNL is inadequate for efficient monitoring of observation wells in the waste-pit area. Chief limitations of the existing trailer-mounted rigs¹⁸ are slow logging speed, insufficient mobility for the terrain, and equipment unreliability and excessive maintenance.

To determine availability of better equipment a literature survey was undertaken, and contacts were made with several companies, other AEC installations, and other branches of the Government. In addition, two well loggers developed by the U.S. Geological Survey and one developed by the New York Operations Office of AEC were obtained for evaluation. All these studies indicated that a well logger is not available which is

adequate for monitoring the wells in the waste-pit area and that a new instrument must be developed.

A study of the requirements for the cable reel and feed mechanism indicated that a unit similar to the one used in the AEC instrument would satisfy the major requirements. Prints of the AEC unit were obtained, and such features as were adaptable were incorporated in a new design. This design incorporates additional features including: an overload clutch; automatic log, retract, and shutoff; resettable level-wind index; optional two-speed hand operation; and a greater range of automatic logging speeds. Scintillation and G-M detectors were evaluated for sensitivity, complexity, and reliability, and a G-M detector was selected for the new well logger.

Loggers with active impedance-matching networks such as cathode followers in the probe present reliability and maintenance problems, while those with passive matching networks present other problems. By utilizing the average current from the G-M tube, rather than the pulse rate, the necessity for impedance-matching networks is eliminated. A further advantage of current operation is greater simplicity in the count-rate meter, since only one vacuum tube is required.

A detailed study of the factors involved in current operation of halogen-quenched G-M tubes has been completed. The study confirmed the feasibility of current operation for this application, and a prototype unit has been laboratory tested. Construction of a field unit, design of a hydrostatic seal on the probe and cable, packaging of the count-rate meter and the power supply, as well as additional laboratory and field tests, are to be completed.

The entire assembly, including the electronic equipment, the reel assembly, and the power source, will be mounted on a frame for operation from a Jeep station wagon; each unit will be separately demountable.

Scintillation spectrometry was considered to be a highly desirable adjunct to gross gamma counting. A U.S. Geological Survey logging spectrometer from Grand Junction, Colorado, was tested in several ORNL wells in which the radiochemical compositions were known. The results of these tests indicated that spectrometry was not practical in this application without further development of the apparatus and technique. Further consideration of spectrometry has been postponed until completion of the gross-gamma-ray logger.

¹⁶J. B. Page and L. D. Baver, *Soil Sci. Soc. Amer. Proc.* 4, 150 (1939).

¹⁷K. E. Cowser and F. L. Parker, *Soil Disposal of Radioactive Wastes at ORNL; Criteria and Techniques of Site Selection and Monitoring*, paper presented at the Annual Health Physics Society Meeting at Pittsburgh, June 17-19, 1957; *Health Phys.* (to be published).

¹⁸J. M. Garner and B. Kahn, *H-P Semiann. Prog. Rep.* Jan. 31, 1954, ORNL-1684, p 22.

LAND BURIAL OF SOLID WASTES

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Selection of New ORNL Burial Ground

As a result of the increased use of the ORNL burial grounds by outside agencies and the increased amounts of solid wastes generated at X-10 and Y-12 over the past years, the average area required for burial has increased from about 1.5 to 5.0 acres per year. Off-site wastes account for about 35% of the volume. With the present rate of burial it appeared that the operating burial ground would be filled in four to six months and that a new burial ground would be needed.

A new burial ground should meet the following requirements:

1. It should be large enough to meet the needs of the Laboratory for the next four or five years.
2. It should be an area of gentle relief in which operation is easy and which should not be subject to flooding by surface water or a high ground-water table.
3. The soil should be easy to excavate with earth-moving equipment and firm enough to stand in steep cuts.
4. The area should lend itself to the inexpensive construction of private roads for hauling.
5. It should be located in Melton Valley, preferably in the White Oak Creek drainage basin.

The location of the present operating burial ground is indicated in Fig. 16, which is part of a detailed topographic map of the Melton Valley. Area E is the location of the waste disposal pits. Four areas were selected for a preliminary field survey. Areas A and A', believed to be the most suitable for a new burial ground, comprise 20-25 acres of gently to moderately sloping land. Area A' is a separate part of the proposed burial site. This area probably will be used for the disposal of high-level beta-gamma solid wastes in order to continue the policy of segregating those wastes from alpha-contaminated solids. While area B could be used for burial, it also meets the topographic requirements for seepage-pit disposal. Area C may be used as an extension to burial-ground operations in areas A and A'. However, area C consists of about 5 acres and does not have the burial capacity for a four- to five-year period. Area D is divided into small sections by

gulleys, several of which are deep. These gulleys are not indicated on the topographic map.

A series of auger holes (Fig. 17) were drilled as observation wells in areas A and A' for determining the depth to the water table and the character of the residual cover. The information will be used in the selection of the most suitable sites. Wells of greater depth will be drilled for determining the occurrence and circulation of ground water at greater depth.

ECOLOGICAL RESEARCH

S. I. Auerbach

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Ecological studies during the past year have been conducted in areas contaminated by waste disposal operations, with major emphasis devoted to determining the concentration and distribution of fission products in the soil, plant, and animal components of these environments. The bed of the former White Oak Lake and the forest areas immediately surrounding the radioactive waste pits are the environments under investigation. In connection with these studies, effort has been expended on the estimation of animal populations and community structure in these contaminated areas and in similar uncontaminated areas in order to provide a basis for more refined work on the turnover of radionuclides by these trophic levels of the ecosystem.

In addition to these studies on the fate of fission products in the environments, preliminary investigations were started on programs concerned with the long-term ecological effects of radiation. Included are studies of mammal populations in areas of the White Oak Lake bed which have an exposure rate of about 500 mr per day. In Melton Valley, which contains the contaminated areas, an analysis of the summer bird populations was performed.²²

¹⁹Consultant, University of Tennessee.

²⁰Temporary employee.

²¹ORINS research participant.

²²J. C. Howell, *Long-Range Ecological Study of the Oak Ridge Area: I. Observations on the Summer Birds in Melton Valley*, ORNL CF-58-6-14 (June 3, 1958).

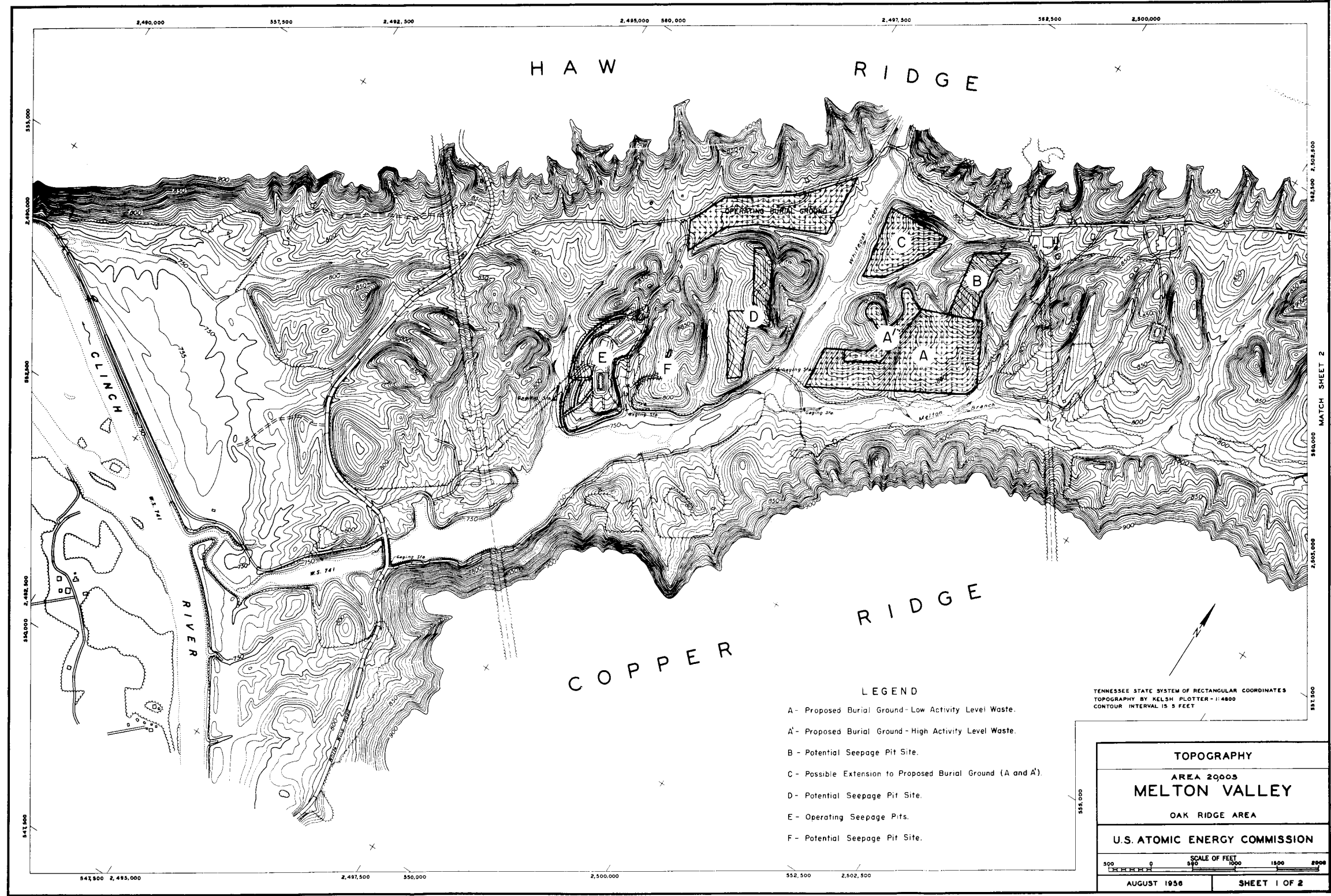


Fig. 16. Topographic Map of Melton Valley.

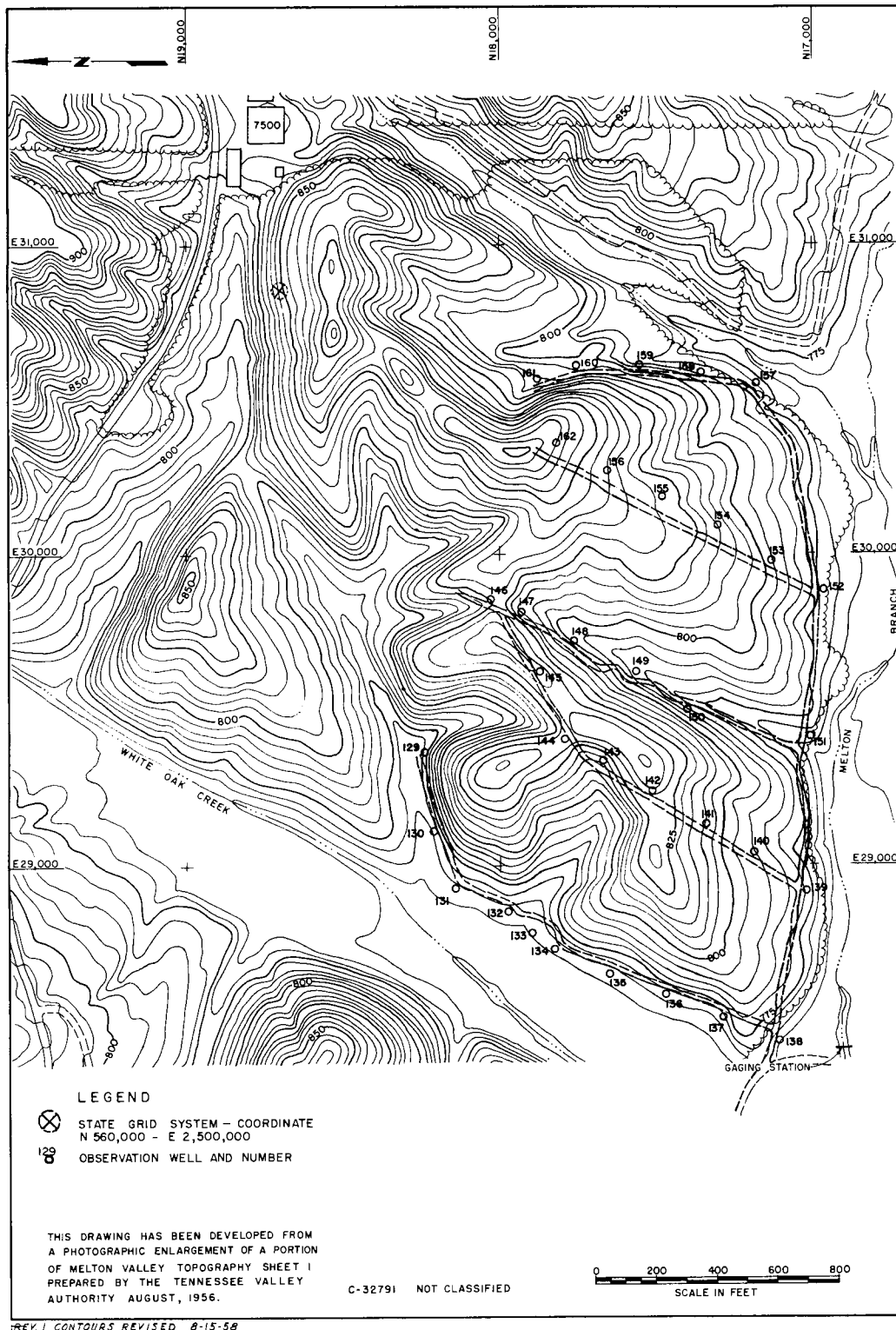


Fig. 17. Topography of Proposed Burial Ground and Location of Auger Holes.

Laboratory experiments on the uptake of Cs^{137} by algae typical of the Clinch River and its tributaries were performed and have been reported in the open literature.²³ Summer survey work on the plankton of the Clinch River and its local tributaries was continued.²⁴

White Oak Lake Bed Studies

Vegetation. – There was considerable change in the pattern of vegetation during the past year. This is to be expected because the area will tend to undergo plant succession in the succeeding years until the biota reaches an equilibrium in the local climate. The general pattern of vegetation in the ecology study grid is illustrated in Fig. 18. Instead of an almost pure stand of smartweed (*Polygonum* spp.), which was characteristic of the first growing season, there is present a variety of new plant associations. Among the more abundant of these associations is the black willow (*Salix nigra*), which is rapidly invading the

lake bed and which can be expected to dominate most of this area within a few years.

Soil Studies. – Additional analyses of the soils for fission products and common nutrient elements were made. Total fission product analyses were made at five locations in the lower part of the lake bed. These analyses, together with extrapolations to an acre basis, are shown in Table 8. Additional measures of fission products at other sites in the same general area are shown in Fig. 18. The differences exemplify the heterogeneity of distribution of fission products in this soil. In addition to these analyses more detailed studies were made in a 50- by 50-ft agricultural plot which was prepared in the upper part of the lake bed (Fig. 19). The plot was subdivided into rows and columns 12 ft in width. One hundred forty-four stations were marked, and a soil sample for analysis was taken from each just prior to seeding with corn (*Zea mays*).

The general chemical properties of the soil in this plot were typical of most of the lake bed. The water pH ranged from 6.86 to 7.50 and the salt pH from 6.66 to 7.21. The per cent carbonate ranged from 0.22 to 0.86. Aliquots of the 144 soil

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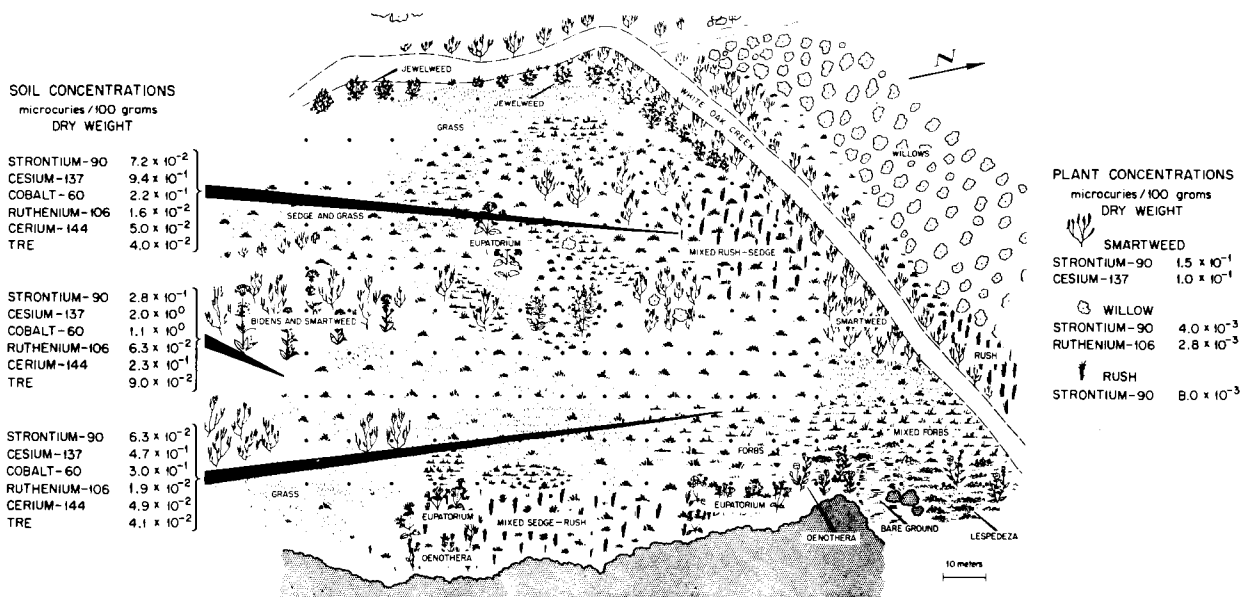


Fig. 18. Distribution of Vegetation in the Ecology Study Area of White Oak Lake Bed, July 1957.

Table 8. Concentrations of Fission Products in the Lower Part of White Oak Lake Bed
(microcuries per 100 g of dry soil)

Sample Station	Cs ¹³⁷	Sr ⁹⁰	Co ⁶⁰	Ce ¹⁴⁴ -Pr ¹⁴⁴	Ru ¹⁰⁶ -Rh ¹⁰⁶	Trivalent Rare Earths
ST-8	1.79	0.28	1.22	0.26	0.07	0.26
ST-9	1.92	0.28	1.22	0.16	0.05	0.16
ST-19	2.22	0.30	1.11	0.30	0.08	0.30
ST-7C	0.47	0.06	0.30	0.05	0.02	0.04
ST-10G	1.91	0.12	0.46	0.11	0.07	0.05
UT-1	0.94	0.07	0.22	0.05	0.02	0.04
Average Curies per Acre, 6-in. Depth						
	14.0	2.0	7.0	2.0	0.5	1.0



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Fig. 19. View of the Agricultural Plot in White Oak Lake Bed, June 1957.

samples were leached with 0.1 N HCl. Calcium, potassium, magnesium, and sodium concentrations were determined by means of a flame photometer.²⁵ Phosphorus was determined by the method of Bray.^{25,26} Table 9 gives the results of these analyses. Ammonium acetate (1 N) extractions of exchangeable calcium varied from 14.8 to 15.8 milliequivalents (meq) per 100 g of soil, exchangeable magnesium from 1.1 to 1.47 meq, potassium from 0.15 to 0.17 meq, and sodium from 0.14 to 0.16 meq.

An additional 48 aliquots of the soil samples were analyzed for total Cs¹³⁷ and Co⁶⁰ by gamma spectrometry (Table 10). Table 10 also contains the results of the Sr⁹⁰ analyses. These represent 1 N ammonium acetate leachings analyzed by the method of Kahn and Goldin.²⁷ Ammonium acetate is the standard reagent for extracting the exchangeable components in soil. In order to get a more complete characterization of the Sr⁹⁰ relationships in the soil of this plot, ten samples were leached consecutively with distilled water, dilute acid (0.1 N HCl), and strong acid (6 N HCl). These three extractions give the water-soluble component (readily available for plant uptake); the dilute-acid-extractable component, which includes the exchangeable fraction and sparingly soluble salts; the fraction extractable with strong acid, which would not be available; and the total of these. The analytical procedure for Sr⁹⁰ determination was a modification of that of Farabee.²⁸ The results are shown in Table 11. According to these data and those of the ammonium acetate extractions (Table 10), about 5.1% of the Sr⁹⁰ was in water-soluble form, 85.5% was in dilute-acid-extractable form, and about 9.3% was in strong-acid-extractable form. Exchangeable Sr⁹⁰ comprised 32.4% of the total, based on the average of the total Sr⁹⁰ shown in Table 11 and the mean concentration shown in Table 10.

The two extracting agents (0.1 N HCl and 1 N NH₄OAc) are among those commonly used

in soils analysis. For our purposes, the dilute-acid extraction seemed more feasible in that it permitted rapid analyses of large numbers of samples. Also, the dilute-acid-extract analyses gave larger estimates and thus were more conservative for studies of uptake by plants, since uptake would tend to be underestimated rather than overestimated. Dilute-acid values for calcium, potassium, and magnesium were 100% higher than the ammonium acetate extractions; for sodium, the dilute-acid extractions were 60% higher.

The agricultural plot was laid out in a 4 × 4 Latin square. This arrangement provided a method for evaluating the horizontal distribution of elements in the plot. A statistically significant difference between arithmetic means for rows could be demonstrated for most of the elements, but differences between arithmetic means for columns were not found. The rows were laid out parallel to the former shore of White Oak Lake. It is likely that the differences were the result of differential deposition of silt. From Tables 9 and 10 it can be seen that the differences between means for rows, although significant, were not large. However, the adjacent rows 3 and 4 contained the lowest and highest means for most of the elements, which indicates that microgeographic distribution must be considered when evaluating field-plot data. Comparison of these detailed analyses of soils with those obtained the previous summer from other parts of the lake bed shows that values for nutrient elements in the agricultural plot are somewhat lower. Possibly, this lowering is due to leaching during the winter and to the nonreplacement of these elements on the exchange complex after their removal by the vegetation of the previous season.

Plant Studies. — Samples of the natural vegetation on the lake bed were collected during the summer of 1957 and analyzed for radiostrontium, radiocesium, and for nutrient elements. Radiostrontium concentrations were determined by radiochemical methods, and radiocesium was determined by gamma spectrometry. A flame photometer was used to determine Ca, K, Mg, and Na concentrations.

Among the plant species sampled were *Solidago* spp., *Rhus glabra* L., *Salix nigra* Marsh., *Juncus* spp., *Fraxinus* sp., *Impatiens pallida* Nutt., *Eupatorium serotinum* Michx., *Polygonum lapathifolium* L., *Oenothera biennis* L., *Bidens frondosa* L., and *Phytolacca americana* L. The samples

²⁵Analyses performed by Special Analyses Group, Analytical Chemistry Division.

²⁶R. H. Bray, *Diagnostic Techniques for Soils and Crops* (ed. by H. B. Kitchem), p 53-86, American Potash Institute, Washington, 1948.

²⁷B. Kahn and A. S. Goldin, *J. Amer. Water Works Assoc.* 49, 767-71 (1957).

²⁸L. B. Farabee, *Procedure for the Radiochemical Analysis of Strontium and Barium in Human Urine*, ORNL-1932, p 1-19 (Feb. 6, 1955).

Table 9. Analyses of Nutrient Elements in White Oak Lake Bed Agricultural Plot, Spring 1957

	Element	Column 1	Column 2	Column 3	Column 4	Row Means		
Row 1	Ca ^(a)	33.37	33.27	34.43	33.57	33.66		
	Na ^(a)	0.145	0.146	0.120	0.118	0.132		
	K ^(a)	0.247	0.250	0.233	0.241	0.243		
	Mg ^(a)	1.20	1.48	1.50	1.51	1.44		
	P ^(b)	0.52	0.84	0.61	0.70	0.67		
Row 2	Ca	33.09	36.61	28.30	26.93	31.23		
	Na	0.123	0.124	0.116	0.101	0.116		
	K	0.225	0.225	0.196	0.191	0.209		
	Mg	1.45	1.44	1.37	1.37	1.39		
	P	0.60	1.52	1.08	1.22	1.10		
Row 3	Ca	30.30	31.51	26.76	23.33	27.98		
	Na	0.111	0.102	0.108	0.099	0.105		
	K	0.206	0.209	0.204	0.165	0.196		
	Mg	1.46	1.40	1.31	1.30	1.37		
	P	1.02	1.02	1.22	0.96	1.06		
Row 4	Ca	35.52	39.71	38.28	42.27	38.94		
	Na	0.138	0.158	0.169	0.178	0.161		
	K	0.224	0.225	0.224	0.240	0.228		
	Mg	1.41	1.45	1.50	1.82	1.55		
	P	1.40	1.24	1.28	1.28	1.30		
						Grand Means		
						$\bar{x}^{(c)}$	$s^{(d)}$	$n^{(e)}$
Column means	Ca	33.07	35.28	31.94	31.53	32.95	7.76	144
	Na	0.129	0.133	0.128	0.124	0.128	0.021	144
	K	0.226	0.227	0.214	0.209	0.219	0.028	144
	Mg	1.38	1.44	1.43	1.49	1.44	0.21	144
	P	0.89	1.16	1.06	1.04	1.04	0.32	143

(a) meq per 100 g of soil.

(b) ppm.

(c) Mean.

(d) Standard deviation.

(e) Number of replicates.

Table 10. Concentrations of Important Fission Products in White Oak Lake Bed Agricultural Plot, Spring 1957
(microcuries per 100 g of soil)

	Element	Column 1	Column 2	Column 3	Column 4	Row Means		
Row 1	Co ⁶⁰ (a)	0.34	0.33	0.35	0.41	0.36		
	Cs ¹³⁷ (a)	1.59	1.65	1.34	2.05	1.66		
	Sr ⁹⁰ (b)	0.039	0.032	0.028	0.031	0.032		
Row 2	Co ⁶⁰	0.43	0.43	0.35	0.29	0.38		
	Cs ¹³⁷	1.78	1.53	1.46	1.43	1.55		
	Sr ⁹⁰	0.026	0.028	0.035	0.031	0.030		
Row 3	Co ⁶⁰	0.35	0.35	0.28	0.21	0.30		
	Cs ¹³⁷	1.15	1.35	1.04	0.71	1.06		
	Sr ⁹⁰	0.021	0.031	0.027	0.018	0.024		
Row 4	Co ⁶⁰	0.31	0.46	0.39	0.46	0.41		
	Cs ¹³⁷	1.03	1.87	1.76	1.84	1.63		
	Sr ⁹⁰	0.039	0.039	0.035	0.038	0.038		
						Grand Means		
						$\bar{x}^{(c)}$	$s^{(d)}$	$n^{(e)}$
Column means	Co ⁶⁰	0.36	0.39	0.35	0.34	0.36	0.13	48
	Cs ¹³⁷	1.39	1.60	1.40	1.51	1.47	0.47	48
	Sr ⁹⁰	0.031	0.033	0.031	0.029	0.031	0.0066	48

(a) Total concentrations analyzed by gamma spectrametric analysis.

(b) 1 M ammonium acetate extracts analyzed radiochemically.

(c) Mean.

(d) Standard deviation.

(e) Number of replicates.

were taken from various areas of the lake bed at the end of the growing season. The sampling was random, and the number of replicates ranged from two to five. Different plant parts (leaves, stems, petioles, flowers, and berries) were analyzed separately, where possible.

Concentrations of Sr⁹⁰, of Cs¹³⁷, and of nutrient elements in typical species of natural vegetation are given in Table 12. Differences between species of plants, as shown here, may not be significant since no consideration was given to the habitat of the individual plant. Highest radiocesium concentrations were found in *Solidago* spp. and *Impatiens pallida* Nutt. Comparison of plant

parts indicates that leaves and flowers tend to contain higher concentrations of radionuclides and nutrient elements than do the stems. Radiocesium values for petioles (two species) and berries (*Phytolacca* only) approximated those for leaves and flowers.

The Sr⁹⁰-to-calcium and the Cs¹³⁷-to-potassium relationships in these plant samples were examined. Radiostrontium varied directly with calcium both intra- and interspecifically. Intraspecifically, radiocesium varied independently of potassium. When the average values for all species were used, the concentration of Cs¹³⁷ was found to be proportional to the concentration of potassium.

Table 11. Comparison of Sr^{90} Concentrations in White Oak Lake Bed Agricultural Plot Obtained by Different Extracting Agents*
($\mu\text{c} \times 10^{-3}$ per 100 g of soil, dry wt)

Sample No.	H_2O	0.1 N HCl	6 N HCl	Total
1	4.3	59.9	11.7	75.9
2	4.4	76.8	2.7	83.9
3	3.9	85.6	6.6	96.1
4	4.8	63.7	10.2	78.7
5	2.8	73.5	4.4	80.7
6	2.4	57.0	9.4	68.8
7	5.3	82.0	3.6	90.9
8	4.6	60.0	9.9	74.5
9	3.8	74.8	4.7	83.3
10	6.2	73.2	13.8	93.2
Av	4.2	70.6	7.7	82.6
Av % of total	5.1	85.5	9.3	99.9

*The availability of radiostrontium to plants is represented by the differential extractability of strontium by leaching solutions of different hydrogen ion concentration. The distilled-water and dilute-acid (0.1 N HCl) extracts can be considered available for plant uptake, while the strong-acid (6.0 N HCl) extract can be considered unavailable.

Thus, it would appear that those species which accumulate potassium or calcium also tend to accumulate cesium or strontium, respectively. No relationship was found between concentrations of radiostrontium and concentrations of potassium or magnesium nor between radiocesium and concentrations of calcium or magnesium.

In order to obtain more explicit information on these soil-plant relationships a corn (*Zea mays*) crop was planted in the agricultural plot. Four common varieties, namely, Aristogold Hybrid, Country Gentleman, Golden Bantam, and Hickory King were planted in May 1957. Sampling was done in three stages: leaf samples were taken separately at the appearance of the ear for each variety, ear samples were taken at maturity, and stem and flower samples were taken at the end of the season. The samples were dried, ground, and analyzed for chemical content by standard

methods. For radioanalysis, plant samples were dry-ashed at 450°C ; Cs^{137} was determined by gamma spectrometry, and Sr^{90} was determined by using a modification of the method of Farabee.²⁸

Successful growth and yield were obtained for the two field-corn varieties (Hickory King and Aristogold Hybrid). The garden corn varieties were severely stunted. This differential success was attributed to a combination of climatic (extreme rainfall followed by a period of drought) and soil (phosphorus deficiency) conditions. As a result of this unequal growth, sampling of the corn could not be conducted according to the Latin square design which had been laid out. Data for the radionuclides and important nutrient elements are summarized by plant part in Table 13. Comparisons of varieties were not possible because of the inadequate growth and thus inadequate replication. Such values as were obtained for the garden corn fell well within the range of those for the field corn.

Both radiocesium and radiostrontium were found in higher concentrations in leaves and flowers than in the other plant parts sampled (Table 13). Potassium, calcium, sodium, and magnesium concentrations appeared higher in leaves than in other parts. Analyses of variance for these elements show that the means for plant parts, as given in Table 13, were significantly different. Concentrations of Sr^{90} in the samples were found to be directly related to the concentrations of calcium in these samples. Cesium-137 concentrations were not directly related to potassium concentrations; flower samples had high cesium values, but low potassium values. In the translocation of strontium and cesium to the higher aerial organs, there does not seem to be stepwise discrimination against these elements (except in the case of grain); in fact, the reverse seems to be true.

The uptake of radiostrontium and radiocesium is currently being evaluated by comparison with the uptake of calcium and potassium, respectively. Strontium/calcium ratios for the wild-plant species (averages of plant parts) varied around 7.0×10^4 μc of Sr^{90} per 1000 g of calcium. The range of the ratios was 5.5 to 8.2×10^4 . With the use of an average ratio for the soil (0.1 N HCl extract) of 5.0×10^4 , discrimination factors slightly greater than 1 were obtained for these plants. Strontium/calcium ratios in the more detailed corn

Table 12. Concentrations of Elements in Natural Vegetation

Values are means of two to five replicates

Part of Plant	Constituent	Plant						
		<i>Solidago</i>	<i>Rhus</i>	<i>Eupatorium</i>	<i>Polygonum</i>	<i>Bidens</i>	<i>Impatiens</i>	<i>Fraxinus</i>
Flowers	Sr ^{90(a)}	0.028		0.041	0.024	0.031		
	Cs ^{137(a)}	0.040		0.014	0.011	0.012		
	K ^(b)	1.47		1.58	1.13	1.82		
	Ca ^(b)	0.61		0.79	0.32	0.75		
	Mg ^(b)	0.04		0.09	0.09	0.11		
Stems	Sr ⁹⁰	0.052	0.114	0.028	0.062	0.024		
	Cs ¹³⁷	0.018	0.006	0.005	0.005	0.005	0.064	0.007
	K	0.78	0.55	0.87	2.13	0.44	2.34	0.37
	Ca	0.54	1.19	0.35	1.02	0.64	3.91	0.67
	Mg	0.11	0.10	0.19	0.33	0.09	0.32	0.07
Leaves	Sr ⁹⁰	0.094	0.112	0.098	0.104	0.051		0.082
	Cs ¹³⁷	0.279	0.019	0.008	0.018	0.008	0.074	0.023
	K	1.37	1.09	1.02	1.14	1.18	1.32	0.79
	Ca	1.55	1.84	2.05	1.80	2.44	2.74	1.74
	Mg	0.25	0.13	0.20	0.53	0.31	0.21	0.25

^(a) μC per 100 g, dry wt.^(b) g per 100 g, dry wt.Table 13. Concentrations of Elements in Corn (*Zea mays*)

The numbers represent mean values

Constituent	Part of Plant						Least Significant Difference
	Leaf	Husk	Grain	Cob	Stem	Flower	
Sr ^{90(a)}	39.7	6.3	2.5	4.4	12.1	28.4	11.00
Cs ^{137(a)}	49.1	21.9	12.7	19.5	17.1	69.1	6.38
K ^(b)	1.54	0.98	1.21	0.94	0.67	0.64	0.25
Ca ^(b)	0.568	0.111	0.107	0.0568	0.165	0.228	0.047
Na ^(b)	0.0133	0.0049	0.0041	0.0034	0.0028	0.0054	0.0041
P ^(b)	0.163	0.153	0.174	0.154	0.186	0.071	0.056
Mg ^(b)	0.166	0.0656	0.0707	0.0437	0.0895	0.111	0.041

^(a) $\mu\text{C} \times 10^{-3}$ per 100 g, dry wt.^(b) g per 100 g, dry wt.

studies averaged 7.1×10^4 ; for plant parts, these ranged from 2.3×10^4 (grain) to 12.5×10^4 (flower). Discrimination factors in the corn varied around 1; for plant parts these were from 0.5 (grain) to 2.8 (flower). These discrimination factors are similar to those reported by others²⁹ for different species of plants. It should be noted that even though the calcium level in the corn plot was high, there was little or no discrimination against radiostrontium. This reinforces the view of others^{29,30} that beyond a certain concentration, calcium has little or no depressant effect upon the uptake of Sr^{90} . The use of the ratio of Cs^{137} to potassium has not been as widespread nor is it as well founded as the strontium/calcium ratio. For comparative purposes, the ratio expressed as millimicrocuries of Cs^{137} per 1000 g of potassium is used here. The Cs^{137}/K ratios of the several species of native plants (averages of plant parts) ranged from 0.8 to 8.4×10^4 . Using an over-all average value for the soil of 1.4×10^5 , discrimination factors varied from 0.06 to 0.60. In the corn, the radiocesium/potassium ratio averaged 2.7×10^4 ; plant parts ranged from 1.0×10^4 (grain) to 10.8×10^4 (flowers). Discrimination factors based on dilute-acid extracts of the agricultural plot soil (ratio 1.4×10^5) varied from 0.07 to 0.77. A tenfold difference in discrimination factors exists both between species and between plant parts. The combination of these suggests that differences of two orders of magnitude might exist between plant parts of different species. Such differences are seen in our wild-plant samples; for example, *Solidago* spp. leaves had 203.0×10^3 m μc of Cs^{137} per 1000 g of potassium, while *Polygonum* stems had 2.0×10^3 . In spite of the discrimination against Cs^{137} , this nuclide was present in the corn plants in concentrations higher than those of Sr^{90} (Table 13). By comparing the corn data (Table 13) with those for the native plants (Table 12), it can be seen that the calcium and the Sr^{90} values for the corn are lower than those for the wild species.

High leaf values for both Cs^{137} and Sr^{90} , compared with stems and fruits, have been reported

by others for various species of plants.³¹ Few workers have given flower concentrations; those reported are lower than for leaves. In both corn and natural vegetation from the lake bed, flowers had concentrations of Sr^{90} and Cs^{137} equal to or greater than the concentrations in leaves. The genetic implications of high flower concentrations are obvious.

Paired soil-plant measurements on the corn plot would have permitted a more careful evaluation of radiostrontium-calcium and radiocesium-potassium relationships. Because of the partial failure of the crop, only a few such paired values for the various elements could be obtained. No relationships could be seen between plant and soil values for either the radioactive or the stable elements. Possibly, soil variation was too slight to be reflected in the corn plants; also, the soil sampling method may not have given sufficient basis for pairing with individual plants.

Mean values for the corn crop and the samples of natural vegetation are sufficiently similar to suggest that these averages are reliable, but accurate measures of the extent of variation in the vegetation will require further study.

Insect Studies. — In the summer of 1956, a series of samples of the insect fauna on vegetation of the lake bed was taken. The purposes of the program were to ascertain summer population size and biomass relationships for these insects so that estimates could be made of the amount of uptake and turnover of fission products in these levels of the ecological food web.

The insect species and their relations to the major vegetation types (smartweed, sedge-rush, and willow) have been investigated. Estimates of the standing crop of herbivorous insects per square meter of vegetation averaged approximately 200 mg dry weight for the sedge-rush and smartweed areas and 100 mg dry weight for the willow areas. Most of the samples of herbivorous insects had gamma counting rates higher than background; samples taken later in the summer had progressively higher counting rates.

Estimates of the maximum concentrations of Cs^{137} in the herbivorous insects were made by assuming that the majority of the gamma counts

²⁹C. L. Comar, R. S. Russell, and R. H. Wasserman, *Science* 126, 485-92 (1957).

³⁰H. Nishita and K. H. Larson, *Summary of Certain Trends in Soil-Plant Relationship Studies of the Biological Availability of Fall-Out Debris*, UCLA-401 (July 28, 1957).

³¹R. S. Russell, H. M. Squire, and R. P. Martin, *The Effects of Operation Hurricane on Plants and Soils*, AERE-SPAR-3, p 1-74 (July 1955).

were due to this nuclide. These data are given in Table 14. Concentrations of Cs^{137} in the leaves are of the same order of magnitude as the concentrations in these insects. Current research effort is concerned with the further clarification of these relationships by studies of the uptake of fission products by some of the more abundant species of insects on the lake bed.

Table 14. Amounts of Cs^{137} in Nine Samples of Herbivorous Insects Collected on Three Host Plants ($\mu\text{c} \times 10^{-4}$ per 200 mg, dry wt, estimated by assuming that cesium is responsible for majority of gamma counts)

Week Collected	Amount of Cs^{137}		
	Smartweed	Sedge	Willow
2	0.38		
3	0.81		
5		0.42	
6	2.56		
7		1.24	
9	3.26		
10	3.04	1.22	6.00

Soil Microarthropod Studies. — A sampling program for soil arthropods on White Oak Lake bed was begun in October 1956 and continued during 1957. The objectives are to identify the species and estimate the numbers of arthropods in lake bed soils, to follow the succession of arthropods as the lake bed soils and vegetation mature, and, eventually, to estimate the role of these arthropods in the turnover of radionuclides in the lake bed ecosystem.

The sampling technique and early results were given in a previous report.³² The present report extends these data and gives results on species composition, community structure, and influence of soil moisture on the soil population of arthropods.

At least forty species of mites (Acarina) are known to occur in the soils of White Oak Lake bed. Most have been identified as to genus, but

specific identifications have been made for only a few. Table 15 lists these mites by genus, and gives food habits, abundance of individuals, and frequency of occurrence in samples for the abundant genera. Collembola were collected, but have not yet been identified beyond the family.

The abundance of the mite genera, Table 15, is given as the percentage of the total number of individuals (2722) collected during the study. Genera with less than 1% abundance are listed as "rare." Frequency of observation, the per cent of the 85 samples that contained the species, is given for those with abundances of 1% or more. Ten genera have abundances of 1% or more; together these constitute 90% of the total number of individuals. *Eupodes* (42%), *Tarsonemus* (25%), and *Oppia* (10%) are clearly the most abundant mite genera in lake bed soils; these genera also occur with high frequency (from 48% for *Oppia* to 71% for *Eupodes*).

The food habits given in Table 15 were gleaned from the literature. The food habits of most of the mites are not definitely known, but have been deduced from the habits of a few similar species and from comparisons of morphology and habitat. The term "reducer," used for the sarcoptiform mites, requires some clarification. These mites are associated with decaying plant materials, and whether they feed on the coarse litter, the organic products of decomposition, or the fungi associated with decomposition is not known. Therefore the term "reducer" is used in contrast to the other food habits listed in Table 15.

The considerations of frequency and abundance over the entire period of study do not give an adequate representation of the soil fauna, since the community structure changes with time. Table 16 gives estimates of the numbers of dominant soil microarthropods per square meter in six sampling periods. Initially, the phytophagous prostigmatids (*Tarsonemus* and *Eupodes*) were predominant; in later samples these mites declined as *Oppia* increased. The trend is probably a successional one; *Oppia* is a litter inhabitant, and, seemingly, the increase in number of this mite is related to the increasing amount of litter.

One of the factors affecting microarthropod populations is soil moisture. Moisture in the soil varies extremely on the lake bed because of the periodic flooding by runoff water from the surrounding environs and the lack of sufficient plant

³²R. M. Anderson *et al.*, H-P Ann. Prog. Rep. July 31, 1957, ORNL-2384, p 20.

Table 15. Mite Fauna of White Oak Lake Bed Soils, October 1956 to February 1958

Mite	Food Habits	Abundance ^a	Frequency ^b
Suborder Trombidiformes			
<i>Alicorhagia</i>	Predator	Rare ^c	
<i>Bdella</i>	Predator	Rare	
<i>Coccotydeus</i>	Predator	Rare	
<i>Ereynetes</i>	Predator	Rare	
Eupodidae genus	?	Rare	
<i>Eupodes</i>	Herbivore	42	71
<i>Nanorchestes</i>	Predator	2	4
<i>Pachygnathus</i>	Predator	Rare	
Pyemotidae genus	?	Rare	
Raphignathidae genus	Predator	Rare	
<i>Rhagidia</i>	Predator	Rare	
Scutacaridae genus	Parasite	3	37
<i>Tarsonemus</i>	Herbivore	25	68
<i>Tydeus</i>	Predator	1	17
Suborder Mesostigmata			
Aceosejidae genus	Predator	1	19
<i>Amblyseius</i>	Predator	Rare	
<i>Arctoseius</i>	Predator	2	26
Ascaidae genus	Predator	Rare	
<i>Hypoaspis</i>	Predator	Rare	
Laelaptidae genus	?	Rare	
Macrochelidae genus	Predator	Rare	
Neoparasitidae genus	Predator	Rare	
Parasitidae genus	Predator	Rare	
<i>Platyseius</i>	Predator	Rare	
<i>Rhodacarus</i>	Predator	Rare	
<i>Typhlodromus</i>	Predator	Rare	
Suborder Sarcoptiformes			
Acaridae genus	Reducer	3	19
Eremaeidae genus	Reducer	Rare	
Hypochthoniidae genus	Reducer	Rare	
<i>Oppia</i>	Reducer	10	48
<i>Pelops</i>	Reducer	1	17
<i>Platynothrus</i>	Reducer	Rare	

^aPercentage of total number of individuals.^bPercentage of samples containing mites of abundant species.^cLess than 1%.

Table 16. Dominant Soil Arthropods of White Oak Lake Bed
Each figure based on 10–15 samples

Arthropod	Estimated Number per Square Meter					
	October 1956	January 1957	May 1957	July 1957	December 1957	February 1958
Insecta						
Entomobryidae	3,300	2,600	3,600	6,800	3,100	600
Sminthuridae	1,000	1,600	200	1,300	6,400	6,000
Acarina						
<i>Eupodes</i>	32,600	2,100	10,900	13,000	300	100
<i>Tarsonemus</i>	8,600	22,500	1,900	5,700	900	200
<i>Oppia</i>	600	500	3,000	3,600	600	4,900
Acaridae	2,200	700	100	100	1,000	0

cover to prevent extreme drying during drought. Moisture content of the soil samples was measured by weighing the samples before and after drying in the Berlese apparatus; the weight change was expressed as percentage moisture.

That higher soil moistures have a depressant effect on the number of microarthropods is shown in Fig. 20. Association between the amount of soil moisture and the numbers of animals was tested by the "corner test" of Olmstead and Tukey³³ and found to be present ($P = 0.01$). However, it may be seen from Fig. 20 that the trend is a weak one. Figure 21 shows the influence of soil moisture on the presence of three different arthropods, *Eupodes*, *Oppia*, and Sminthuridae, in soil samples. Frequencies of these arthropod species in the samples are expressed as proportions of the total number of soil samples at each soil-moisture level. It is seen that Sminthuridae increase in frequency at higher moisture levels, *Oppia* is little affected, and *Eupodes* decreases at higher levels of soil moisture. *Tarsonemus* (not illustrated) also decreases with high soil moistures. Little relationship could be detected for the other, less numerous species. Thus, it appears that the extremes of soil moisture may exert a marked influence on the composition of the soil microarthropod fauna. As succession continues, the extremes of soil moisture

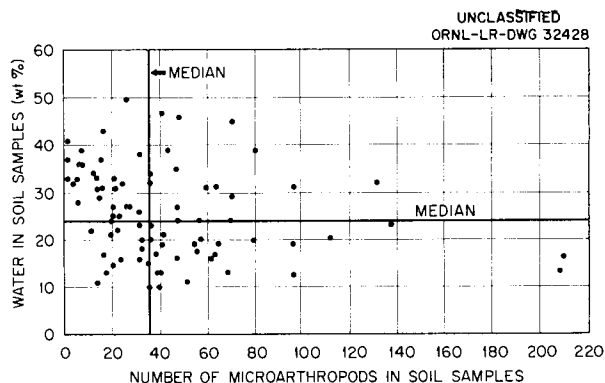


Fig. 20. Number of Microarthropods and Weight Per Cent of Water for 85 Soil Samples.

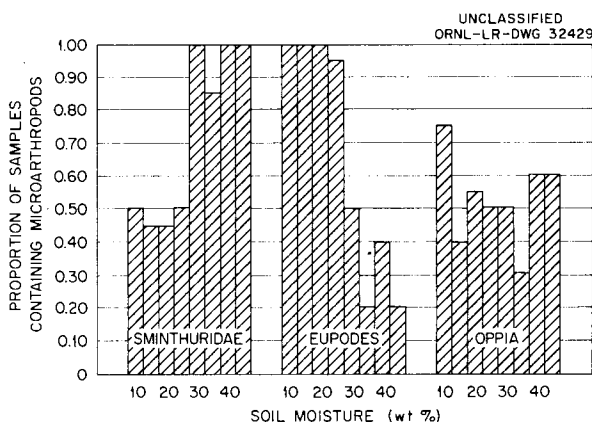


Fig. 21. Effect of Soil Moisture on the Presence of Three Species of Microarthropods in Soil Samples.

³³P. S. Olmstead and J. W. Tukey, *Ann. Math. Statistics* 18, 495–513 (1947).

may become the principal limiting factor for the White Oak Lake bed microarthropods in the soil.

Small-Mammal Program. — A small-mammal program was started in December 1957. The purpose of this program is to conduct long-term studies of small mammals (e.g., rats, mice, and shrews) in areas contaminated with low-level radioactive wastes. Essentially, this program is directed toward the problems of low-level chronic exposure, both internal and external, and is related to many aspects of fallout. Small mammals are especially useful in studies of this kind because they are prolific, easily handled, and may spend their entire lives within a study area of a few acres. These animals in the radioactive areas are subjected to constant external radiation (~ 20 mr/hr) and are continually ingesting various radionuclides. In order to determine the effects of these exposures it may be necessary to examine many individuals over a long period.

Before sound deductions can be made about the mammals, much basic information is necessary. Prerequisite information includes facts about the individuals (species, sex, weight, age, and reproduction) and, as a result, facts about the populations (density, structure, and change).

Most of the field work so far has been done in two areas, one on the dry bed of the former impoundment known as White Oak Lake and the other, a control area, located approximately one-quarter mile below the confluence of the Clinch River and White Oak Creek. The lake bed trapping area contains about 2.5 acres, but eventually will include about 9.3 acres. The control area covers 10.9 acres. Trapping stations, marked by stakes, are placed in the two areas in a grid pattern with 10 meters between stations, forming squares having an area of 1 are with stations at each corner.

In the White Oak Lake area from the first of April through June, cotton rats (*Sigmodon hispidus*), rice rats (*Oryzomys palustris*), white-footed mice (*Peromyscus leucopus*), and house mice (*Mus musculus*) were caught. During the same period pine mice (*Pitymys pinetorum*), white-footed mice, harvest mice (*Reithrodontomys humulis*), and house mice were caught in the control area. Mammal succession in the White Oak Lake area is indicated in Fig. 22 for the times shown. The 1956 and 1957 work was exploratory, and consequently the data are not extensive. As may be seen, the house mice were present when the area was first trapped, increased to a fairly high population

during the last month of 1956 and first half of 1957, and decreased until only an occasional individual was taken in 1958. The cotton rats appeared during the summer of 1957 and rapidly built up to a high population that is still present. Rice rats were first recorded in the April 1958 trapping but probably appeared during the interim between September 1957 and April 1958. White-footed mice occur as occasional visitors from nearby areas, and the short-tailed shrew (*Blarina brevicauda*), least shrew (*Cryptotis parva*), and Norway rat (*Rattus norvegicus*) occurred only as wandering individuals.

Waste Pit Studies

Tree Sampling Program. — The initial studies on contamination of trees by fission products from the radioactive waste pits were continued during June and July 1957, and additional samples were taken in April, May, and June 1958. The trees sampled included most of the monitor trees chosen in 1957. Because of new patterns of contamination, particularly those caused by the appearance of Cs^{137} in the trees, additional trees were added this year. The species of trees, together with their location in relation to the nearest core well, are listed in Table 17. The locations of the trees in relation to the waste pits are shown in Fig. 23.

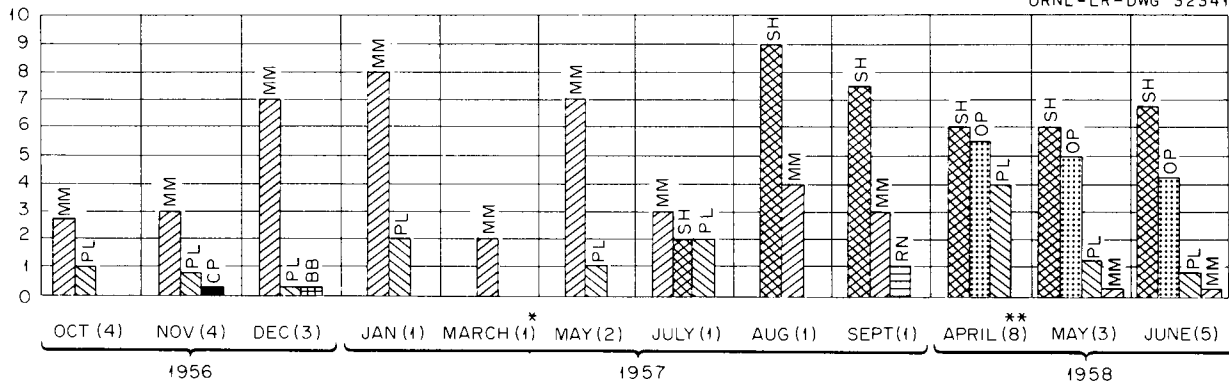
The monitor trees were divided into two groups. Those growing on the western portion of the pit area are designated as "west seep" trees and included Nos. 20, 24, 26, 38, 39, and 40. Those trees growing on the eastern portion are designated as "east seep" and include trees 10, 15, 16, 19, 29, 31, 33, 34, and 41.

Leaves and twigs were collected at various heights up to 24 ft. Litter samples were collected from immediately beneath the tree. All samples were dried, ground in a Wiley mill, and dry-ashed at temperatures ranging from 350–450°C. Samples taken in June and July 1957 were analyzed radiochemically for Ru^{106} . All subsequent samples were analyzed by gamma spectrometry.³⁴

Concentrations of Ru^{106} found in various parts of these trees, including ground litter, are summarized in Table 18. The concentrations of Cs^{137} , Zr^{95} - Nb^{95} , and Co^{60} found in these trees are summarized in Table 19. In addition, Ce^{144} has been detected in many of the samples.

³⁴ Analyses performed by Radiochemistry Group, Research and Development Section, Analytical Chemistry Division.

HEALTH PHYSICS ANNUAL PROGRESS REPORT

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* AREA MUDDY FROM FLOOD AND RAINS

** FIVE COTTON RATS AND SIX RICE RATS DROWNED IN TRAPS ON LAST NIGHT THAT TRAPS WERE SET

- [Cross-hatched] SHORT-TAILED SHREW (*BLARINA BREVICAUDA*)
 [Solid black] LEAST SHREW (*CRYPTOTIS PARVA*)
 [Diagonal lines] HOUSE MOUSE (*MUS MUSCULUS*)
 [Dotted] RICE RAT (*ORYZOMYS PALUSTRIS*)
 [Horizontal lines] WHITE-FOOTED MOUSE (*PEROMYSCUS LEUCOPUS*)
 [Vertical lines] NORWAY RAT (*RATTUS NORVEGICUS*)
 [Cross-hatched] COTTON RAT (*SIGMODON HISPIDUS*)

Fig. 22. Small Mammals Present in White Oak Lake Bed in 1956, 1957, and 1958. The number in parentheses after the month indicates the number of nights in that month that traps were set. The numbers indicated by the bars are derived by dividing the number of captures during the month by the number of nights that traps were set.

Table 17. Names and Locations of Trees Sampled for Radionuclides

Tree No.	Common Name	Scientific Name	Location	
			Direction	Near Well No.
10	Sassafras	<i>Sassafras variifolium</i> Ktze.	NW	85 (across road)
15	Sycamore	<i>Platanus occidentalis</i> L.	NW	106
16	Sweet gum	<i>Liquidambar styraciflua</i> L.	SE	83
19	Black walnut	<i>Juglans nigra</i> L.	S	84
20	Tulip poplar	<i>Liriodendron tulipifera</i> L.	W	95 (across seep)
24	White pine	<i>Pinus strobus</i> L.	S	106
26	Scrub pine	<i>Pinus virginiana</i> Mill.	NE	106
29	Black willow	<i>Salix nigra</i> Marsh.	S	97
31	Red maple	<i>Acer rubrum</i> L.	S	97
33	Pokeberry	<i>Phytolacca americana</i> L.	S	83 (edge of road)
34	Sycamore	<i>Platanus occidentalis</i> L.	NW	85
38	Sweet gum	<i>Liquidambar styraciflua</i> L.	W	118 (across road)
39	Cork elm	<i>Ulmus thomasi</i> Sarg.	W	116 (across road)
40	White oak	<i>Quercus alba</i> L.	S	116
41	Sycamore	<i>Platanus occidentalis</i> L.	E	97A

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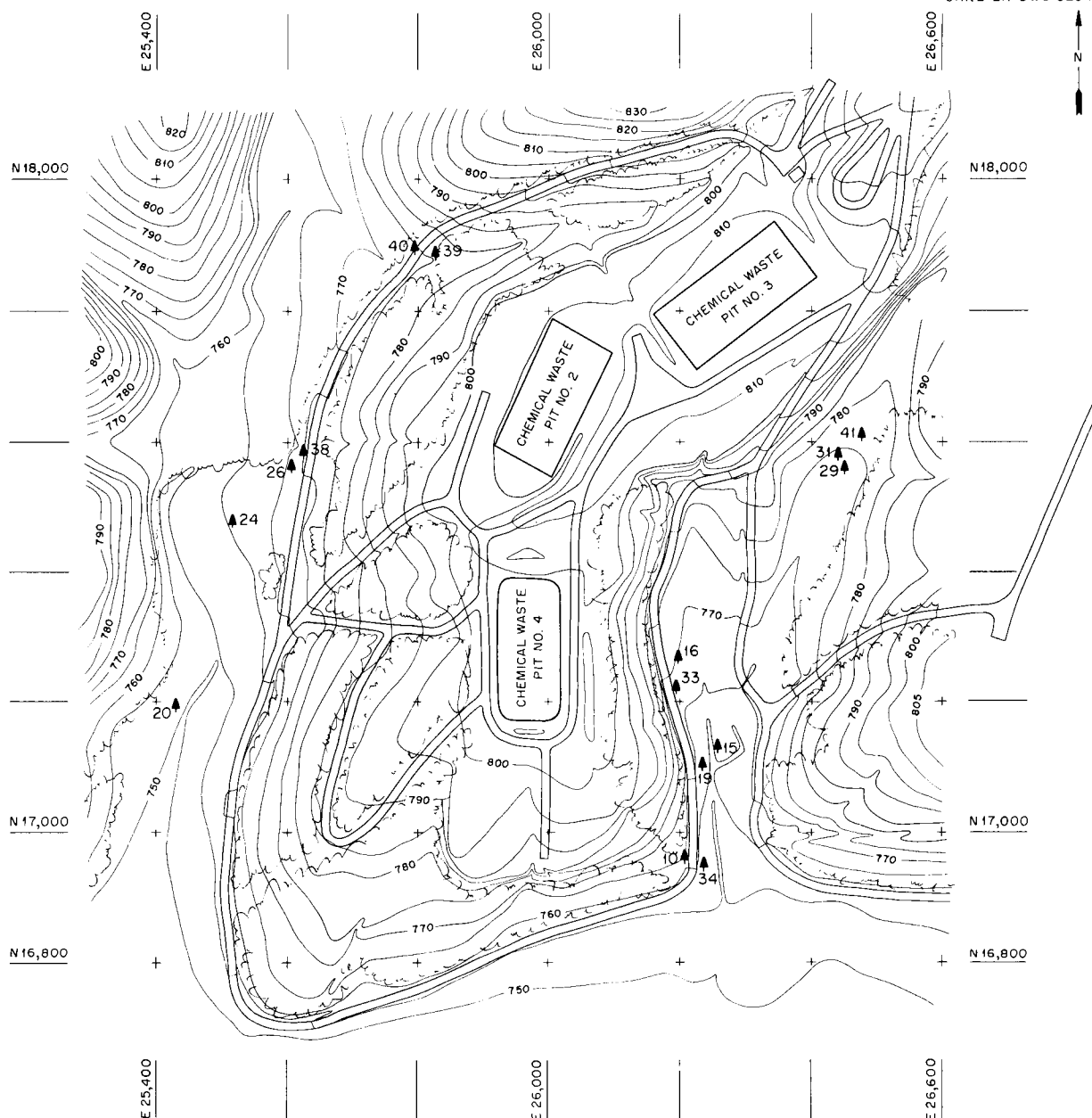


Fig. 23. Locations of Trees Sampled for Radionuclides.

Table 18. Ruthenium-106 ($\mu\text{c/g}$, dry wt) in Trees Around the Waste Pits

Tree No.	Part	Height (ft)	Concentration			
			June 1957	July 1957	April 1958	May 1958
West Seep						
26	Litter				2.4×10^{-4}	4.5×10^{-4}
	Twigs		5.0×10^{-4}		4.1×10^{-4}	2.9×10^{-4}
	Leaves		1.3×10^{-3}	1.2×10^{-3}	1.6×10^{-3}	1.2×10^{-3}
24	Litter				2.2×10^{-4}	1.7×10^{-4}
	Twigs				8.1×10^{-3}	2.3×10^{-3}
	Leaves		4.9×10^{-4}	6.5×10^{-4}	1.2×10^{-4}	5.4×10^{-5}
20	Litter		*	*	4.0×10^{-4}	*
	Twigs		*	*	6.8×10^{-5}	*
	Leaves		*	*		
38	Litter		**	**	**	3.8×10^{-5}
	Twigs	8				*
		12				*
		16				2.7×10^{-5}
		20				*
	Leaves	8				*
		12				*
		16				*
		20				*
39	Twigs		**	**	**	2.5×10^{-5}
	Leaves					3.6×10^{-5}
40	Twigs		**	**	**	2.2×10^{-5}
	Leaves					6.3×10^{-6}
East Seep						
10	Litter				3.6×10^{-4}	
	Twigs		2.0×10^{-5}	**	*	
		20	**	**	**	1.3×10^{-5}
		24	**	**	**	4.5×10^{-5}
	Leaves		1.0×10^{-5}	1.2×10^{-5}	**	
		20	**	**	**	2.2×10^{-5}
		24	**	**	**	1.7×10^{-5}

Table 18 (continued)

Tree No.	Part	Height (ft)	Concentration			
			June 1957	July 1957	April 1958	May 1958
15	Litter		**	**	1.8×10^{-3}	**
	Twigs		1.2×10^{-3}	**	7.2×10^{-4}	4.4×10^{-4}
		12	**	**	**	6.7×10^{-4}
		20	**	**	**	1.0×10^{-3}
	Leaves		9.4×10^{-4}	2.6×10^{-3}	**	7.2×10^{-4}
		12	**	**	**	1.2×10^{-4}
		20	**	**	**	9.9×10^{-5}
16	Twigs		1.1×10^{-2}	1.3×10^{-2}	**	**
	Leaves		2.0×10^{-2}			
19	Twigs	8	**	**	**	1.1×10^{-5}
		12				8.5×10^{-5}
		20				4.2×10^{-5}
	Leaves	8	**	**	**	5.8×10^{-6}
		12				5.4×10^{-5}
		20				*
33	Leaves		1.7×10^{-2}	2.0×10^{-2}	**	7.2×10^{-4}
	Twigs		**	**	**	8.9×10^{-5}
34	Litter		**	**	7.2×10^{-3}	
	Twigs		2.8×10^{-4}		6.7×10^{-4}	7.2×10^{-4}
		12	**	**	**	4.5×10^{-4}
		20	**	**	**	5.8×10^{-4}
	Leaves		1.3×10^{-3}	1.0×10^{-3}	**	7.6×10^{-4}
		12	**	**	**	2.8×10^{-4}
		20	**	**	**	3.1×10^{-4}

*Not detected.

**No samples taken on this date.

Table 19. Cesium-137, Zr⁹⁵-Nb⁹⁵, and Co⁶⁰ ($\mu\text{c/g}$, dry wt) in Trees Around Waste Pits

Tree No.	Part	Height (ft)	Concentration					
			Cs ¹³⁷		Zr ⁹⁵ -Nb ⁹⁵		Co ⁶⁰	
			April 1958	May 1958	April 1958	May 1958	April 1958	May 1958
West Seep								
26	Litter		1.6×10^{-4}	9.9×10^{-5}	1.1×10^{-5}	3.2×10^{-5}	5.0×10^{-5}	9.4×10^{-5}
	Twigs		3.1×10^{-5}	3.6×10^{-5}	2.1×10^{-5}	3.4×10^{-5}	1.4×10^{-4}	2.7×10^{-5}
	Leaves		*	4.5×10^{-5}	3.3×10^{-5}	6.5×10^{-5}	4.5×10^{-4}	4.7×10^{-4}
24	Litter		1.6×10^{-4}	1.6×10^{-4}	3.9×10^{-5}	4.4×10^{-5}	4.5×10^{-5}	6.7×10^{-5}
	Twigs		4.3×10^{-6}	3.7×10^{-6}	1.1×10^{-5}	1.1×10^{-5}	3.1×10^{-5}	1.8×10^{-5}
	Leaves		1.2×10^{-5}	1.0×10^{-5}	6.3×10^{-5}	3.7×10^{-5}	7.2×10^{-5}	5.4×10^{-5}
20	Litter		4.9×10^{-5}	**	4.2×10^{-5}	**	5.4×10^{-5}	**
	Twigs		6.3×10^{-6}	**	1.3×10^{-5}	**	1.8×10^{-5}	**
40	Twigs		**	6.7×10^{-6}		5.4×10^{-6}		*
	Leaves			1.1×10^{-5}		9.9×10^{-6}		*
39	Twigs			6.3×10^{-6}		1.3×10^{-5}		*
	Leaves			8.6×10^{-6}		1.9×10^{-5}		5.8×10^{-6}
38	Litter		**	6.3×10^{-5}	**	5.0×10^{-5}	**	6.3×10^{-6}
	Twigs	8		7.2×10^{-5}		1.1×10^{-5}		*
		12		1.5×10^{-5}		1.8×10^{-5}		*
		16		7.2×10^{-6}		1.1×10^{-5}		*
		20		1.5×10^{-5}		1.6×10^{-5}		*
	Leaves	8		8.5×10^{-5}		1.3×10^{-5}		*
		12		*		1.8×10^{-5}		*
		16		*		2.5×10^{-5}		
		20		7.6×10^{-6}		2.7×10^{-5}		*
East Seep								
10	Litter		2.1×10^{-4}	**	6.3×10^{-5}	**	1.4×10^{-5}	**
	Twigs		1.9×10^{-5}	2.6×10^{-5}	2.1×10^{-5}	2.0×10^{-5}	*	*
		24	**	2.2×10^{-5}	**	1.8×10^{-5}	**	*
	Leaves		**	1.2×10^{-5}	**	1.5×10^{-5}	**	*
		24	**	4.5×10^{-5}	**	1.0×10^{-5}	**	*
15	Litter		3.2×10^{-4}	**	9.4×10^{-5}	**	1.6×10^{-4}	
	Twigs		2.6×10^{-5}	1.7×10^{-5}	1.6×10^{-5}	8.5×10^{-6}	8.5×10^{-5}	1.0×10^{-4}
		12	**	*	**	3.3×10^{-5}	**	5.8×10^{-5}
		20	**	*	**	1.3×10^{-5}	**	7.6×10^{-5}

Table 19 (continued)

Tree No.	Part	Height (ft)	Concentration					
			Cs^{137}		$\text{Zr}^{95}\text{-Nb}^{95}$		Co^{60}	
			April 1958	May 1958	April 1958	May 1958	April 1958	May 1958
15	Leaves	8	**	*	**	2.5×10^{-5}	**	2.1×10^{-5}
		12	**	5.4×10^{-6}	**	3.9×10^{-5}	**	2.1×10^{-5}
		20	**	1.7×10^{-5}	**	8.1×10^{-5}	**	*
19	Twigs	8	**	1.8×10^{-5}	**	1.3×10^{-5}	**	*
		12	**	9.0×10^{-5}	**	3.4×10^{-5}	**	1.2×10^{-5}
	Leaves	8	**	5.8×10^{-6}	**	4.0×10^{-5}	**	*
		12	**	7.8×10^{-6}	**	2.6×10^{-6}	**	9.4×10^{-6}
29A	Twigs	10	**	2.4×10^{-5}	**	9.4×10^{-6}	**	1.2×10^{-5}
		20	**	2.2×10^{-5}	**	1.4×10^{-5}	**	1.7×10^{-5}
	Leaves	10	**	4.5×10^{-5}	**	3.6×10^{-5}	**	3.1×10^{-5}
		20	**	4.3×10^{-5}	**	2.5×10^{-5}	**	2.2×10^{-5}
31	Twigs	5	**	1.0×10^{-5}	**	7.2×10^{-6}	**	*
		12	**	7.6×10^{-6}	**	8.5×10^{-6}	**	4.5×10^{-6}
	Leaves	5	**	1.4×10^{-5}	**	5.4×10^{-5}	**	8.1×10^{-6}
		12	**	8.5×10^{-6}	**	5.8×10^{-5}	**	1.6×10^{-5}
34	Litter		1.9×10^{-4}	**	7.2×10^{-5}	**	4.5×10^{-4}	**
	Twigs		3.2×10^{-5}	*	1.7×10^{-5}	1.5×10^{-5}	7.2×10^{-5}	6.7×10^{-5}
		12	**	*	**	1.7×10^{-5}	**	5.8×10^{-5}
	Leaves	8	**	*	**	2.4×10^{-5}	**	1.4×10^{-4}
		12	**	*	**	4.5×10^{-5}	**	7.2×10^{-5}
41	Twigs	6	**	3.1×10^{-5}	**	1.5×10^{-5}	**	5.4×10^{-5}
		12	**	1.6×10^{-5}	**	2.2×10^{-5}	**	1.8×10^{-5}
	Leaves	6	**	7.2×10^{-6}	**	2.6×10^{-5}	**	5.8×10^{-5}
		12	**	2.2×10^{-5}	**	7.2×10^{-5}	**	4.5×10^{-5}

*Not detected or masked by Ru^{106} .
 **No samples taken on this date.

The original intents of this investigation were (1) to evaluate the concentrations of Ru^{106} in trees in relation to its concentration in the monitor core wells as an aid in evaluating its dispersion through soil and (2) to ascertain whether there was accumulation of Ru^{106} by trees. The resolution of these objectives has been rendered more difficult by the presence of other fission products. The hypothesis that fission products are reaching the trees by means of ground seepage only no longer appears to be completely tenable. Cowser³⁵ has demonstrated that significant amounts of radioactivity above that due to fallout can be obtained on gummed paper trays placed around the perimeter of the pits within 30 to 50 ft. The evidence indicates that the movement may be due to wind action on the exposed sides of the pits during periods of low liquid levels. Although no gummed paper stations have yet been placed in the sites of tree sampling, the presence of Cs^{137} , Co^{60} , and $\text{Zr}^{95}\text{-Nb}^{95}$ in trees not in contact with ground seeps (cf. trees 20 and 38) indicates that some mode of contamination other than ground seepage is involved. The possibility of tree accumulation of Ru^{106} from ground seepage cannot be entirely discounted. Tree No. 26 is situated on a seepage line which is easily located with a G-M survey meter. The leaves, twigs, and litter of this tree contained significant concentrations of Ru^{106} ranging from 2.1×10^{-4} to 1.6×10^{-3} $\mu\text{C}/100$ g (Table 18). Approximately 40 ft north-east of tree 26 (scrub pine) is located tree 38 (sweet gum). Samples from the sweet gum tree taken in May 1958 (Table 18) contained Ru^{106} in only two of the nine samples taken. These two samples had concentrations of 2.7×10^{-5} to 3.8×10^{-5} $\mu\text{C}/100$ g, which were almost two orders of magnitude less than those of tree No. 26. Both trees had similar concentrations of Cs^{137} and $\text{Zr}^{95}\text{-Nb}^{95}$ (Table 18). These data indicate that ground seepage of Ru^{106} is responsible for the large concentrations found in those trees whose roots are in known seepage lines. In the case of those trees which are not growing in seepage lines, the Ru^{106} contamination of their parts probably is due to air-borne contamination. Examination of Tables 18 and 19 indicates a similar pattern of contamination for Co^{60} .

³⁵K. E. Cowser, personal communication.

Comparison of this year's data with those reported last year³⁶ shows no evidence of a buildup in Ru^{106} concentrations. The 1957 concentrations of Ru^{106} in ground litter in the east seep area ranged from 1.5×10^{-3} to 3.8×10^{-3} $\mu\text{C}/\text{g}$ dry wt. In the west seep area, the concentrations ranged from 1.2×10^{-4} to 9.6×10^{-4} $\mu\text{C}/\text{g}$ dry wt. In the spring of 1958, litter at east seep stations ranged from 3.6×10^{-4} to 7.2×10^{-3} $\mu\text{C}/\text{g}$ dry wt. At west seep stations, Ru^{106} concentrations in litter ranged from 1.7×10^{-4} to 4.5×10^{-4} $\mu\text{C}/\text{g}$ dry wt. Leaf and twig concentrations of all the fission products from samples analyzed to date do not appear to differ from those obtained the previous year.

As in 1957, leaf concentrations of Ru^{106} are approaching litter quantities as the season progresses. An exception to this pattern is tree 26, for which leaf values have exceeded litter values. Tree 26 is dying, and this change in physiological condition may account for the increase in leaf concentrations. Last year, tree 16 underwent the same pattern of Ru^{106} increase in the leaves as the tree showed symptoms of dying. After death the concentration of Ru^{106} decreased in the tree.

Fission product concentrations in the ground litter did not show an increase over the previous year even though an additional increment of leaves was added this year. Pending more complete studies, it is assumed that this may be due to rapid leaching and biological breakdown of the leaf material resulting in the return of the radionuclides to the upper layers of soil.

Microarthropods of Pine-Needle Litter. — Preliminary studies have been conducted on the microarthropod fauna of short-leaf pine tree litter. This tree species was chosen because it comprises one of the dominant forest stands adjacent to the radioactive waste pits and is also dominant in various parts of Melton Valley. The aim of these studies was to characterize this habitat in terms of its microarthropod fauna as a prelude to studies on the accumulation and turnover of radionuclides by these animals. Emphasis was placed on the selection of abundant species, particularly of oribatid mites, and also on local variation of oribatid mite populations in short-leaf pine litter.

³⁶S. I. Auerbach *et al.*, *H-P Ann. Prog. Rep.* July 31, 1957, ORNL-2384, p 35-36, Tables 27 and 28.

In Melton Valley, three areas similar in topography and general plant cover were selected for study. Three permanent collecting stations, 63 cm square, were selected in each of the three areas. Three samples were removed from each station at weekly intervals for eight consecutive weeks (June 29 to August 17), and a total of 215 litter-samples were taken. These samples were round cores, 4.1 cm in diameter by 7.6 cm long. Arthropods were extracted from the samples in a Tullgren funnel and were counted.

Results of counts for different arthropod groups in litter samples from the three areas are given in Table 20. The three areas have very similar faunal structures. The mites (Acarina) account for more than 80% of the animals collected; mites and Collembola combined account for 95% of the animals collected.

Special consideration was given to the oribatid mites, since these were the most numerous animal group in the litter samples. Only adults were considered, because it was not possible to identify the immature stages.

Sixty species of oribatid mites were identified in the litter samples. However, only a few species were abundant. Table 21 gives the numbers of mites collected and the percentage of the total oribatids for the nine most abundant species in this study. Also, a cumulative percentage is given. The two most abundant species (*Oppia* sp. and *Cultroribula* sp.) alone account for almost half the adult oribatid mites collected. It is seen from Table 21 that the percentages fall sharply from the most abundant to the less abundant species; however, there is no logical basis for separating a group of "abundant" species from the other species. Thus, the inclusion of *Zygoribatula*, for example, among the abundant species is arbitrary. Figure 24 shows the frequency distribution of the species in the 215 samples. While more than half of the 60 species were absent from most of the 215 samples, there is no clear separation of a group of "rare" species. Thus, while the species listed as "abundant" will form the basis for further discussion, these are not necessarily the only important ones in the litter.

Each of the species of abundant oribatids showed clumping in their distributions of individuals in the samples. This is illustrated in Fig. 25 for *Cultroribula* sp., in which the frequency distribution of mites in litter samples is shown

together with a Poisson distribution having the same mean (2.47 mites per sample). Clearly, the distribution of the mites shows aggregation, since the number of samples containing no mites (92) greatly exceeds the expected number (18), and there are a number of samples with 10 or more individuals where virtually no individuals are expected. While it is possible that these distributions are the results of nonrandom sampling, it seems more likely that there is some biological cause, since these mites would build up populations only in those environmental situations which were favorable to their reproduction.

An analysis of variance was performed for each of the abundant oribatid species, to investigate the distribution of the mites among the sample areas. Table 22 illustrates the design (provided by M. Kastenbaum of the Mathematics Panel) for this analysis and presents data for *Cultroribula* sp. as an example. Since this arrangement is in part hierarchic (stations in areas) and in part orthogonal (weeks) the tests of variance ratios were not straightforward. In general, the main effects were tested over the appropriate interactions when the latter were significant; where interactions were not significant, the station and area effects were treated as hierarchic. In Table 22, the mean squares are significant ($P = 0.01$) for stations in areas, weeks, and the stations in areas by week interaction. Thus, differences exist between the average numbers of mites per station within each area and between the average numbers of mites per week; the significant interaction shows that the relationship between means in the stations was not constant through time. In this example, differences between the areas are no greater than differences between stations within areas.

In Table 23, results of variance ratio tests are summarized for an additional six of the abundant species. It is seen that generalizations cannot be made concerning differences between areas or stations in areas, since the species investigated differ in their distributions. The area differences which do exist probably have little biological meaning, except as evidence that local variation in population size is large.

The investigations summarized here form a sufficient groundwork for studies on the role of the microarthropod fauna in the release of fission products from contaminated litter. Such studies are now in progress.

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Table 20. Number of Arthropods Obtained from Three Areas in 215 Samples of Litter

Arthropod Group	Number of Individuals			Total, All Areas
	Area I*	Area II*	Area III**	
Arachnida				
Pseudoscorpionida	13	28	20	61
Araneida	59	77	25	161
Acarina	7,466	7,964	9,746	25,176
Mesostigmata	274	178	218	670
Trombidiformes	1,810	1,669	2,614	6,093
Sarcoptiformes	5,382	6,117	6,914	18,413
Paupoda	12	23	20	55
Diplopoda	3	3	2	8
Chilopoda	10	3	10	23
Symphyla	7	58	15	80
Insecta	1,630	2,057	1,120	4,807
Protura	75	90	114	279
Thysanura			1	1
Entotrophi (Japygidae)	16	4	2	22
Collembola	1,334	1,665	721	3,720
Poduridae	44	47	45	136
Onychiuridae	329	524	84	937
Isotomidae	135	271	311	717
Entomobryidae	657	644	203	1,504
Sminthuridae	169	179	78	426
Orthoptera (Blattidae)	0	2	0	2
Isoptera	0	0	38	38
Corrodentia	6	1	5	12
Thysanoptera	52	43	38	133
Hemiptera	0	2	1	3
Adults		1	1	
Nymphs		1		
Homoptera	10	22	39	71
Adults	6	8	16	
Nymphs	4	14	23	
Lepidoptera (larvae)	2	2	3	7
Diptera	47	48	37	132
Adults	12	21	20	
Larvae	35	27	14	
Pupae			3	
Coleoptera	52	58	45	155
Adults	22	33	23	
Larvae	30	25	22	
Hymenoptera	36	120	76	232
Wasps	24	51	50	
Formicidae	12	69	26	
Total	9,200	10,213	10,958	30,371

*72 samples.

**71 samples.

Table 21. Numbers of the Most Abundant Oribatid Mites in Short-Leaf Pine Litter

Species	Number of Mites Collected	Percentage of Total	Cumulative Percentage of Total
<i>Oppia</i> sp.	2715	41.2	41.2
<i>Cultroribula</i> sp.	530	8.0	49.2
<i>Tectocepheus velatus</i>	356	5.4	54.6
<i>Galumna</i> sp.	244	3.7	58.3
<i>Scheloribates</i> sp.	208	3.2	61.5
<i>Trhypochthonius americanus</i>	205	3.1	64.6
<i>Peloribates</i> sp.	179	2.7	67.3
<i>Suctobelba</i> sp.	176	2.7	70.0
<i>Zygoribatula</i> sp.	138	2.1	72.1
Remaining 51 species	1838	27.9	100.0
Total	6589	100.0	

Table 22. Analysis of Variance: *Cultroribula* sp. Counts Transformed by $(X + 0.5)^{1/2}$

Source	Sum of Squares	Degrees of Freedom	Mean Square
Areas	22.9294	2	11.4647
Stations in areas	38.5526	6	6.4254*
Weeks	20.6939	7	2.9563*
Areas by weeks interaction	5.3705	14	0.3836
Stations in areas by weeks interaction	31.0688	42	0.7397*
Error	47.0987	143	0.3294
Total	165.7139	214	

*Significant at $P = 0.01$.

Table 23. Results of Variance Ratio Tests in Analysis of Variance

Source	Value of P at Which Differences Are Significant					
	<i>Scheloribates</i> sp.	<i>Zygoribatula</i> sp.	<i>Tectocepheus</i> <i>velatus</i>	<i>Galumna</i> sp.	<i>Oppia</i> sp.	<i>Trhypochthonius</i> <i>americanus</i>
Areas	0.01	0.05	0.01	n.s.*	n.s.	n.s.
Stations in areas	n.s.	0.01	0.01	0.01	0.01	0.01
Weeks	n.s.	n.s.	n.s.	0.01	0.01	0.01
Areas by weeks interaction	0.05	n.s.	n.s.	n.s.	0.01	0.05
Stations in areas by weeks interaction	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.

*Not significant.

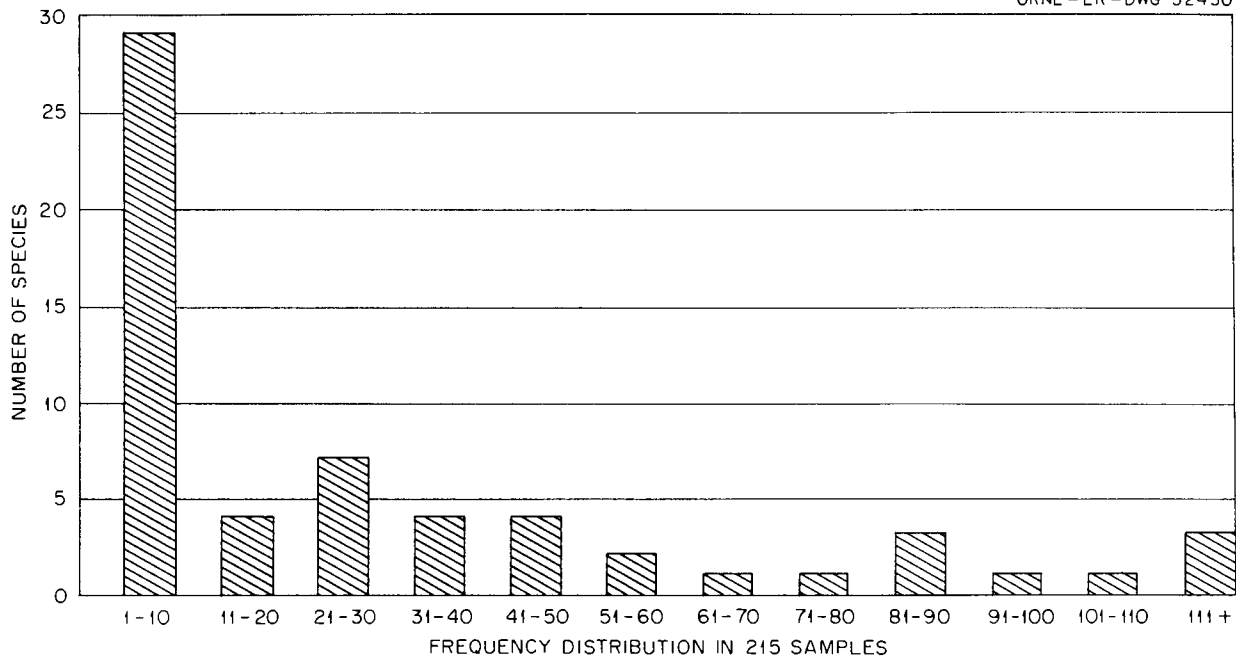
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ORNL - LR - DWG 32430

Fig. 24. Frequency Distribution of Species of Oribatid Mites in 215 Samples of Pine-Needle Litter.

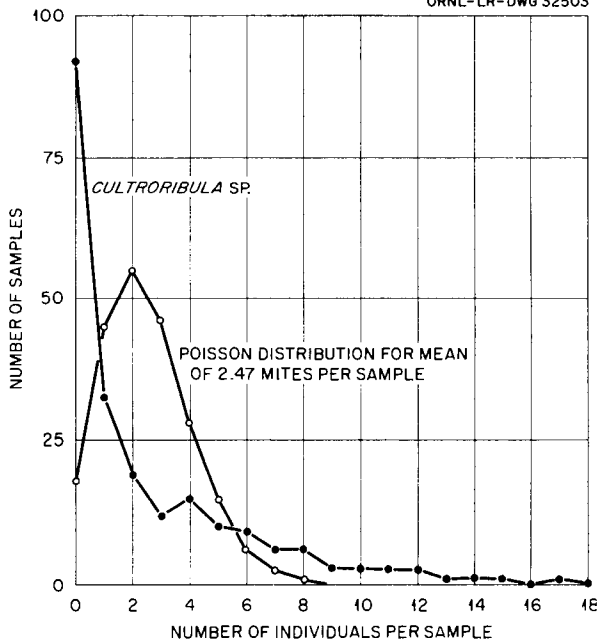
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Fig. 25. Frequency Distribution of Individuals of *Cultroribula* sp. in 215 Samplings.

CHARACTERIZATION OF HIGH-LEVEL FUEL REPROCESSING WASTES

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Synthesis of Typical Wastes for Laboratory Study

At present a number of fuel element designs are being evaluated, and since the differences in the materials used in fabrication of the fuel elements make it necessary that different separations processes be used, the waste produced is of great variety. To establish standardized compositions and make it possible for investigators at different sites to work with comparable wastes, it is desirable to assign compositions and suggested procedures for the preparation of synthetic wastes. Therefore, representative waste types were defined and procedures were developed for their preparation, as part of the Health Physics Division-Chemical Technology Division program in waste disposal. Through consultation with members of the Chemical Technology Division's Long-Range Planning Committee the five wastes chosen were:

1. Purex (a nitric acid waste from aluminum-jacketed natural uranium fuel),
2. TBP-25 (an aluminum nitrate, nitric acid waste from enriched-uranium-aluminum alloy elements),
3. STR (zirconium and aluminum nitrates in hydrofluoric acid),
4. Darex (stainless steel nitrates in nitric acid),
5. SIR (stainless steel nitrates in nitric and sulfuric acids).

These wastes are all raffinates from solvent extraction separations of uranium (natural or enriched) from its fission products and/or alloying elements. The compositions given do not include fission products, but they may be added for radioactive work. Table 24 is the prescription for preparing synthetic wastes.

The assumptions determining the curies per gallon and the heat produced by decay are the following:

1. The fuels are irradiated to 4000 megawatt days per ton at a neutron flux of 5×10^{13} neutrons/cm²/sec and cooled for 100 days after discharge from the reactor.
2. The burnup for U²³⁵-enriched fuel elements is 53%.
3. The energy distribution is approximately 50% gamma and 50% beta.

These simulated waste solutions can be prepared from stock chemicals. The amount of each chemical required is shown in Table 24. The chemicals are dissolved in a sufficient quantity of distilled water [except for Zr(OH)₄ and Sn] and are poured into a 1-liter volumetric flask. Each of the acids is diluted, then mixed with the salt solutions in the flask; finally the whole is diluted to 1 liter.

The simulated zirconium-hydrofluoric acid waste (STR) is prepared in a different fashion. In this preparation it is necessary to dissolve the zirconium hydroxide in about 300 ml of 9 M HF. As aqueous hydrofluoric acid readily attacks glass, a polyethylene beaker, or better still, a magnesium beaker which may be heated on a hot plate, must be used.

The mossy tin should be dissolved with part of the nitric acid before being added to the solution in the volumetric flask. This waste must be stored in a polyethylene container, but the other waste solutions may be stored in glass bottles.

The Purex waste solution described here is the evaporated concentrate. It is evaporated to reduce the volume for storage and to recover nitric acid. In field operations, this concentrated waste is neutralized with sodium hydroxide before being stored in mild steel tanks.

The first-cycle aqueous raffinate from the Purex process is 1 M in nitric acid, about 0.03 M in ferric sulfate, and contains traces of sodium and mercuric nitrates. This preparation of synthetic waste represents about 99% of the bulk salts. However, no attempt was made to add the minute amounts of various chemicals or corrosion products that may be important in a specific test. For much of the research on waste disposal, simulated wastes are entirely satisfactory.

Physical and Chemical Properties of Wastes

Physical and chemical properties of synthetic wastes were investigated from the following standpoints:

1. evaporation of acid wastes,
2. neutralization of acidic waste followed by solid-liquid separation,
3. the effect of a flocculating agent on settling rates of the neutralized slurries,
4. further volume reduction by evaporation of slurries and supernatants resulting from neutralizations.

Evaporation was considered to be the simplest means of volume reduction for wastes. The mechanical problems of bumping and spattering did not give serious trouble in the evaporation of acid waste. The results of tests of evaporation of acid waste are presented in Table 25.

Calcination of the dried residues at temperatures up to 900°C gave further weight reduction. Calcined weights were 57, 91, 66, 42, and 26% of the dried

Table 24. Constituents of Synthetic Chemical Processing Waste Solutions

Synthetic Waste Chemical	Amount Used per Liter	Final Concentration, M
Purex, Concentrated		
$\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	202 g	0.5
Na_2SO_4	21.3 g	0.15
H_2SO_4	35.5 ml of 18 M	0.60
HNO_3	353 ml of 15.6 M	5.5
TBP-25		
$\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	600 g	1.6
$\text{Fe}_2(\text{SO}_4)_3 \cdot x\text{H}_2\text{O}$	0.8 g [as $\text{Fe}_2(\text{SO}_4)_3$]	0.002
$(\text{NH}_4)_2\text{SO}_4$	2.64 g	0.02
$\text{Hg}(\text{NO}_3)_2$	3.25 g	0.01
NH_4NO_3	2.4 g	0.03
HNO_3	33 ml of 15.6 M	0.5
STR		
$\text{Zr}(\text{OH})_4 \cdot 12\text{H}_2\text{O}$	206 g	0.55
$\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	273 g	0.75
$\text{Cr}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	6.4 g	0.016
Sn (mossy)	1.43 g	0.012
HF	107 ml of 28 M	3.0
HNO_3	83 ml of 15.6 M	1.3
Darex		
$\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	275 g	0.68
$\text{Cr}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	64 g	0.16
$\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	19.1 g	0.075
$\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	46.1 g	0.123
HCl	1 ml of 1.0 M	0.001
HNO_3	189 ml of 15.6 M	2.95
SIR		
$\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	28.0 g	0.07
$\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	2.3 g	0.008
$\text{Cr}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	13.0 g	0.018
$\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	4.4 g	0.017
$\text{Mn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	0.119 g	0.001
H_2SO_4	28 ml of 18 M	0.51
HNO_3	131 ml of 15.6 M	2.35
Radioactive Nature of Wastes		
Waste	Radioactivity (curies/gal)	Heat of Decay (Btu/hr/gal)
Purex	1300	22.7
TBP-25	2500-5100	44-88.6
STR	100	1.71
Darex	1000	17.6
SIR	1500	25.0

Table 25. Physical Properties of Evaporated Unneutralized Waste

Volume Evaporated: 250 ml

Waste	Crystallization Volume (% of original)	Solid Residue			Remarks
		Weight (g)	Volume (ml)	Volume (% of original)	
Purex	40	26.8	17	6.8	
TBP-25	24	47.0	60	24	Syrupy liquid which solidified on cooling
STR	50	38.1	38	15.2	
Darex	40	23	23	9.3	
SIR	7	6.6	8	3.2	Glassy solid formed without crystallization

residue for Purex, Darex, STR, TBP-25, and SIR, respectively. The nitrate content of the residue varied directly with weight loss during calcination.

The wastes were neutralized with sodium hydroxide solution and with dry calcium hydroxide in order to evaluate the difficulty of separating the resulting solids from the supernatant, which could then receive further treatment. Filtration rates of all the neutralized wastes were prohibitively low on fritted glass or sintered stainless steel. Settling rates varied over an extremely wide range, with precipitates from Purex, TBP-25, and Darex wastes settling very slowly. Precipitates from SIR waste settled most rapidly, with those from STR waste next. Settling rates for SIR precipitates are higher than for Purex, TBP-25, and Darex precipitates by as much as a factor of 20. Neutralization with $\text{Ca}(\text{OH})_2$ gave precipitates which settled more rapidly than those from NaOH neutralization.

A flocculating agent, IFA-313, made by the Illinois Water Treatment Company, Rockford, Ill., was used in order to improve settling rates. The $\text{Ca}(\text{OH})_2$ -neutralized slurries are more likely to be improved than the NaOH -neutralized slurries. The flocculating agent does not affect the final volume of precipitate. Table 26 presents selected data showing the effect of IFA-313 addition.

If volume reduction is desired following neutralization, it is possible to evaporate the slurry if the operation is carried out in a system which prevents bumping, such as an agitated vessel or a circulating evaporator. Table 27 presents the results of

evaporation of unfiltered, neutralized wastes. After a solid-liquid separation was effected, the resulting supernatant could be evaporated without precautions taken against bumping. The crystalline residue, NaNO_3 or $\text{Ca}(\text{NO}_3)_2$ with other salts, had volumes of 25–65% of the original waste volumes. The volume variation was proportional to the solid content of the original waste solution.

FIXATION OF HIGH-LEVEL WASTES BY SINTERING

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Pilot Pit 2

Pilot Pit 2 was a large-scale sintering furnace designed to produce a sintered clinker 6 ft in diameter by 2 ft thick. A charge of mix 15 (aluminum nitrate waste, Conasauga shale, limestone, and soda ash) was heated with an immersion heater built of Calrod heaters distributed through an "egg crate" assembly of stainless steel sheet. Tracer activity in the form of approximately 100 millicuries of mixed fission products was added to the charge.

Primary objectives of the experiment were to produce a large sintered clinker by using mix 15 as the raw material, and to test the effectiveness

³⁷Chemical Technology Division.

Table 26. Effect of a Flocculating Agent (IFA-313) on Settling Rates of Neutralized Synthetic Wastes

Waste	Neutralizing Agent	IFA-313 (ml/liter)	Settling Rate (mm/min)		Remarks
			5 min	10 min	
Purex	NaOH	0	0.5	0.5	
		2.8	1.6	1.4	
	Ca(OH) ₂	0	0.9	1.6	Concentration threshold for IFA-313 effectiveness
		0.9	1.9	2.2	Concentration threshold for IFA-313 effectiveness
		3.3	4.4	5.6	
Darex	NaOH	0	0.4	0.4	
		5.6	0.4	0.4	
	Ca(OH) ₂	0	1.3	1.6	
		5.6	1.5	1.6	
SIR	NaOH	0	1.0	0.8	
		6.3	1.3	2.5	
	Ca(OH) ₂	0	14.4	11.2	
		0.1	19.4	11.9	
		6.0	24.0	13.0	
STR	NaOH	0	0	0.2	
		5.4	0.3	0.3	
	Ca(OH) ₂	0	2.8	2.8	Maximum settling rate of the Ca(OH) ₂ -precipitated STR waste
		1.6	8.5	6.9	
		5.4	1.9	2.0	

of the off-gas facility for condensing the evolved moist aerosols and for filtering out the solid aerosols. Drawings and photographs of the furnace, the immersion heater, and the mixer for preparation of the charge have been presented elsewhere.³⁸

Sintering experience gained with immersion-type electrical resistance heaters had shown that great care must be exercised in the fabrication of such units in order to secure reliability. Following the installation and hookup of the furnace, a detailed check was made of the location and continuity of

each heater and the approximately 30 thermocouples installed at various points in the grid.

The large batch mixer (a vertical, cylindrical tank equipped with a 15-hp flat-blade turbine agitator) functioned satisfactorily to produce a 720-gal batch. A trial mix was made before the actual test, and the gel was discharged to waste rather than to the furnace. It was possible to carry through the mixing procedure on the basis of a time cycle determined during the trial mix, thus avoiding frequent observations of the radioactive slurry.

The slightly radioactive mass (~100 mc) was heated with energy input at a decreasing rate corresponding to radioactive decay for material

³⁸W. J. Boegly *et al.*, *H-P Ann. Prog. Rep.* July 31, 1957, ORNL-2384, p 58.

21 days out of the reactor at the beginning of the test. Figure 26 shows the variation of power input with time. The experiment was designed to achieve a temperature of 900°C in the mix at the end of 22 days. However, because of failure of individual Calrod units in the immersion heater, it was impossible to continue along the designed power curve. Also, because of failure of the

heaters in an irregular pattern, the power was not evenly distributed through the cake.

Maximum temperatures of approximately 550°C were reached in the clinker about 20 days after startup. Figure 27 is a plot of temperatures vs time of operation; location of the thermocouples is shown in the inset. After the maximum temperatures were reached the temperatures decreased with failure of additional heaters. Figure 28 shows a section of the furnace and clinker and the location of maximum temperatures.

The off-gas system was effective in condensing the evaporated water and in filtering out particulate matter. However, carbon steel components of the off-gas system failed before completion of the experiment because of nitric acid corrosion. Because of failure of the components and difficulty with the exhausters, it was not possible to secure data on the effectiveness of the glass-fiber filter.

Further work to be done in evaluation of the experiment includes disassembly, inspection, leaching tests on the clinker, and measurement of the amount of activity contained in the glass-fiber filters.

"Adiabatic" Self-Sintering Experiment

The experience gained from the Pilot Pit 2 experiment indicated that one of the most complex

Table 27. Volume Reduction Effected by Evaporation of Unfiltered Neutralized Wastes

Waste	Neutralizing Agent	Volume of the Solid Residue	
		(ml)	(% of waste)
Purex	NaOH	14	56
	Ca(OH) ₂	12	48
Darex	NaOH	4	16
	Ca(OH) ₂	5	20
STR	NaOH	5	20
	Ca(OH) ₂	5	20
SIR	NaOH	1	4
	Ca(OH) ₂	2	8
TBP-25	NaOH	4	16
	Ca(OH) ₂	8	32

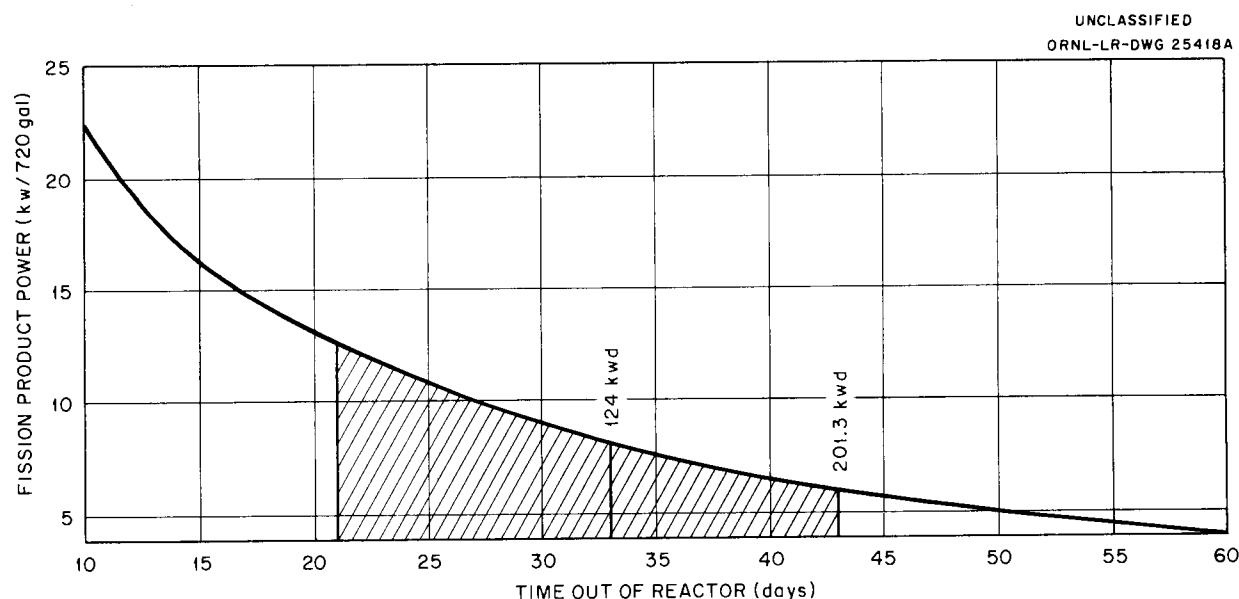


Fig. 26. Fission Product Power, As a Function of Time of Decay, in 24 MTR Assemblies (200 g of U²³⁵) Irradiated 16 Days at 2×10^{14} neutrons/cm²/sec.

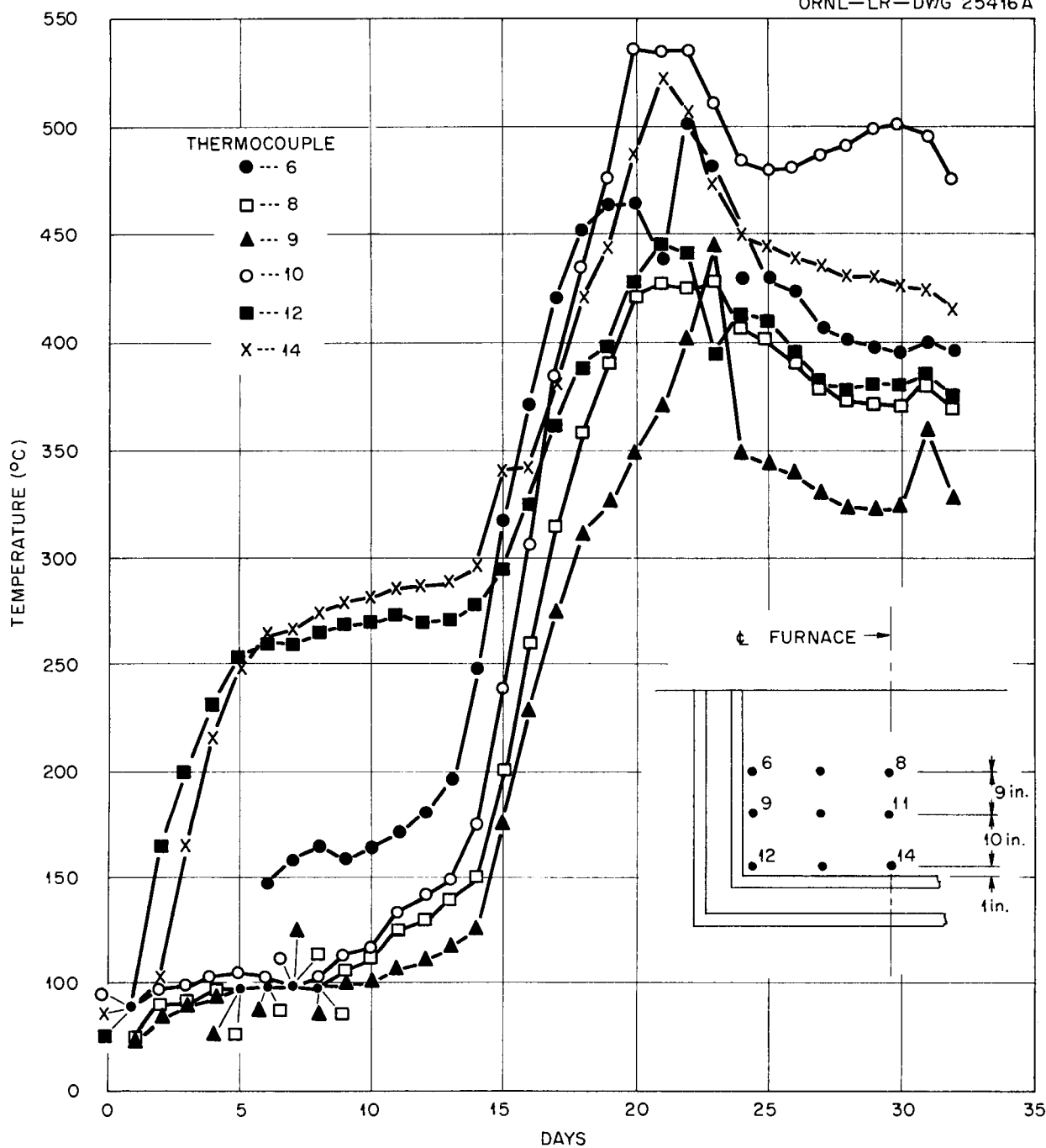
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Fig. 27. Temperature of Clinker at Selected Sites, Pilot Pit 2.

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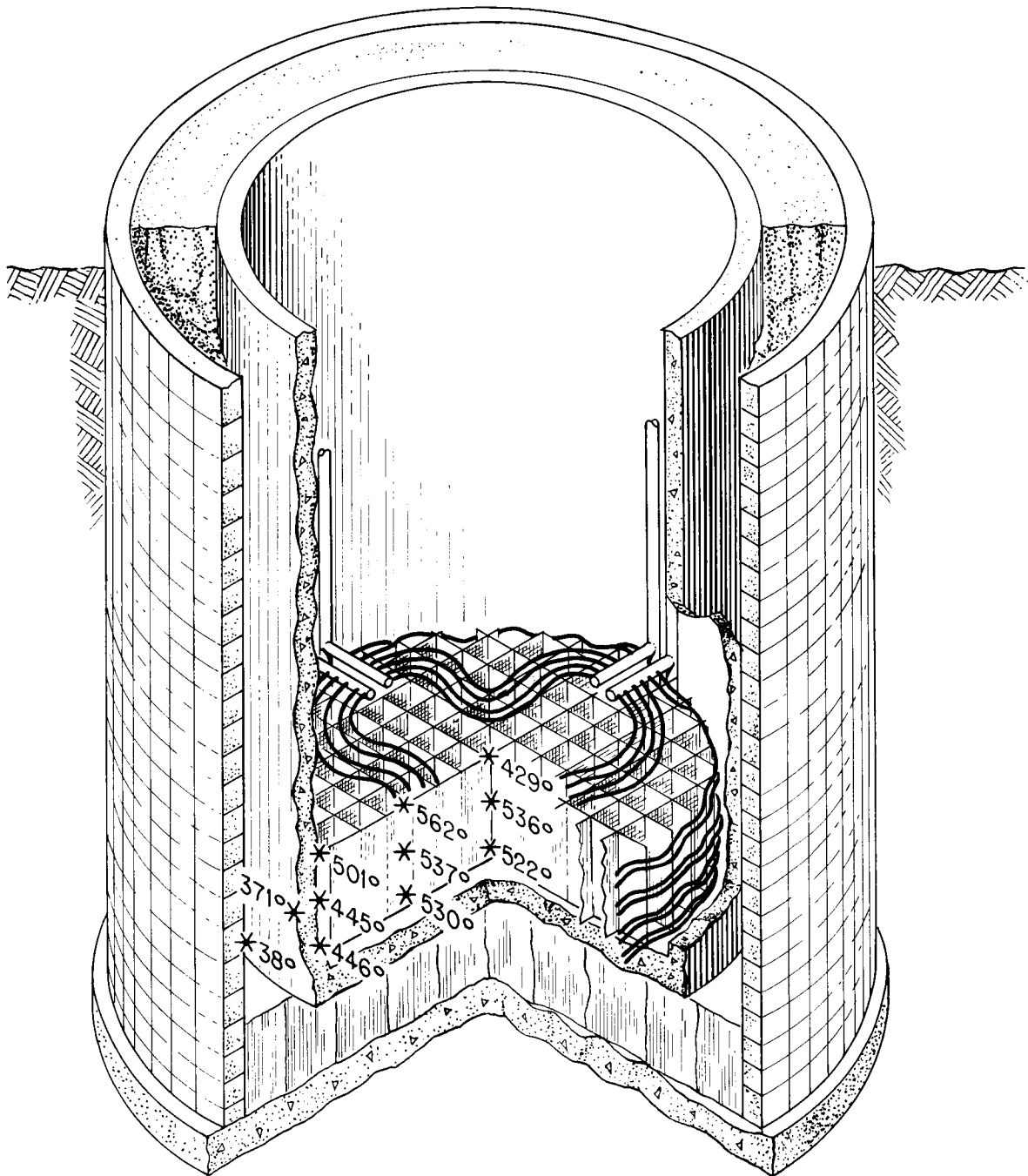


Fig. 28. Maximum Sinter Temperatures ($^{\circ}\text{C}$), Pilot Pit 2.

engineering problems in the sintering program is the simulation of a fission product heat source by means of artificial heaters. The "adiabatic" experiment was proposed³⁹ in order to bridge the gap between pilot-scale experiments using artificial heating and experiments in which all the required heat was available from radioactive decay.

The adiabatic experiment was designed to use 30 gal of high-level waste produced by the dissolution of one conventionally irradiated and cooled 200-g MTR fuel element. As this fuel element (irradiated at a constant power level corresponding to an initial thermal neutron flux of 2.2×10^{14} neutrons/cm²/sec for 42 days) would provide only 375 w of fission product power after cooling for 90 days, sufficient power would not be available to meet the sintering requirements because of the inherent heat loss. For this reason the experimental furnace was equipped with a heat barrier to reduce the heat losses from the furnace so that the major portion of the power would be available for self-sintering. The 90-day-cooled waste will contain approximately 10^5 curies of fission product activity.

Design of the adiabatic experiment was initiated in August 1957; the site selected for the experiment was the field facility used for Pilot Pit 2. The equipment is composed of three major parts: (1) furnace and mixer assembly, (2) off-gas cleaning system, and (3) thermal measurement and power control house.

Furnace and Mixing Assembly. — To ensure containment of the fission products, the entire mixing and furnace assembly is located in a single shielded underground vessel shown in Fig. 29. The containment vessel is composed of two concrete liners; the outer liner of Silo block is 10 ft in diameter and 10 ft deep, and the inner liner is a concrete pipe, 6 ft in diameter and $9\frac{1}{2}$ ft deep, with the annulus being filled with earth for shielding and structural purposes. A biological shield, 5 ft thick, covers the liners. Charging is accomplished by means of a remotely operated valve in the line from the mixer located directly above the furnace. The furnace consists of two concentric cylindrical stainless steel cans 24 in. in diameter and 42 in. deep; the outer can is used for structural and safety purposes, and the inner can is used to retain the fired material (Fig. 30).

An insulated cover is bolted to the inner furnace can and contains lifting hooks for removing the inner can, which will contain the final product. The cover is equipped with an off-gas line, a slip joint for the mixer discharge pipe, and connectors for thermocouples. All components which come into contact with the waste solution are made of stainless steel.

The most important part of this furnace is the heat barrier, or "adiabatic heater." The installed heater can be seen in Fig. 31 and consists of embedded ceramic heater elements (Fig. 32). The function of this heat barrier is to reduce heat losses from the furnace during self-sintering. By operating a furnace of this type with a heat barrier it is possible to self-heat waste-clay-flux slurries with waste solutions containing smaller amounts of activity. When the heat barrier is used in a self-heating experiment, the thermal controls are preset to allow small thermal gradients from the furnace to the surrounding insulation, but at no time to provide heat to the furnace.

Off-Gas Cleaning System. — Gases, vapors, and particulates released during mixing and firing are exhausted to the off-gas system. The system is constructed in two shielded concrete tanks 7 ft in diameter by 10 ft deep and consists of four units: (1) a condensate storage vessel, (2) a packed tower containing a 20% solution of NaOH, (3) a recirculating caustic scrubber, and (4) a multibed filter system (Fig. 33). Since the off-gas vessels are located about 50 ft from the furnace, it has been calculated that vapor released during the slurry dewatering stage will condense in the off-gas line and drain to the condensate vessel. From this vessel the gases will then enter the packed tower for removal of CO₂ and fission product gases. Gases leaving the packed tower will be passed through a jet mixer and continuously contacted with 20% caustic scrub solution until essentially all the noxious inert gases and fission products have been removed. Any gas remaining (mainly the chemically inert fission gases) will be passed through a multibed filter vessel (composed of layers of sand, soda lime, and activated carbon) before being released to the atmosphere. The vacuum required to exhaust the gases from the furnace and mixer is provided by the jet mixer on the recirculating caustic scrubber. In order to evaluate the performance of each unit or combination of units, bypass lines have been installed

³⁹ *ibid.*, p 64.

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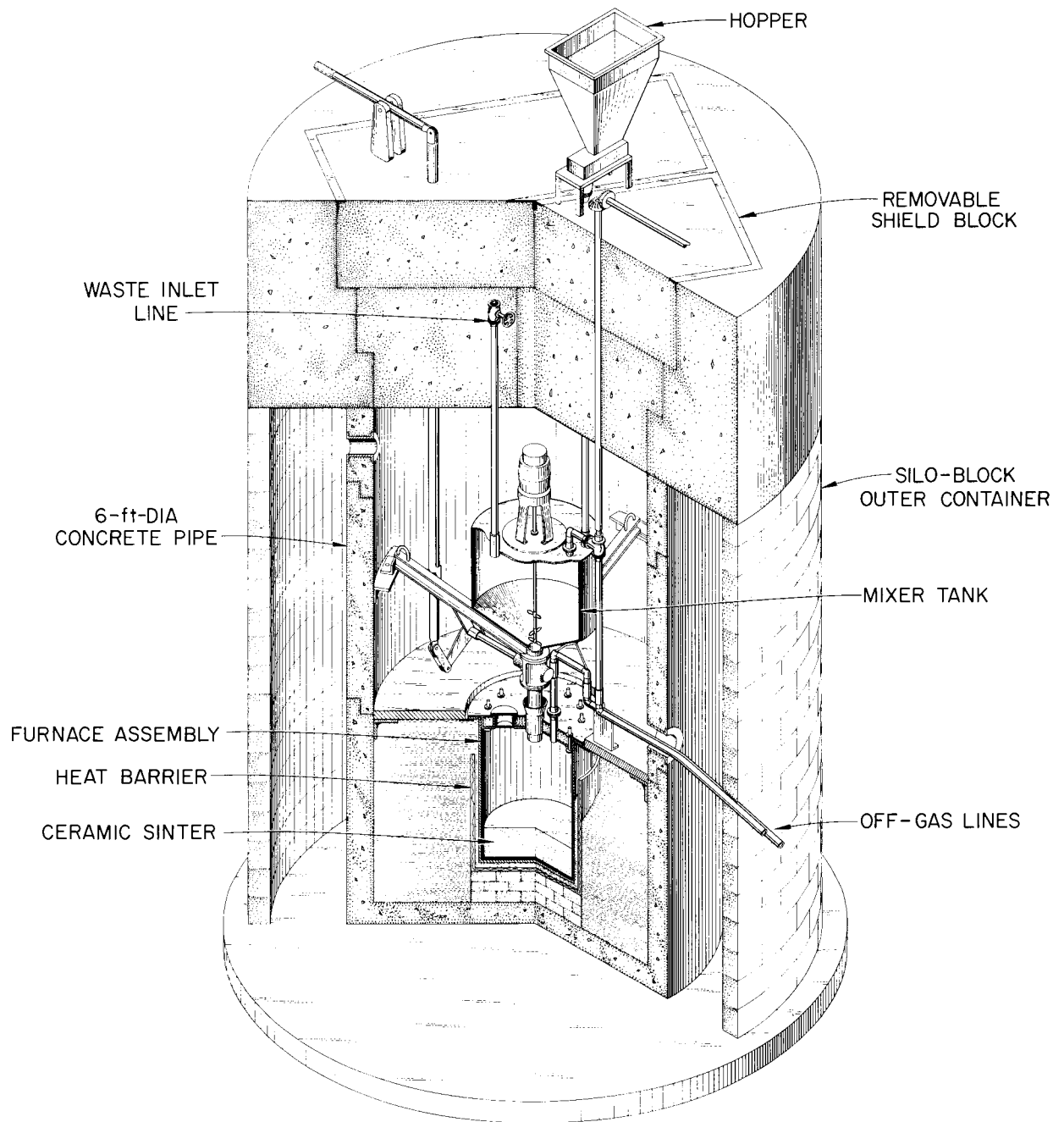
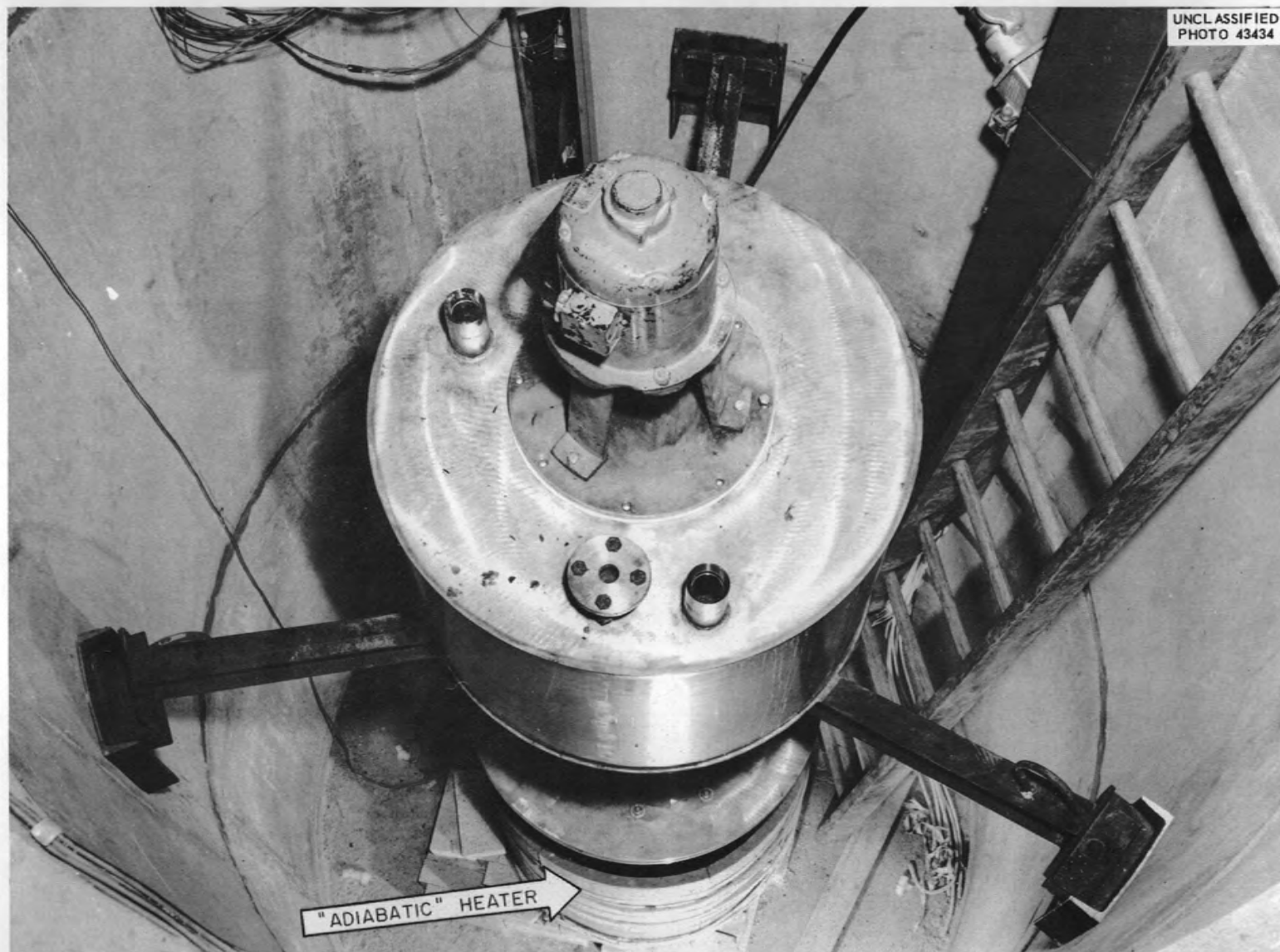


Fig. 29. Experimental Facility for High-Level Self-Sintering.



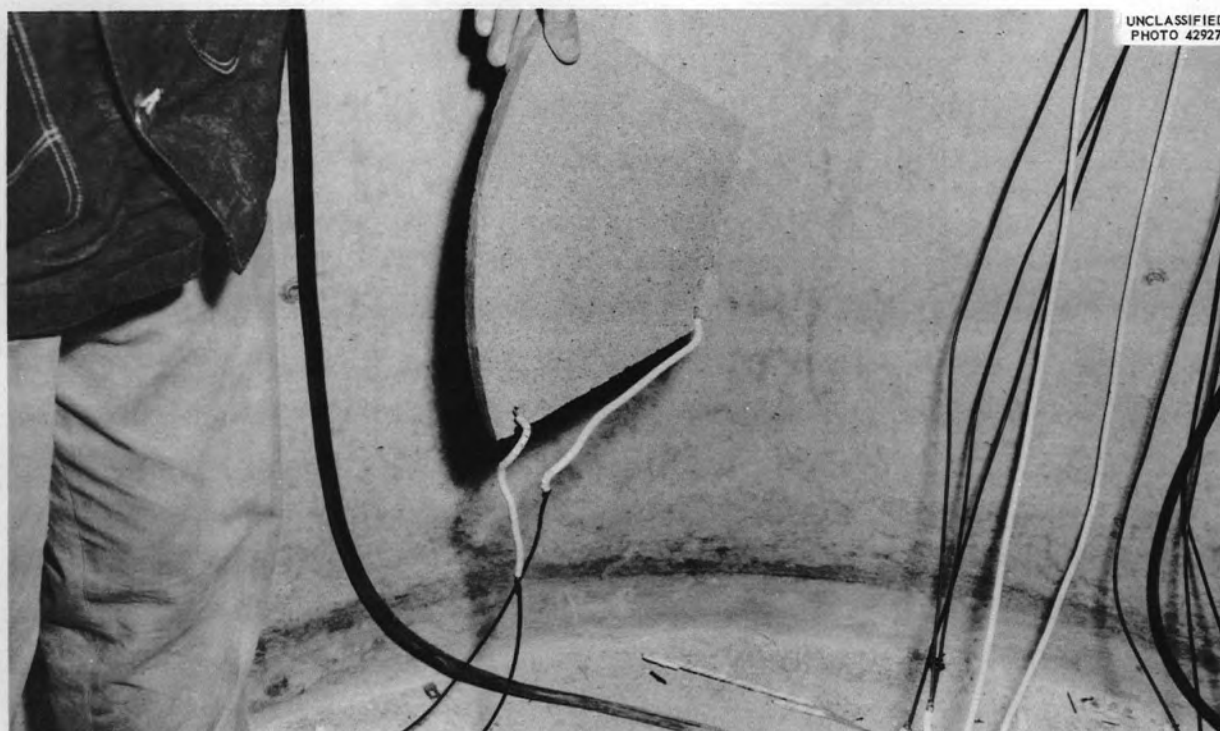
Fig. 30. "Adiabatic" Furnace, Showing Inner Container and Cover.



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Fig. 31. Top View of Assembled Furnace and Mixer, Showing "Adiabatic" Heater.

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Fig. 32. Typical Heat-Barrier Element.

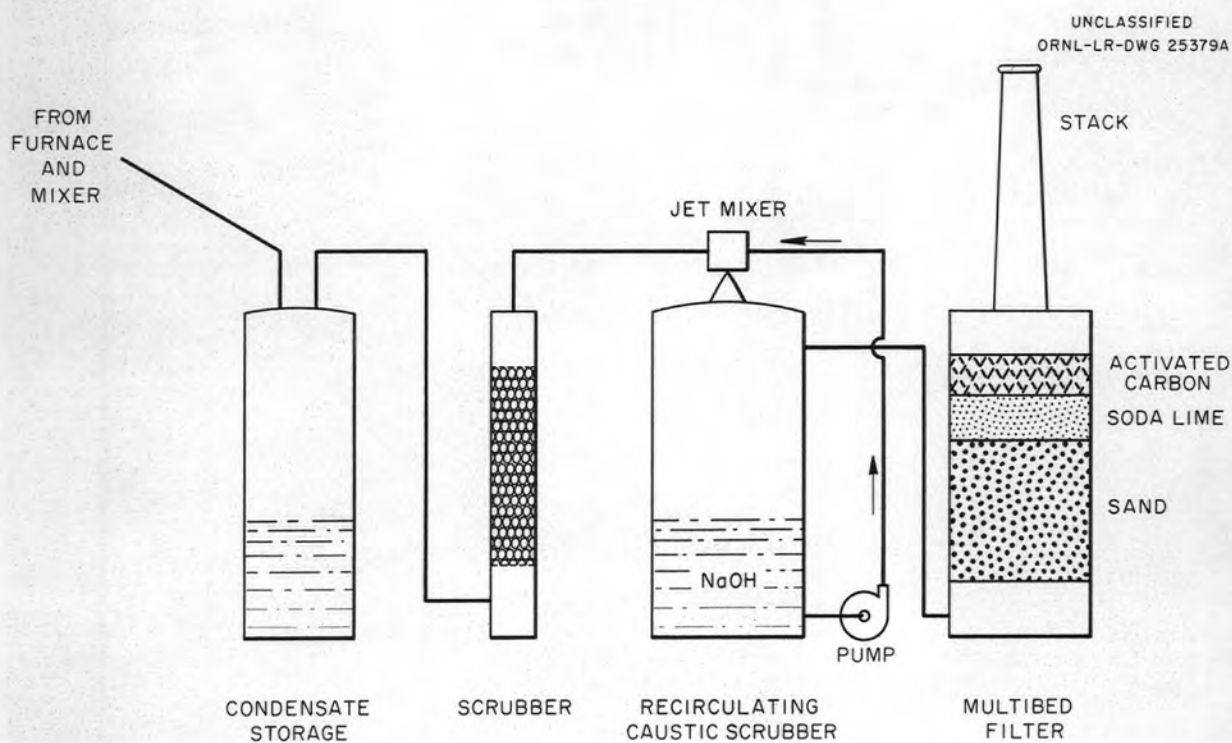


Fig. 33. Schematic Diagram of Off-Gas System.

around all vessels except the condensate storage. Isokinetic sampling devices have been installed in the gas lines on both sides of each vessel in order to determine the efficiency of each unit or combinations of units. Provisions will be made to separate the sampled gas stream into condensable gases, noncondensable gases, and particulates. The central gas sampling station will be located in a shielded cubicle adjacent to the off-gas system.

Thermal Measurement and Power Control. - All recording instruments and power controls are located in one building. A small structure adjoining this building houses the devices for power regulation. Provisions have been made to obtain a complete thermal history of each experiment.

Thermocouples are located in the ground, the concrete liner annulus, the furnace insulation, and inside the furnace. Continuous temperature recording devices for 36 thermocouples are provided (Fig. 34). In addition, thermocouples are located in the heat barrier to control the surface temperature of the heating elements. There are six separate control zones in the heat barrier: two on the bottom, three on the sides, and one on the furnace cover. Each thermocouple on the heat barrier has a corresponding thermocouple located adjacent to it on the furnace liner (Fig. 35). These paired thermocouples are used to control the heater temperature. Figure 36 is a diagrammatic sketch of the control system. When the paired thermocouples show a thermal gradient

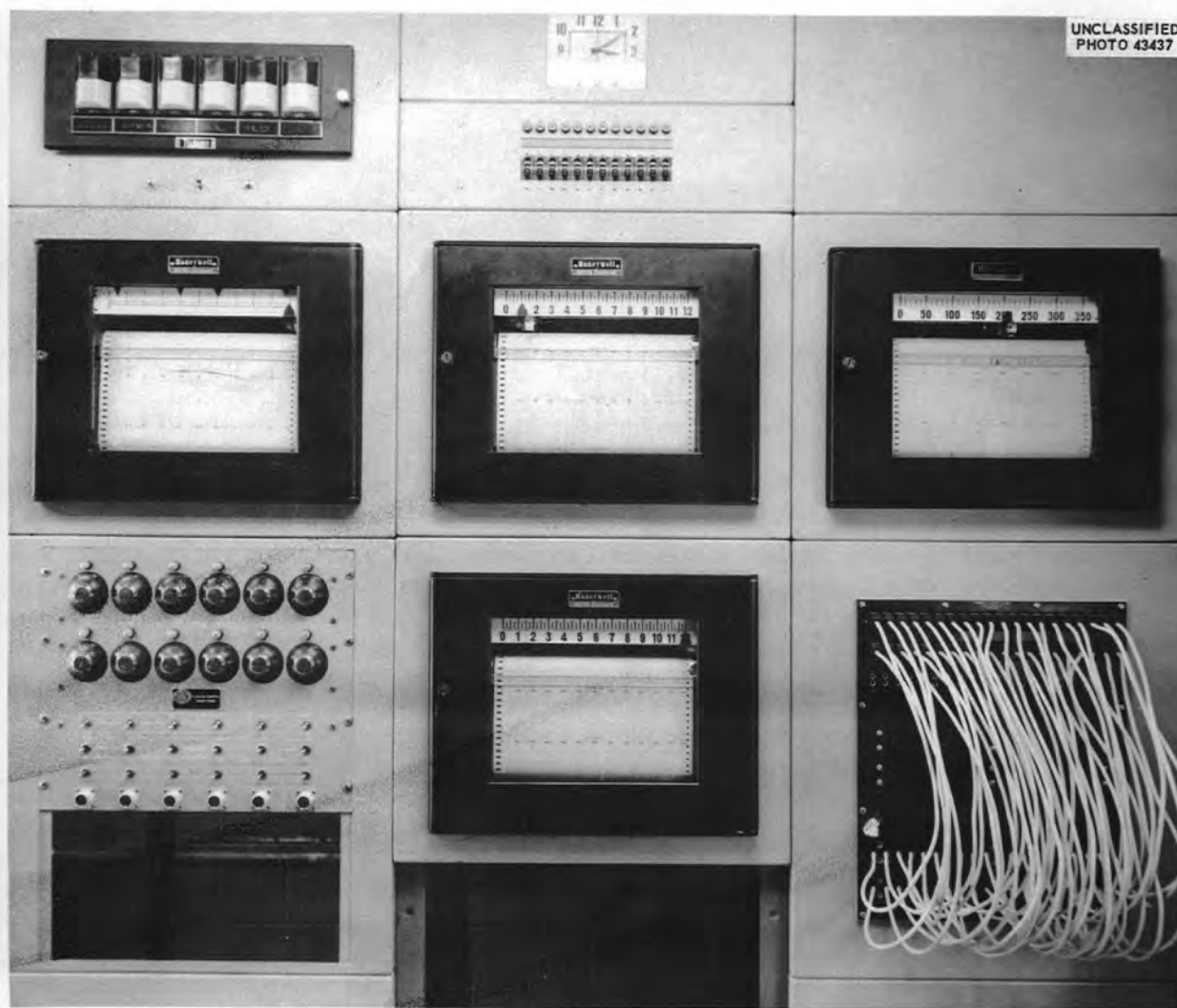


Fig. 34. Temperature Measurement and Power Control Panel.

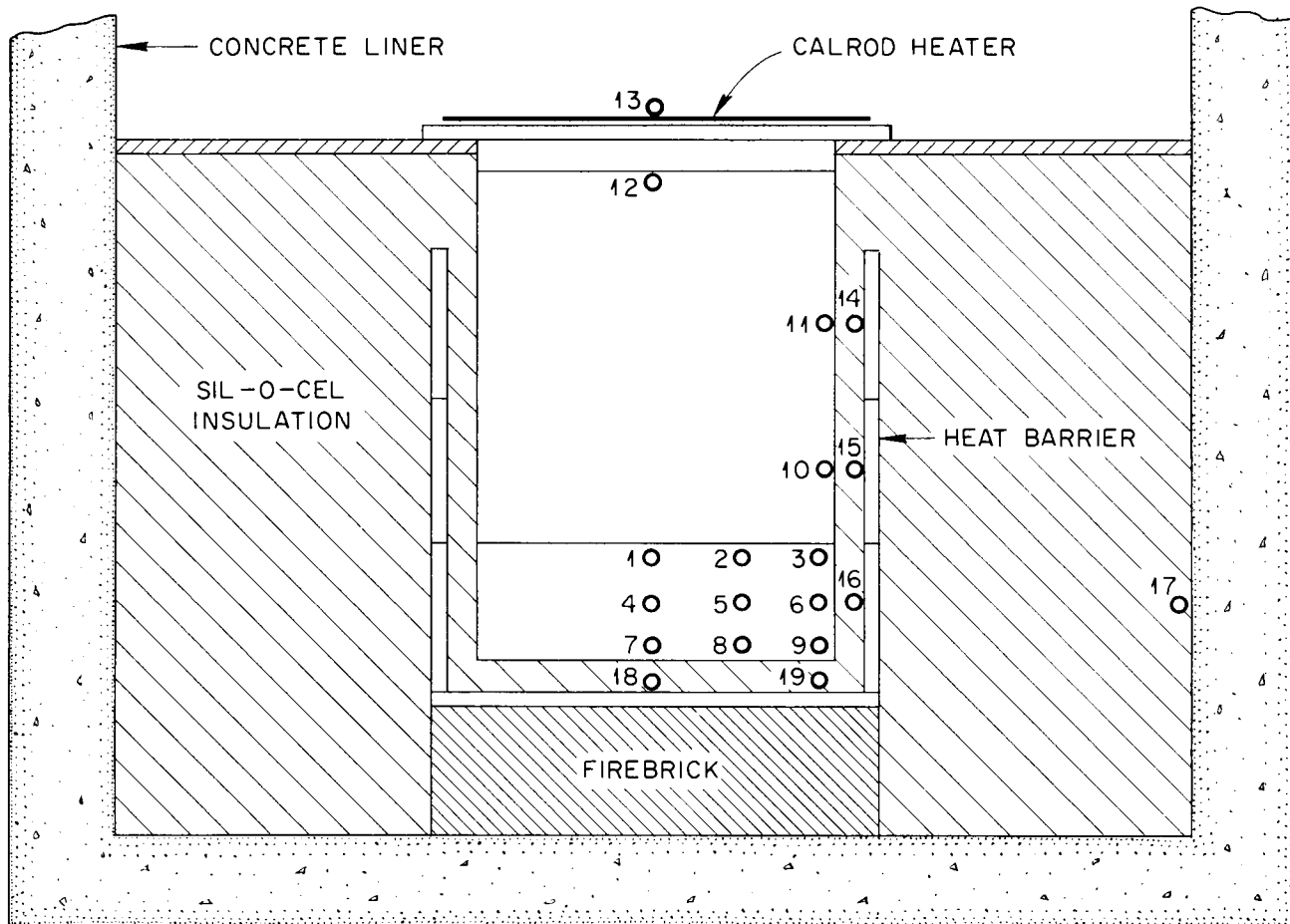


Fig. 35. Location of Thermocouples for Furnace and Heat Barrier.

greater than the preset value, a signal is set to the power regulators and a correction is made in the power applied to the heat barrier. This process continues until the thermocouples are again in balance. A complete scanning of the six control zones is made every 4 min.

Power measurement is provided by wattmeters on each control zone. In addition, an instrument records the total instantaneous power. Flexibility in operation of the heat barrier has been provided by bringing the leads from each heating element to the control house, where changes in heater units can be made by using jumpers and switches (Fig. 37).

Method of Operation. - Before charging the mixer with waste, all components will be checked for operational performance. When this has been

completed, three of the four shield plugs will be placed over the furnace and mixer vessel, providing from 3 to 5 ft of biological shielding. Then, waste solution from a carrier will be jetted to the mixer through a temporary shielded line connected to the valve shown in Fig. 29. When the waste transfer has been completed, this line will be disconnected and the remaining shield plug positioned. From this point in the procedure, the waste will have been isolated inside this vessel. Solid ingredients will be added from the outside by means of the solids feeder, and the waste and solids will be mixed. During this process the mixer will be vented to the off-gas system for evacuation of gases. When mixing has been completed the slurry will be released to the furnace and the heating cycle will start. As soon as a



Upon completion of the firing period, the material in the furnace will cool slowly to ambient temperature. After a period of time sufficient to allow thermal and radioactive cooling, the fired product can be sampled and/or removed for further testing. These operations will be performed after removing the shield plugs and the mixer. If desired, sampling of the final product can be performed by lowering a core-drilling assembly into the mixer brackets and drilling through the mixer discharge hole, or by removing the complete inner furnace assembly (furnace can and cover) and loading it into a shielded container for transfer to a hot-cell facility.

Present Status. - Design and construction of the adiabatic experiment are approximately 90% complete. Since the results obtained in the hot-cell experiments show that the mix 15 sinter will

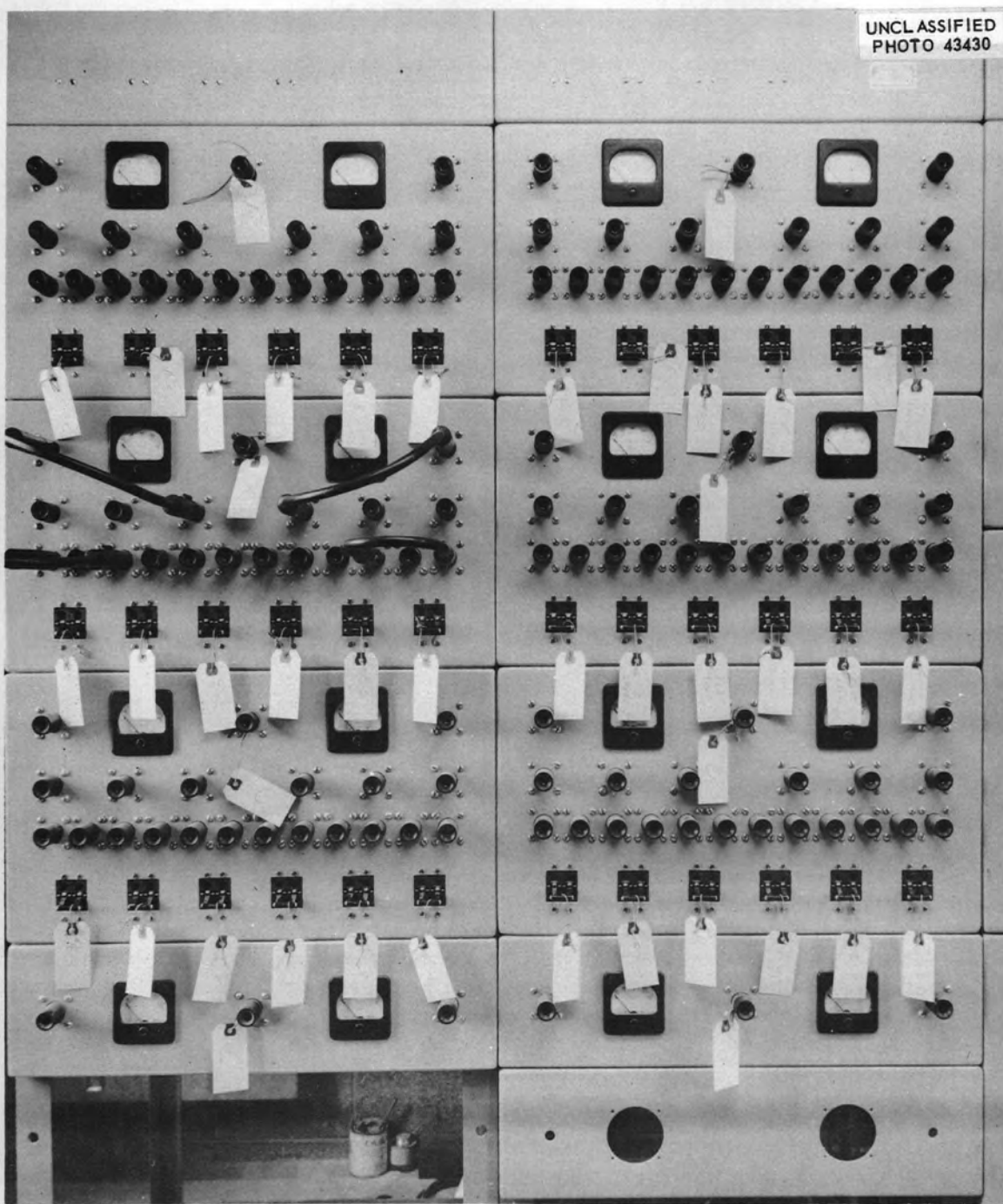


Fig. 37. Power Connections to Heat Barrier, Showing Jumper Connections.

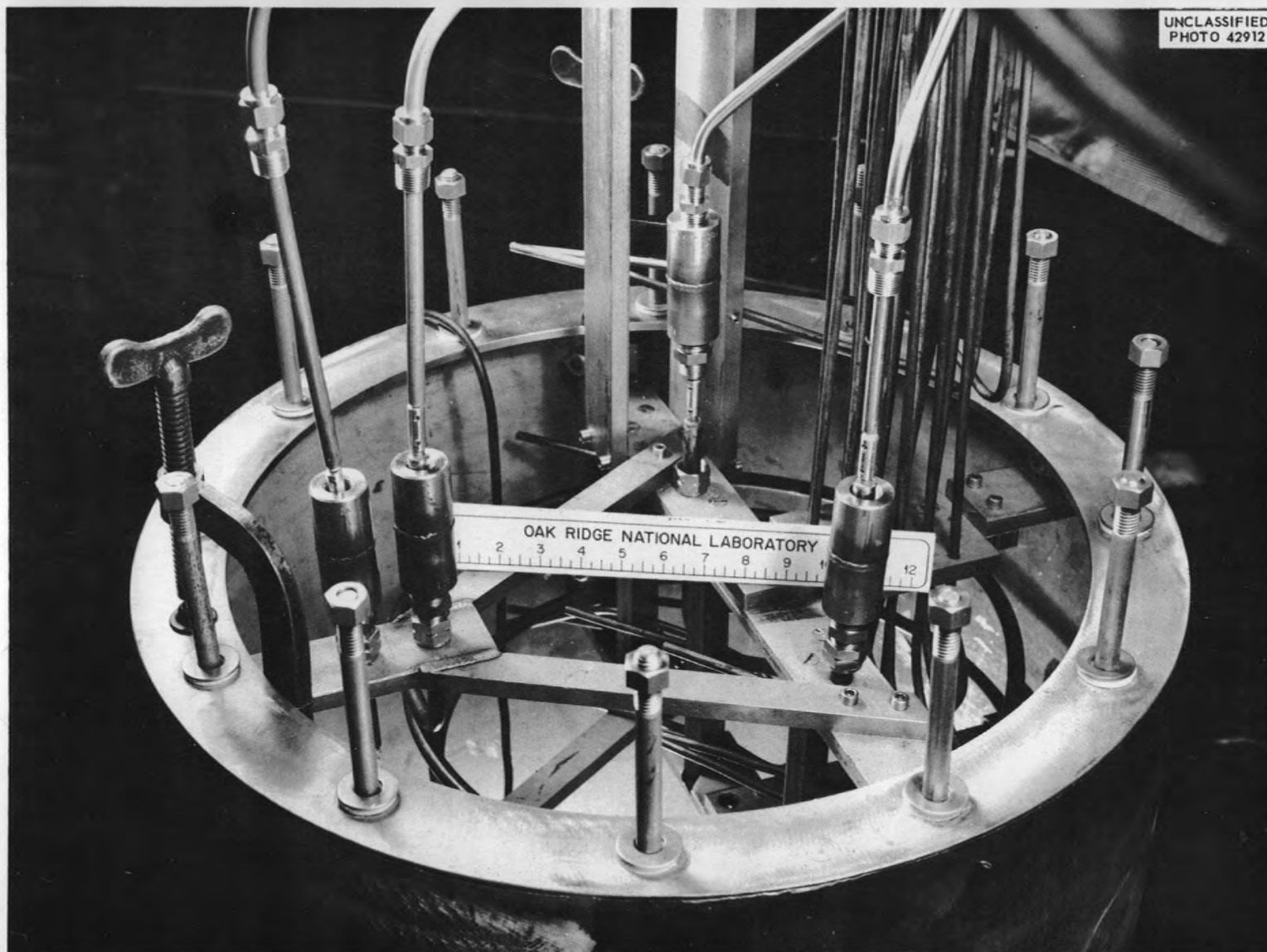


Fig. 38. Furnace with Pretest Heater and Thermocouples Installed.

not retain cesium and strontium to a sufficiently high degree, it has been decided to postpone the completion of this experiment until a new mixture has been obtained. During the interim, the construction of the equipment will be completed and field testing of the components will be started.

Theoretical Calculation of Maximum Temperature During Self-Sintering. — A mathematical program was written for the IBM 704 computer to obtain information on the maximum temperature achieved during self-sintering and the temperature distribution in the final product. A. B. Newey, W. S. Henderson, Jr., D. A. Madden, and W. A. Peter, Jr., of the MIT Practice School, prepared the program and obtained computer data for one set of conditions. The program is so written that the important parameters can be varied. Major variables are the material surrounding the pit (viz., salt or earth), type and amount of insulation, pit dimensions, and initial power generation rate in the waste solution.

The initial calculation was in terms of a pit 6 ft in diameter by 10 ft deep, containing a final ceramic clinker 2 ft thick, located in the ground with the furnace surrounded by $1\frac{1}{2}$ ft of Sil-O-Cel insulation. The initial rate of heat generation was 17.7 w/gal, a value similar to that used in the Pilot Pit 2 experiment.

Although the data have not been completely analyzed at this time, the maximum temperature calculated was 2796°F (1535°C), which is in excess of the temperature required for sintering ($\sim 800^\circ\text{C}$). This temperature would have been achieved 40.35 days after the experiment was started.

Another calculation was attempted by use of the Oracle computer, but, during the time interval available, the program could not be debugged and run. The program will be completed at a later date.

Preliminary Evaluation of Hazards of the "Adiabatic" Self-Sintering Experiment

The hazards associated with the normal and routine operation derive from two sources: (1) the radiation fields existing in the various areas occupied by operating personnel and (2) air-borne contamination resulting from off-gas emission. Such hazards are controllable because the radiation fields are determined by the type and thickness

of the biological shields provided for the experiment, while the air-borne contamination is determined by the efficiency of the off-gas system.

The calculated dose rates in all accessible areas in the vicinity of the furnace and mixer assembly and in the control room will be less than 1 mr/hr. During the charging operation, gamma ray streaming will lead to dose rates up to 1.5 r/hr directly above the two loading pipes penetrating the shield. Similar calculations will be performed to determine the dose rates in the vicinity of the off-gas system as soon as additional data are obtained on the expected source distribution within this system.

In the absence of the final operating procedures it is impossible to perform a hazards study by investigating the probability of a significant release of activity. Study has been limited to an analysis of the theoretical consequences of releases of a specific nature without regard to the mechanism of the release itself.

Subsurface Release. — A failure of the inner and outer shells could result in a total loss of the contained activity. If this release were to occur below the ground surface there would be some justification for comparing this situation with that existing beneath the chemical-waste pits.⁴⁰ With the possible exception of some of the shorter-lived nuclides, such as Zr^{95} and Sr^{89} , these pits have received over the past several years activities comparable to those proposed for use in this experiment with regard both to type and amount. As has been shown,⁴⁰ only negligible amounts of this activity escape from the immediate vicinity of the pits. In the case of a partial release, the fraction of activity not escaping into the ground would add to the difficulty of dismantling the experiment, but this would not be expected to increase the small and inherent hazard of that operation by a significant amount. In either case, other than an abortive ending to the experiment, no unusual problems would be expected.

Surface Release. — A release in which all or a portion of the activity reaches the surface of the ground represents one of the most serious accidents

⁴⁰K. E. Cowser and F. L. Parker, *Soil Disposal of Radioactive Wastes at ORNL; Criteria and Techniques of Site Selection and Monitoring*, paper presented at the Annual Health Physics Meeting at Pittsburgh, June 17-19, 1957; *Health Phys.* (to be published).

that could occur. Dose rates in the vicinity of such a release would be sufficiently high to deliver a severe exposure to anyone within the immediate area even though area evacuation is rapid. In the event of such a release most of the activity would be retained in the first few inches of the soil wetted by the waste solution. Although the ion exchange properties of the soil would cause some degree of fractionation of the activity retained at any particular depth it is assumed for this case that all the released activity is retained homogeneously within a finite depth. This assumption is made for reasons of computational simplicity. The dependency of the dose rate on the area and thickness of a zone contaminated with the entire 10^5 curies has been calculated and is shown in Fig. 39. The progressively smaller reduction in dose rate with increases in the thickness of the contaminated zone indicates that only the upper few inches of the active zone

contribute significantly to the dose rate. This suggests the possibility of using chemical agents to strip the activity from the soil in this zone, thus allowing the activity to penetrate to a greater depth to take advantage of the shielding properties of the uppermost soil layer.

Atmospheric Release. — While the preliminary analysis of a direct release to the atmosphere has not been completed, it is not expected that any other than a sudden release of a major portion of the activity would result in dangerous levels of air contamination in areas remote to the point of release. An analysis of a typical off-gas escape, assuming a release of 2×10^4 curies over a 100-hr period, indicates a maximum downwind concentration of 10^{-6} to 10^{-5} $\mu\text{C}/\text{cm}^3$ at a distance of 500 m.

Small-Scale Experiments in 4507 Hot Cell

Tests were conducted on the retentivity of the sinter of mix 15 for fission products from actual waste solution.⁴¹ The activity of the waste used was about 1 curie/ml. The initial activity of the waste solution used in each run is shown in Table 28. Although the volume of the actual waste solution is lower in the first two runs, the total volume was brought to 12 cc by the addition of synthetic waste solution. The waste-clay mixture was slowly heated to approximately 800°C and was allowed to cool to room temperature. A known volume of water was added to the sinter, and the gross gamma and beta activities in the water were determined (Table 28). The sinter produced in run 5 exhibited the highest retention of fission products. The sinter produced in run 5 also differed in appearance from those of the two previous runs, being hard and dense in contrast to the much softer sinters formed in runs 3 and 4. Furthermore, in runs 3 and 4 the sinter had risen to the rim of the stainless steel container. The heating period was approximately 40 hr for runs 4 and 5, compared with 14 hr for runs 1, 2, and 3. Though the reason for the different sinter characteristics for the runs is not definitely known, observations indicate that the rate of heating and the moisture content of the mix are important variables.

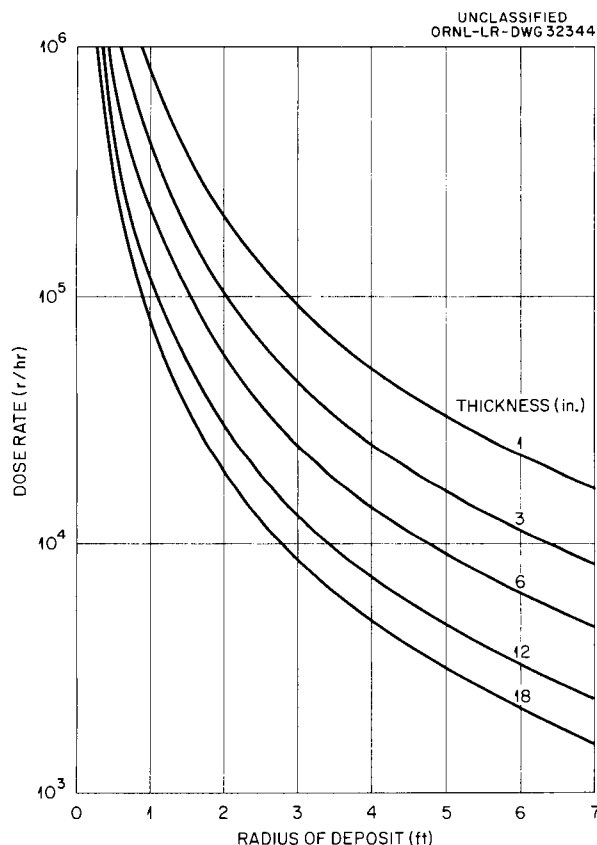


Fig. 39. Assumed Ground-Level Dose at Center of Contaminated Zone, as a Function of Radius and Thickness.

⁴¹For a description of mix 15 see T. V. McVay, R. L. Hammer, and M. P. Hayden, *Sanitary Engineering Aspects of the Atomic Energy Industry. A Seminar Sponsored by the AEC and the Public Health Service, Held at the Robert A. Taft Engineering Center, Cincinnati, Dec. 6-9, 1955, TID-7517(Pt. 1b), p 336.*

Table 28. Leaching Tests of a Sintered Ceramic Mass Made from Simulated Aluminum Nitrate Waste and Mix 15

	Run 1	Run 2	Run 3	Run 4	Run 5
Volume of actual waste solution (ml)	2	6	12	12	12
Radioactivity of waste					
Gross γ , counts/min	1.6×10^9	4.8×10^9	9.6×10^9	9.0×10^{10}	9.0×10^{10}
Gross β , counts/min	4.0×10^9	1.2×10^{10}	2.4×10^{10}	4.9×10^{11}	4.9×10^{11}
Volume of leaching water (ml)	40	40	55	55	55
Time of contact (hr)	6	6	6	16	6
Activity of leach water					
Gross gamma (counts/min)	3.4×10^8	8.9×10^8	1.8×10^9	3.3×10^9	4.4×10^8
Gross beta (counts/min)	1.5×10^8	3.1×10^8	7.3×10^8	2.8×10^9	2.2×10^8
Per cent of gamma leached	21	18	20	3.7	0.5
Per cent of beta leached	4	3	3	0.6	0.04

The fraction of the total activity found in the leaching water was determined (Table 28), but corrections were not made for the loss of activity during heating. The results in Table 29 show that the activity lost during heating is relatively low; thus, the error introduced by neglecting this loss is not significant.

Radiochemical analyses were made of the initial waste solution and of the water used to leach the sinter. The data for strontium and cesium retention are given in Table 30. The element most completely leached by water is cesium. Strontium retention, though unsatisfactory from the standpoint of safe disposal, is better than that of cesium. The percentage of the individual isotopes leached was determined by dividing the activity of that isotope in the leaching water by the total activity of that isotope in the system. Note, in particular, the decreased activity of cesium in the leaching water after 72 hr contact time as compared to the activity after 6 hr. An explanation for this behavior is possible: in runs 1, 2, and 3 the heating period was limited to 14 hr and this duration of heating may not be sufficient to complete the reactions occurring in the waste-clay mixture. One of the reaction products is a sodium silicate glass. The glass on contact with water could decompose, releasing any entrapped cesium.

Since all components are left in the water system, it is possible for them to recombine to form new phases which have ion exchange properties. Such phases could then sorb the cesium in the solution.

Inspection of Tables 28 and 30 will show that over 90% of the gross gamma activity in the leaching water is due to cesium. The results of these tests strongly suggested re-evaluation of mix design with emphasis on the conditions involved in fission product retention and, in particular, the conditions controlling cesium retention.

In order to determine the nature of the off-gases released during sintering, a series of experiments were performed. Figure 40 shows the equipment used in the first four runs; in run 5 the first caustic bubbler was replaced with a multibed filter⁴² (sand, soda lime, activated carbon). Sampling of the off-gas system was performed at various predetermined temperature values (usually 90, 300, 500, and 800°C). The results of the five runs are presented in Table 29.

The samples removed from the off-gas system were analyzed for various important fission products. Table 31 shows the concentrations of fission

⁴²W. J. Boegly *et al.*, *H-P Ann. Prog. Rep.* July 31, 1957, ORNL-2384, p 62.

Table 29. Off-Gas Results from Hot-Cell Runs 1 Through 5

	Run 1	Run 2	Run 3	Run 4	Run 5
Volume of "hot" waste (cc)	2	6	12	12	12
Waste activity					
Gross γ , counts/min	1.6×10^9	4.8×10^9	9.6×10^9	9.0×10^{10}	9.0×10^{10}
Gross β , counts/min	4.0×10^9	1.2×10^{10}	2.4×10^{10}	4.9×10^{11}	4.9×10^{11}
Off-gas activity					
Condensate, gross γ , counts/min	2.5×10^5	3.8×10^5	1.4×10^6	2.8×10^6	2.5×10^6
First caustic scrubber, gross γ , counts/min	4.8×10^6	1.8×10^6	3.0×10^7	6.6×10^7	Multibed filter
Second caustic scrubber, gross γ , counts/min	8.0×10^6	7.2×10^6	1.3×10^7	1.4×10^8	5.6×10^6
Total gross γ , counts/min	1.5×10^7	9.4×10^6	4.4×10^7	2.1×10^8	
Condensate, gross β , counts/min	6.2×10^6	2.4×10^6	9.0×10^6	7.3×10^6	4.4×10^6
First caustic scrubber, gross β , counts/min	1.4×10^6	6.1×10^5	6.5×10^6	1.0×10^7	Multibed filter
Second caustic scrubber, gross β , counts/min	2.5×10^6	8.0×10^6	6.4×10^6	1.8×10^7	3.7×10^6
Total gross β , counts/min	1.0×10^7	1.1×10^7	2.2×10^7	3.5×10^7	
Gross γ , per cent of feed	0.94	0.20	0.46	0.23	
Gross β , per cent of feed	0.25	0.09	0.09	0.007	

products in the bubblers and traps at various sampling temperatures during run 4.

In general the results of the five runs indicate that the amount of activity released to the off-gas system decreases as the firing temperature increases. The amount of activity collected in the off-gas system varied from 0.20 to 0.94% for the gamma counts and from 0.007 to 0.25% for the beta counts. In run 5 it was not possible to analyze the multibed filter for radioactivity, but a comparison of the results of this run with run 4 indicates that a removal of 50 to 90% of the activity passing the condensate trap may have been accomplished in the filter. Further studies will be required to more adequately determine the efficiency of this filtering unit. Of all five runs, run 4 is felt to be the most representative in terms of off-gas production.

Laboratory Studies of Fixation Mechanisms

The hot-cell results suggested the investigation of the probable mechanism(s) involved in the retention of cesium by ceramic sinters. The

difficulty of retaining cesium in solid form has been reported by workers in other laboratories.^{43,44} Although numerous clay materials have been investigated for retention of fission product wastes, the shale used in mix 15 had not been characterized sufficiently. Therefore mineralogic studies were made of the shale by use of x-ray diffraction techniques. From the x-ray, chemical, and base exchange data, the shale was found to contain 35% quartz, 25% vermiculite, and 40% illite.

Shale and pure clay minerals available in the laboratory were used to study the sorption of cesium from 1.67 M $\text{Al}(\text{NO}_3)_3$ solutions. The results in Fig. 41 show that the shale has the highest affinity for cesium in the concentration range studied. Samples of illite were obtained

⁴³A. B. Amphlett and D. T. Warren, *Fixation of Activity in Solid Form by Absorption on Soils*, AERE-C/R-2202 (April 1957).

⁴⁴M. I. Goldman et al., *Report of Working Meeting on Fixation of Radioactivity in Stable, Solid Media at the Johns Hopkins University, June 19-21, 1957*, TID-7550, p 7.

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Table 30. Retention of Strontium and Cesium in Sinters Produced from High-Level Fission Product Waste and Mix 15

	Run 1	Run 2	Run 3	Run 4	Run 5
Initial activity in waste solution (counts/min)					
Sr	3.0×10^8	9.0×10^8	1.8×10^9	3.4×10^{10}	3.4×10^{10}
Cs	2.4×10^8	7.2×10^8	1.4×10^9	1.7×10^{10}	1.7×10^{10}
Volume of leaching water used (ml)					
	40	40	55	55	55
Activity of leach water					
Sr					
6-hr contact time	1.7×10^6	8.4×10^6	4.6×10^7	5.2×10^7	3.6×10^7
72-hr contact time	1.0×10^6	1.3×10^7	3.2×10^7	4.6×10^7	
Cs					
6-hr contact time	3.2×10^8	6.4×10^8	5.2×10^8	3.2×10^9	4.3×10^8
72-hr contact time	1.0×10^8	1.7×10^8	4.3×10^8	5.2×10^9	
Per cent Sr leached in:					
6-hr contact time	0.57	0.93	0.26	0.15	0.11
72-hr contact time	0.33	1.4	0.18	0.14	
Per cent Cs leached in:					
6-hr contact time	100	89	37	19	2.5
72-hr contact time	42	24	31	30	

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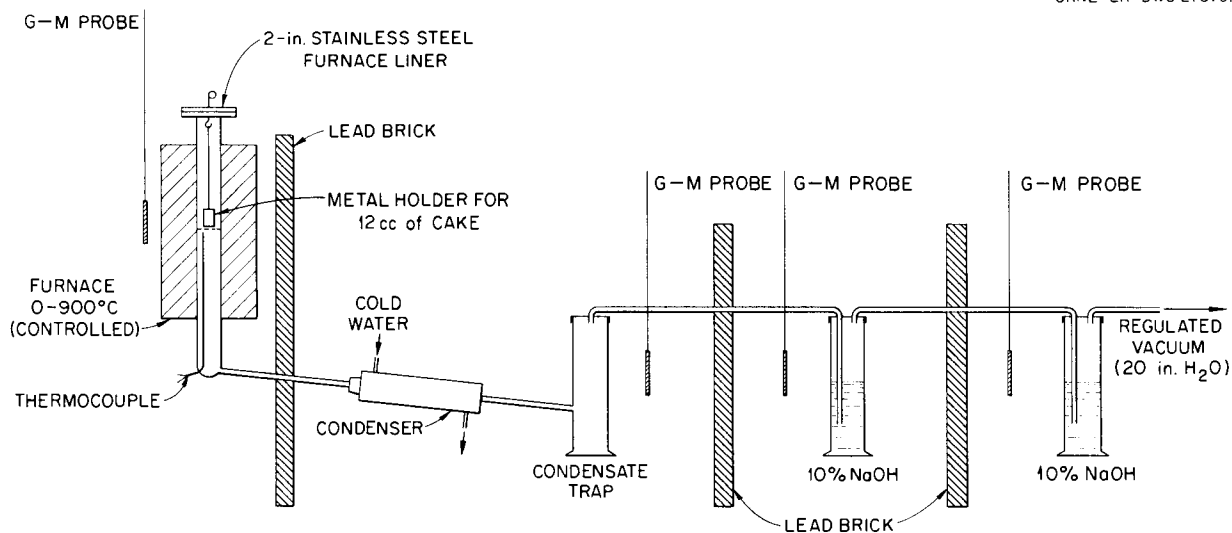


Fig. 40. Sintering and Off-Gas Equipment for High-Activity Experiments.

Table 31. Fission Product Activity Collected in Condensate and Caustic Scrubber, Run 4

Vessel	Temperature (°C)	Activity (counts/min)							
		Gross β	Gross γ	Zr-Nb γ	Cs γ	Sr β	Ce β	TRE β	Ru γ
Feed		4.9×10^{11}	9.0×10^{10}	8.8×10^9	1.7×10^{10}	3.4×10^{10}	2.6×10^{11}	6.7×10^{10}	2.1×10^{10}
Condensate	90	2.5×10^6	5.5×10^5	2.0×10^5	3.0×10^5	2.7×10^5	1.5×10^6	4.8×10^5	7.8×10^4
	300	6.5×10^5	7.9×10^5		4.0×10^5				6.3×10^4
	500	9.7×10^5	2.7×10^5		8.5×10^4	2.6×10^4			1.0×10^5
	800	3.2×10^6	1.2×10^6	6.2×10^4	2.2×10^5	3.2×10^5	1.5×10^6	4.3×10^5	8.3×10^5
First caustic scrubber	90	2.5×10^6	2.0×10^7	1.9×10^5	2.4×10^5	8.2×10^4	3.0×10^5	9.8×10^4	1.0×10^6
	300	5.6×10^6	3.3×10^7	2.9×10^7	2.7×10^5	2.7×10^5	1.1×10^6	3.5×10^5	3.1×10^6
	500	1.5×10^6	6.8×10^6			1.4×10^5	4.6×10^5	1.7×10^5	8.6×10^6
	800	4.9×10^5	6.1×10^6	5.4×10^6	2.2×10^4	1.6×10^4	6×10^4	2.8×10^4	2.8×10^5
Second caustic scrubber	90	9.2×10^6	9.8×10^7	9.8×10^7	4.8×10^5	3.0×10^5	1.9×10^6	5.0×10^5	3.6×10^6
	300	5.2×10^6	2.3×10^7	2.2×10^7	5.1×10^5	3.7×10^5	1.8×10^6	5.6×10^5	2.4×10^6
	500	2.1×10^6	1.6×10^7	1.5×10^7	1.8×10^5	2.2×10^5	5.2×10^5	1.8×10^5	1.6×10^6
	800	1.2×10^6	5.2×10^6	4.2×10^6		4.8×10^4		6.9×10^4	7.8×10^5

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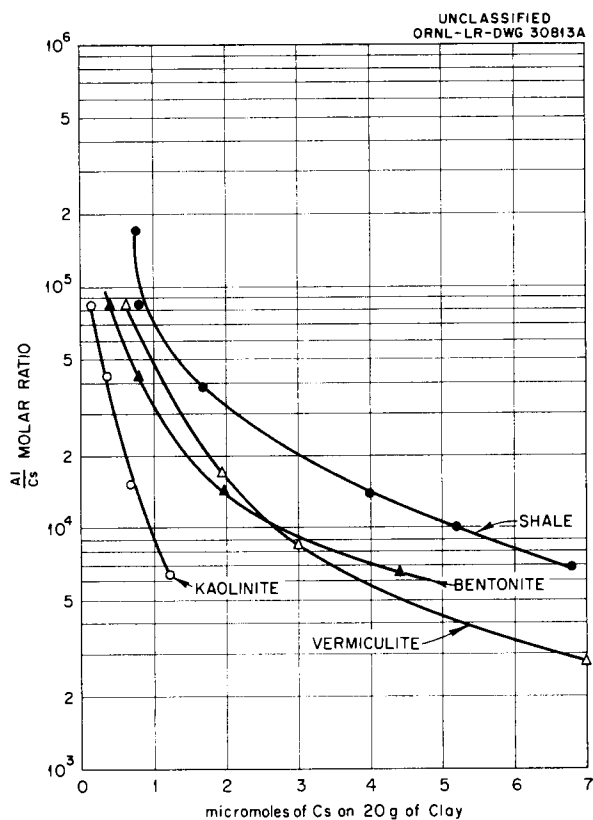


Fig. 41. The Sorption of Cesium by Clays and Shale from 50 ml of 1.67 M Aluminum Nitrate Waste Solution.

from Ward's Natural Science Establishment, and sorption studies confirmed the suspicion that the strong affinity for cesium by the shale was due to the illite component. Results on the cesium sorption by illite and other selected materials are shown in Figs. 42 and 43.

In order to test the hypothesis that the affinity for cesium is related to the potash-containing mica or illite (in contrast to any base exchange relationship), synthetic potassium and sodium fluorophlogopites were ground and treated with synthetic Purex waste neutralized with sodium hydroxide. The quantitative sorption by these synthetic micas is shown in Fig. 44. The dotted line for the sodium mica denotes insufficient experimental data for accurate slope measurement. These studies show that the preferential sorption of cesium is related to structural features of clays more than to base exchange characteristics.

Sorption of Strontium by Clays. — Although existing concepts of clay mineral behavior do not favor

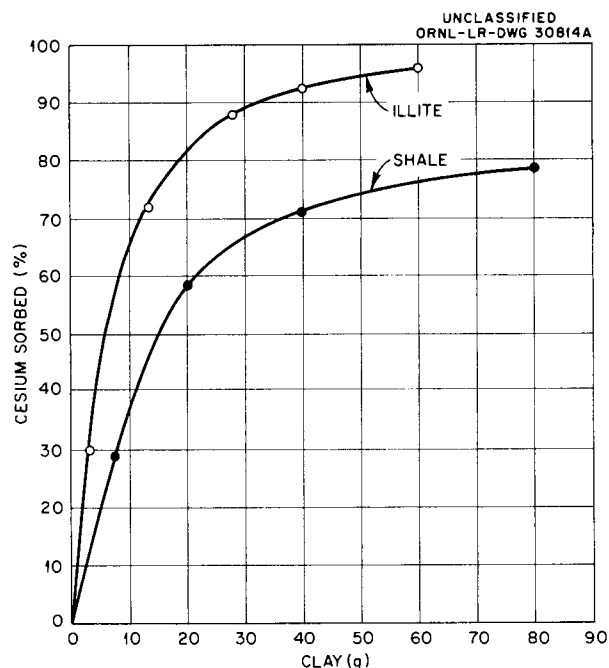


Fig. 42. Cesium Sorption by Fithian Illite and Conasauga Shale. Fifty milliliters of 1.67 M aluminum nitrate solution containing 50 mg of cesium per liter was used.

the sorption of strontium from the concentrated $\text{Al}(\text{NO}_3)_3$ waste solution, several clays were tested. When vermiculite and shale are used, the data in Table 32 support the theory. Vermiculite was selected because this mineral is usually found to be calcium-saturated in the natural state, and therefore suggests an affinity for alkaline earth elements. The significant feature of these results is that the increased valency of strontium, in comparison with cesium, did not result in preferential sorption as might be predicted from sorption studies on organic exchange resins.⁴⁵

Cesium Retention After Heat Treatment. — Although sorption can play a dominant role in removing cesium from solutions at room temperatures, this is no guarantee that the same process is significant at all temperatures. In order to test the retention of cesium as a function of temperature, different materials, after mixing with synthetic Purex waste neutralized with sodium hydroxide (Table 33), were heated in a combustion-type

⁴⁵R. Kunin and R. J. Myers, *Ion Exchange Resins*, Wiley, New York, 1950.

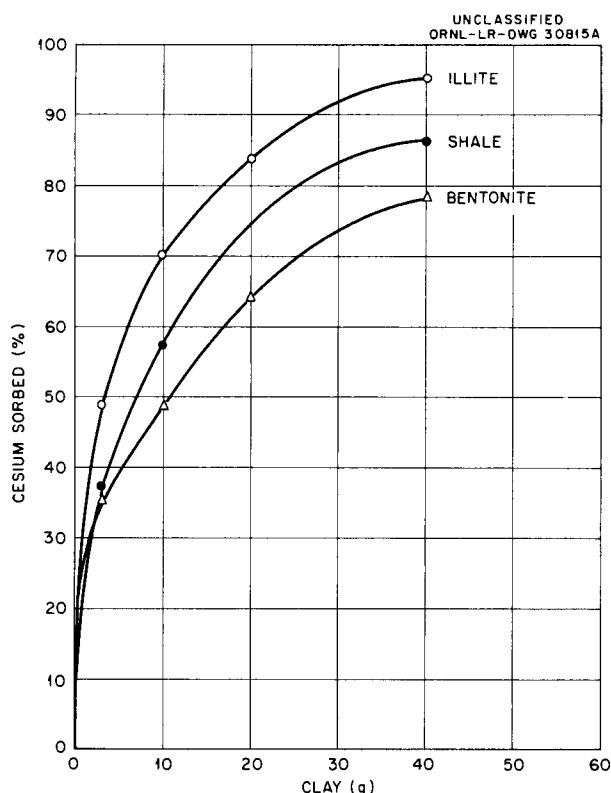


Fig. 43. Cesium Sorption by Fithian Illite, Wyoming Bentonite, and Conasauga Shale. Fifty milliliters of sodium hydroxide neutralized nitric acid waste containing 50 mg of cesium per liter was used.

furnace and samples were taken at 100, 300, 500 or 600, and 800°C. Ten milliliters of distilled water per gram of solid was added to the samples, and the activity of the water was determined at different times. Up to 300°C, sorption plays a major role in the retention of cesium. The data in Table 33 refer to the cesium activity in the water after one week contact. After being heated to higher temperatures and cooled, particularly after 800°C heating, the clays and feldspar show higher retention characteristics. The synthetic mica sample showed excellent sorptive characteristics after being heated at the lower temperatures but poor retention after being heated to 800°C. The feldspar sample, on the other hand, showed poor sorptive characteristics after being heated to rather low temperatures; but retention appears to improve in the samples which were subjected to higher temperatures. The retention of cesium by the feldspar and synthetic mica

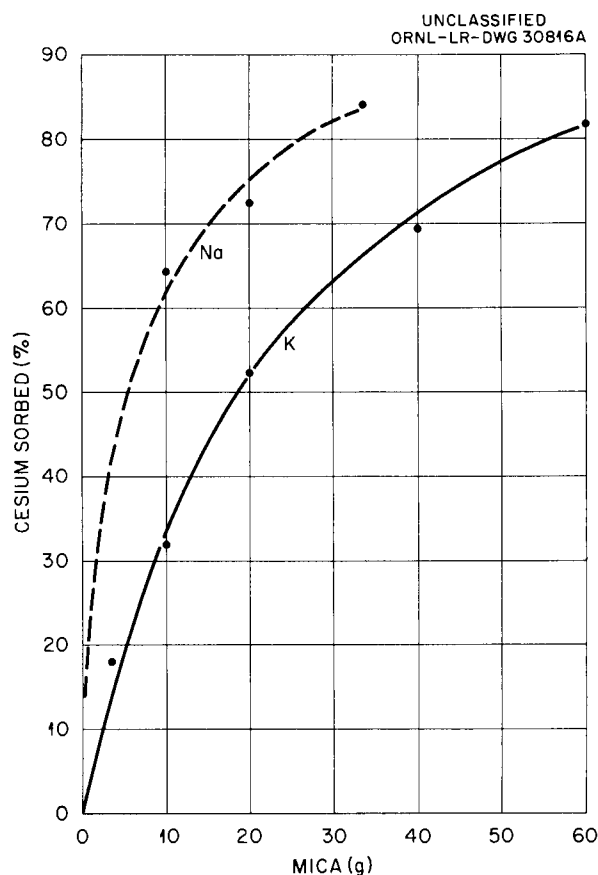


Fig. 44. Cesium Sorption by Synthetic Sodium- and Potassium Fluorophlogopites. Fifty milliliters of sodium hydroxide neutralized nitric acid waste containing 100 mg of cesium per liter.

samples after heating must be evaluated with caution, because the amount of cesium lost during the heating process was not determined.

In Fig. 45 the leaching characteristics of fired bentonite and illite clay are shown. Note that the samples heated at the lower temperature (600°C) release cesium continuously; the activity in the water from the glassy samples (800°C) increases to a maximum value, then decreases with time. This behavior was also noted in studies of actual waste solutions containing cesium, and a possible explanation was given. Although limited data do not justify any positive conclusions, the results on the amount of cesium lost during firing of the waste-clay mixtures are worthy of mention. With increasing amounts of cesium sorbed on the clays,

Table 32. Uptake of Strontium by Conasauga Shale and Vermiculite

Weight of solvent material: 20 g

Solution: 50 ml of 1.6 M $\text{Al}(\text{NO}_3)_3$ with soda ash added and with Sr^{89} tracer

Solvent Material	Strontium Concentration (milligrams per 50 ml of solution)	Initial Activity of $\text{Al}(\text{NO}_3)_3$ Solution (counts/min/ml)	Final Activity of $\text{Al}(\text{NO}_3)_3$ Solution (counts/min/ml)
Shale	0.128	2612	2701
	0.640	2698	2649
	1.28	2660	2608
	2.56	2564	2645
Vermiculite	0.128	2742	2648
	0.640	2700	2635
	1.28	2662	2535
	2.56	2578	2613

the amount of cesium collected in the off-gas train is reduced.

One of the major difficulties associated with leaching tests is the method used for determining the leaching of fission products. Results differed, and depended on the leaching agent, temperature, and mode of leaching. In order to standardize the leaching test, recommendations were made at the Working Meeting held at the Johns Hopkins University.⁴⁶ An apparatus incorporating most of the suggestions by the committee appointed at the meeting has been designed and constructed. Preliminary testing of this apparatus is in progress. Further studies are being made on preparation and evaluation of sinters by increasing the temperature of firing and by changing the compositions of the waste-clay mixtures.

DISPOSAL OF HIGH-LEVEL WASTES INTO ROCK SALT FORMATIONS

The proposed use of salt formations as a disposal medium for radioactive wastes is based upon:

1. the impermeability of salt to the passage of

water, though salt is 1100 times as soluble as limestone,

2. the widespread geographic distribution of salt, Fig. 46,
3. the extremely large quantities of salt available, greater than 6×10^{13} tons as of 1943,
4. the structural strength of salt, 5000 psi,
5. the relatively high thermal conductivity of salt, approximately 3 times as great as that of any other formation,
6. the possibility of recovery at some future time of the wastes injected into the salt,
7. the relative ease of excavating cavities in salt by mining and the even greater ease and lower cost of developing solution cavities in salt, one to ten cents per gallon,
8. the low seismicity in the areas of major salt deposits.

In spite of these favorable factors, the degree to which the structural properties of salt are affected by chemical interaction, pressure, temperature, and radiation will determine the role of salt in the disposal of radioactive wastes.

Heat Calculations

The plasticity of salt and its structural strength change to a considerable degree with changes in temperature and pressure. Therefore, it is

⁴⁶H. C. Thomas, *Report of Working Meeting on Fixation of Radioactivity in Stable, Solid Media at the Johns Hopkins University, June 19-21, 1957, TID-7550, p 87.*

Table 33. Effect of Heat Treatment on Retention of Cesium by Sinters

Sinters prepared by mixing materials with 50 ml of synthetic Purex waste, tagged with Cs¹³⁷ tracer and neutralized with sodium hydroxide, and heating to temperature given.

Run No.	Material	Weight (g)	Cesium Concentration (grams of Cs per liter of solution)	Temperature (°C)	Initial Activity in Waste Solution (counts/min/g)	Activity in Leach Solution (counts/min/g)	Retention (%)
11	Wyoming bentonite	40	0.087	100	2.31×10^5	5.74×10^4	75.2
				300	2.62×10^5	8.23×10^4	68.6
				600	3.62×10^5	6.58×10^4	71.8
				800	3.38×10^5	6.00×10^2	99.8
13	Fithian illite	20	0.087	100	2.42×10^5	1.93×10^4	92.0
				300	3.69×10^5	1.73×10^4	95.3
				500	3.33×10^5	4.63×10^4	86.1
				800	3.95×10^5	5.04×10^3	98.7
6	Fithian illite	60	0.087	800	1.09×10^5	3.30×10^2	99.7
2	Fithian illite	20	0.044	500	5.07×10^4	2.24×10^3	95.6
				800	5.10×10^4	2.80×10^2	99.4
14	Potash feldspar	40	0.087	100	2.63×10^5	2.24×10^5	14.8
				300	3.19×10^5	2.34×10^5	24.6
				600	3.24×10^5	5.54×10^4	82.9
				800	4.35×10^5	1.40×10^4	96.8
17	Sodium fluorophlogopite (synthetic mica)	20	0.087	100	1.77×10^5	8.50×10^2	99.5
				300	2.63×10^5	9.50×10^2	99.6
				500	2.25×10^5	1.15×10^4	94.9
				800	3.59×10^5	7.30×10^4	79.7

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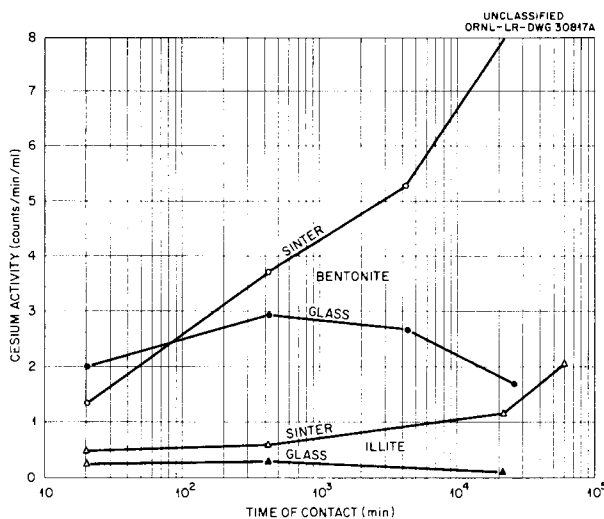


Fig. 45. The Leaching Characteristics of Two Clays, Fired at 600°C (a Sinter) and at 800°C (a Glass).

necessary to determine the temperature rise caused by the storage of radioactive wastes in salt cavities. Initial attempts to compute the maximum possible temperature in a salt cavity were confined to the simpler geometric cases — plane, sphere, and cylinder.

To further simplify the calculations, the heat source (decaying fission products) was assumed to be constant. The thermal conductivity, density, and specific heat of salt were assumed to be constant and independent of the temperature for a particular calculation. Finally, all the heat was assumed to be dissipated by conduction, no changes in state were allowed, no convection currents were permitted in the wastes, and the salt surrounding the cavity was assumed to be infinite in extent. All these assumptions, except the last, are on the safe side: the actual temperature will be lower. In the case of a spherical cavity, the solution for the steady state, with the thermal conductivity of salt equal to the thermal conductivity of the wastes, is

$$T = \frac{A_0 a^2}{2K}.$$

If the waste is considered to be infinitely well stirred, then

$$T = \frac{A_0 a^2}{3K}.$$

In the case of these and the following equations,

r = radial distance from center of sphere, ft,

T = temperature, °F,

K = thermal conductivity,

$$\frac{\text{Btu}}{\text{hr}} \frac{\text{ft}}{\text{ft}^2 \text{ } ^\circ\text{F}} = \frac{\text{Btu}}{\text{hr ft } ^\circ\text{F}},$$

$$A_0 = \text{heat generating source, } \frac{\text{Btu}}{\text{hr ft}^3},$$

a = radius of sphere, ft,

b = radius of salt medium, ft,

t = time, hr,

$k = K/\rho c$ = thermal diffusivity, ft²/hr,

c = specific heat, $\frac{\text{Btu}}{\text{lb } ^\circ\text{F}},$

ρ = density, lb/ft³,

$C = c\rho$ = specific heat per unit volume,

$$\frac{\text{Btu}}{^\circ\text{F ft}^3},$$

$$\text{erfc } x = 1 - \text{erf } x = \frac{2}{\sqrt{\pi}} \int_x^\infty e^{-\xi^2} d\xi,$$

$$i^n \text{erfc } x = \int_x^\infty i^{n-1} \text{erf } \xi d\xi, \quad n = 1, 2, \dots,$$

ξ = variable of integration,

λ = decay constant, hr⁻¹.

If the maximum temperatures permitted at the wall of the sphere are fixed, then the maximum amounts of fission products allowed in spheres of various sizes so as not to exceed the predetermined temperatures are as shown in Fig. 47. If it is assumed that thermal convection occurs in the wastes and that all other conditions remain the same, the permissible concentrations of fission products in spheres are as shown in Fig. 48.

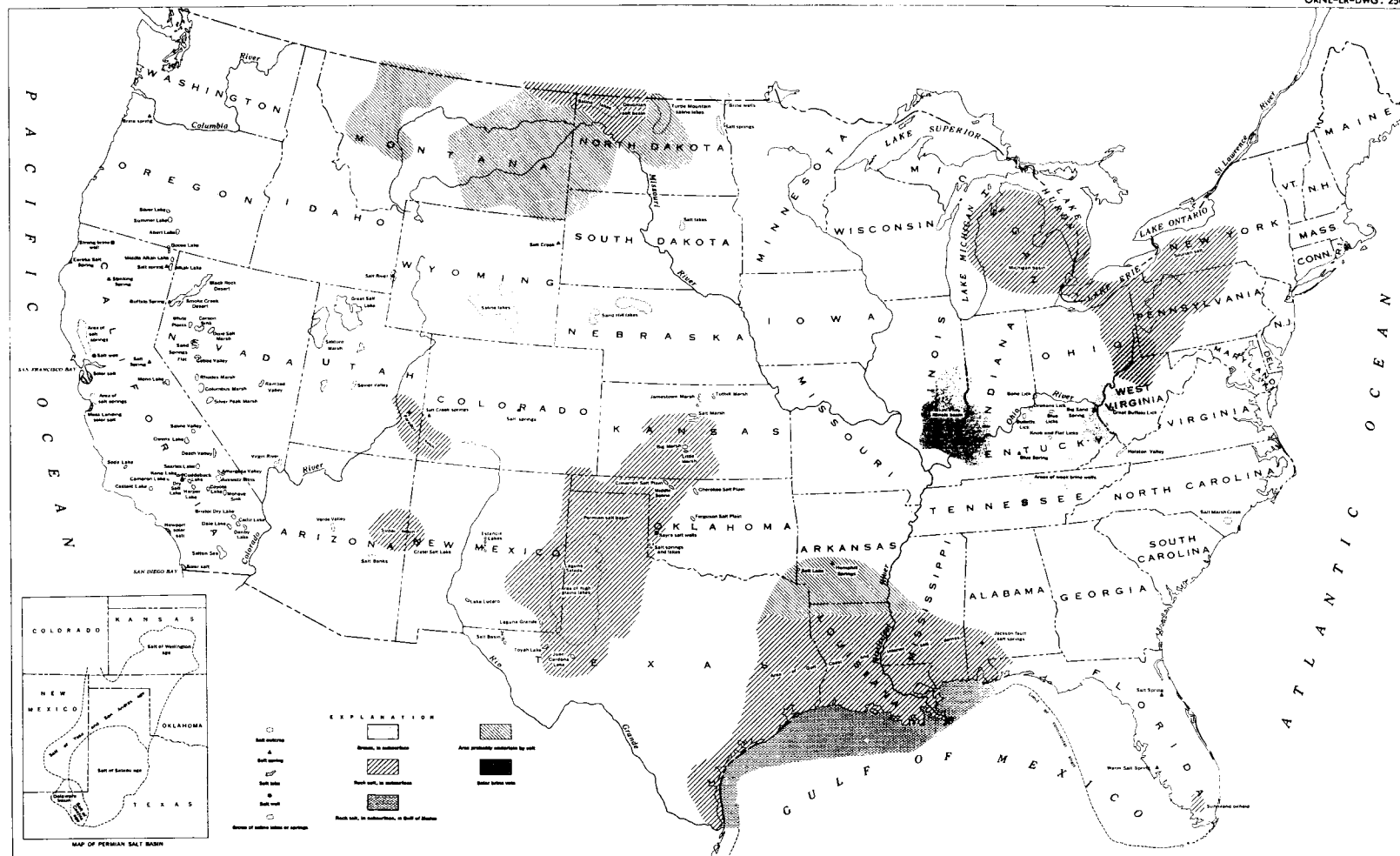


Fig. 46. Index Map of Salt Occurrences in the United States.

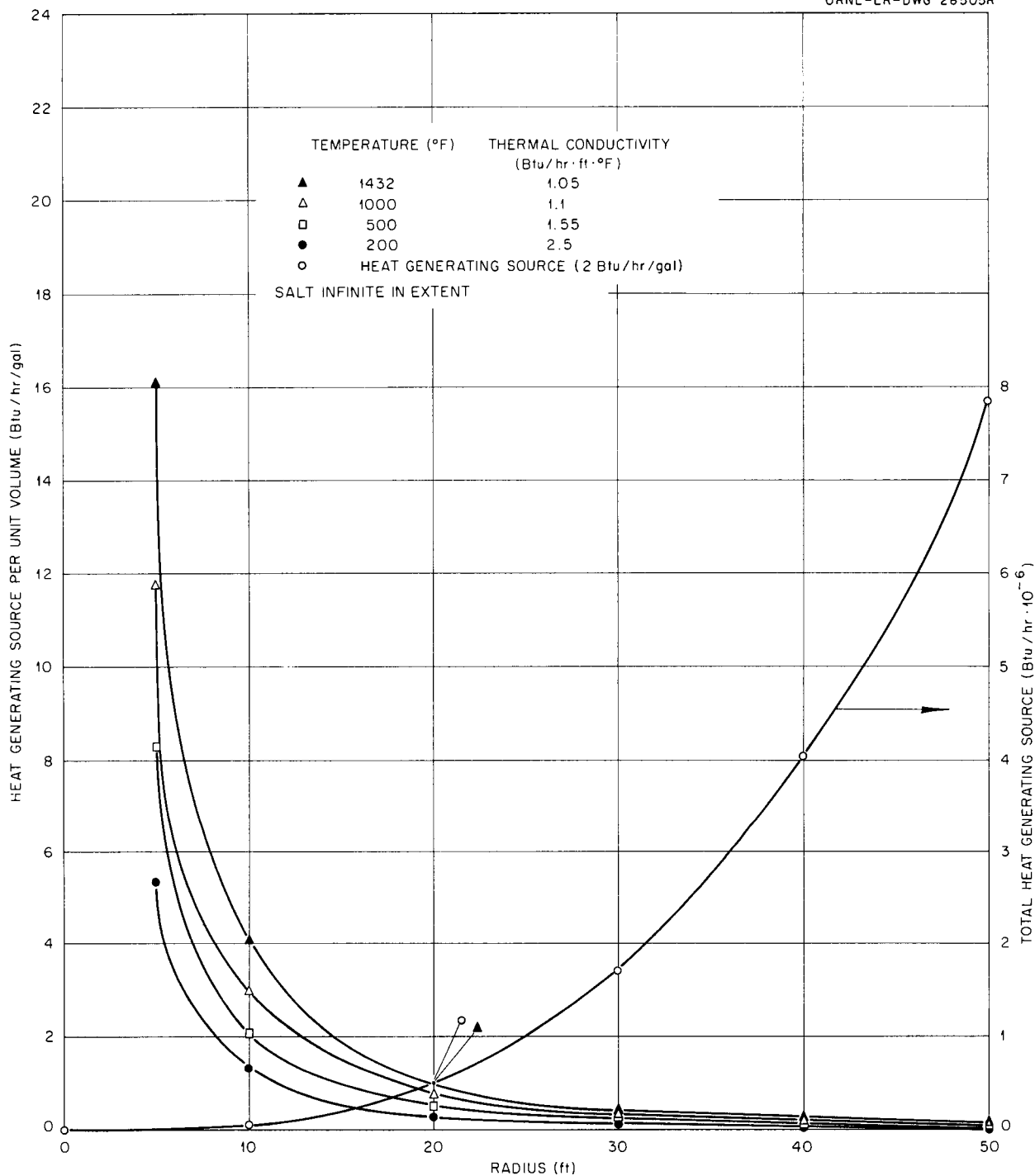
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Fig. 47. Heat Source Required To Achieve Steady-State Temperature in a Sphere; Thermal Conductivity of Salt and Waste Equal; No Thermal Convection.

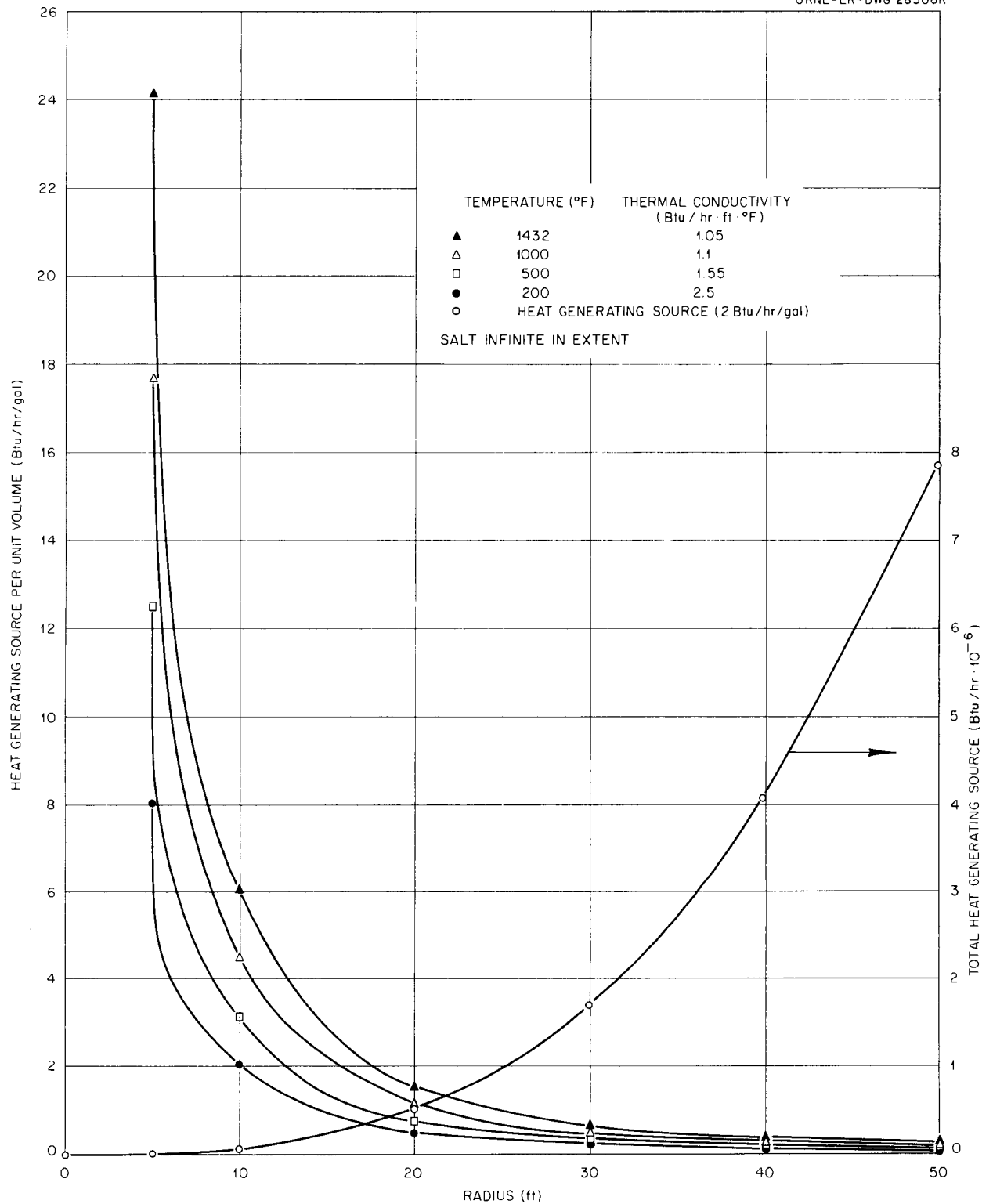
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Fig. 48. Heat Source Required To Achieve Steady-State Temperature in a Sphere; Thermal Conductivity of Waste Infinite; Thermal Convection.

Birch⁴⁷ solved analytically the transient problem for a flat plate and a sphere. He assumed a constant heat source, uniform heat conductivity and specific heat for the waste and the salt, and infinite conductivity of the waste. In the case of the sphere, for $0 \leq r < a$,

$$T(r,t) = \frac{A_0 t}{C} \left[1 + \frac{2a}{r} \left(i^2 \operatorname{erfc} \frac{a+r}{\sqrt{4kt}} - i^2 \operatorname{erfc} \frac{a-r}{\sqrt{4kt}} \right) + 2 \frac{\sqrt{4kt}}{r} \left(i^3 \operatorname{erfc} \frac{a+r}{\sqrt{4kt}} - i^3 \operatorname{erfc} \frac{a-r}{\sqrt{4kt}} \right) \right] ;$$

and for $r > a$,

$$T(r,t) = \frac{2A_0 a t}{Cr} \left[i^2 \operatorname{erfc} \frac{r-a}{\sqrt{4kt}} + i^2 \operatorname{erfc} \frac{r+a}{\sqrt{4kt}} + \frac{\sqrt{4kt}}{a} \left(i^3 \operatorname{erfc} \frac{r+a}{\sqrt{4kt}} - i^3 \operatorname{erfc} \frac{r-a}{\sqrt{4kt}} \right) \right] .$$

These reduce to the results obtained by Crowell⁴⁸ when the steady state is reached (i.e., when $t \rightarrow \infty$). When the heat generating source is assumed to decay exponentially, Birch derives the following solutions. In the case of the sphere, for $r = 0$,

$$T(0,t) = \frac{A_0 a^2}{2Ck} \left\{ e^{-\lambda t} \int_{a/\sqrt{4kt}}^{\infty} e^{\lambda a^2 / 4k\beta^2} \left(\operatorname{erf} \beta - \frac{2\beta}{\sqrt{\pi}} e^{-\beta^2} \right) \frac{d\beta}{\beta^3} \right\} ;$$

and for $r = a$,

$$T(a,t) = \frac{A_0 a^2}{3Ck} \left\{ 3e^{-\lambda t} \int_{a/\sqrt{kt}}^{\infty} e^{\lambda a^2 / k\beta^2} \left[\operatorname{erf} \beta - \frac{2}{\beta\sqrt{\pi}} (1 - e^{-\beta^2}) \right] \frac{d\beta}{\beta^3} \right\} .$$

The functions in the last two equations are not tabulated and therefore must be integrated numerically. Numerical integration for a number of cases would be quite tedious. However, Schecter⁴⁹ independently solved the above equations, assuming a decay rate varying with time. Later, the equations were programed for the Oracle (high-speed digital computer at ORNL), and the maximum temperatures for given waste concentrations in any size sphere for two-, six-, and ten-

year-cooled wastes were reported. However, the time-space distribution of the temperature is not given. The proper size of a test cavity that would show temperature changes in a reasonable time is determined by the temperature-time-space relationship of a six-year-cooled waste (2 Btu/hr/gal), with a constant heat source.

According to Birch, "heating the liquid some hundred degrees (Centigrade) or so would not produce drastic physical changes, and . . . plastic deformation of shale or evaporite layers, if sufficiently thick might continue to provide a tight container." For this reason it was decided to limit the temperature at the wall of the sphere to 200°F. This would eliminate any necessary corrections due to changes in state, since the temperature is kept below the boiling point of the

⁴⁷F. Birch, "Thermal Considerations in the Deep Disposal of Radioactive Waste," *Report for the Committee on Waste Disposal of the Division of Earth Sciences*, NAS-NRC Pub. 588 (in press).

⁴⁸J. C. Crowell, unpublished results, January 1958.

⁴⁹R. S. Schecter, unpublished calculations, January 1958.

wastes. According to previous data it was found that a 2-Btu/hr/gal waste in a sphere 10 ft in radius would not reach a temperature greater than 200°F for the case in which the thermal conductivity of the waste is infinite.

By use of the equations of Birch and Goldenberg,⁵⁰ the time-space distribution of the temperature was calculated for a 20-ft-dia sphere containing a constant heat source, with the thermal conductivity of the waste equal to the thermal conductivity of the salt. The results are shown in Fig. 49.

Computations are in progress to determine the time-space distribution of temperature for the case in which the heat generating source is decaying as a function of time and the thermal conductivity in the sphere is equal to infinity.

Experimental work is progressing to determine the differences in thermal conductivity, if any, between dome and bedded salt deposits. The bedded deposits will be tested with the heat flow parallel or perpendicular to the bedding planes.

Laboratory Studies of Chemical Compatibility

Efforts have been devoted primarily to determining the quantity and chemical composition of the gases evolved after mixing synthetic Purex wastes and acid aluminum nitrate wastes with rock salt and to devising experiments which will indicate the direction and nature of migration of these wastes in natural salt formations. Blocks of rock salt used for these experiments were obtained from Grand Saline Salt Mine, Grand Saline, Texas. Samples for chemical analysis and chemical interaction studies were removed from 1-ft cubes, crushed, sieved, and quartered several times to produce representative samples.

⁵⁰H. Goldenberg, *Brit. J. Appl. Phys.* 2, 235 (1951).

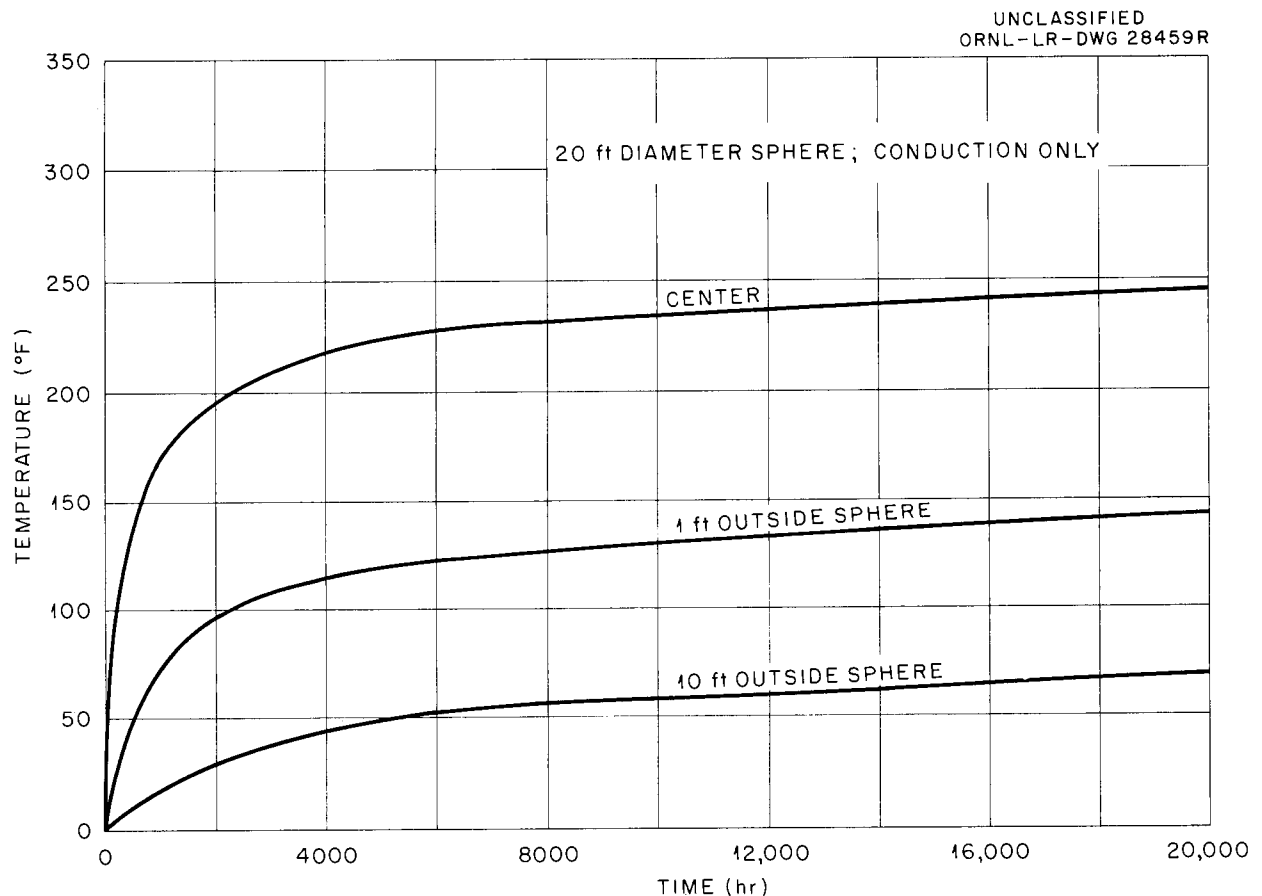


Fig. 49. Time-Space Temperature Distribution of Wastes Stored in Salt Cavities.

Some idea of the chemical purity of this material may be had by comparing the chemical analyses of Grand Saline rock salt and Mallinckrodt analytical grade sodium chloride (Table 34). Spectrographic analysis of the above samples indicated that Ba, Ca, K, Li, Mg, and Sr are present in rock salt and Mallinckrodt salt in concentrations less than 0.01%, while Cu, Fe, and Si were found in concentrations less than 0.001%.

Table 34. Comparative Analysis of Grand Saline Rock Salt and Mallinckrodt Reagent Grade Sodium Chloride

Constituent	Grand Saline Rock Salt	Mallinckrodt Reagent Grade Sodium Chloride
Br ⁻ , mg/kg	200	200
I ⁻ , mg/kg	280	70
NO ₃ ⁻ , mg/kg	10	5
SO ₄ ²⁻ , mg/kg	4440	60
CO ₃ ²⁻ , %	0.1	0.1
H ₂ O, %	0.1	0.1
NaCl*, %	94.7	97.0

*NaCl equivalent of determined Cl⁻.

Chemical reactions of natural rock salt with aluminum nitrate waste solution and with Purex waste solution were studied by saturating the waste solutions with salt and measuring the gas produced at various temperatures and waste concentrations. In practice, 500-ml quantities of the salt-waste mixtures were placed in 500-ml Erlenmeyer flasks equipped with thermometer and gas vent tubes and located in a temperature-controlled water bath. Pressure and volume of the gases produced in the reaction flask were measured by means of a water manometer and gas buret. Cumulative gas production values for both waste types indicated that 2 to 3 ml of gas was produced per 500 ml of solution per day.

Following the above studies, a number of waste-salt solutions were aerated. In these experiments

air was pulled through a reaction vessel (containing 500 ml of waste-salt mixture) at approximately 1 liter/min. The reaction vessel was connected in series with fritted glass gas scrubbers. Absorption and/or colorimetric reagents in 100-ml volumes were placed in the gas scrubbers and arranged in such a manner as to indicate the type of gas being produced. These experiments were designed to indicate the influence of air as a sweep gas and to provide some information relative to the type of analytical procedures necessary for comprehensive gas analysis. Primary attention for these experiments centered about Purex waste types, since several investigators⁵¹ have indicated that NaCl-HNO₃ mixtures exhibit properties similar to aqua regia and evolve NO₂Cl. Thus, several attempts were made to produce this gas in sufficient volume for absorption analysis. Ten-molar sodium hydroxide, methylamine hydrochloride, Griess reagent (sulfanilic acid and naphthylamine), ortho-tolidine, and silver nitrate were used as absorption and colorimetric reagents in the gas scrubbers in an attempt to identify the gaseous products of the waste-salt mixtures. Sodium hydroxide decomposes NO₂Cl and NOCl to yield NO₂ or NO and Cl₂; thus, by forcing the gas stream through the concentrated alkali and decomposing the gas, NO₂ would be available for reactions with methylamine hydrochloride or Griess reagent, whereas the Cl₂ liberated by the decomposition would remain in the NaOH solution. Ortho-tolidine hydrochloride in dilute solutions acts as an oxidation-reduction indicator and may be used for the colorimetric analysis of chlorine, NO₂, NO₂Cl, and other oxidizing agents. This reagent was employed in this experiment to indicate the length of time required for color production, that is, as a qualitative reaction indicator. Results of this study indicated that relatively small amounts of air (5 to 10 liters) were sufficient to promote oxidation-reduction reactions between synthetic waste and salt, resulting in the production of NO₂ and Cl₂ and traces of NO₂Cl.

The influence of beta and gamma radiation on the reaction of aqueous fuel-recovery wastes and natural rock salt can best be studied by

⁵¹J. W. Mellor, *A Comprehensive Treatise on Inorganic and Theoretical Chemistry*, vol 8, p 613, Longmans, Green, London, 1940.

putting radioactive wastes into natural rock-salt formations. However, the hazards of such an experiment must first be determined by small-scale laboratory experiments. If waste solutions containing fission products in representative amounts are used, the identification of reaction products is not readily adaptable to routine analytical chemical methods. In order to overcome this difficulty and to obtain information concerning the role of gamma radiation on the chemical reactions and subsequent gas production, irradiation by means of Co^{60} was used. Waste-salt mixtures, similar to the solutions used in the above study, were prepared in duplicate and placed in 1-liter Pyrex vessels equipped with a vent tube and located in the Co^{60} radiation unit. The radiation facility provided for this study is operated by the Isotopes Division and consists of 1-ft³ radiation chamber surrounded by aluminum-clad cobalt cylinders of varying length, providing a 3.5×10^6 r/hr gamma field. The device was further adapted for this study by connecting the sample containers to the gas measurement apparatus with small-diameter plastic tubing through two thermocouple wells. Gas measurement during the radiation period was accomplished by connecting the plastic off-gas tube to a mercury-filled gas buret provided with a water manometer. Temperature measurements were taken of the gas in the vessel containing the waste-salt mixture, the radiation chamber surrounding the vessel, and the outside air. These temperatures were recorded by means of thermocouples connected to a multi-point Brown recorder for the two pilot experiments. It was not possible to record temperature and gas production for all duplicate radiation experiments, because only two thermocouple wells provide access to the radiation chamber. By correlating the recorded temperatures from the above pilot experiments with the temperature measurements of cooling air circulated through the radiation facility, the temperature of the gas can be calculated when future experiments are performed.

Gas production rates in these experiments were significantly different from those obtained with the earlier nonirradiated salt-waste solutions and thus prompted an attempt to determine the nature of gases being evolved by the irradiation-induced reactions. An Orsat volumetric gas analysis system employing liquid absorbents was used to analyze the gas collected from selected radiation

experiments. The solutions employed in the Orsat apparatus included 15 M NaOH, methylamine hydrochloride, potassium permanganate, ferrous sulfate, and alkaline pyrogallol solutions. These solutions were selected in accordance with the results of the air-mixing experiments referred to earlier. A copper oxide column and electric heater were used to determine hydrogen. The analytical procedure involved collection of the entire amount of gas produced by the solution.

Chemical composition, volumes, and cumulative gas production are shown in Table 35 and Fig. 50. The large difference in volumes of gas produced by acid and neutralized waste may be due either to the difference in acidity or to the presence of hydrous iron oxides produced by neutralization of the waste. Analysis of the gas shows that the mechanism of reaction may be only an accelerated radiolysis of the aqueous media or the prevention of recombination of the decomposition products of water (Tables 36 and 37). The major gases are nitrogen and oxygen. In the acidic 8 M Purex sample, the nitrogen-to-oxygen ratio is relatively constant, whereas the neutralized Purex sample shows an increase in the oxygen content with exposure time.

The high yield per unit volume of solution in run 14 was the result of an increased surface area of salt which, in turn, increased the number of secondary electrons available for reaction in the vessel. This condition indicates that gas production can be increased by a factor of 10 by increasing the surface area of salt exposed to the waste.

Ionic Migration. — Migration of the major ionic constituents and the fission products in fuel-recovery wastes through salt formations may well be influenced by several variables. The migration and resulting contamination may be directly dependent upon the integrity of the formation. During the mining of samples of rock salt for laboratory evaluation and experimentation, small cracks are introduced into the sample which in all probability will affect the results of laboratory testing.

To provide some basis for the above statements, a series of 3-in. salt cubes were removed by means of a handsaw from a large 1.5-ft cube sample. After removal, the small cubes were prepared for leak testing by drilling a 0.25-in.-dia hole in the center of the top face to a depth of 1 in. The samples were placed on a clean stainless steel

Table 35. Cumulative Gas Production of Waste and Waste-Salt Mixtures Exposed to Gamma Radiation

			Dosage, 3.5×10^6 r/hr				
			Initial Sample Volume, 500 ml				
Run	Description of Solution	Saturated with Salt	Cumulative Gas Production (cc)				
			Irradiation Time				
			5.0 hr	10.0 hr	15.0 hr	20.0 hr	30.0 hr
3A	8 M Purex, acid	Yes	143.7	220.7	328.3	420.9	562.0
4A	8 M Purex, adjusted to pH 10.0	No	275.4	443.6	622.1	811.9	1116.8
5A	8 M Purex, adjusted to pH 10.0	Yes	355.9	555.8	686.1	798.3	1086.2
6A	8 M Purex, acid	No	248.4	279.1	295.0	320.4	382.8
7A	4 M Purex, acid	Yes	221.7	230.3	234.8	248.9	
8A	4 M Purex, adjusted to pH 10.0	Yes	290.4	430.7	549.2	689.8	925.2
9A	1.88 M Purex, acid	Yes	215.6	239.9	279.1	317.1	344.2
10A	8 M Purex, adjusted to pH 10.0	Yes	264.1	420.9	547.9	651.1	902.2
11A	5 M Purex, acid	Yes	263.6	300.0	311.0	319.4	396.7
12A	6.27 M Purex, acid	Yes					
13A	8 M Purex, acid	Yes	291.6	349.8	360.6	381.0	479.0
14A	Salt slurry, 8 M Purex, acid	Yes	459.5	505.3			
15	8 M Purex, acid	No	292.8	322.1	339.3	402.7	506.0
16	8 M Purex, adjusted to pH 10.0	Yes					

sheet, and 1 ml of synthetic waste solution was introduced into the drilled cavity. Then, one electrode of an ohmmeter was placed in the solution. The metal plate supporting the salt sample served as the other electrode. Thus, when the solution reached the metal plate, the sample would conduct and deflect the needle of the meter. In general, all the samples indicated that about 3 min was required for the solution to complete the distance through the block. Of the fifteen samples tested by this procedure only two failed to indicate conductance. The same testing procedure has been applied to salt blocks of larger size, and the results are similar.

One method of determining the migration of fission products and associated waste material in salt structures is to immerse the test specimen in saturated salt solution. This testing method tends to decrease the importance of surface faults and provides a fluid medium suitable for radiochemical and quantitative chemical analysis. Migration study 1 represents the first effort to

incorporate the above factors into a laboratory experiment. The salt specimen selected for this study was originally part of the samples used in the conductivity experiments. The 20-cm-square by 30-cm-high salt specimen was prepared by boring a hole in the block, 10 cm in diameter and 17.5 cm deep. One liter of fission product tracer solution, prepared by adding 0.1 mc of mixed-fission-product solution to 1 liter of synthetic acid Purex waste solution, was added to the cylindrical cavity. The drilled block was then placed in a glass jar containing 7.5 liters of saturated salt solution. The assembly was placed in a 40°C water bath; 1-ml samples of the saturated salt solution bath were collected periodically and counted in a deep-well gamma scintillation counter. Additional amounts of salt solution were added to the bath daily to compensate for the volume lost by evaporation. This experiment is still in progress; however, results to date indicate that this procedure, with certain modifications, may be useful for future studies. This experiment has shown

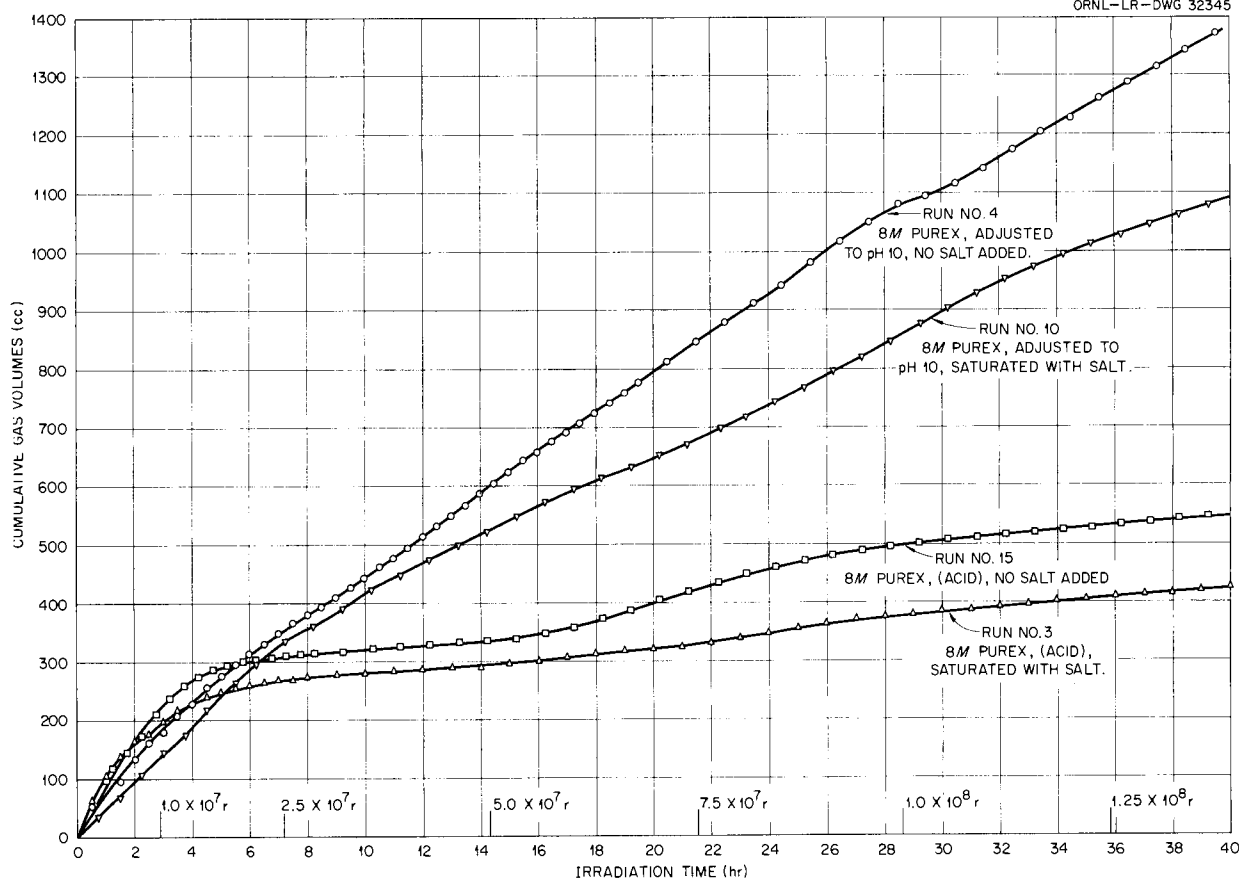
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Fig. 50. Gas Production of Purex-Type Waste Subjected to Gammo Irradiation.

Table 36. Chemical Composition of Gas Produced During Run 15

8 M Purex, Acid, No Salt Added

Sampling Period		Time Elapsed (hr)	Sample Volume (cc)	Composition (%)				
From	To			Cl ₂	NO ₂	O ₂	H ₂	N ₂
1545	1615	0.5	59.5	1.2	1.6	21.6	0.3	75.2
1615	1800	2.25	98.7	0.9	0.2	20.6	0.0	78.3
1800	2030	4.75	97.0	0.4	0.2	20.9	0.1	78.4
2030	0900	17.25	96.2	0.4	0.2	21.8	2.9	74.6
0900	1400	22.25	67.8	0.4	0.9	24.9	1.2	72.6
1400	2000	28.25	81.9	1.8	1.7	26.3	2.3	67.9
2000	0900	41.25	56.5	1.2	1.4	25.0	2.3	70.2
0900	1621	48.60	43.3	1.6	1.0	25.5	3.0	69.1

Table 37. Chemical Composition of Gas Produced During Run 10B

8 M Purex, adjusted to pH 10, saturated with salt

Sampling Period		Time Elapsed (hr)	Sample Volume (cc)	Composition (%)				
From	To			Cl ₂	NO ₂	O ₂	H ₂	N ₂
0940	1030	0.83	60.3	0	0	19.6	0	80.4
1030	1330	3.83	97.0	0	0	21.1	0	78.9
1330	1515	5.58	99.0	0	0	28.2	0	71.8
1515	1800	8.33	95.8	0.5	0	28.9	0.6	69.9
1800	2100	11.33	94.3	0	0	34.0	0.5	65.4
2100	0200	16.33	94.8	0	0	41.0	1.2	57.9
0200	0600	20.33	80.3	Trace	0.5	41.5	1.6	56.3
0600	1200	26.33	39.8	0	0.2	53.0	2.3	44.5
1200	1500	29.33	68.4	0.3	3.8	57.2	3.6	35.1
1500	2100	35.33	98.2	Trace	1.1	70.1	3.6	25.0
2100	0500	43.33	94.8	0	7.5	59.4	3.9	29.3
0500	0815	46.58	58.9	0	13.7	41.5	1.4	41.5
0815	1500	53.33	88.7	0	3.2	70.1	4.5	22.2
1500	0400	66.33	97.6	0	5.0	70.7	4.0	20.3
0400	0815	70.75	96.2	0	1.0	44.1	1.9	52.6

that the migration of fission products reaches a maximum in about 20 days and remains constant thereafter. One difficulty which accompanied the above experiment was recrystallization and crystal growth from the central cavity to the surrounding bath. Samples of the recrystallized fission-product-salt mixture taken from the block indicate that the fission product activity was probably introduced into the surrounding saturated salt solution by successive recrystallization of the waste-salt mixture rather than by migration through the salt. Subsequent experiments in which recrystallization is inhibited are in progress, including separate studies designed to study the extent of fission product migration by means of recrystallization.

Experience with sintered aluminum nitrate waste-clay mixtures has shown that aluminum hydrates effectively reduce the mobility of fission products in aqueous solutions. Thus a series of experiments designed to relate migration or leachability of fired salt mixtures was begun. Synthetic

aluminum nitrate waste solutions, in various concentrations, were mixed with rock salt in the ratio of 10 ml of waste to 10 g of salt. The mixture was transferred to ceramic crucibles and heated for 1 hr at the temperature listed in Table 38. After being heated, the samples were leached in 500 ml of saturated salt solution until quantitative analysis revealed the changes in pH or Al⁺⁺⁺ retention. The experiment was repeated with additional samples in the 200–400°C range. Samples heated to 300–400°C were hard, insoluble, and very resistant to leaching. Sections of this material were kept submerged in tap water for one month without any visible evidence of reaction or particle separation. The results also show that at higher firing temperatures the pH of the water increases. This increase is presumably due to the loss of HCl and the formation of NaNO₃; the latter component decomposes at the higher temperatures forming sodium oxide. Future experiments for evaluating the procedure will be made with the use of radioactive fission products.

Table 38. Leaching of Heated Mixtures of Aluminum Nitrate Waste and Salt

Mixtures were composed of 10 ml of aluminum nitrate waste solution and 10 g of Texas rock salt (Grand Saline mine) heated for 1 hr at the temperature stated
Heated mixtures were leached in 500 ml of saturated salt solution

Sample No.	Heating Temperature of Mixture (°C)	Al ⁺⁺⁺ Concentration in Leaching Solution (ppm)			pH of Leaching Solution			
		Leach Time			Leach Time			
		6 hr	27 hr	150 hr	6 hr	27 hr	53 hr	150 hr
1	103	14.2	14.2	14.2	4.1	3.7	3.3	3.6
2	103	14.0	14.0	14.1	3.7	3.6	3.4	3.7
3	200	0.2	0.9	3.5	4.5	4.3	4.1	4.4
4	200	0.3	0.8	3.7	4.5	4.3	4.0	4.4
4A	300	0.0	0.0	0.0	5.2	5.3	5.0	4.9
4B	300	0.0	0.0	0.0	5.4	5.4	5.3	5.1
4C	350	0.0	0.0	0.0	5.6	5.7	5.8	5.6
4D	350	0.0	0.0	0.0	5.6	5.7	5.8	5.6
5	400	0.0	0.0	0.0	5.6	5.8	5.7	5.8
6	400	0.1	0.0	0.5	5.8	5.9	5.7	5.8
6A	450	0.0	0.1	0.1	6.6	7.8	7.7	7.6
6B	450	0.0	0.1	0.1	6.4	7.3	7.5	7.4
6C	500	0.0	0.2	1.5	7.3	8.5	8.6	9.1
6D	500	0.1	0.1	1.6	7.5	8.5	8.7	9.2
7	600	0.5	3.0	11.7	9.1	9.8	9.6	8.8
8	600	0.4	3.1	9.1	9.0	9.9	9.7	8.8
9	800	9.6	9.6	9.7	9.7	10.3	10.5	9.6
10	800	9.0	9.0	9.3	9.5	10.4	10.5	9.5
11	800	9.5	9.7	9.9	10.1	10.3	10.5	9.4
12	800	9.7	9.8	9.8	9.6	10.5	10.5	9.4

The areas of study covered by this report serve to indicate the complex nature of waste-salt mixtures. Gas production values obtained from gamma radiation of waste-salt mixtures are relative and serve only to indicate the volumes of gas to be expected under a particular set of conditions. Work is in progress to identify more specifically the nature of the gases and to determine the reactions responsible for their production. In addition, a series of experiments has been designed to investigate gas production and associated reactions under more realistic conditions, namely, with larger salt specimens containing significant amounts of radioactive waste solutions.

The mechanism of migration of fission products in salt formations continues to be a problem. Additional bath testing is in progress, and methods of determining acceptable salt specimens for

further tests are being developed. Other factors which may influence the physico-chemical reactions of salt and waste, such as pressure and salt impurities, are being considered.

Site Selection

Criteria in picking a desirable test site are:

1. suitability of the formation — geology well known with adequate thickness of salt, minimum depth to salt, freedom from extraneous shafts or core holes, homogeneous salt beds or domes,
2. accessibility — near paved roads, railroads, water transportation, living accommodations, skilled labor, and sufficient water,
3. distance from large population centers and from areas of active development of natural resources,

4. government ownership of the land, because this would make the property available and ease the problem of the supervision of the site,
5. suitability as a permanent disposal facility if the tests should prove successful.

There is general agreement that there should be initial tests with synthetic wastes and simulated heat sources followed by tests with radioactive wastes. In order to expedite the nonradioactive tests it would be desirable to utilize cavities presently available.

Investigation of the possibilities of radioactive waste storage in salt formations in the north-eastern United States has been performed under subcontract by the Geotechnical Corporation.⁵² The report details the salt formations and mines in the areas of the United States bounded on the north by Canada, on the east by the Atlantic Ocean, on the west by Montana, Wyoming, and Colorado, and on the south by Oklahoma, Arkansas, Tennessee, and North Carolina.

DISPOSAL OF HIGH-LEVEL WASTES IN DEEP WELLS

W. de Laguna W. J. Lacy O. M. Sealand

Cooperation with Other Agencies

The experience of the petroleum industry in brine disposal, in well drilling and operation, and in reservoir engineering has been explored by a committee appointed by the American Petroleum Institute (API). The committee visited Oak Ridge and Hanford to study the waste disposal problem. In turn, several of the ORNL staff, as well as representatives of the AEC, Hanford, and the U.S. Geological Survey, visited representative installations of the petroleum industry. The API committee is preparing its report, and no detailed discussion of their opinions is yet possible. Their general conclusions, however, may be summarized as follows:

A careful program for site selection and exploration will be required, but locations can be found which will permit long-term containment of the liquids underground. Before injection of the waste into these reservoirs, some pretreatment probably will be required, but no serious operating

difficulties are foreseen. Plugging of the injection wells can be controlled, and corrosion of the well casing may be largely an economic problem. The injection plant components, such as pumps, filters, and well-head connections, will offer some novel and initially expensive design problems, but with experience the cost can be reduced. The problem of heat generation in depth after disposal may not be serious. In general, the committee felt that it would be feasible to inject fresher and more concentrated waste with a lower pH than originally was thought possible.

A report by Birch, prepared for the National Academy of Sciences,⁵³ has defined approximate limiting concentrations of long-lived fission products that may be present at the time of injection without causing the subsequent temperature rise to exceed conservatively safe limits. The assumptions are that the waste is injected into a formation of 20% porosity and that all the pores are filled with waste for a great distance out from the injection well. Under these circumstances, if the formation is 80 ft thick, the waste may be permitted to contain roughly 4 curies of strontium or cesium per gallon, and 2 curies if the formation is 240 ft thick. As 20% porosity represents very approximately 1 gal/ft³, it is also possible to say that the disposal formation can hold about 2 to 4 curies of cesium or strontium per cubic foot without exceeding safe thermal limits, although other factors considered by Birch must not be neglected. Some dilution of the waste, therefore, may be required even after aging. The capacity of any deep-well disposal system will probably be large, so that increasing the volume by dilution will not be a problem.

A second area in which progress has been made is in the search for possible test and operating sites. The problems involve judgment and compromise, made difficult by the lack of basic information required for a logical determination. An obvious problem is the choice of depth. Although greater depth does suggest greater safety, the cost of well drilling increases with depth and limits the amount of exploration and monitoring which can be provided. A second problem is

⁵²W. B. Heroy, *Possibilities for the Storage of Radioactive Waste in Salt Cavities in the Northeastern United States*, Geotechnical Corp., Dallas, Feb. 15, 1958.

⁵³F. Birch, "Thermal Considerations in Deep Disposal of Radioactive Waste," *Report for the Committee on Waste Disposal of the Division of Earth Sciences*, NAS-NRC Pub. 588 (in press).

getting the detailed geological information needed for site selection. Accurate, detailed underground information is generally available only in areas that have been actively prospected for oil. A producing oil field is clearly undesirable. However, since the waste for a test will have to be shipped, the amounts used will be limited, and a test site can probably be located in a non-producing area in an oil-producing region.

There are two types of test sites being considered. The first would have a highly permeable formation about 5 ft thick at a depth of roughly 300 ft, and would be used to test the rate and distance of movement of various fission products in tracer quantities. The second type of test site would involve a sand lens of known limited dimensions, surrounded by impermeable shale, at a depth of about 1000 ft, and in which much larger-scale tests of heat dissipation, fission product migration, and well design would be made. These sites can probably be located in areas where the geology is well known because of exploration for oil. Preliminary investigations conducted with the Illinois State Geological Survey suggest that the Illinois Basin is not well suited, but other investigations do give more encouragement for areas in Kansas and Oklahoma.

The requirements for a site suitable for the location of a fuel reprocessing plant are different and more exacting. A determination of the relative merits of disposal into salt formations and disposal into deep wells will require actual operating experience. There would be clear advantages, both in terms of safety and in terms of economy of operation, in locating the first new fuel reprocessing plant at a place where disposal by both methods would be feasible. For example, it may develop that large-volume medium- to low-level waste can best be disposed of in wells, whereas high-level or solid waste may be best disposed of into salt. Also, if one method should develop unexpected dangers, the plant would be able to fall back on the other method. There is good reason to believe that one site, if carefully chosen, should be suitable for both methods.

The U.S. Geological Survey group at Oak Ridge is preparing a report on the general suitability of the major physiographic subdivisions of the United States for waste disposal by the various methods now under consideration. Several of the state geological surveys have been asked to do

more detailed work in their areas. This work will be expanded as the criteria for desirable test and operational sites are formulated.

Laboratory Studies of Chemical Compatibility

Encouraging progress has also been made in the laboratory studies of certain problems related to the disposal of radioactive wastes into deep wells.

Purex-type waste was selected for further study because it comprises over 90% of the total high-level waste available at present. The stored waste is neutralized to reduce corrosion of the tanks, and the waste volume is reduced by evaporation to save tank space. Therefore, it is desirable to use a neutralized and concentrated waste solution for laboratory work.

In the deep-well injection of neutralized waste, it would be necessary to know the identities and approximate concentrations of the fission products. Therefore, the behavior of fission products during neutralization was investigated by using concentrated synthetic Purex waste. Appropriate amounts of carriers of the important high-yield radioactive fission products were added to the waste solutions; they included Sr, Y, Zr, Nb, Te, Re, Cs, Ba, and Ce. Enough radioactive isotope of each important fission product was added to the solution to serve as a tracer. Samples of the solution were neutralized with concentrated NaOH solution and $\text{Ca}(\text{OH})_2$ suspension. The original solution, the supernatant liquid, and the precipitate after neutralization were analyzed radiochemically.

When nitric acid waste is neutralized with sodium hydroxide, several precipitates form and tend to scavenge many of the fission products. The concentration of iron, which is present in actual waste from the addition of ferrous sulfamate, is more than adequate to serve as a scavenger on neutralization. In addition, an appreciable concentration of uranyl ions, present from losses in solvent extraction, helps to scavenge the solution.

A gross fission product decontamination factor (D.F.) of between 2 and 10 was expected from the neutralization. Neutralization and filtration removed most of the scavengeable fission products in one step. The volume reduction achieved by repeating the procedure was limited by the volume of the precipitated iron, uranium, and fission products. The nonscavenged fission products

(mainly cesium and strontium) and detectable quantities of ruthenium remained in the liquid phase.

The sludge and supernatant liquid from neutralization by sodium hydroxide and by calcium hydroxide were radiochemically analyzed, and the results are shown in Table 39. Both hydroxides gave significant decontamination for strontium, zirconium, niobium, and rare earths; but there was very little decontamination of ruthenium and cesium. The gross beta decontamination factors were 6 for sodium and 3 for calcium. The gross gamma decontamination factors were 2.7 times higher, or 15 and 8.7, respectively. These results were within the expected range of removal. Sodium neutralization resulted in a higher D.F. than calcium on Purex wastes, mainly because the floc formed with calcium did not scavenge as much zirconium.

The various radioelements tested, the counting methods used, and the concentrations of activity before and after neutralization with sodium and with calcium are indicated in Table 39. The per cent contribution to the total activity is also shown for each of the radioelements in

the acid waste. The values for the waste neutralized by NaOH and by Ca(OH)_2 represent the concentration of activity remaining in the supernatant liquid, and the values in the adjacent columns represent the decontamination factors for that particular radioelement. The results of this study are in good agreement with values obtained at other laboratories.

Solutions containing from 50 to 280 mg per liter of the nonradioactive carrier (the range of fission product concentration in actual waste) were tested for each radioelement. In the laboratory, the solution was adjusted to pH 7; in production plants, enough caustic is added to raise the pH to about 10.5, at which point the removal of strontium and trivalent rare earths would be two to three times greater. The caustic was added very quickly in this test, and the samples for radiochemical analysis were removed a few hours after the precipitate settled. If the caustic were added more slowly and the precipitate allowed to stand longer, the cesium removal would probably have been only half as much because less cesium would be occluded in the floc. However, the test results from neutralization of the waste do give

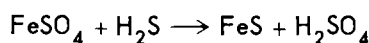
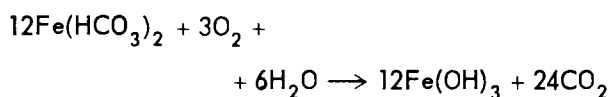
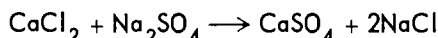
Table 39. Decontamination of Simulated Purex Waste by Neutralization

Radioelement	Radiation Counted	Simulated Waste		Neutralized by NaOH		Neutralized by Ca(OH)_2	
		Activity (counts/min/ml)	Activity (% of total)	Activity (counts/min/ml)	D.F.*	Activity (counts/min/ml)	D.F.*
Strontium	Beta	53.1×10^3	19	12.4×10^3	4	23.7×10^3	2
Zirconium	Gamma	21.5×10^3	7.7	75	2.9×10^3	8.5×10^2	25
Niobium	Gamma	99.5×10^3	35	46	2.1×10^3	15	6.6×10^3
Ruthenium	Gamma	1.1×10^3	0.5	103	10	80	13
Cesium	Gamma	27.7×10^3	10	12.3×10^3	2	17.9×10^3	1.5
Cerium	Beta	2.06×10^3	0.8	24	9	157	13
Trivalent rare earths	Beta	74.2×10^3	27	6.6×10^3	11	1.24×10^3	60
Gross	Beta				6		3
	Gamma				15		8.7
Total		280×10^3		31×10^3	9	44×10^3	6

*D.F. = decontamination factor = $\frac{\text{activity original}}{\text{activity remaining}}$.

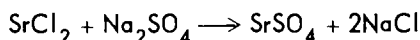
useful values for fission product scavenging by iron. The effectiveness of this scavenging diminishes with the age of the waste because the more easily scavenged fission products generally have short half lives and have already decayed from the older wastes. The more easily removed fission products with short half lives are zirconium, yttrium, niobium, and cerium. Strontium-90 (28-year, principal beta emitter) and Cs¹³⁷ (30-year, principal gamma emitter) have long half lives.

One of the problems in deep-well disposal is plugging of the well (1) by suspended solids, (2) by chemical reaction of the waste with the solid matrix of the aquifer, or (3) by chemical interaction between the waste and interstitial fluids. The following are typical reactions which might bring about the third type of plugging:



An obvious test would consist in mixing a sample of the interstitial water with the waste solution to see whether a precipitate forms. If a precipitate does form, it might be concluded that the waste solution could not be injected without plugging.

A paper by Bernard⁵⁴ reported results contrary to this general conclusion. Some of Bernard's results have been confirmed in this laboratory. A 0.1 M SrCl₂ solution tagged with radioactive Sr⁸⁹ was injected into a column containing 6450 g of clean Ottawa sand; the pores were filled with a solution of 0.1 M Na₂SO₄. The reaction which would occur if these solutions were mixed is



A small quantity (2 mg) of indigo carmine dye was added to the injected solution to serve as a visible tracer of the flow pattern. As the strontium chloride solution displaced the sodium sulfate solution, there was no loss in permeability

and no plugging of the column because there was only a small zone of contact between the two solutions. The data shown in Fig. 51 indicate that during injection there is a displacement of the interstitial solution, and, because the sand acts to prevent the two liquids from mixing, precipitation and eventual plugging of the column are prevented. This indicates that there is little mixing within the sand bed and very little precipitation. These salt concentrations are high compared with the concentration of the interstitial solutions found in most formations.

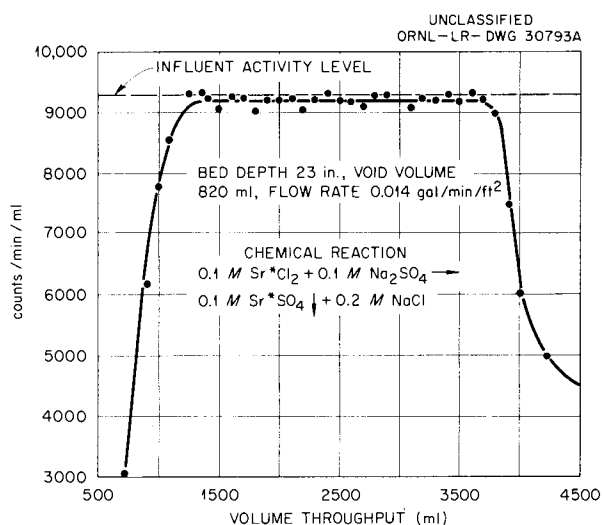


Fig. 51. Interstitial Solution Compatibility Study with Ottawa Sand and Sr⁸⁹.

After 3500 ml of solution had passed through the 23-in. sand bed, the sand was rinsed with distilled water. The column effluent showed a rapid decrease in counting rate, the counterpart of the rapid increase exhibited by the initial breakthrough.

From the foregoing it can be concluded that there is little danger of plugging the formation by injecting a solution that normally forms a precipitate with the interstitial solution. Instead of mixing, there is a pistonlike displacement of the interstitial liquid by the injected waste and only a small zone of precipitation along the contact edge, which forms a barrier to prevent further mixing.

⁵⁴G. G. Bernard, *Illinois State Geol. Survey, Bull.* 80, 98-115 (1956).

Birch⁵⁵ has shown that a formation can contain from 2 to 4 curies of Sr^{90} or Cs^{137} per cubic foot without resulting in a temperature rise exceeding conservative limits. Depending on the porosity of the formation, the waste solution can contain from 2 to 4 curies per gallon of these fission products at the time of injection. However, if cesium or strontium is appreciably sorbed by the rock in or near the well bore, local overheating might result. Because cesium is poorly scavenged during neutralization of the waste, it will probably be the principal source of heat.

The sorption of radiocesium from neutralized Purex waste onto quartz sand and mixtures of quartz, feldspar, mica, and several types of clay was investigated by means of column experiments. The initial tests were made with clean Ottawa sand, either water-rinsed or acid-leached, packed in columns of various sizes. Neutralized Purex waste, 300 to 4000 ml, containing $\sim 10,000$ counts/min/ml of Cs^{137} plus 100 mg/liter of cesium carrier was passed through the sand slowly. Virtually no sorption of radiocesium by either the normal or the acid-leached Ottawa sand was found. Visual observations indicated no change in permeability during the run, and no formation of precipitate in the sand; no coating of the sand grains could be detected under 70X magnification. The untreated sand contained less than 0.1% clay as coating on the grains. The sorption of radioactive material was so slight that no measureable difference in activity could be found between the influent and effluent solutions.

Two feldspars were similarly tested: orthoclase and an oligoclase containing 85% albite and 15% anorthite. Feldspar was added to the quartz sand in known proportions and the above procedure was repeated. During operation there was no visible decrease in flow rate and no precipitate formed within the column. Microscopy indicated no coating of either the quartz or feldspar grains. As in all other tests, a search was made for side effects: precipitation, grain coating, and gas evolution. The results indicated that about $(4.6 \pm 2.0)\%$ of the Cs^{137} was adsorbed from 100 ml of waste solution by a column containing 100 g of

feldspar and 100 g of quartz. As more solution was passed through, sorption decreased; after 200 ml had passed through the column, the column effluent contained approximately the same concentration as the influent solution. There was a slight increase in the pH of the waste solution (from 7.0 to 7.25) after contact with 100 g of orthoclase.

Concentrated Purex waste solution contains about 100 mg/liter of Cs^{137} . The specific activity of Cs^{137} is about 78 curies/g. Therefore, 100 mg of cesium per liter would be equivalent to 7.8 curies/liter or about 29 curies per gallon of concentrated waste. Unconcentrated Purex would contain about $\frac{1}{10}$ this amount of cesium or roughly 3 curies/gal. If the latter waste is injected into a formation with a porosity of 20%, the interstitial concentration of cesium would be 4.49 curies/ft³. The laboratory results indicate that a 1-to-1 sandstone-feldspar mixture may adsorb up to 13.5 mg of cesium per cubic foot. The concentration of the adsorbed cesium would be 13.5 mg/ft³ or about 1.05 curies. This is less than $\frac{1}{4}$ the concentration of activity in the interstitial space. The feldspar showed a concentration factor of cesium to sodium of about 29:1; that is, the ratio of cesium to sodium ions on the feldspar was higher by a factor of 29 than the ratio in the waste.

Vermiculite-quartz mixtures, tested under the same conditions, showed very little concentration of the radiocesium by the vermiculite. Although vermiculite has an exchange capacity of 100 meq per 100 g, only 3% of the cesium was removed from the first 2 or 3 liters of waste solution, and there was virtually no sorption of cesium from the last liter. The 100 g of vermiculite sorbed 9 mg of cesium, so that 10 kg would sorb about 0.75 curie of cesium. The concentration factor for cesium over sodium was about 10:1, assuming purity of the vermiculite. Even though the vermiculite has 100 times greater base exchange capacity than feldspar, the exchange is a calcium replacement system, and calcium has a large hydrated ionic radius similar to sodium; therefore, although cesium normally has preference to sodium in ion exchange, when the ion being replaced is calcium, the preference for cesium over sodium is very slight. The general formula for vermiculite is



with x being 1-1.4, and γ being about 8. The

⁵⁵F. Birch, "Thermal Considerations in Deep Disposal of Radioactive Waste," *Report for the Committee on Waste Disposal of the Division of Earth Sciences*, NAS-NRC Pub. 588 (in press).

Mg^{++} and Ca^{++} are the balancing and largely exchangeable cations.

The sorption of radionuclides, particularly Cs^{137} , by small amounts of clay was investigated. The clay minerals of greatest importance included illite, kaolinite, halloysite, and montmorillonite. The procedure was to add from 0.3 to 20 g of clay to 300 ml of the Purex waste solution and then to stir for 2 hr at 216 rpm. After stirring, the solution was filtered through filter paper, and samples were taken for counting. The counting was performed on a well-type gamma scintillation counter using an NaI(Tl) crystal. Three samples were prepared for counting by placing 1-ml aliquots in 10×75 -mm glass culture tubes. It was possible to determine the number of milligrams of cesium adsorbed per 100 g of clay, the removal of Cs^{137} tracer from solution, and the ratio of radiocesium to stable cesium.

Figure 52 is a graph showing the sorption of radiocesium from a sodium hydroxide-neutralized 6 M Purex waste solution by the clay minerals listed above as well as by shale and bentonite. The Conasauga shale contains a mixture of clay minerals and quartz. Its ability to sorb cesium from the waste solution is large, considering the relatively large content of inert quartz. The bentonite sample is a clay of undetermined composition, from Texas.

Studies were made of the sorption by illite of radiocesium from solutions having varying sodium nitrate concentrations. Sodium nitrate is the salt resulting from neutralization of Purex waste (which contains HNO_3) by sodium hydroxide. Concentrations of sodium nitrate ranged from 6 M to 0.5 M;

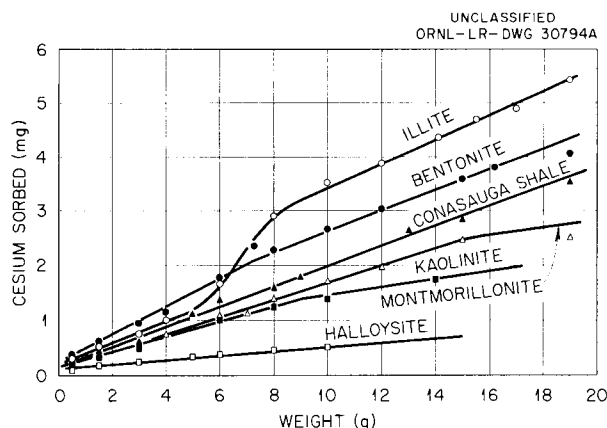


Fig. 52. Efficiency of Various Materials for Removal of Cs from Neutralized Purex Waste.

this range of sodium includes neutralized, concentrated Purex waste, which is about 6 M, and unconcentrated waste, which is 1 M. The results (Fig. 53) show a threefold increase in cesium sorption with a tenfold decrease in sodium ion concentration. Next, the effect of cesium concentration on cesium sorption from 6 M sodium nitrate solution was determined. These concentrations ranged from 10 mg/liter of cesium to 100 mg/liter of cesium. The 10 mg/liter cesium concentration is representative of a diluted 1 N unconcentrated Purex waste, while the 100 mg/liter is slightly more concentrated than the 7.0 M evaporated waste. The results indicate approximately a threefold increase in cesium adsorption with a tenfold increase in cesium concentration, as shown in Fig. 54. The relative effect on the

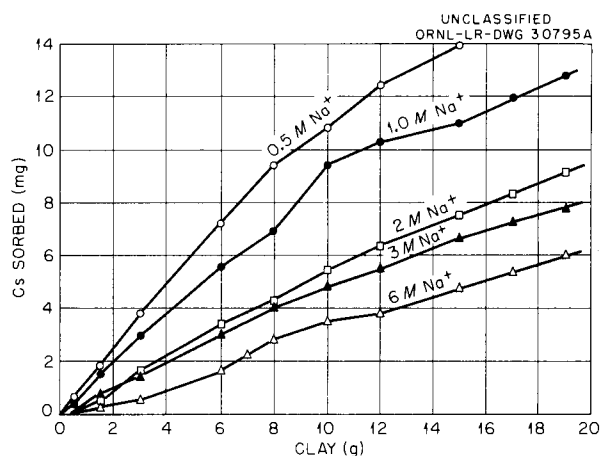


Fig. 53. The Effect of Sodium Ion Concentration on the Sorption of Cesium by Illite Clay. The concentration of cesium was 100 mg/liter in all tests.

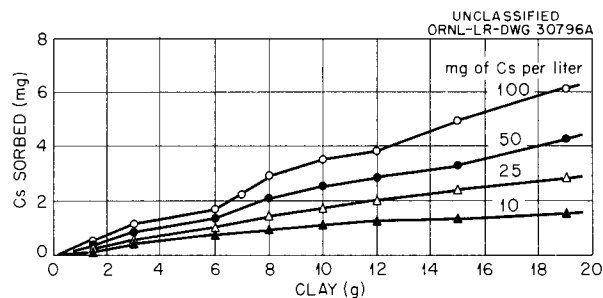


Fig. 54. Effect of Cesium Concentration on the Sorption of Cesium by Illite Clay from a 6 M $NaNO_3$ Solution.

amount of cesium sorbed per gram of clay by increasing the concentration of cesium carrier tenfold or by decreasing the concentration of sodium ion by the same factor was the same. In Fig. 53, the values on the 1 M Na⁺-illite curve for 10- and 12-g quantities of clay are higher than normal, perhaps due to experimental error.

If less concentrated acid waste were neutralized for disposal or if 6 M acid waste were neutralized and diluted with brine, the sorption of cesium would decrease as the ratio of sodium to cesium in solution increased. There was no indication that the concentrations of Cs¹³⁷ would exceed the equilibrium value near the well bore. It should also be remembered that liquid wastes would be diluted, making the concentrations of cesium lower than those investigated here.

The laboratory work done so far suggests that plugging of the disposal formation by precipitates, resulting from chemical interaction between the waste and the brines already in the formation, is not likely to be a problem. This conclusion is in agreement with the experience of the petroleum industry. The possibility of plugging due to chemical reaction between the waste and the solid matrix of the formation is another problem. The results of the sorption studies suggest strongly that undue localized heat production, due to sorption of radioactive materials near the well, can be controlled so that it will not be a serious problem.

RELATED COOPERATIVE PROJECTS

R. M. Richardson ⁵⁶	E. G. Struxness
R. W. Maclay ⁵⁶	W. J. Boegly
W. M. McMaster ⁵⁶	M. A. Cobble
W. de Laguna	L. C. Emerson
R. J. Morton	S. I. Auerbach

A Survey of the Physical Divisions of the United States with Reference to the Land Disposal of Radioactive Wastes

As an aid in selecting sites for the disposal of radioactive wastes on land, the U.S. Geological Survey is preparing a report on the physical divisions of the United States. The report describes briefly the sources, characteristics, and rates of production of radioactive wastes. Possible

methods of waste disposal on land and the problems of underground disposal are considered. The geologic and hydrologic factors affecting the selection of waste disposal sites are discussed. Each of the 25 major physical divisions of the United States is described in some detail, and potentially favorable or unfavorable areas for waste disposal are delineated. The favorable sites include areas underlain by salt deposits at reasonable depth, structural basins containing porous, brine-saturated formations, and areas underlain by thick beds of shale or other impermeable rock.

Geologic Map of the Oak Ridge Reservation

A general-purpose geologic map of the Oak Ridge reservation, which comprises about 59,000 acres, is being prepared by the U.S. Geological Survey. The map, which is on a scale of 2000 ft to the inch, shows the distribution and attitude of the rock formations that underlie the area. A brief accompanying text describes the geology of the reservation. The map and report provide information necessary for the selection of sites for waste disposal and other facilities.

Steering Committee for Joint Congressional Committee Hearings

One member of the Section served as a member of a Steering Committee organized by the Executive Director of the Joint Congressional Committee. The purpose of the Steering Committee was to aid in organizing and preparing testimony for public hearings on problems of industrial radioactive waste disposal. All AEC installations and several government agencies and private industrial groups were invited to participate in the hearings, which will be held in Washington beginning on or about January 15, 1959.

Geneva Exhibits

The Waste Disposal Research and Engineering Section was requested to prepare the design and supervise construction of one panel of the fuel cycle exhibit for the 1958 International Conference on the Peaceful Uses of Atomic Energy. The purpose of this panel is to illustrate waste treatment and disposal practices. As completed, the panel consists of a geologic model showing both present and future waste disposal practices. Located in the center of this model is a cutaway

⁵⁶U.S. Geological Survey.

section of a waste storage tank showing the method used to recirculate sludge and prevent "bumping" of the tank contents. The back face of the panel is a comprehensive flowsheet showing the relationships between various methods of treatment and disposal.

Assistance was also supplied in the preparation of a short movie illustrating fuel reprocessing operations in the United States.

Geneva Review Committee No. 24

One member of the Section served as a member of Geneva Review Committee No. 24 of the Office of International Conference (OIC), Second International Conference on Peaceful Uses of Atomic Energy. This committee reviewed abstracts and papers for the sessions on meteorology, oceanography, and waste disposal. Its responsibility was concluded with recommendations to the OIC on publication of papers in the Proceedings and on oral presentation at the Conference.

Maritime Reactor Waste Disposal

A preliminary study was made of literature pertaining to the use of the oceans as a disposal site for certain radioactive wastes resulting from operation of nuclear powered merchant ships. Early in this study it was concluded that, in spite of the low levels of waste involved, the lack of knowledge in the broad fields of oceanography and marine ecology effectively prevented the early establishment of standards acceptable at the international level. Accordingly, it was proposed to the Maritime Reactors Branch, Atomic Energy Commission, that competent assistance in these areas be brought to bear on this problem. Subsequent to this proposal and at the request of the Maritime Reactors Branch, a committee of

experts working under the auspices of the National Academy of Sciences has undertaken the responsibility of establishing appropriate standards.

American Standards Association, Sectional Committee N5

Two of the waste research staff are members of ASA Sectional Committee N5, "Chemical Engineering for the Nuclear Field," which was organized in June 1957 under the program of the Nuclear Standards Board. They have participated in meetings and the work program of Subcommittee N5.2 on Radioactive Waste Disposal. They are serving as leaders of two of the seven working groups under this subcommittee: (1) Classification of Wastes (gaseous, liquid, and solid); and (2) Permissible Waste Effluent Standards. Information and experience from ORNL studies on radioactive wastes are utilized in the ASA program to develop uniformity in waste disposal practice.

AEC Ecological Review Committee

One member of the Section served on a special ecological advisory committee, which was organized by the AEC Division of Biology and Medicine, Washington, D.C., in cooperation with the American Institute of Biological Sciences for the purpose of reviewing certain aspects of weapons fallout research at the test site in Nevada. The committee met for three days at the Nuclear Test Site, Mercury, Nevada. Here they reviewed the local ecological programs on weapons and fallout effects. Certain hitherto unused areas within the test reservation were inspected for their applicability as sites for large-scale experimental studies on the ecological effects of radioactive fallout. The report of this committee is in preparation.

RADIATION DOSIMETRY

G. S. Hurst

R. H. Ritchie

R. D. Birkhoff

DOSIMETRY APPLICATIONS

J. A. Auxier J. S. Cheka
E. T. Arakawa W. W. Ogg
F. W. Sanders

Ichiban Project

Operation Plumbbob Experiments. – The major accomplishment of the Ichiban project was a successful series of experiments at the Nevada Test Site (NTS) during Operation Plumbbob in the summer of 1957. This included angular distribution measurements for gamma rays and fast neutrons, attenuation factors for two light-frame structures, air dose determinations, and related measurements.

For gamma radiation, the physical quantity that was measured was the first-collision tissue dose; for neutrons, both the energy distribution and the first-collision tissue dose were obtained. The

anhydrous chemical dosimeters developed by Sigoloff¹ were used for gamma radiation measurements, and the threshold detector system developed at ORNL was used for neutron measurements.²⁻⁴

The angular distribution for gamma rays was peaked in the direction defined by a line connecting the point of detonation and the point of measurement, as shown in Fig. 55. This forward peak was also present but less pronounced in the case of fast neutrons (Fig. 56). The fast-neutron angular

¹S. C. Sigoloff, *Nucleonics* 14(10), 54–56 (1956).

²G. S. Hurst *et al.*, *Rev. Sci. Instr.* 27, 153–56 (1956).

³F. J. Davis *et al.*, *H-P Semiann. Prog. Rep.* July 31, 1956, ORNL-2151, p 93.

⁴P. W. Reinhardt and F. J. Davis, "Improvements in the Threshold Detector Method of Fast Neutron Dosimetry," *Health Phys.* (in press).

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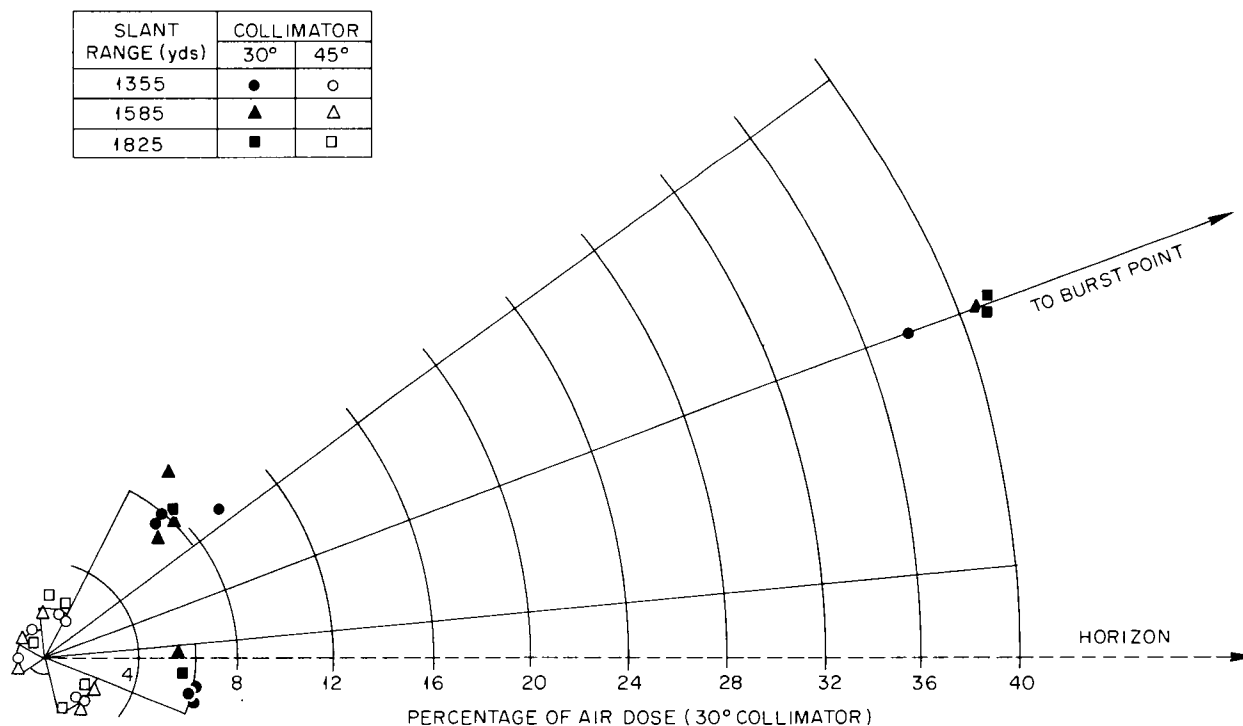


Fig. 55. Gamma Angular Distribution.

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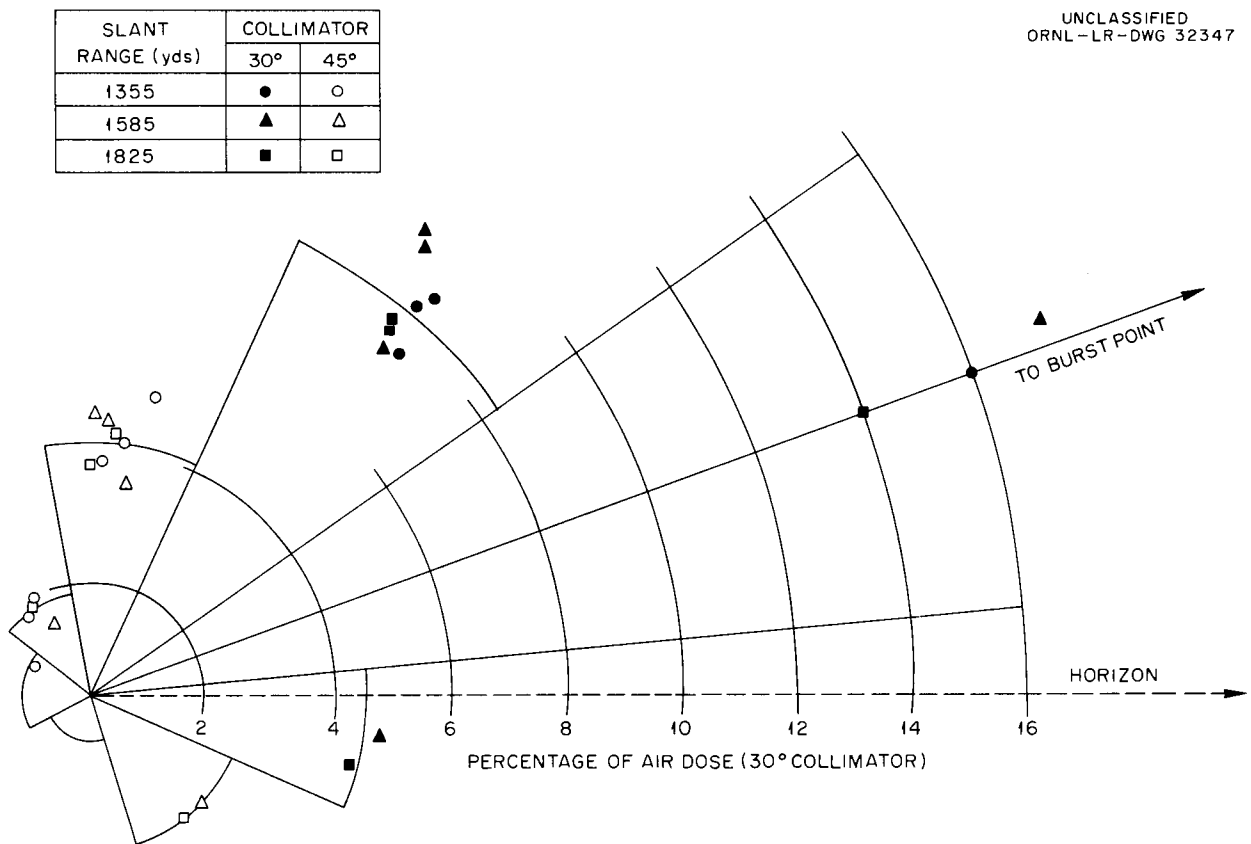


Fig. 56. Neutron Angular Distribution.

distribution can be represented by a spherical distribution and a peak in the forward direction. For both fast neutrons and gamma rays the angular distribution is rather insensitive to type of weapon and to distance from ground zero.

Dose measurements were made of the gamma-ray and fast-neutron dose inside two identical light frame houses at the same distance from the same detonation. Both the neutron- and gamma-ray dose in the house varied by a factor of 2, depending on the exact location of a point within the house (see Figs. 57 and 58). The dose within the house correlated rather well with a single simple factor — the straight line distance as measured from the point of entry of the ray into the house to the point of measurement on the ray path from the burst point. In most cases the spread of individual points from the curve drawn to represent this correlation was less than $\pm 10\%$.

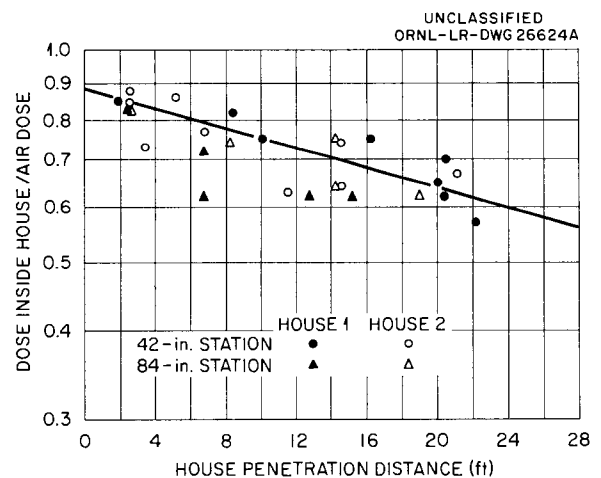


Fig. 57. Attenuation of Gamma Radiation by Typical Single-Story Japanese Houses.

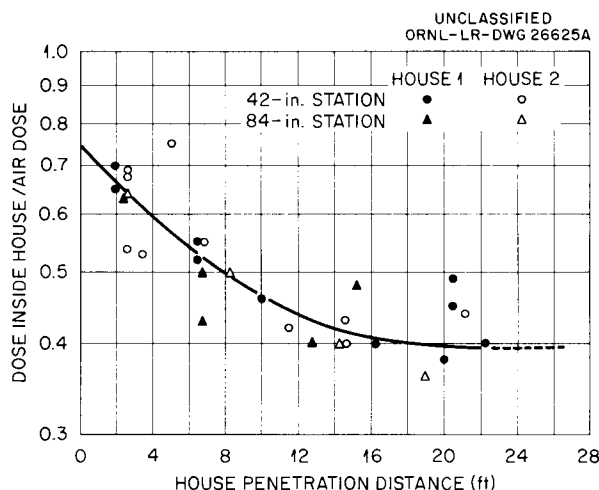


Fig. 58. Attenuation of Fast Neutrons by Typical Single-Story Japanese Houses.

The greatest uncertainty in determining the doses received by survivors of the Hiroshima and Nagasaki bombings is the evaluation of the air dose. The air doses (from neutrons and gamma rays) for the Hiroshima and Nagasaki weapons have been calculated by York⁵ and are presented in Figs. 59 and 60. The dotted curves show the range of estimated probable errors.

Liaison with ABCC. – Liaison with the Atomic Bomb Casualty Commission has continued successfully with one ORNL representative on a two-year assignment in Japan. With the guidance this representative provides, the shielding histories taken for survivors have been improved, and the rate at which the histories are accumulated has increased. The accumulation and study of records concerning bomb-burst data, physical measurements of houses, aerial photographic interpretation, and other shielding considerations are continuing.

Civil Effects Exercise 58.1

Experimental Evaluation of the Radiation Protection Afforded by Residential Structures from Distributed Sources. – There has been a need for experimental data which can be used to evaluate the protection afforded by residences against radiation due to fallout. The H-P Division has cooperated with personnel from the Atomic Energy

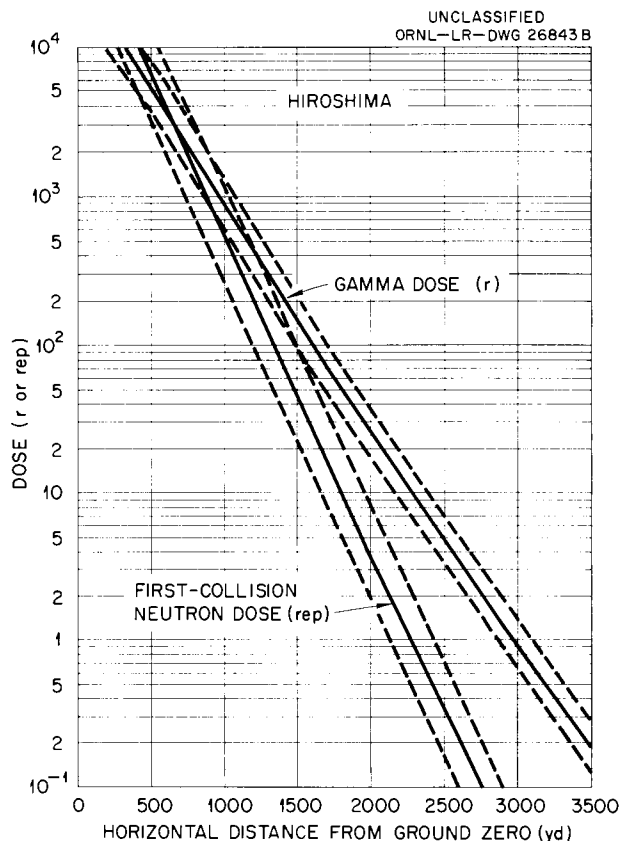


Fig. 59. Dose vs Horizontal Distance from Ground Zero.

Commission's Civil Effects Test Group, Federal Civil Defense Administration, the National Bureau of Standards, Project Civil of the University of California, and Tracerlab, Inc., in an experiment at the Nevada Test Site to obtain some of the needed information. Fallout was simulated by arrays of Co^{60} (400 sources of 4.15 mc each) and Cs^{137} (20 sources of 120 mc each). In addition, one large Co^{60} source (2.1 curies) and one large Cs^{137} source (1.8 curies) were placed inside some houses for measurements of point sources for evaluation of reciprocity.

The final conclusions that may be drawn from the experiment must, in general, await a detailed analysis of the data. However, certain early conclusions may be reached after a cursory examination of the results and from preliminary analyses. Some of these conclusions follow:

1. As expected, the best shielded areas of a house are in the basement; typically, they are better by a factor of 10 than other regions of a house.

⁵Communicated by M. Morgan, *Dose Calculations for Hiroshima and Nagasaki*, ORNL CF-57-11-144 (Nov. 29, 1957).

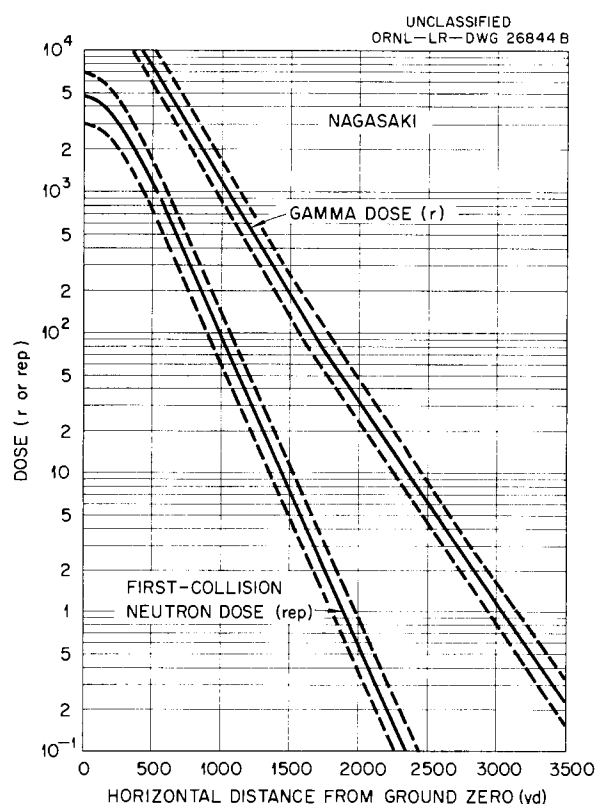


Fig. 60. Dose vs Horizontal Distance from Ground Zero.

2. The dose rate on the first and second floors of a light frame structure, due to uniform ground and roof contamination, is largely from the ground sources (75 to 90%).

3. Radiation shadows cast by brick chimneys, floor joists, floors, and other structural components of a house reduce the dose rate significantly, on the average to one half.

4. Measurements when a point source is within the house and the detector is on the roof indicate that the total attenuation may vary by a factor as great as 4 when the detector is moved distances less than 1 ft. This large variation for negligible changes in source-detector distances is caused by inhomogeneities in floor, joists, and roof members.

5. The walls of a two-story brick house provide a factor of 4 greater shielding from Co^{60} on the ground than the two-story frame house, which provides a factor of about 2 in protection.

6. Dose rates in a wood rambler, from roof sources, are about the same as on the second floor of a two-story frame house. Dose rates from

ground sources are somewhat lowered (10–20%) by plastered walls, as compared with $\frac{1}{4}$ -in. plywood.

7. Streaming of radiation through basement windows was reduced, on the average, by 25% when sand bags were placed in the window wells and over the window. The reduction in various parts of the hut ranged from 5 to 400%.

A presentation of data and a description of the experiment are in press as a CEX (Civil Effects Exercise) report. A comprehensive analysis of the data is being made.

Energy and Angular Response of Pocket Ionization Chambers

Since pocket ionization chambers are known to be energy dependent in response,⁶ tests have been made on modifications that should minimize such dependence. The modifications consisted of tin sleeves of various thicknesses covering all or part of the chambers. Figure 61 summarizes the results of these tests. It appears that a 0.040-in.-thick coat of tin, covering 3.5 cm of the charging end of the chamber, gave the flattest response over the largest energy range. The deviation was minimal down to 50 kev (eff.), with the chamber reading low below this energy.

While the above-mentioned tests were in progress, the effect on the response of the chambers to the orientation of the chambers was also measured. Figure 62 shows the angular dependence of response for a bare and a sleeved chamber to two energies: 250- and 100-kev x rays of 196.5- and 69.0-kev effective energies, respectively.

PHYSICS OF TISSUE DAMAGE

H. P. Yockey J. S. Mendell⁷
D. R. Nelson

The papers presented at the Symposium on Information Theory in Health Physics and Radiobiology were edited and will be published by Pergamon Press in the fall of 1958 under the title *Symposium on Information Theory in Biology*.

An analysis was made of the "Greenhouse" data on the radiation aging of mice from the point of view of the theory of aging given in the above-mentioned volume. It was shown that life shortening applies to all members of the exposed group of

⁶F. H. Day, *Ra-Det* 3(4), 1 (1950).

⁷AEC Fellow.

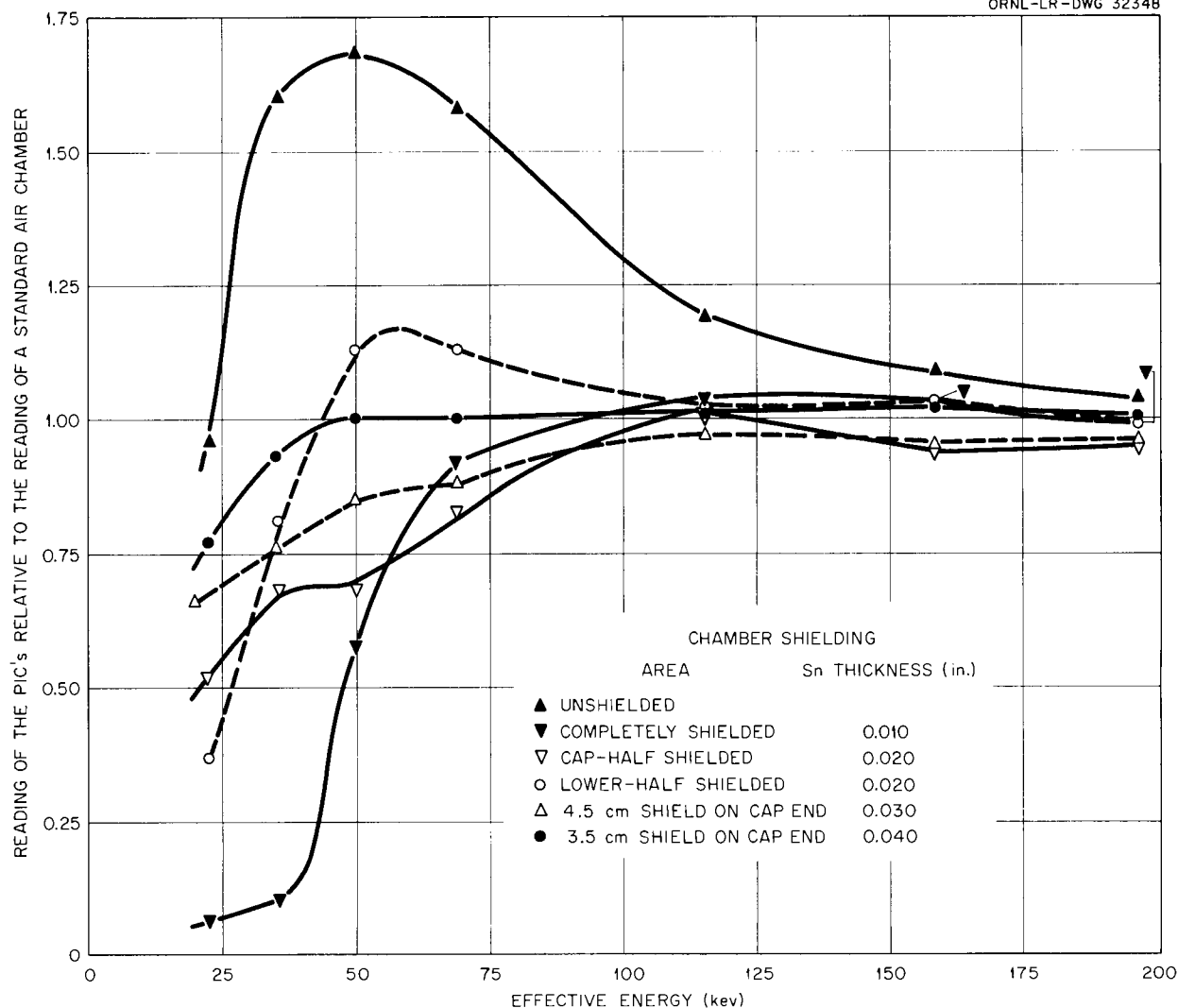
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Fig. 61. Energy Dependence of Pocket Ionization Chambers (PIC's).

animals uniformly, regardless of time of death. More explicitly, this means that although *some* irradiated mice live longer than *some* controls, all irradiated mice were damaged and their lives were shortened.

The program with Clevite Research Center in connection with radiation damage to water-soluble piezoelectric crystals is making progress. The problems surrounding the proper mounting of the crystals so that the low-temperature irradiations

can be made have been solved, and the irradiations are proceeding.

Several explosions have occurred in connection with liquid-nitrogen cryostats in the reactor. They may be due to the accumulation of oxygen in the cryostat because of fractionation of the liquid nitrogen. Our apparatus eliminates this difficulty by using a closed loop of circulating liquid nitrogen as a heat transfer fluid. This liquid is cooled in boiling liquid nitrogen by a heat exchanger which is outside the reactor.

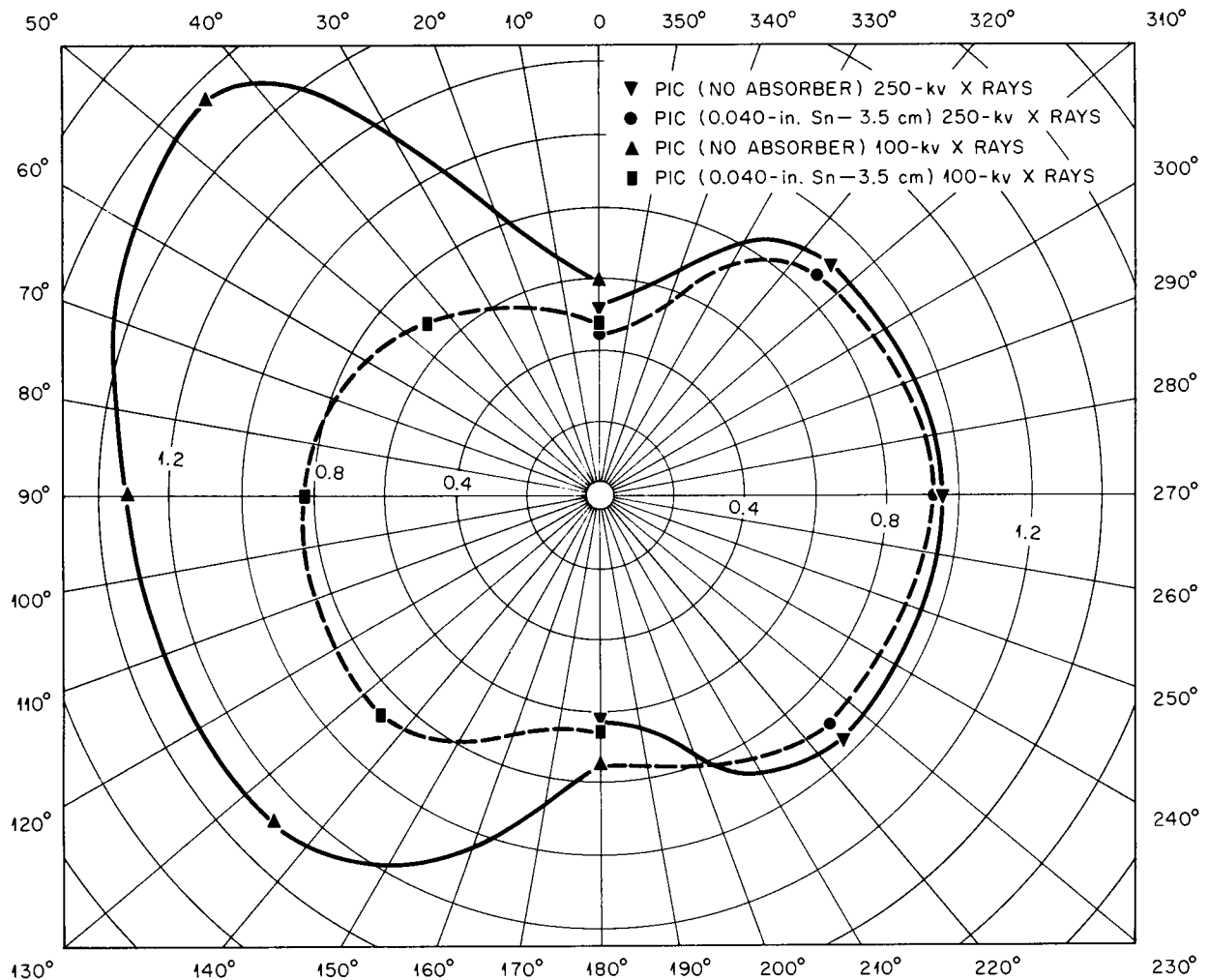
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Fig. 62. Angular Response of Pocket Ionization Chambers (PIC's).

THEORETICAL PHYSICS OF DOSIMETRY

J. Neufeld R. H. Ritchie
W. S. Snyder

Neutron-Dose Studies in Small Phantoms

The maximum dose absorbed by a convex phantom from a beam of neutrons must be at least as large as the first-collision dose at the irradiated surface of the phantom and cannot exceed the maximum dose in a slab which contains the phantom.⁸ Thus

$$1 \leq \frac{\text{Maximum dose in phantom}}{\text{First-collision dose}} \leq \frac{\text{Maximum dose in slab}}{\text{First-collision dose}}$$

Here the first-collision dose represents the energy absorbed in a small volume of tissue which effectively absorbs the recoil atoms and protons produced but does not absorb the gamma rays appreciably.

Small-sample Monte Carlo estimates of the fraction on the right in the above equation have been calculated for tissue slabs with thicknesses of 5–1 cm and for neutron energies from 10 Mev to thermal. These preliminary estimates indicate that the fraction does not exceed 1.5 for neutron energies in the range 0.1–10 Mev (see Fig. 63). For lower energies it is much higher, especially for the thinner slabs, because the energy of the photons

⁸W. S. Snyder, *Health Phys.* 1(1), 51 (1958).

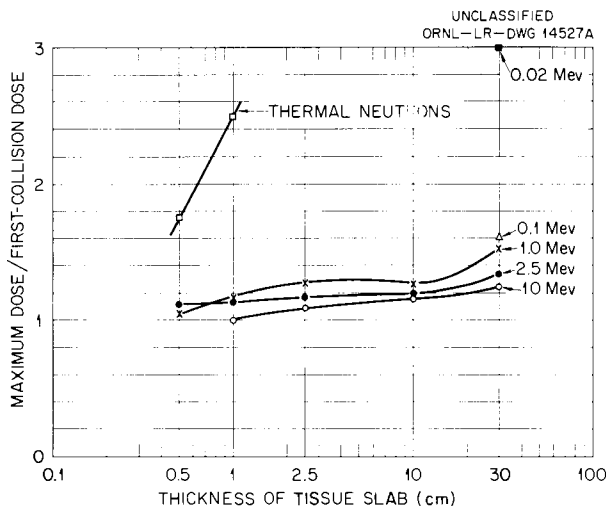


Fig. 63. Ratio of Maximum Dose and First-Collision Dose.

produced by the (n, γ) reaction with hydrogen is unrepresented in the first-collision dose but contributes appreciably to the dose in the slab (see Fig. 64). Further studies of this ratio for small phantoms of various shapes are planned for the coming year.

Interaction Cross Section of Charged Particles in a Degenerate Fermi-Dirac Electron Gas

To treat the complex many-body problem of the interaction of electrons in a degenerate Fermi-Dirac electron gas, Lindhard⁹ and Hubbard¹⁰ have independently developed a quantum theory of the dielectric constant of an assembly of free electrons. The electric field in the medium is assumed to be classically prescribed, although the electronic motion in the plasma is treated by quantum perturbation theory.

A generalization of the work of the above-mentioned authors has been found in which the concept of a prescribed field is unnecessary and the interaction of charged particles with the electron gas is made consistent in first-order perturbation theory.¹¹ This generalization allows a treatment, in a very simple manner, of such

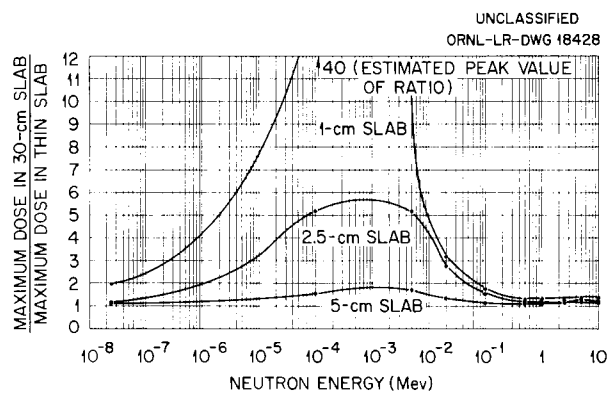


Fig. 64. Small-Sample Estimate of Ratio of Maximum Dose as a Function of Slab Thickness and Neutron Energy.

problems as the interaction of fast electrons with plasma and the correlation energy of the free electron gas.¹²

A field-quantization method has been applied to plasma problems by Bohm and Pines^{13,14} and others. However, inherent in this approach is a wave-vector cutoff procedure, which divides the region of collective interaction of plasma electrons from the region of individual interactions. Separate treatments are necessary in the two regions. In the present theory all interactions are treated on the same basis, and the cutoff wave-vector assumption is unnecessary.

Following Lindhard,⁹ it can be assumed that the wave function, ψ_i , of the i th plasma electron obeys a Hartree one-particle equation in which the self-consistent potential, $\phi(\vec{r}, t)$, causes transitions between plane wave-states. Solve for the transition amplitude by first-order perturbation theory and substitute into

$$(\epsilon_{\vec{k}, \omega} - 1) k^2 \phi_{\vec{k}, \omega} = 4\pi e \sum_i |\psi_i|^2_{\vec{k}, \omega},$$

where the subscript $\vec{k}, \omega$ indicates the $\vec{k}, \omega$ Fourier component, and $\epsilon_{\vec{k}, \omega}$ is the generalized dielectric constant. The right-hand side is proportional to $\phi_{\vec{k}, \omega}$, and thus $\epsilon_{\vec{k}, \omega}$ may be solved for explicitly.

⁹J. Lindhard, *Kgl. Danske Videnskab. Selskab Mat. fys. Medd.* **28**, No. 8 (1954).

¹⁰J. Hubbard, *Proc. Phys. Soc. (London)* **A68**, 978 (1955).

¹¹R. H. Ritchie, *Phys. Rev.* **106**, 874, esp 881, "Note added in proof" (1957).

¹²A formulation similar to this has been given independently by J. J. Quinn and R. A. Ferrell, *Bull. Am. Phys. Soc.* **3**, 202 (1958).

¹³D. Bohm and D. Pines, *Phys. Rev.* **92**, 609 (1953).

¹⁴D. Pines, *Phys. Rev.* **92**, 626 (1953).

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The generalized dielectric constant $\epsilon_{\vec{k},\omega}$ is an operator in configuration space, since it describes the polarization of the medium at arbitrary points in space-time due to perturbations in the medium. Thus by analogy with field theory, a "plasma propagator" function may be defined as

$$V(\vec{r}_2, t_2; \vec{r}_1, t_1) = \frac{4\pi}{L^3 T} \sum_{\vec{k}, \omega} k^{-2} \epsilon_{\vec{k}, \omega}^{-1} \exp [i\vec{k} \cdot (\vec{r}_2 - \vec{r}_1) - i\omega(t_2 - t_1)] ,$$

which gives the potential at $\vec{r}_2, t_2$ due to a unit charge appearing at $\vec{r}_1, t_1$. If $\epsilon \rightarrow 1$, V reduces to the instantaneous Coulomb potential, and $V(2,1)$ may then be regarded as completely specifying nonrelativistic electron interactions in the dielectric medium to the order e^2 .

An alternative procedure for obtaining the plasma propagation function and the dielectric constant, which is more rigorous than the method of Lindhard and which may be extended more easily to a higher order of perturbation, is based on Hubbard's¹⁵ S-matrix formulation of interactions in the Fermi electron gas. By the use of rules for writing matrix elements from Feynman graphs, which are quite similar to the corresponding rules in quantum electrodynamics,¹⁶ sum over primitive graphs representing processes in which an electron-hole pair interacts with an incident electron after the excitation and subsequent de-excitation of zero, one, two, etc., pairs previously. The excitation in turn is passed on from one pair to another. The dielectric constant obtained is identical with the Lindhard result, except that it is symmetric in ω , while Lindhard's is antisymmetric in ω . The self-energy of an incident electron may be calculated from the Feynman graph¹⁷ of Fig. 65, where $K_i(2,1)$ is the propagation function of a free electron. It can be found for the transition rate, Γ , from an initial state with momentum $\hbar\vec{k}_0$, that

$$\Gamma = - \frac{8\pi e^2}{\hbar L^3} \sum_{\vec{k}_f} \frac{1}{k^2} \text{Im} \left(\frac{1}{\epsilon_{\vec{k}, \omega}} \right) \Bigg|_{\substack{\omega = \omega_0 - \omega_f \\ \vec{k} = \vec{k}_0 - \vec{k}_f}} ,$$

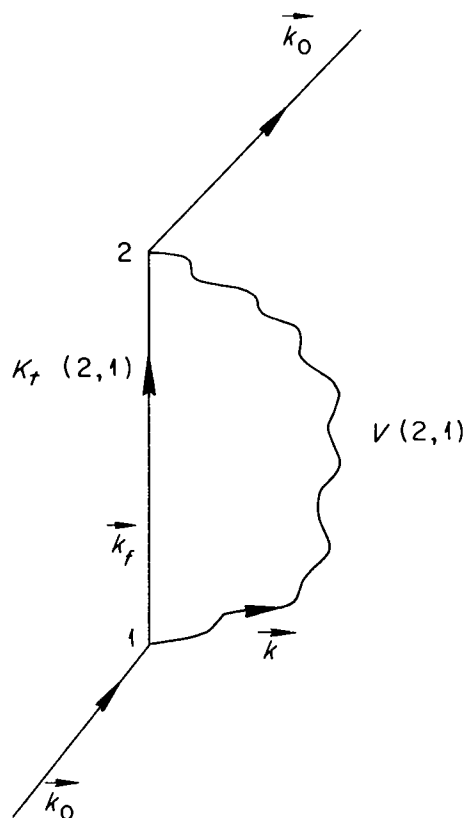


Fig. 65. Feynman Diagram for Calculating the Self Energy of an Electron in a Plasma.

where the sum runs over final states

$$\vec{k}_f, \omega_{0f} = \frac{\hbar}{2m} k_{0,f}^2 ,$$

and L^3 is the normalization volume.

From this, the interaction probability per unit path length that an incident electron of velocity $\vec{v}_0$ shall lose energy $\hbar\omega$ and be scattered into the

¹⁵J. Hubbard, *Proc. Roy. Soc. (London)* **A240**, 539 (1957); **A243**, 336 (1958).

¹⁶S. S. Schweber, H. A. Bethe, and F. De Hoffmann, *Mesons and Fields*, vol I, Row, Peterson, New York, 1955.

¹⁷R. P. Feynman, *Phys. Rev.* **76**, 749, 769 (1949).

element of solid angle $d\Omega$ about θ (see Fig. 66) is given by

$$\tau(\theta, \omega_f) d\Omega d\omega_f = \frac{me^2}{\hbar^3 \pi^2 v_0} \frac{k_f}{k_0} \frac{x^2}{4k_f^2} \left\{ \frac{f_2(x_1 z)}{[z^2 + x^2 f_1(x_1 z)]^2 + x^4 f_2} \right\},$$

where $\bar{k} = \bar{k}_0 - \bar{k}_f$ and

$$\omega = \frac{\hbar}{2m} (k_0^2 - k_f^2),$$

and

$$f_1 = \frac{1}{2} + \frac{1}{8z} \left[1 - \left(z - \frac{x}{4z} \right)^2 \right] \ln \left| \frac{z - \frac{x}{4z} + 1}{z - \frac{x}{4z} - 1} \right| + \frac{1}{8z} \left[1 - \left(z + \frac{x}{4z} \right)^2 \right] \ln \left| \frac{z + \frac{x}{4z} + 1}{z + \frac{x}{4z} - 1} \right|,$$

$$f_2 = \begin{cases} \frac{\pi}{8} \frac{x}{z}, & \text{region I,} \\ \frac{\pi}{8z} \left[1 - \left(z - \frac{x}{4z} \right)^2 \right], & \text{region II,} \\ \sigma, & \text{region III,} \end{cases}$$

where regions I, II, and III are shown in Fig. 67, and

$$x^2 = \frac{e^2}{\pi \hbar v_f}, \quad z = \frac{k}{2k_f}, \quad x = \frac{\hbar \omega}{E_f},$$

and it is understood that $\sigma \rightarrow 0$ after all integrations are performed. In the above expressions, v_f , k_f , E_f are the velocity, wave vector, and energy, respectively, of the most energetic electrons in the Fermi distribution. In region I, interactions occur predominantly between the incident electron and individual plasma electrons through a static shielded potential of the Debye-Hückel form. In region II, for large x and z , interactions are primarily of the Coulomb form between the incident and individual electrons in the plasma. In region III,

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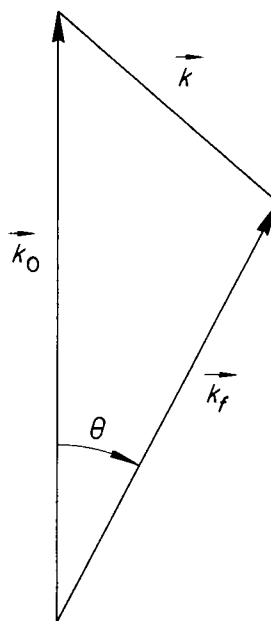


Fig. 66. Momentum Diagram for Scattered Electrons.

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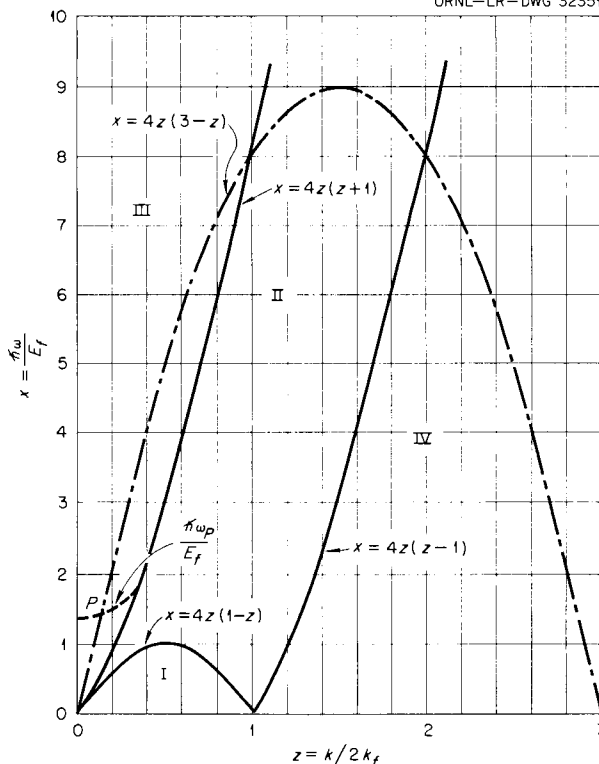


Fig. 67. Regions in the x - z Plane for Which Different Analytical Representations of τ Obtain.

on the line marked P , the phenomenon of plasma oscillations occurs. These oscillations are in the form of localized "hot spots" in the plasma and persist until dissipated by electron-electron or electron-ion collisions. The dashed line of Fig. 67 encloses the region in x - z space which is permitted by energy-momentum conservation for an electron of energy $E_0 = 9E_f$.

If we assume that the energy and momentum absorbed by the plasma are very large compared with E_f and mv_f , respectively, and integrate over ω ,

$$\tau(\theta) d\theta = \frac{2\pi ne^4}{E_0^2} \frac{\cos \theta d\theta}{\sin^3 \theta},$$

which is just the Born approximation for the scattering of electrons on free electrons in the laboratory system of coordinates.

It may be shown that for an electron of energy E_0 , if $E_0 - E_f \ll E_f$, one has, approximately,

$$\begin{aligned} \tau &= \int d\omega d\Omega \tau(\omega, \theta) \\ &= \frac{1}{32a_0\alpha^2} (\alpha^2 - 1) \left[\frac{1}{1 + x^2/2} + \frac{1}{x(1 - x^2/2)^{1/2}} \tan^{-1} \left(\frac{1 - x^2/2}{x^2} \right)^{1/2} \right], \end{aligned}$$

where a_0 is the Bohr radius, and $\alpha^2 = E_0/E_f$.

A slow positron with velocity $v_0 \ll v_f$ and incident on the Fermi gas may be shown to have an interaction probability of

$$\tau = \int d\Omega d\omega_f \tau(\omega, \theta) = \frac{\pi m^2 v_0^5}{105 \hbar v_f^2},$$

which agrees with the result of Lee-Whiting¹⁸ in the same limit. He assumed a screened Coulomb interaction between the positron and plasma electrons and calculated τ in the Born approximation, taking explicit account of the exclusion principle in the scattering process. The present formulation automatically yields this result in the low-energy limit and should be considerably more accurate than the results of Lee-Whiting at larger

energies, since it takes into account the dynamical properties of plasma.

A numerical evaluation of $\tau(\theta, \omega)$ has been carried out for an electronic density corresponding to $x^2 = 0.344$. This is appropriate to the conduction band in aluminum. The quantity

$$T(x, z) = \frac{\hbar v_0^2}{e^2} \tau(x, z)$$

is plotted in Fig. 68, which shows contours of equal intensity in T in the x - z plane. The plasma resonance line P beginning at $\hbar\omega/E_f = 1.36$ has a high intensity and vanishingly small width in the limit of zero damping. There is a quasi-resonance in regions I and II which has a much lower intensity.

The solid line of Fig. 69 shows a plot of $T(x) = \int T(x, z) d\Omega$ for an electron of energy $E_0 \gg E_f$. The dashed line gives a plot of $\hbar v_0^2/e^2$ times the probability that the incident particle shall lose energy to one-particle states (does not include excitation of oscillations). It is planned that these interaction probabilities will be used to

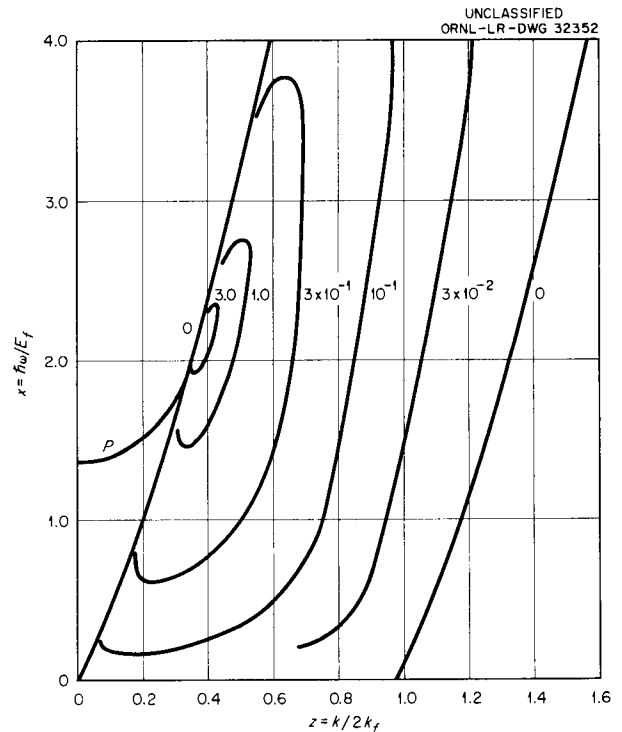


Fig. 68. Contours of Equal Values of $\frac{\hbar v_0^2}{e^2} \tau(x, z)$ in the x - z Plane.

¹⁸G. E. Lee-Whiting, *Phys. Rev.* 97, 1557 (1955).

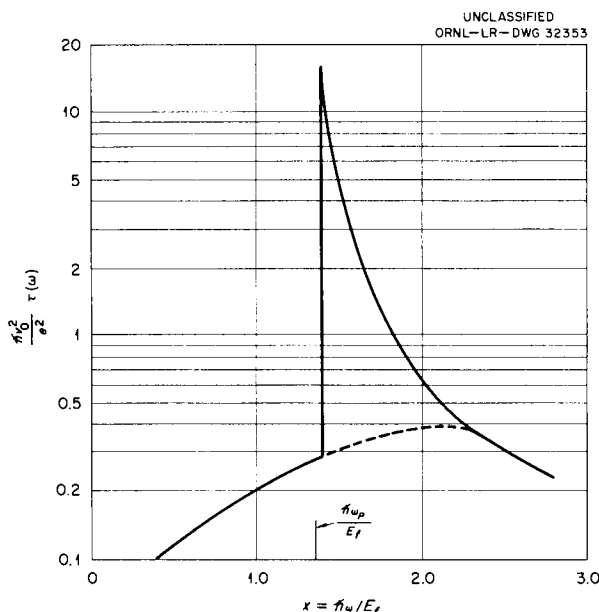


Fig. 69. Interaction Probability in a Free-Electron Gas as a Function of Energy Loss, $\hbar\omega$.

calculate the slowing-down spectrum of electrons in a Fermi gas.¹⁹

Low-Energy Collisions with a Fermi-Dirac Gas

Goldberger²⁰ has treated the problem of the interaction of neutrons and heavy nuclei with the aid of the statistical model of the nucleus. He has considered collisions between incident neutrons and individual nucleons and takes specific account of the action of the exclusion principle in reducing the collision cross section. These techniques have been applied recently in extensive calculations of intranuclear cascades.^{21,22} In a particular case, Goldberger has determined the effective mean free path for isotropic center-of-mass scattering with constant cross section on a degenerate Fermi gas and finds that the cross section is reduced by a factor of $1 - 7/5P_0^2$ due to the exclusion principle if $P_0 > \sqrt{2}$, where P_0 is the momentum of the incident particle in units of

¹⁹Wolff has made a calculation of the spectrum of secondary electrons generated by fast electrons in a Fermi gas but used a rather unrealistic cross section derived from the static Debye shielded Coulomb potential. See P. A. Wolff, *Phys. Rev.* 95, 56 (1954).

²⁰M. L. Goldberger, *Phys. Rev.* 74, 1269 (1948).

²¹N. Metropolis *et al.*, *Phys. Rev.* 110, 185 (1958).

²²N. Metropolis *et al.*, *Phys. Rev.* 110, 204 (1958).

the Fermi momentum. No result is given for $P_0 < \sqrt{2}$. Goldberger's result has been used by Wolff²³ in the calculation of secondary-electron cascades in metals.

In using Wolff's theory, the authors had occasion to work out the effective cross section for the case $P_0 < \sqrt{2}$ and believe that the result is of sufficient interest to be made generally available.

Following Goldberger, we let the momentum of the incident particle be $\vec{P}_0$ and that of one of the target particles be $\vec{P}_1$. All momenta will be measured in units of the Fermi momentum.

Referring to Fig. 70, let $\vec{R}_0$ and $\vec{R}_1$ be the position vectors in the laboratory system. Then we may write in the case of equal masses that the position vector $\vec{R}$ of the center of mass of the system is

$$\vec{R} = \frac{1}{2}(\vec{R}_1 + \vec{R}_0)$$

or

$$\vec{V} = \frac{1}{2M}(\vec{P}_1 + \vec{P}_0),$$

where $\vec{V}$ is the center-of-mass velocity. The momentum of the incident particle in the C.M. system must be

$$\vec{p}_0 = \vec{P}_0 - M\vec{V} = \frac{1}{2}(\vec{P}_0 - \vec{P}_1),$$

²³P. A. Wolff, *Phys. Rev.* 95, 56 (1954).

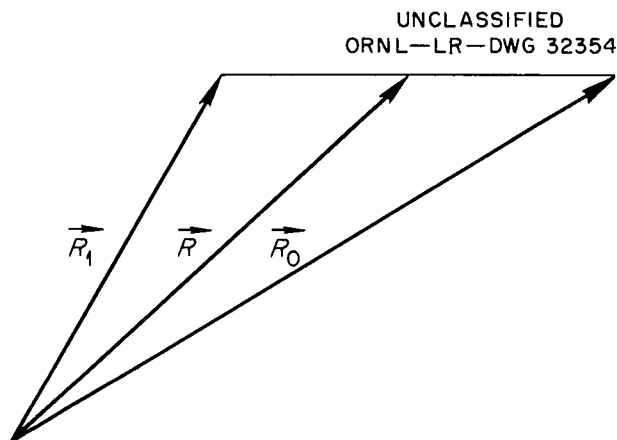


Fig. 70. Position Vectors of Incident and Target Particles in the Laboratory System.

and the momentum of the target particle must be equal to $-\vec{p}_0$ in the same system. If the incident particle suffers a momentum change $\vec{g}$, then, clearly, the struck particle undergoes a change of $-\vec{g}$, and the final momentum of the incident particle $\vec{p}_f$ is given by

$$\vec{p}_f = \vec{p}_0 + \vec{g}.$$

The cross section for interaction of the particles in the C.M. system may generally be written as a function only of $\vec{p}_0$ and the angle between $\vec{p}_0$ and $\vec{p}_f$. Assume that the cross section for scatter into an element of solid angle $d\Omega_f$ is $\sigma d\Omega_f$. Following Goldberger, $d\Omega_f$ may be expressed in terms of a volume element in $\vec{p}_f$ space;

$$\sigma d\Omega_f = \sigma \frac{\delta(p_f - p_0)}{p_f^2} d\vec{p}_f = \sigma \frac{\delta(p_f - p_0)}{p_0^2} d\vec{g},$$

where the δ -function relation follows from the conservation of energy. We assume that $\vec{p}_f$ and $\vec{p}_0$ are to be expressed as functions of $\vec{g}$ and $\vec{p}_1$. The effective cross section σ is then given by integration of $\sigma d\Omega_f$ over the allowed values of $\vec{p}_1$ times the relative velocity of the two particles $1/M |\vec{p}_0 - \vec{p}_1|$ divided by the incident velocity P_0/M :

$$\bar{\sigma} = \frac{1}{P_0} \int d\vec{g} \int d\vec{p}_1 N(\vec{p}_1) |\vec{p}_0 - \vec{p}_1| \sigma(p_0, \vec{p}_0 \cdot \vec{p}_f) \frac{1}{p_0^2} \delta(\vec{p}_0 - \vec{p}) ,$$

where $N(\vec{p}_1)$ is the density of target particles in momentum space. The integration is restricted by the condition that the final momentum vectors of both particles must be outside the occupied sphere in momentum space.

We choose $\sigma(p_0, \vec{p}_0 \cdot \vec{p}_f) = \sigma/4\pi$, where σ is independent of energy; that is, we assume isotropic center-of-mass scattering.

The coordinate system chosen by Goldberger for carrying out the integral over $\vec{p}_1$ is as shown in Fig. 71. The origin of coordinates is taken at

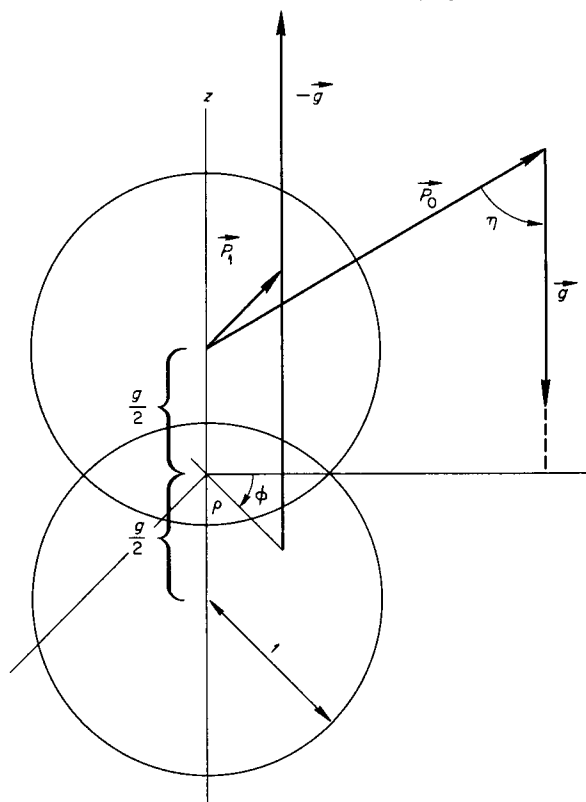


Fig. 71. Coordinate System Used in Calculating Collision Cross Section.

$\vec{g}/2$ from the center of the filled sphere in momentum space, and the $+z$ direction is taken in the $-\vec{g}$ direction. In order that the struck particle shall receive momentum sufficient to carry it into the allowed region, its initial momentum vector $\vec{p}_1$ must lie outside the auxiliary sphere, with the center at $\vec{g}$ relative to the original sphere, and, of course, $\vec{p}_1$ must lie inside the original sphere. We will carry out the integral over $\vec{p}_1$ first, holding $\vec{g}$ constant. All quantities must be expressed in

terms of $\vec{g}$ and $\vec{P}_1$. In the new coordinate system ρ, ϕ, z , we may write

$$\bar{\sigma} = \frac{1}{P_0} \int d\vec{g} \int \rho d\rho \int dz \left| \vec{P}_0 - \vec{P}_1 \right| \frac{1}{p_0^2} \delta(p_0 - p_f),$$

with the restrictions on the ρ, ϕ, z integrations as stated above.

If $p_0 - p_f$ is written in terms of the variables ρ, ϕ , and z ,

$$p_0 - p_f = \frac{1}{2} \left[\left| P_0 - P_1 \right| - \left| P_0 - P_1 + 2\vec{g} \right| \right] \\ = \frac{1}{2} \left[\sqrt{\left(P_0 \cos \eta + \frac{g}{2} - z \right)^2 + (P_0 \sin \eta - \rho \cos \phi)^2 + \rho^2 \sin^2 \phi} - \sqrt{\left(P_0 \cos \eta + \frac{g}{2} - z - 2g \right)^2 + (P_0 \sin \eta - \rho \cos \phi)^2 + \rho^2 \sin^2 \phi} \right].$$

Now

$$\delta(p_0 - p_f) dz \rho_f d\phi \\ = \delta(p_0 - p_f) \frac{d(p_0 - p_f)}{d(p_0 - p_f)/dz} \rho d\rho d\phi,$$

and

$$\frac{d}{dz} (p_0 - p_f) \\ = -\frac{1}{2} \left[\frac{P_0 \cos \eta + \frac{g}{2} - z}{\left| \vec{P}_0 - \vec{P}_1 \right|} - \frac{\left(P_0 \cos \eta - \frac{3g}{2} - z \right)}{\left| \vec{P}_0 - \vec{P}_1 + 2\vec{g} \right|} \right].$$

The integral over z may be carried out immediately: if z_0 is the value of z at which $p_0 - p_f = 0$,

$$z_0 = P_0 \cos \eta - \frac{g}{2},$$

$$\left[\frac{d(p_0 - p_f)}{dz} \right]_{z=z_0} = -\frac{g}{\left| P_0 - P_f \right|}.$$

Then, setting

$$N(\vec{P}_1) = \frac{3}{4\pi} N,$$

where N is the total density of particles in configuration space,

$$\bar{\sigma} = \frac{\sigma_T N}{4\pi} \int d\vec{g} \int_{\rho_1}^{\rho_2} \rho d\rho \int_0^{2\pi} d\phi \frac{3}{\pi g} = \frac{N\sigma_T}{4\pi} \int d\vec{g} F(g),$$

where the ρ integration runs over the allowed region included between the main sphere and the auxiliary sphere in momentum space. There are

three special cases depending on the value of g and z_0 :

- (1) $g \leq 2, \quad 0 \leq z_0 \leq 1 - \frac{g}{2},$
- (2) $g \leq 2, \quad 1 - \frac{g}{2} \leq z_0 \leq 1 + \frac{g}{2},$
- (3) $g \geq 2, \quad \frac{g}{2} - 1 \leq z_0 \leq \frac{g}{2} + 1.$

The following values of $F(g)$ obtain:

- (1) $F(g) = \frac{3}{P_0} (2P_0 \cos \eta - g),$
- (2) and (3) $F(g) = \frac{3}{P_0 g} [1 - (g - P_0 \cos \eta)^2],$

and it is required that the final momentum vector of the projectile, $\vec{P}_f = \vec{P}_0 + \vec{g}$, must lie outside the Fermi sphere:

$$P_0 - 1 \leq g \leq P_0 + 1,$$

$$\cos \eta \leq \frac{P_0^2 - g^2 - 1}{2P_0 g}.$$

The integration over $\vec{g}$ is tedious and must be carried out in five different intervals. We write

$d\vec{g} = 2\pi g^2 dg d(\cos \eta)$ and integrate first over $\mu = \cos \eta$:

Region I $2 \leq g \leq P_0 + 1$

$$S_I = \frac{2P_0}{3N\sigma_T} \bar{\sigma}_I = \int_3^{P_0+1} g^2 dg \int_{g-1/P_0}^{(g^2+P_0^2-1)/2gP_0} d\mu f_1$$

Region II $1 + \sqrt{2 - P_0^2} \leq g \leq 2$

$$S_{II} = \int_{1-\sqrt{2-P_0^2}}^{1+\sqrt{2-P_0^2}} g^2 dg \left[\int_{g/2P_0}^{1/P_0} d\mu f_2 + \int_{1/P_0}^{(g^2+P_0^2-1)/2gP_0} d\mu f_1 \right]$$

Region III $1 - \sqrt{2 - P_0^2} \leq g \leq 1 + \sqrt{2 - P_0^2}$

$$S_{III} = \int_{1-\sqrt{2-P_0^2}}^{1+\sqrt{2-P_0^2}} g^2 dg \int_{g/2P_0}^{(g^2+P_0^2-1)/2gP_0} d\mu f_2$$

Region IV $P_0 - 1 \leq g \leq 1 - \sqrt{2 - P_0^2}$

$$S_{IV} = \int_{P_0-1}^{1-\sqrt{2-P_0^2}} g^2 dg \left[\int_{1/P_0}^{(g^2+P_0^2-1)/2gP_0} f_1 d\mu + \int_{g/2P_0}^{1/P_0} f_2 d\mu \right]$$

Region V $0 \leq g \leq P_0 - 1$

$$S_V = \int_0^{P_0-1} g^2 dg \left[\int_{g/2P_0}^{1/P_0} d\mu f_2 + \int_{1/P_0}^{(1+g)/P_0} d\mu f_1 \right]$$

where

$$f_1 = \frac{1}{g} [1 - (P_0 \mu - g)^2] ,$$

$$f_2 = 2 P_0 \mu - g .$$

Using the relation $P_0^2 = E/E_F$, where E is the kinetic energy of the incident particle, and E_F is the Fermi energy, we finally obtain, after reduction,

$$\frac{1}{N\sigma_T} \bar{\sigma} = 1 - \frac{7}{5} \frac{E_F}{E} + \frac{2}{5} \frac{E_F}{E} \left(2 - \frac{E}{E_F} \right)^{5/2},$$

$$1 \leq \frac{E}{E_F} \leq 2,$$

and, as found by Goldberger,

$$\frac{1}{N\sigma_T} \bar{\sigma} = 1 - \frac{7}{5} \frac{E_F}{E}, \quad 2 \leq \frac{E}{E_F}.$$

Figure 72 shows a plot of the quantity $\bar{\sigma}/N\sigma_T$ as a function of E/E_F .

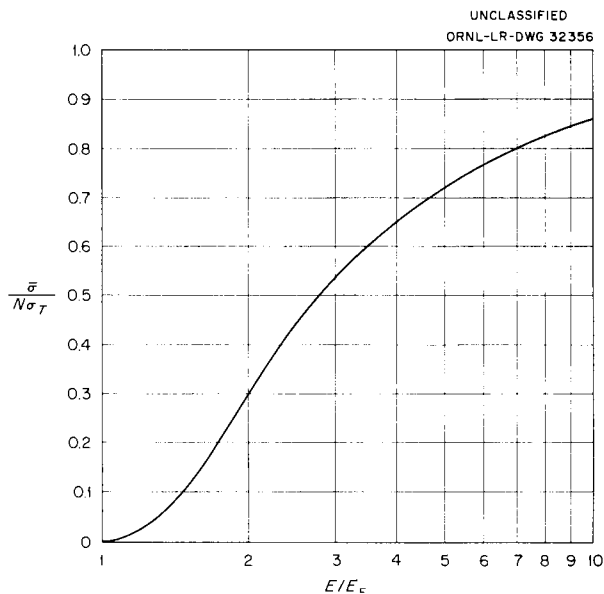


Fig. 72. Exclusion Correction to the Scattering Cross-Section as a Function of the Energy of the Incident Particle in Units of the Fermi Energy.

EXPERIMENTAL PHYSICS OF DOSIMETRY

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A Small Personnel Dosimeter for Beta and Gamma Radiation

The problem of measuring beta-ray exposures to personnel has become increasingly acute because of the growing use of beta-ray-emitting isotopes and because of the variable beta-to-gamma dose ratio from fallout from weapons tests. This variation makes it difficult to estimate beta exposure from gamma-ray dose measurements. In addition, there is a need for a device to measure dose to the hands.

The requirements of a satisfactory dosimeter for this purpose may be summarized as follows:

1. It should record any beta dose that can penetrate the minimal dead layer of the epidermis, nominally taken to be 7 mg/cm².
2. Its response to beta rays at a depth of 7 mg/cm² should be 50% of that to gamma rays, because NBS Handbook 59 allows maximum permissible beta dose to be approximately twice

the maximum permissible gamma dose at this depth.

3. If an ionization-type instrument is used, it should be charged and read on existing equipment, such as the Victoreen Minometer.

4. Its response to x rays and gamma rays should be a true measure of the dose in roentgens, independent of radiation energy.

5. Its electrical leakage should be very low in order to avoid false readings.

6. It should be small and convenient to carry.

7. It should be cheap to manufacture and repair, and strong enough to withstand reasonably rough treatment.

Several years ago our group designed and tested a dosimeter which fulfilled nearly all the requirements, but it was not sufficiently cheap nor rugged and was not suitable for use on the hands.

Description of the New Dosimeter. — The general interest in the device led us to design another type of dosimeter which satisfies all the design criteria mentioned. The basic structure is that of a parallel-plate ionization chamber in the shape of a small pill box, shown in Figure 73. The top and sides are 50% open so as to give a 50% response to beta rays, compared with gamma rays, from all directions within a hemisphere. The dosimeter is 1½ in. in over-all diameter and ¾ in. high, or not

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²⁵USAF.

²⁶USPHS.

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Fig. 73. Beta-Ray-Sensitive Dosimeter.

much larger than a wrist watch. The dosimeter is intended to be mounted on either a wrist strap or in a small holder which clips onto the wearer's collar.

The outer cap is made of nylon, which provides strength, ease of molding, a response to x and gamma rays independent of the radiation energy. The base is of fluorothene for high electrical insulation and appropriate dielectric constant. Inside the cap is a plastic film having a surface density of 7 mg/cm^2 , which approximates the dead layer of the epidermis. The film may be made of a conducting plastic or it may be coated inside with Aquadag to render it electrically conducting.

The structure is shown in Fig. 74, which gives top and cross-section drawings. The lower insulated electrode is a layer of Aquadag painted on the inside of the base and is charged through

a small aluminum rivet in the center. The rivet is normally protected from accidental discharge by a plastic plug. The grounded electrode is formed by another layer of Aquadag painted on the inside of the cap and connected to the aluminum base.

The sensitive volume, 5.3 cm^3 , is the same as that of the Victoreen pencil chamber, model 362. The capacitance of the dosimeter is the sum of the capacitance between the insulated electrode and the inside of the cap and the capacitance between the insulated electrode through the plastic base to the metal base. The total capacitance is also the same as that of the Victoreen, $4 \mu\text{f}$. Hence the sensitivity is the same as that of the Victoreen, and the dosimeter can be charged and read on the same Minometer electroscope. The capacitance is adjusted by altering the thickness of the base, its dielectric constant, and by machining a cavity of proper size in the base. The hollow is

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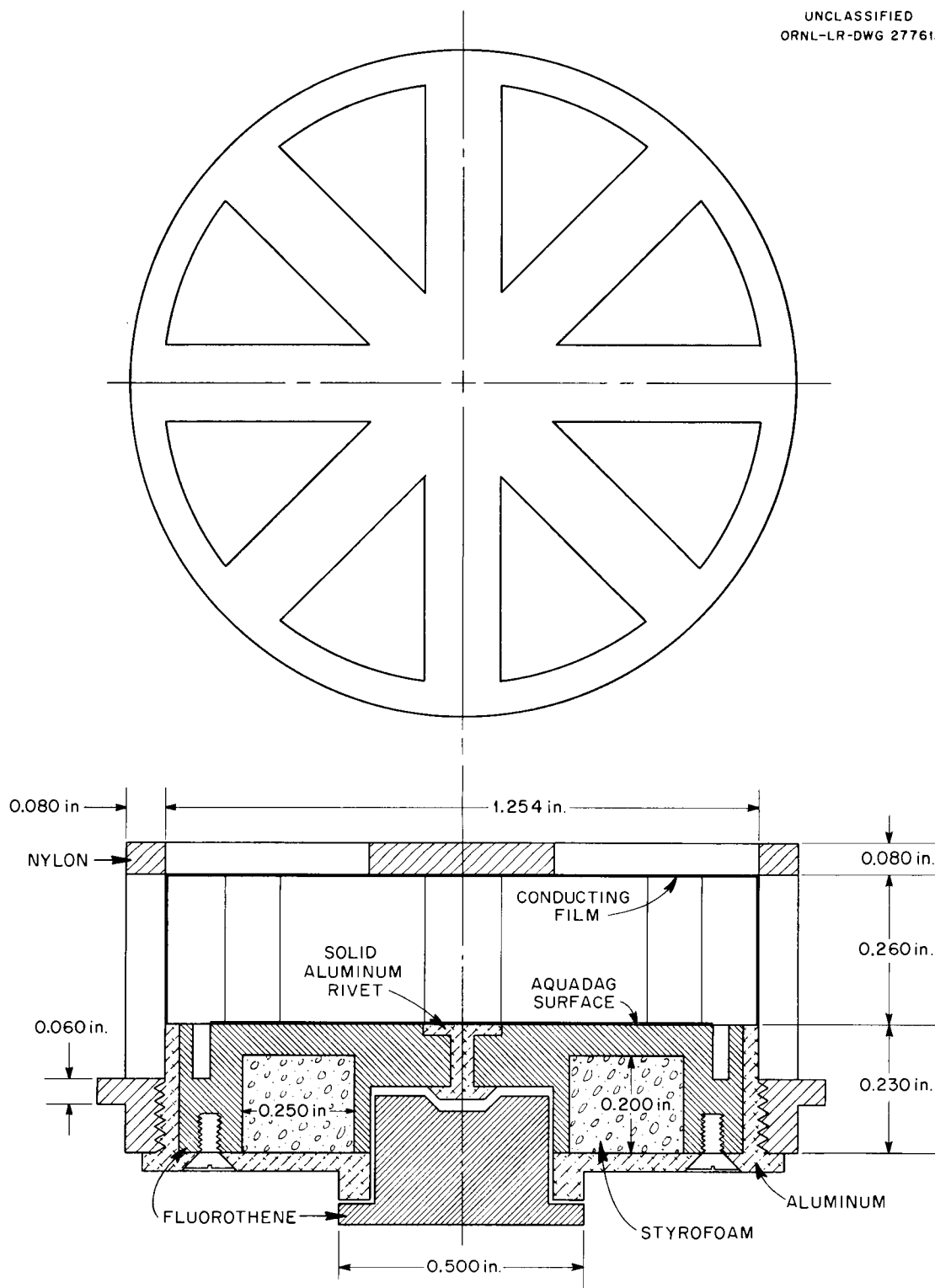


Fig. 74. Beta-Ray-Sensitive Dosimeter, Top View and Cross Section.

filled with Styrofoam to prevent ions from migrating to the surfaces and falsifying the readings.

The dimensions were chosen from calculations of the minimum value of the over-all thickness as the diameter and thickness of the base were varied subject to the condition of fixed volume in the air chamber and fixed total capacitance. At this time the hollow in the base was neglected. If the effective radius of the insulated electrode is r cm and the height of the cylindrical air chamber is x cm, the volume in cm^3 is

$$(1) \quad V = \pi r^2 x .$$

The capacitance C in μmf is the sum of that in the base, whose thickness is y cm and whose dielectric constant is K , and that in the air chamber; hence

$$(2) \quad C = \pi \epsilon_0 r^2 \left(\frac{1}{x} + \frac{K}{y} \right) ,$$

where ϵ_0 is the dielectric constant of a vacuum or air in the units $8.85 \times 10^{-2} \mu\text{mf}/\text{cm}$. If the radius r is expressed as a function of the total thickness $z = x + y$, dz/dr may be determined and set equal to zero in order to find the radius for minimum total thickness. Figure 75 indicates the trends of the various dimensions and shows that the minimum in the total thickness occurs for a diameter of about 3 cm.

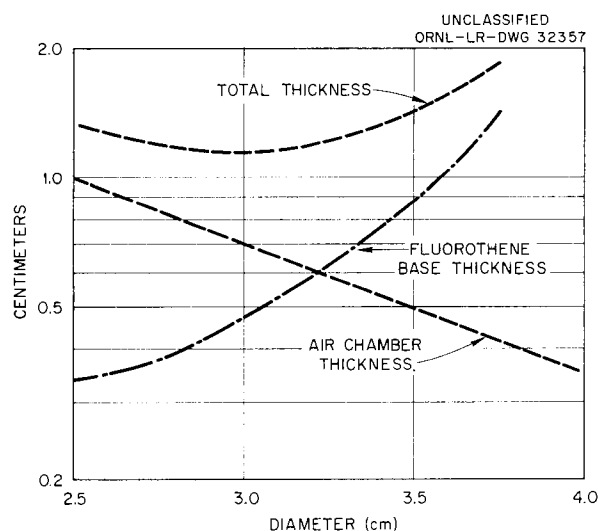


Fig. 75. Variation of Dosimeter Parameters as Chamber Diameter is Changed. Read left scale as ratio for top curve; read in centimeters for the other curves.

The electric field in the chamber must be large enough to collect all the ions formed at any reasonable dose rate. Since Gupton²⁷ has shown that the fields in the Victoreen condenser "r" meter are adequate, the new dosimeter was designed so that the minimum electric field after a 200-mr exposure was at least 120 v/cm, the lowest value in the Victoreen. This condition limits the maximum value of the air chamber thickness, x_m . The dimensions were then chosen for a minimum total thickness z and a value of the air chamber thickness $x < x_m$.

The dosimeters were tested first for their response to x and gamma rays, with the results shown in Fig. 76. The abscissa represents the effective energy of the heavily filtered x rays; the ordinate represents the ratio of the measured reading per roentgen of exposure to the reading at high energies, such as those for 250-kv heavily filtered x rays or radium gammas. Several cap materials were tried, and the results for aluminum, fluorothene, and nylon are shown in Fig. 76 and compared with the results for the Victoreen pocket meter. The main point to notice is that the caps free of metals and heavy atoms give a response which is nearly uniform over the useful range of energies. Nylon was one of the best cap materials and was used in the dosimeter for that reason as well as for its strength. Aside from the somewhat smaller energy-dependent response at low energies, the over-all gamma response is the same as that of the Victoreen pencil: 200 mr for full-scale reading on the Minometer.

Calibration of the Dosimeter. — The dosimeter was calibrated with beta rays by using a method which was developed for the pencil chamber and which has been reported in the literature.²⁸ Briefly, the method depends on surrounding the dosimeter with water solutions of several beta-emitting isotopes. The active layer is thicker than the range of any of the beta rays; so the spectrum is completely degraded and can be calculated according to the theory of Spencer and Fano.²⁹

²⁷E. D. Gupton, *Recombination Losses in Pocket Ionization Chambers*, Masters' thesis, Vanderbilt University (March 1955).

²⁸H. H. Hubbell, Jr., R. M. Johnson, and R. D. Birkhoff, *Nucleonics* 15(2), 85 (1957); *Radiology* 69, 268 (1957); *Pocket Ion Chambers for Beta Radiation Dose*, ORNL-2158 (April 26, 1957).

²⁹L. V. Spencer and U. Fano, *Phys. Rev.* 93, 1172 (1954); H. H. Hubbell, Jr., R. M. Johnson, and R. D. Birkhoff, *Nucleonics* 15(2), 85 (1957).

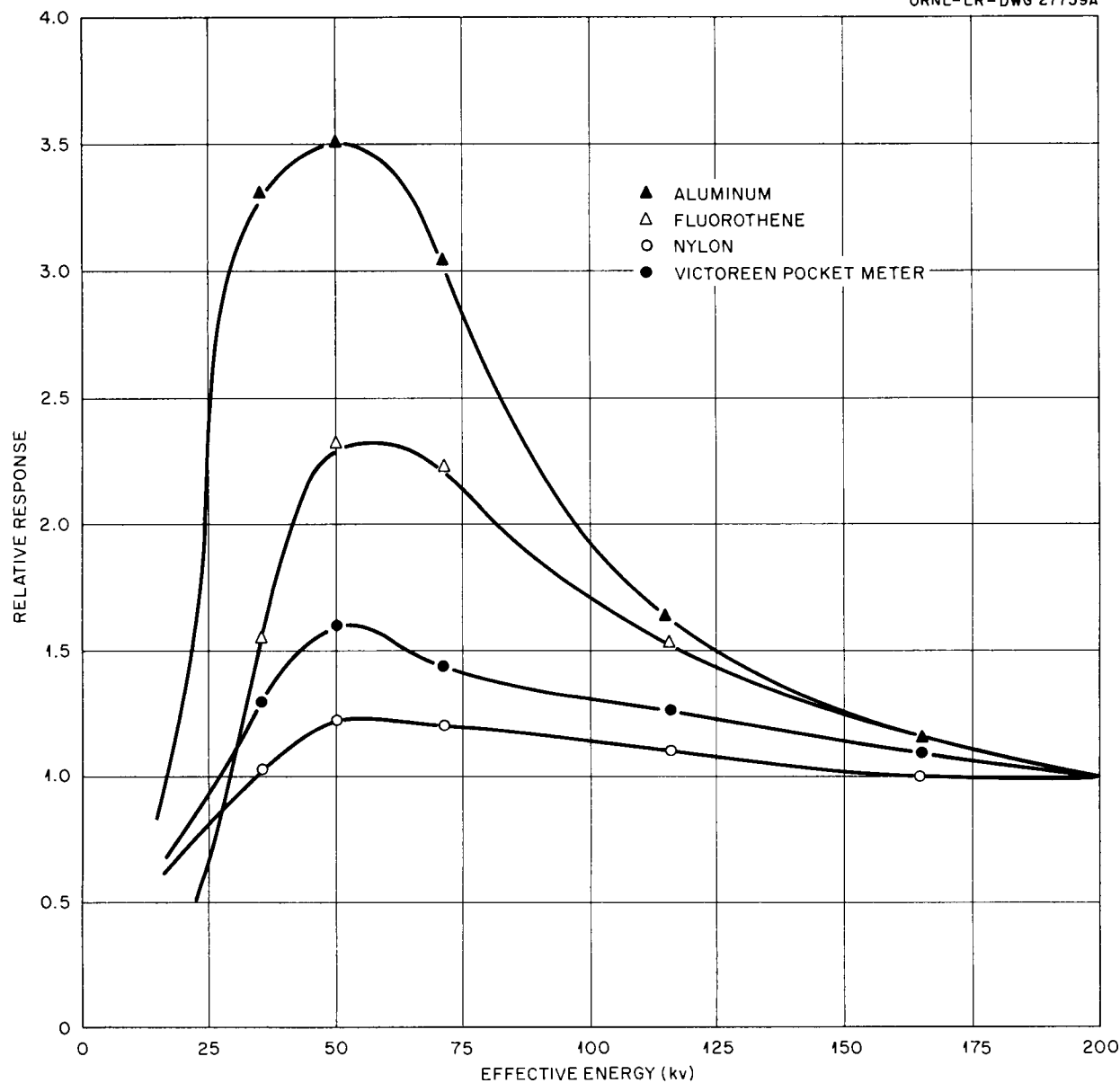
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Fig. 76. Variation of Response to X-Ray Energy for Victoreen Pocket Chamber and for New Dosimeter Made of Various Wall Materials.

A typical spectrum is shown in Fig. 77. The dotted curve represents the thin-source beta-ray spectrum, and the solid curve is the calculated flux spectrum as degraded in water. The average energy of the degraded spectrum in water gives a convenient parameter against which to plot the variation in dosimeter response to beta rays of different energy.

In order to provide a liquid beta source having no wall surrounding the dosimeter, the solution was placed in a small dish and rotated about the vertical axis, thus forming a paraboloidal cavity into which the dosimeter could be inserted.³⁰

³⁰W. M. Hurst, *Monitoring of Liquids for Radioactivity*, ORNL-1155 (Feb. 26, 1952).

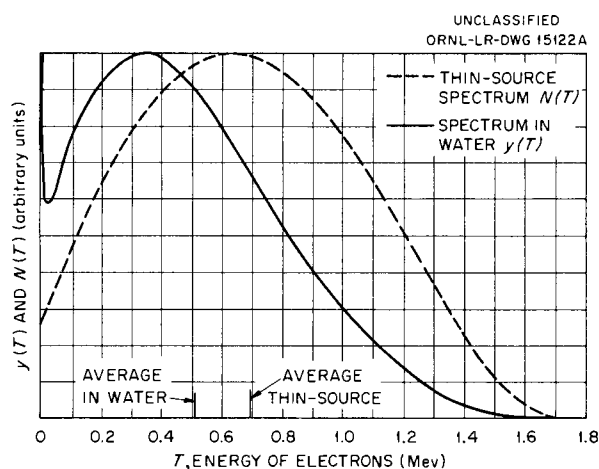


Fig. 77. Phosphorus-32 Spectra Obtained by Two Methods.

Radiation then reached it isotropically within a 2π solid angle, and the dosimeter was not wetted or otherwise contaminated by the liquid beta source. The proper speed of rotation is simply computed, since the equation of the surface is

$$(3) \quad y = \frac{4\pi^2 f^2}{2g} x^2 + C,$$

where f is the angular rotational speed, and g is the acceleration of gravity.

The boundary conditions on the depth and radius of the vessel were determined by the requirement that the minimum thickness of the water solution be equal to the range of the most energetic beta particle in the solution, about 1.3 cm. The rotational speed was determined so that the liquid surface would present an angle of 45 deg opposite the corner of the dosimeter, in order to provide minimum air spacing around the dosimeter to reduce air absorption. Accordingly, $f = 214$ rpm.

The exposure device, including the rotating solution cup and a hinged holder for the dosimeter, is shown in Fig. 78. The performance was entirely satisfactory, and none of the dosimeters were ever wetted or contaminated.

The response curves obtained are shown in Fig. 79, in which the dose read on the device is compared with that calculated from the assay of the

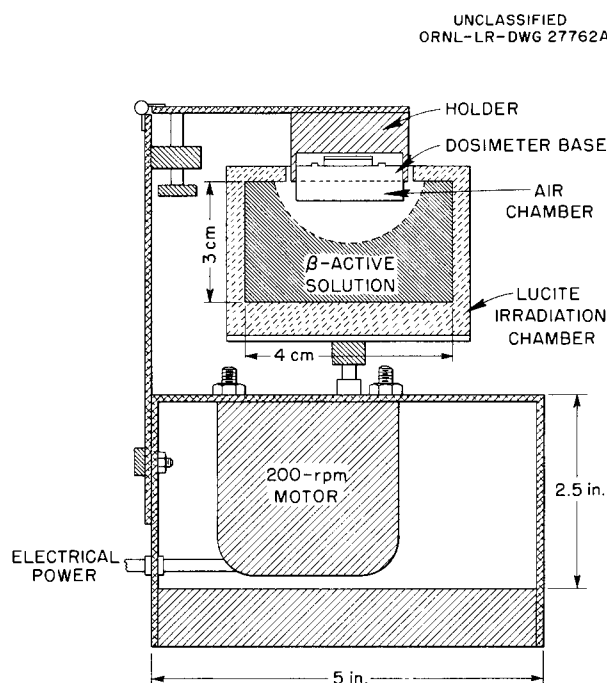


Fig. 78. Device for Exposing Dosimeters to Radioactive Solutions.

solution. The lower curve represents the corresponding response for the Victoreen pocket chamber, which, of course, is not intended to measure beta rays. Seven isotopes with average electron energies in water ranging from 52 kev for S^{35} to 694 kev for Y^{90} were used. These were chosen because they emitted no gamma rays and covered the whole range of beta-ray energies which must be monitored for external exposure.

Figure 80 shows a comparison of the same response with that obtained by a chamber having a 7-mg/cm^2 wall but no protecting ribs. The ordinate is therefore the ratio of the dose measured by the device to that received by the living skin cells. It is nearly flat at 50% response, as it was designed to be.

We therefore feel that this dosimeter is a satisfactory answer to the beta dosimetry problem.

This project was a cooperative one between the Dosimetry Section and the Applied Health Physics Section; ideas and efforts were contributed by A. P. Hull, H. H. Hubbell, Jr., R. D. Birkhoff, J. C. Hart, E. D. Gupton, and D. M. Davis.

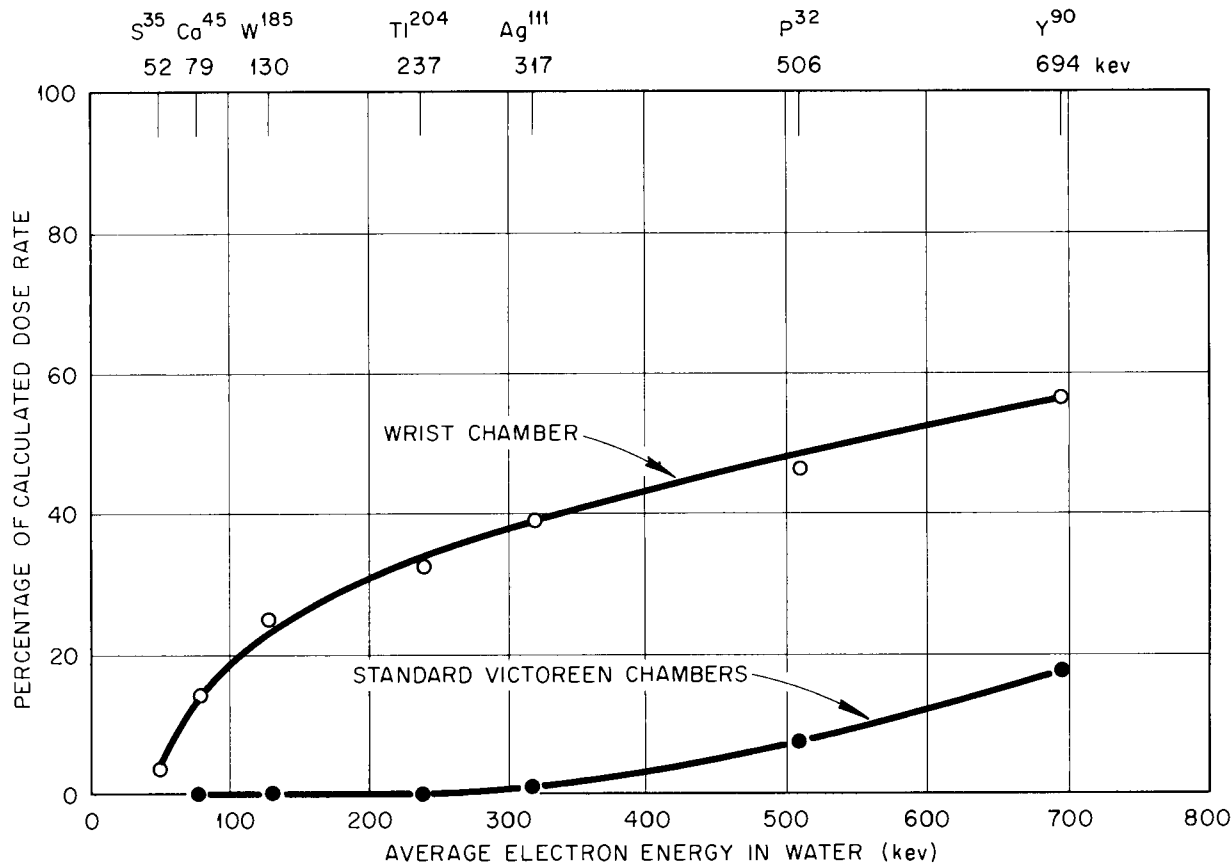
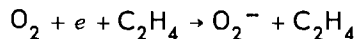
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Fig. 79. Comparative Response of Chambers to Calculated Dose Rate.

Electron Attachment in Mixtures Containing Oxygen

O₂-C₂H₄ Mixtures. — Electron attachment has been studied in O₂-C₂H₄ mixtures by using methods already described.^{31,32} Figure 81 shows that for $E/P = 0.10$, α , the probability of capture per cm in the field direction and per mm O₂, depends on the pressure of C₂H₄ but is independent of the oxygen pressure, $f_1 P$. These results may be expressed by $\alpha_e = \alpha_0 + k' f_2 P$, where α_0 is the value for α_e when $f_2 P \rightarrow 0$, and k' is a constant. Values of α_0 and k' were determined as a function of E/P (see Fig. 82).

The results may be interpreted with the model described in ORNL-2384,³² when the following reaction is added:



Thus O₂⁻ is formed in a stable configuration, and collisional stabilization is not necessary. In the case of O₂-N₂ (ref 32) this process was unimportant; essentially all of the O₂⁻ is formed first in an unstable configuration.

O₂-CO₂ Mixtures. — Some experimental data have been obtained for O₂-CO₂ mixtures, but these results have not yet been analyzed for the probability of capture.

Modification of the Attachment Apparatus. — Because a collimated alpha source is used to generate the free electrons, the range of pressures is limited. A new apparatus has been designed

³¹G. S. Hurst *et al.*, *H-P Semiann. Prog. Rep.* July 31, 1956, ORNL-2151, p 43.

³²R. D. Birkhoff *et al.*, *H-P Ann. Prog. Rep.* July 31, 1957, ORNL-2384, p 97.

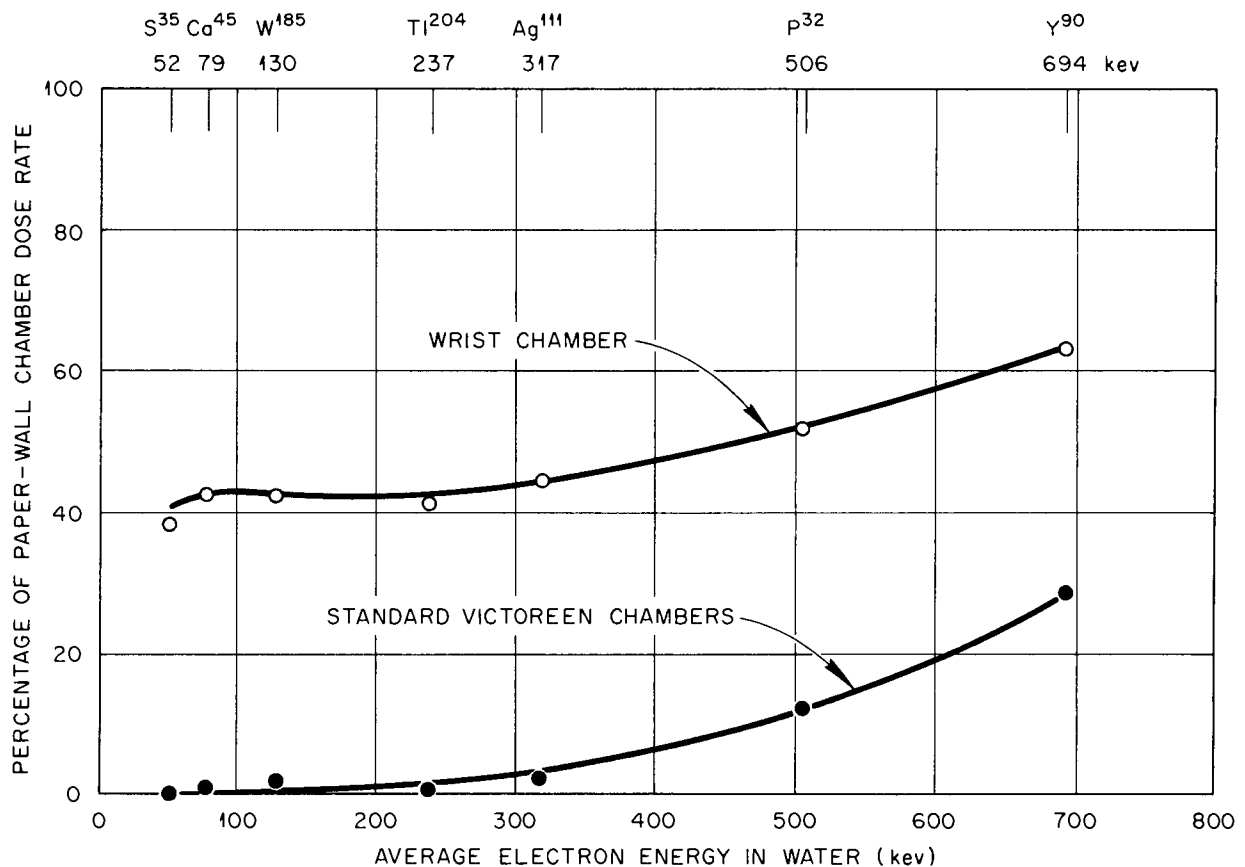
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Fig. 80. Response of Chambers Compared with Paper-Wall Chamber.

to overcome the pressure limitation by using a photoelectric source. The principal difficulty now being encountered stems from the instability of the ultraviolet source.

Measurement of Electron Stopping Power at Low Energies

A knowledge of the stopping power of matter for low-energy electrons is essential to the setting of maximum permissible limits of exposure to beta rays and to the evaluation of the secondary electron flux produced in media subjected to radiation. For electrons the only data presently available at low energies concern the energy loss of 148-kev electrons in aluminum. There apparently is no experimental information for lower energies where the Bethe formula predicts a great increase in stopping power.

The present experiments avoid the problem which has so far limited the measurements of stopping power to higher energies, namely, the absorption and scattering in the window which separates the vacuum of the accelerator from the gas-filled detector (ion chamber or counter). Two calorimetric methods were developed: the first measured the heat radiated by a thin foil which was being heated by the passage of the electron beam, and the second method measured directly the temperature of the foil.

For the first technique, calibration of the radiation detector was accomplished by heating the foil with an electric current running from edge to edge. The stopping power could be calculated from the watts of electric power required to cause the same detector response as that found with beam heating. A blackened thermistor in a Wheatstone

bridge circuit when used as a radiation detector gave a sensitivity of 6 cm of galvanometer deflection per milliwatt of foil power. In comparison, a 10-kev electron beam of $1 \mu\text{a}$ strength which lost 10% of its energy in the foil produced 1 mw of heating and, hence, 6 cm of deflection. While this was satisfactory sensitivity for the measurements desired, the lengthy time constant of the system (several minutes) made data gathering tedious and severely limited the accuracy because of interfering

fluctuations in the room temperature during the observation period. Although the sensitivity could have been improved by more sophisticated detectors (i.e., beam choppers, thermopiles, a-c amplifiers, etc.), the limitation imposed by the time constant, a result of the massive nature of the clamps needed to conduct current to the foil during calibration, was thought to be the fundamental difficulty. Even if the foil had been mounted in a manner to minimize heat transfer to the surroundings and if a detector calibration had been made by use of the complete-beam absorption technique described below, this method would still have had a considerably lower sensitivity than a system in which the temperature-sensitive element was in direct contact with the foil.

In the second method the foil was mounted on a ring about 1 cm in diameter, and the temperature of the foil was measured directly with an array of six thermocouples connected in series electrically and spaced around the periphery of the ring. When a calibrating electron beam, of such low energy that the range was less than the foil thickness, was completely absorbed in the foil, the drift rate of the galvanometer connected in series with these thermocouples was found to be directly proportional to the power expended in the foil. The calibration constant K_1 was found to depend only slightly on the atomic number and thickness of the foil and was typically about $25 \text{ cm} \cdot \text{min}^{-1} \cdot \text{mw}^{-1}$.

An identical independent set of thermocouples similarly positioned provided a means of cooling the foil by Peltier cooling before an experimental run so that the rate of drift could be obtained as the foil passed through room temperature, thus eliminating the need for corrections due to heat transfer to the surroundings. That these losses could be neglected can be seen from the differential equation which describes this system:

$$(1) \quad \dot{T} = K_1 P_B - (T - T_0) K_2$$

where P_B is the beam power delivered to the foil in mw, T_0 is room temperature, and K_2 is the heat transfer constant, with a value of about 0.33 min^{-1} . The solution for a foil initially at a temperature T_i (below room temperature) is readily shown to be

$$(2) \quad T = \left(\frac{K_1}{K_2} P_B + T_0 \right) (1 - e^{-K_2 t}) + T_i e^{-K_2 t}$$

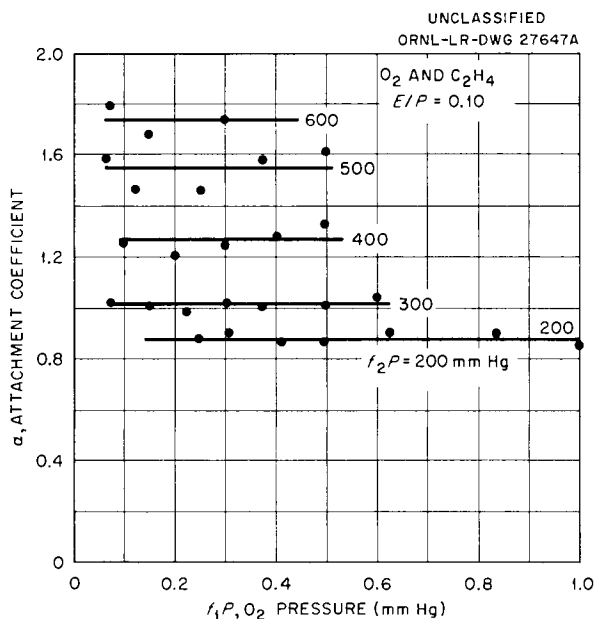


Fig. 81. Variation of Attachment Coefficient, α , with Pressure of Oxygen and Ethylene.

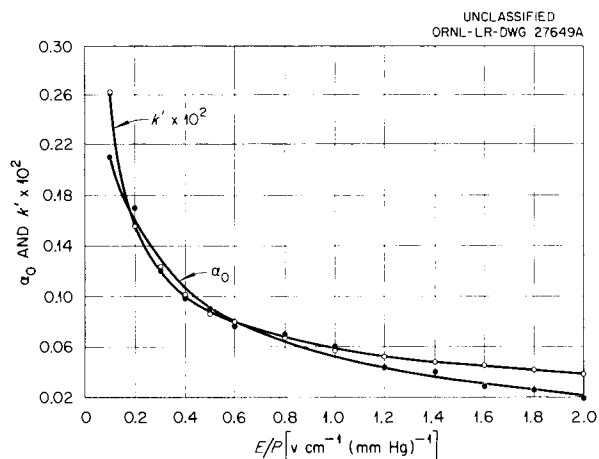


Fig. 82. Values of α_0 and k' as a Function of E/P .

A sketch of the curve for temperature vs time is shown in Fig. 83, where the temperature

$$T^{\infty} = \frac{K_1}{K_2} P_B + T_0 ,$$

and T_+ and T_- , which occur at times $t_0 + \Delta t$ and $t_0 - \Delta t$, respectively, are shown. If the slope is taken in the vicinity of room temperature, from the experimental data it may be shown from Eq. 2 that

$$(3) \quad \text{Slope} \equiv \frac{T_+ - T_-}{2\Delta t} = \frac{K_1}{K_2} P_B \frac{\sinh K_2 \Delta t}{\Delta t} .$$

If the quantity $K_2 \Delta t$ is small enough, the $\sinh$ function may be approximated by its argument, and the slope equals $K_1 P_B$ (i.e., the heat losses may be neglected). From the series expansion of the $\sinh$ function, the mathematical condition for this approximation is that $(K_2 \Delta t)^2 \ll 6$. The experimental observations were always limited to a Δt of 0.5 min so that heat losses could be neglected.

The stopping power for a thin foil of aluminum of thickness $50.3 \mu\text{g}/\text{cm}^2$ is shown in Fig. 84. The Bethe stopping power is plotted also for comparison, and agreement is good. The point at 12 kev has been corrected for scattering in the foil, which causes a path length greater than the foil thickness and hence a greater amount of energy deposition. The correction used involved a modification of the Yang³³ theory of the path length distribution in foils. The Yang theory is valid only for high-energy particles; thus it was necessary to introduce the more appropriate multiple-scattering theory of Molière³⁴ into the Yang schematization before it could be applied to the experiments here.

A Study of Preacceleration in Beta-Ray Spectroscopy

The experimental analysis of the low-energy regions of beta spectra has proved to be quite difficult because these electrons have insufficient energy to penetrate most ordinary counter windows. The problem has been attacked both by going to thinner windows and by adding energy to the electrons. Very thin windows have been made, but there is a lower limit on the practical thickness

imposed by conditions of strength and durability. Furthermore, even a windowless counter shows an effective window thickness due to escaping counter gas.

Postacceleration (addition of energy between the energy analyzer and the counter) has been simple to achieve and is useful, but the technique is limited to accelerating voltages which are less than the cutoff energy of the counter window. With voltages above this, a large increase in background is noted.

Preacceleration (addition of energy between the source and the energy analyzer) has been used by several investigators in an effort to avoid the high background associated with postacceleration. These investigators have generally been successful in getting electrons through counter windows. However, in many cases the work is in doubt because of the lack or incompleteness of knowledge of the action of the accelerator.

The essential features of the preacceleration technique may be readily determined. For relativistic electrons which leave the source with a kinetic energy T and momentum p and which receive an additional energy ΔE , the following equalities hold:

$$(1) \quad T + m_0 c^2 + \Delta E = \sqrt{p^2 c^2 + (m_0 c^2)^2} + \Delta E \\ = \sqrt{(p')^2 c^2 + (m_0 c^2)^2} ,$$

where p' is the momentum after acceleration. The relationship between the incremental momenta, dp and dp' , obtained from the above equation, is involved in considerations of resolution and transmission:

$$(2) \quad \frac{dp'}{dp} = \frac{p}{p'} \left[1 + \frac{\Delta E}{T + m_0 c^2} \right] .$$

The momentum resolution obtainable for the spectrum at the source dp/p is thus related to the momentum resolution inherent in the machine, dp'/p' , by the following:

$$(3) \quad \frac{dp}{p} = \frac{dp'}{p'} \left(\frac{I_{p'}}{I_p} \right)^2 \frac{1}{[1 + \Delta E/(T + m_0 c^2)]} ,$$

where $I_{p'}$ and I_p are the magnet currents corresponding to the transmission of momenta p' and p and are proportional to these momenta. Since the

³³C. N. Yang, *Phys. Rev.* **84**, 599 (1951).

³⁴G. Molière, *Z. Naturforsch.* **32**, 78 (1948).

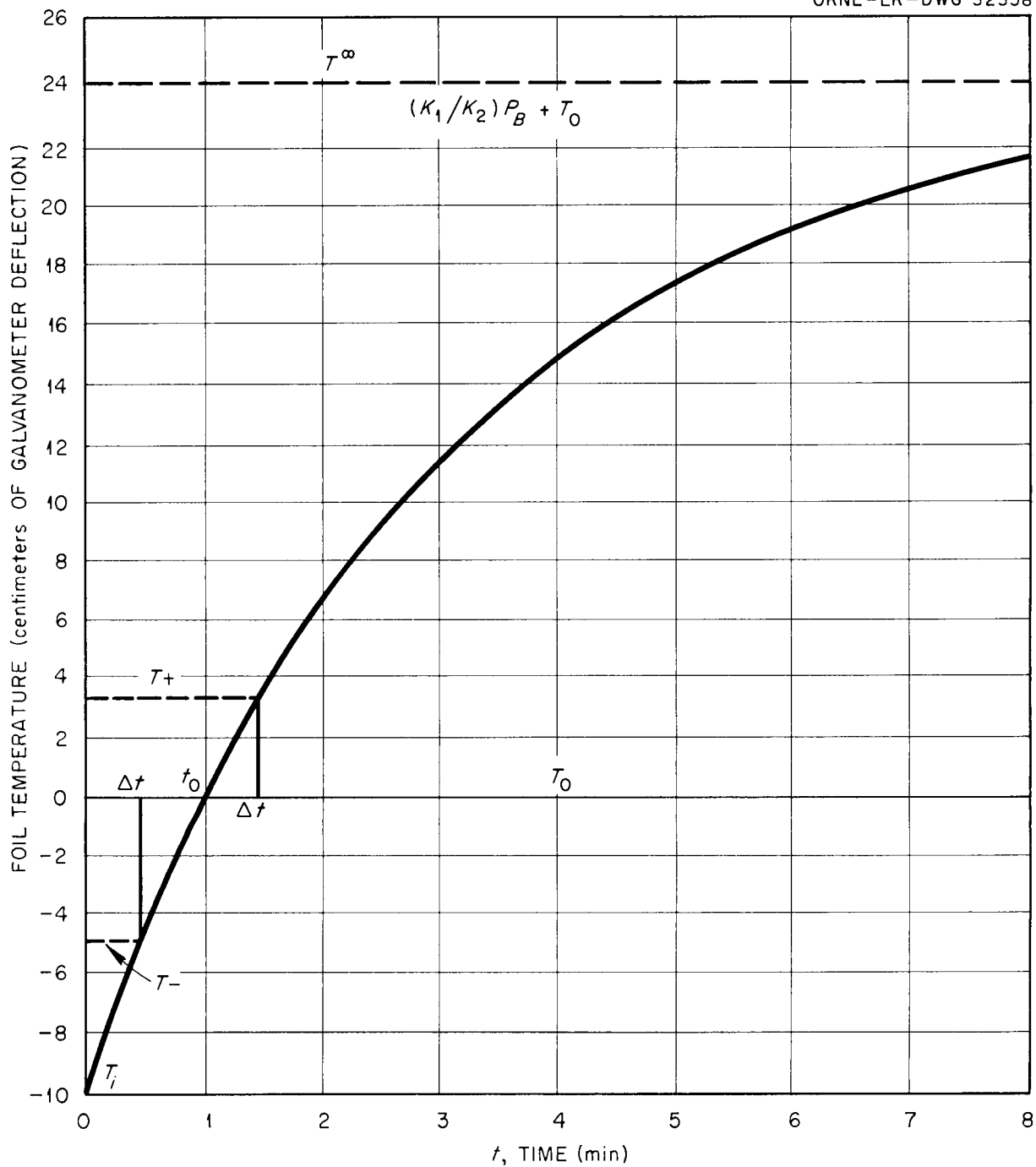
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Fig. 83. Temperature-Time Curve, According to Eq. 2, for an Initial Temperature (T_i) of a 10-cm Galvanometer Deflection and an Absorbed Power (P_B) of 0.3 mw.

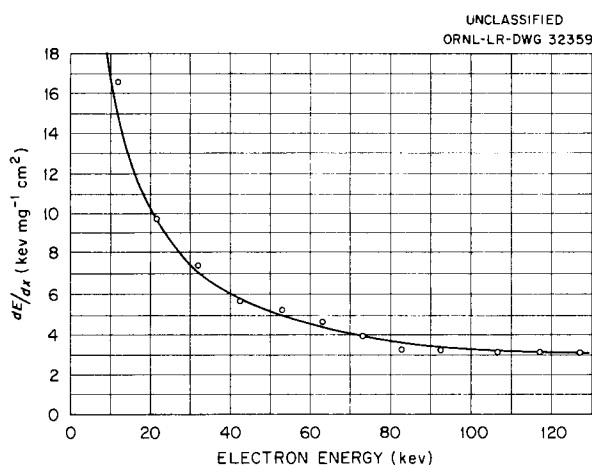


Fig. 84. Stopping Power of Aluminum for Low-Energy Electrons. The solid line represents the Bethe stopping power and is included for comparison.

bracketed quantity in Eq. 3 is very nearly unity in any practical situation and since $I_p' > I_p$, it is immediately clear that the momentum resolution is impaired ($dp/p > dp'/p'$) by preacceleration. The ratio of dp/p to dp'/p' is shown in Fig. 85 as a function of the energy at the source T for ΔE equal to 10, 20, and 30 keV.

A somewhat more subtle characteristic of preacceleration is the distortion to the shape of the momentum spectrum produced by this artificial widening of the momentum increment from the source which is transmitted by the spectrometer. If $N_1(p)$ is the momentum spectrum at the source and $N_2(p')$ is the momentum distribution transmitted by the spectrometer after preacceleration, then

$$(4) \quad N_1(p) dp = N_2(p') dp' = \left[N_2(p') \frac{dp'}{dp} \right] dp$$

The distribution $N_2(p')$ is obtained by dividing the observed counts per minute (c/m) by the momentum p' (or the current I_p' which is proportional to it) in the usual manner. Substitution of Eq. 2 in Eq. 4 then yields

$$(5) \quad N_1(p) = \frac{c/m}{(I_p')^2} I_p \left[1 + \frac{\Delta E}{T + m_0 c^2} \right],$$

which provides a prescription for converting counts per minute to a momentum distribution. This procedure yields a distribution identical with the one which would have been observed in the

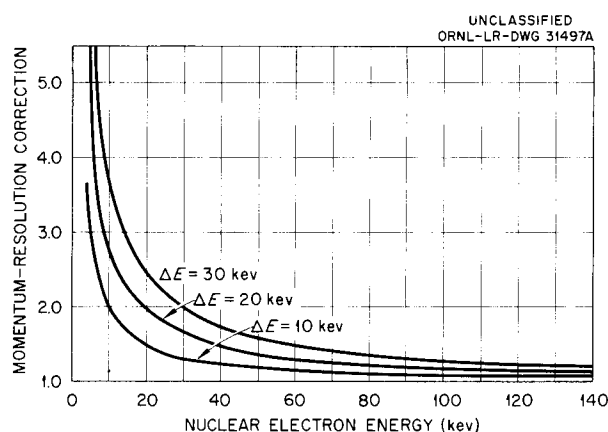


Fig. 85. Momentum-Resolution Correction for Added Energies of 10, 20, and 30 keV.

absence of preacceleration. It should be noted that the bracketed factor in Eq. 5 is a consequence of the relativistic relationship between energy and momentum (Eq. 1). This factor may be shown to be unity if the nonrelativistic equation,

$$(6) \quad \frac{p^2}{2m} + \Delta E = \frac{(p')^2}{2m},$$

is used in the above analysis instead of Eq. 1. The factor $I_p/(I_p')^2$ in Eq. 5 has a simple physical interpretation. The momentum spectrum, $(c/m)/I_p'$, is erroneously large because of the enlarged momentum increment from the source which is transmitted by the spectrometer. This spectrum may be reduced in magnitude to correspond to a correct momentum increment by multiplying by the ratio of the momentum increments ($\propto I_p/I_p'$) proportional to I_p/I_p' .

Although the primary purpose of preacceleration is to permit the counting of electrons whose energy is below the cutoff energy of the counter window, it has another effect which can be advantageous: that of effectively opening the momentum window of the spectrometer. This effect counteracts the shrinking of the window, which, being proportional to the momentum for fixed-slit magnetic spectrometers, approaches zero as the momentum of the electrons approaches zero.

To illustrate this effect, a graph of count rate vs energy has been plotted in Fig. 86 with and without acceleration (30 kv) for an isotope with a linear Fermi plot (Pm^{147} with an end point at 225 keV).

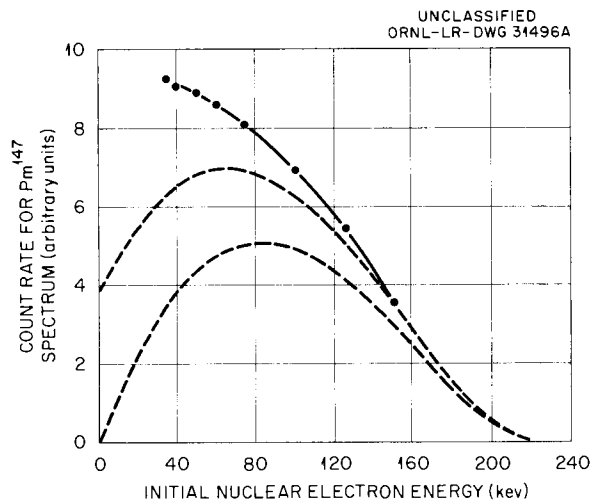


Fig. 86. Counts for an Allowed Beta Spectrum Without Acceleration (Lower Curve), with 30-kv Preacceleration (Middle Curve), and Observed Counts Per Minutes (Points). Differences between latter two curves are due to focusing action of accelerating electrodes.

The lower curve is a plot of the count rate observed when the spectrum is taken with a spectrometer in which the momentum window is proportional to the momentum. Since the Fermi line is a graph of $\sqrt{N(p)/f}$, squaring this and multiplying by f gives $N(p)$. Multiplying $N(p)$ by the momentum gives the counts per minute which would be observed at that momentum.

The middle curve is a graph of the count rate observed when the same spectrum is taken with 30 kv preacceleration, assuming no focusing. This is calculated by multiplying $N(p)$ by

$$(7) \quad \frac{(I_p)^2}{I_p} \left[1 + \frac{\Delta E}{T + m_0 c^2} \right]^{-1}$$

from Eq. 5 in order to obtain c/m .

The upper curve shows how focusing by the electric field may further enhance the count rate. The points shown are experimental and represent data from a ring source having a 3-mm axial collimating aperture.

The focusing noted in Fig. 86 is not found until the added energy ΔE is an appreciable fraction of the initial energy T . For example, in Fig. 87 the K conversion line from Au^{198} is shown with various amounts of energy added. The line is displaced an amount proportional to the added

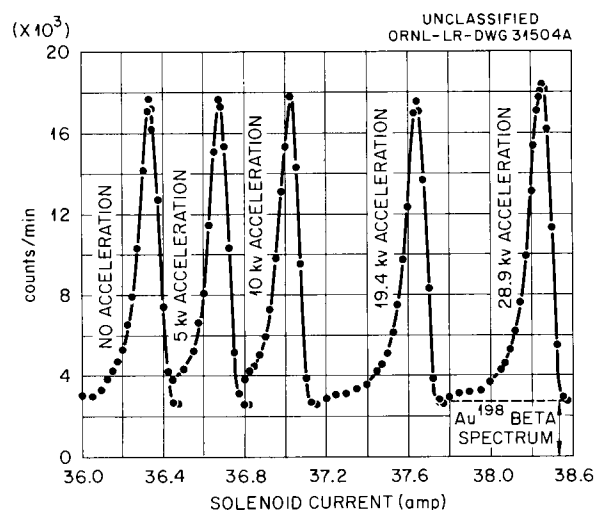


Fig. 87. Shift of the Au^{198} K-Conversion Line with Preacceleration Voltage.

energy, and its shape and size are seen not to be altered by a 10% increase in energy.

When relatively more energy is added, however, a definite increase in transmission is noted, even when the increase predicted by Eq. 5 is removed from the spectrum. This additional increase (denoted by F) is shown in Fig. 88 as a function of source energy T and for accelerating voltages of 10, 19.6, and 28.9 kv.

The trajectory for a particle in combined electric and magnetic fields is described by a well-known set of coupled differential equations. Unfortunately, they are not soluble in the situation presented here, which involves the superposition of a uniform magnetic field and an inverse-square electric field. Some insight into the behavior of an electron of initial energy T and accelerated by an amount of energy ΔE may be gained by contrasting its trajectory with that of an electron of initial energy $T + \Delta E$ which receives no additional energy. The motions differ principally in the direction which is normal to both the magnetic field and to the radial direction (in the cylindrical sense) from the magnet axis. It may be readily shown that the velocity gained in this direction is the same for the two electrons, and when they have reached the outer sphere is given by

$$(8) \quad \dot{z}(r_2) = \frac{eBr_2 \sin \theta}{m}$$

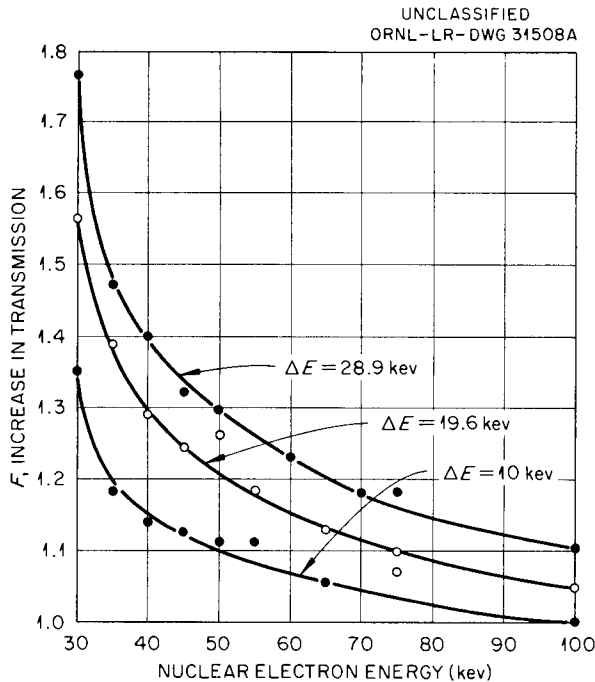


Fig. 88. Increase in Transmission Due to Focusing Action of the Preaccelerator for Applied Voltages of 10, 19.6, and 28.9 kev.

Here, θ is the takeoff angle at the source, measured from the spectrometer axis, and B is the magnetic field associated with the transmission of electrons of energy $T + \Delta E$. The displacements for the electrons in the z direction differ in general by some amount Δz which may be computed in the form of the ratio of Δz to the usual $z(r_2)$ from the equation

$$(9) \quad \frac{\Delta z}{z(r_2)} = \nu^2 \sqrt{1 + \frac{\Delta E}{T}} - 1 + 2 \sqrt{1 + \frac{\Delta E}{T}} \int_{\nu}^1 \frac{\mu d\mu}{\sqrt{\frac{\Delta E}{T} (\nu)^{-1} - (\mu)^{-1} + 1}}$$

where $\nu = r_1/r_2$, and $\mu = r/r_2$.

Although the integral in Eq. 9 may be solved exactly, the result is rather involved algebraically, and the evaluation may be more easily made by numerical integration. The result for various values of ν is shown in Fig. 89. It is seen that $\Delta z/z(r_2)$ increases as both $\Delta E/T$ and ν increase.

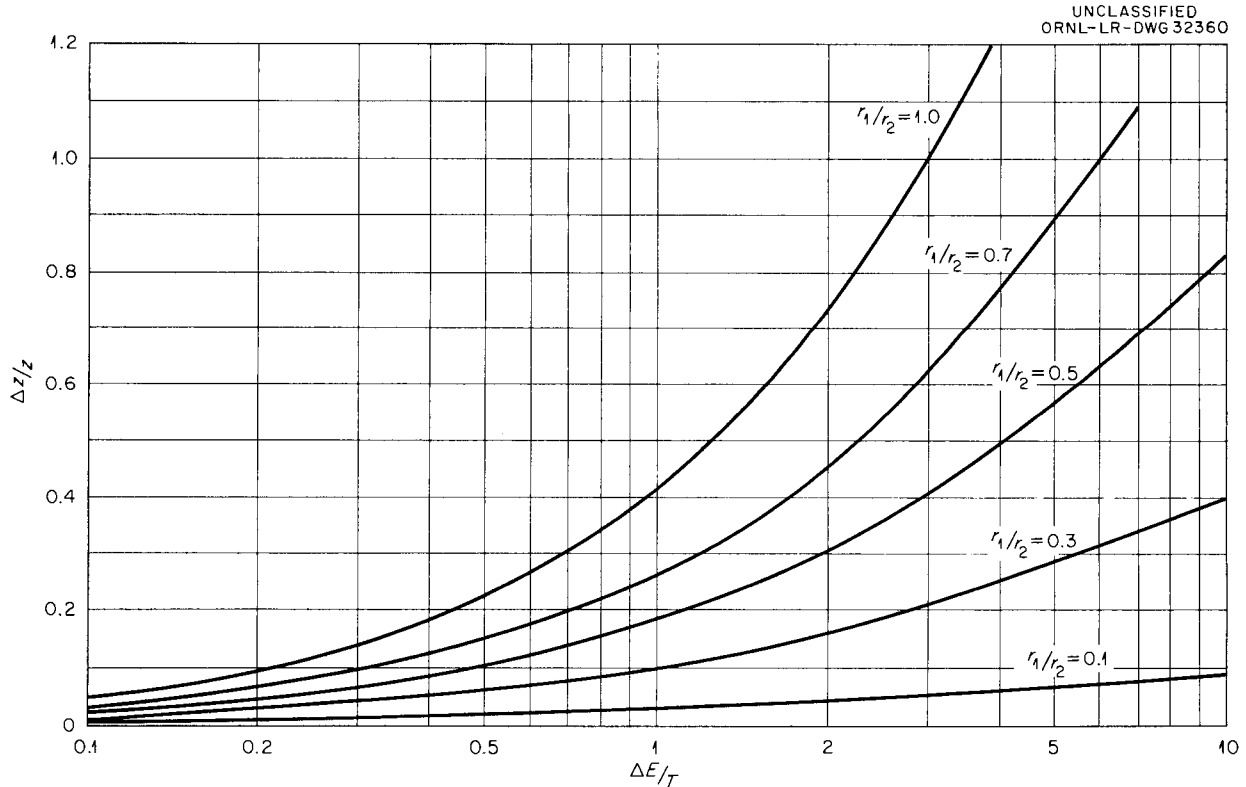


Fig. 89. Fractional Displacement of Electron Beam in Lateral Direction as a Function of Ratio of Accelerating Sphere Radii and Fractional Energy Added by Accelerator.

Although neither the magnitude nor the sign of the focusing effect may be predicted from this analysis, Eq. 9 may be useful in predicting the conditions under which a focusing action may be anticipated. For example, in Fig. 87 the focusing for $\nu = 0.5$ (used in this research) is seen to be negligible when $\Delta E/T \approx 0.02$. However, when $\Delta E/T \approx 1$ as in Fig. 88 and thus $\Delta z/z(r_2) \approx 0.2$, a definite focusing effect is noted.

It is apparent that focusing may be avoided by using a sufficiently small value of ν . This, however, causes a higher electric field at the surface of the inner sphere and may induce cold cathode emission. The value $\nu = 0.5$ was chosen here in order to minimize this field for a given applied voltage V and outer sphere radius r_2 . That is, if the electric field at $r = r_1$, and

$$(10) \quad E = \frac{V}{r_1(1 - r_1/r_2)}$$

is differentiated with respect to r_1 and the result equated to zero, r_1 is found to equal $r_2/2$. Apparently, a compromise value of ν will be necessary to maintain a moderate field at the surface of the inner sphere and, at the same time, to keep focusing effects small.

Finite Source Considerations for the Spherical Condenser Spectrometer

The resolution and transmission of the spherical condenser spectrometer (the "Keplertron") have been calculated theoretically for a point source on the axis of symmetry at the surface of the inner sphere, and the results have appeared in the literature.^{35,36} For this simple case the transmission T (defined as the fraction of all mono-energetic particles which leave the source and which succeed in reaching the detector) is given by

$$(1) \quad T = \frac{\chi}{4},$$

where χ is the supplement of the central angle through which the particle rotates while in the inverse-square electric field and is measured in radians. The energy resolution $\delta_{1/2}$ is defined

as the width of the line profile at half height divided by the energy and has the simple form

$$(2) \quad \delta_{1/2} = \frac{\chi^2}{16}.$$

It should be noted that the useful range of χ may vary from $\chi = 1$, where high transmission (25%) and relatively low energy resolution (6.3%) are desired, to a value of $\chi = 0.2$, where these figures are 5% and 0.25%, respectively. The relationship between transmission and resolution is independent of χ and may be obtained from the above equations:

$$(3) \quad T = \sqrt{\delta_{1/2}}.$$

The calculations described above have been extended³⁷ to include sources on the inner sphere but displaced slightly from the axis of symmetry. Such displacements are found to affect the transmission and resolution far less than displacements away from the inner sphere in the radial direction along the axis. The transmission for off-axis sources on the inner sphere was found to be expressible in terms of inverse cosine functions and incomplete elliptic integrals of the second kind. Line profiles were calculated for source displacements η/χ of 0.08, 0.16, 0.22, and 0.30, where η is the displacement around the sphere in units of the inner sphere radius. These line profiles were combined to obtain line shapes for disk sources. A typical example is shown in Fig. 90, where $4\pi T/\chi$ times the average transmission has been plotted for a point source ($\eta/\chi = 0$) and for a disk source whose radius in units of χ is 0.3. The abscissa is the fractional deviation in energy δ/χ^2 (in units of χ^2) from the energy for which the spectrometer is adjusted. It may be noted that if $\chi = 1$, the resolution has been reduced from 6.3 to 8.15%, and the transmission T from 25% ($4\pi T/\chi = 3.14$) to 18.4% ($4\pi T/\chi = 2.31$). However, the source may now be very large; for example, if the inner sphere were 20 cm in radius, the source would be 6 cm in radius. Such large sources make possible beta spectroscopy at low energy with sources of low specific activity and low surface density of activity.

³⁵R. D. Birkhoff *et al.*, *H-P Ann. Prog. Rep.* July 31, 1957, ORNL-2384, p 98.

³⁶R. D. Birkhoff, R. H. Ritchie, and J. S. Cheka, *Bull. Am. Phys. Soc.* 2, 174 (1957).

³⁷R. D. Birkhoff and R. H. Ritchie, *Bull. Am. Phys. Soc.* 3, 181 (1958).

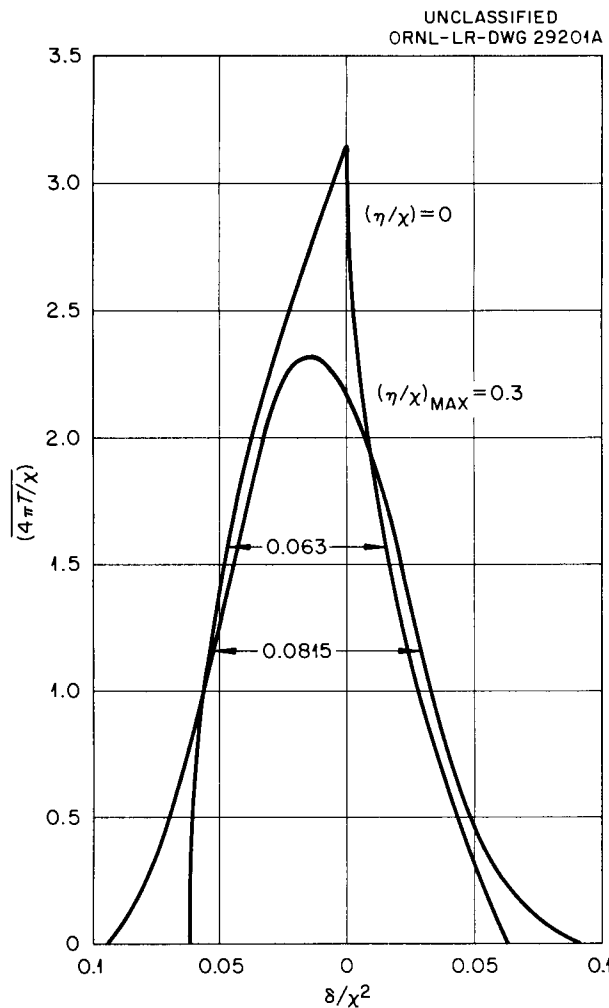


Fig. 90. Line Profile Shapes for Point Source, $(\eta/\chi) = 0$, and for Large-Disk Source, $(\eta/\chi)_{\max} 0.3$.

For a χ of 0.2 radian, the resolution with the disk source would drop from 0.25% to 0.33%, and the transmission from 5% to 3.7%. The source radius for a 20-cm inner sphere would now be 1.2 cm.

The transmission and resolution obtainable for disk sources of any radii $(\eta/\chi)_{\max}$ are shown in Fig. 91. Again, the average transmission T times $4\pi/\chi$ is shown. The energy resolution $(\delta/\chi^2)_{1/2}$ in units of χ^2 is shown on the right ordinate. Apparently, this spectrometer design possesses not only a very useful transmission and resolution, but a high luminosity (product of transmission and source area) as well.

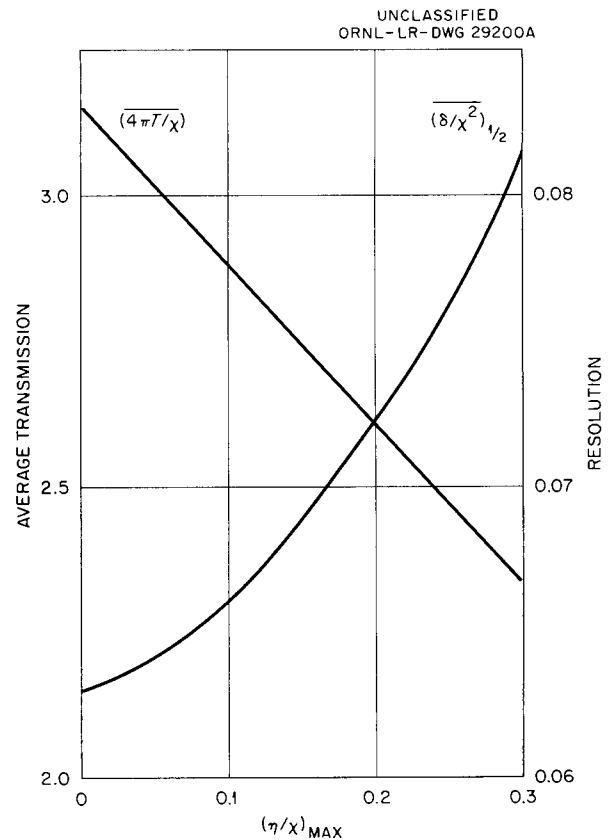


Fig. 91. Average Transmission and Resolution as a Function of Size of Disk Source.

Comparison of Theoretical and Filtered Experimental X-Ray Spectra

The satisfactory evaluation of x-ray exposures in both physical and biological experiments frequently requires a knowledge of the spectral composition of the x-ray beams used. In radiation chemistry and radiation damage work it is often necessary to know the energy delivered to a material as a function of depth. This can be calculated only if the spectral energy distribution in the beam is known. Similarly, this distribution must be known in order to determine the relative amounts of energy delivered to bone and tissue in biological work. In dosimetry, since x-ray dose is measured in roentgens, and the roentgen is defined in terms of air ionization, it is necessary to know whether the response of any dosimeter for all x-ray energies is the same as that of an air ionization chamber. Hence the calibration of

any dosimeter involves its exposure to x-ray beams, each containing only a narrow and accurately known range of energies. It was pointed out by Taylor³⁸ and others that neither the effective energy (or kilovolts) of an x-ray beam nor its peak kilovoltage and half-value layer are precise or even unique specifications of the energy distribution in the spectrum, so there is further need for spectral energy distribution measurements for many uses of x rays.

The x-ray facility in the Health Physics Division is frequently used for calibrating new dosimeters, so that an accurate knowledge of the energies and energy spreads in the x-ray beams available is essential. These quantities were determined in this work. However, the experimental methods for determining the spectral distributions in the beams and the technique for correcting the data are of general applicability. A more complete account of the work described here is available.³⁹

The shape of the spectrum to be expected may be calculated from the work of Kramers,⁴⁰ who in 1923 showed, on the basis of semiclassical assumptions, that the continuous bremsstrahlung spectrum from a thick target could be expressed in a form which may be written as

$$(1) \quad I(E) dE = -C(E - E_0) dE, \quad E < E_0, \\ = 0, \quad E > E_0,$$

where $I(E) dE$ is the intensity of the x rays per unit energy interval E , E_0 is the applied voltage, and C is a constant. The correctness of this expression had been demonstrated experimentally by Wagner and Kulenkampff⁴¹ the year before. Sommerfeld⁴² showed that essentially the same results followed from a wave-mechanical approach. By combining Kramers' result with the usual exponential absorption law and the mass absorption coefficients recently recalculated by Grodstein,⁴³

we can calculate the shape of the spectrum after transmission through the inherent filtration of the tube and any added filters:

$$(2) \quad I(E) dE \\ = -C(E - E_0) \prod_i \exp \left\{ - \left[\frac{\mu}{\rho}(E) \right]_i \rho_i x_i \right\}.$$

In this expression, $(\mu/\rho)(E)_i$ is the mass absorption coefficient of the i th filter of density ρ_i and thickness x_i .

The result is the well-known filtered x-ray spectrum which cuts off on the high-energy end at the Duane-Hunt limit and rises to a maximum at some lower energy. At energies below the maximum, the high absorption of lower-energy x rays in the filters may be made to reduce the intensity to negligible values as zero energy is approached.

As pointed out by Ehrlich⁴⁴ and by Ramm and Stein,⁴⁵ the simple Kramers' calculation makes no allowance for photon absorption or electron back-scattering in the x-ray target, nor for the characteristic x-ray emission lines of the target. However, these authors remark, and Greening⁴⁶ has confirmed, that the effects of these factors are small for the heavily filtered radiation used here. Theoretical spectra were therefore calculated from Eq. 2 for comparison with the experimental spectra.

Direct measurement of the spectra was made with a scintillation spectrometer consisting of an NaI(Tl) crystal, 3.6 cm in diameter by 2.62 cm high, covered on top with approximately 60 mg/cm² of MgO for light reflection and a 6-mg/cm² aluminum foil to seal out moisture. The voltage supply for the multiplier, the linear amplifier, and the differential pulse-height analyzer were conventional. The x rays were collimated by several lead diaphragms, so that the beam was parallel and the spot on the crystal was only 5 mm in diameter; hence, edge effects were entirely negligible. The crystal thickness was sufficient to ensure nearly complete absorption of the beam (94% at 200 kev and 99.5% at 150 kev). The scale

³⁸L. S. Taylor, *Radiology* 29, 22 (1937).

³⁹J. C. Villforth, R. D. Birkhoff, and H. H. Hubbell, Jr., *Comparison of Theoretical and Experimental Filtered X-Ray Spectra*, ORNL-2529 (July 1, 1958).

⁴⁰H. A. Kramers, *Phil. Mag.* 46, 836 (1923).

⁴¹E. Wagner and H. Kulenkampff, *Ann. Physik* [4] 68, 369 (1922).

⁴²A. Sommerfeld, *Proc. Natl. Acad. Sci. U.S.* 15, 393 (1929); *Ann. Physik* [5] 11, 257 (1931).

⁴³G. W. Grodstein, *Natl. Bur. Standards Circ. No.* 583 (1957).

⁴⁴M. Ehrlich, *J. Research Natl. Bur. Standards* 54, 107 (1955); *Radiation Research* 3, 223 (1955).

⁴⁵W. J. Ramm and M. N. Stein, *Phys. Rev.* 92, 1081 (1953).

⁴⁶J. R. Greening, *Brit. J. Radiol.* 20, 71 (1947); 21, 75 (1948); *Proc. Phys. Soc. (London)* A63, 1227 (1950).

of pulse heights, H , was calibrated by the use of gamma and x rays from known radioisotopes and characteristic x rays from a fluorescent radiator. Such a calibration is shown in Fig. 92. The large number of digits given in the least-squares equations is carried for computational consistency and has no physical significance.

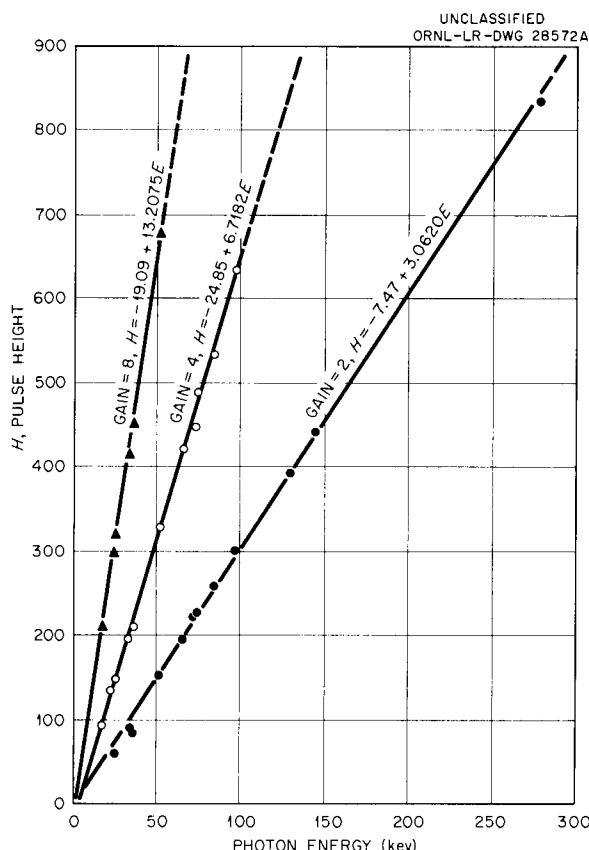


Fig. 92. Calibration Curves for the Scintillation Spectrometer.

The fundamental difficulty of poor resolution arises from the small number of photoelectrons ejected from the photocathode. The average number of these is accurately proportional to the energy of the photon incident on the crystal, but for successive absorptions of monoenergetic photons the number of photoelectrons in each pulse is statistically distributed about the average number. Thus the spectrometer takes a spectrum line and spreads it out into a distribution about the original energy.

Let $L(E, E')$ be the resolution curve of the spectrometer, that is, the fraction of the number of photons of energy E' hitting the crystal which are counted at a setting E . A normalized Gaussian distribution was used for L to take account of the fact that the resolution varies with $E^{-1/2}$:

$$(3) \quad L(E, E') = \frac{1}{a\sqrt{\pi}} \exp \left(-\frac{(E - E')^2}{a^2} \right),$$

where $a^2 = CE$.

The values of the width constant a were determined from measured pulse-height spectra of sharp gamma- and x-ray lines. To show how this is done, Eq. 3 may be rewritten in the form

$$(4) \quad \log L(E, E') = \log \frac{1}{a\sqrt{\pi}} - (\log_{10} e) \left(\frac{E - E'}{a} \right)^2.$$

For a fixed incident energy E' , if $\Delta E = E - E'$, the difference between the incident photon energy and the energy for which the analyzer is set, Eq. 4 becomes

$$(5) \quad \log L = C - D \left(\frac{\Delta E}{a} \right)^2.$$

Since the pulse height H is directly proportional to the energy E ,

$$(6) \quad \log L = C - F \left(\frac{\Delta H}{a} \right)^2.$$

The number of counts per minute, which is proportional to the function L , may then be plotted vs $(\Delta H)^2$ on semilog paper as shown in Fig. 93, and a^2 may be determined from the slope by a least-squares analysis. The relation between a^2 and energy is linear, as shown in Fig. 94.

Only four readily available radioactive isotopes were found having single gamma rays in the region of interest. The K x-ray peaks gave additional points for the curves for pulse height vs energy and resolution and for a^2 vs energy. Additional points were obtained by using the fluorescent K x rays from elements irradiated with x rays from a commercial machine.

The K series of x rays consists of four principal lines: the K_{α_1} , the K_{α_2} doublet, and, somewhat higher in energy, the K_{β_1} and the K_{β_2} . If the

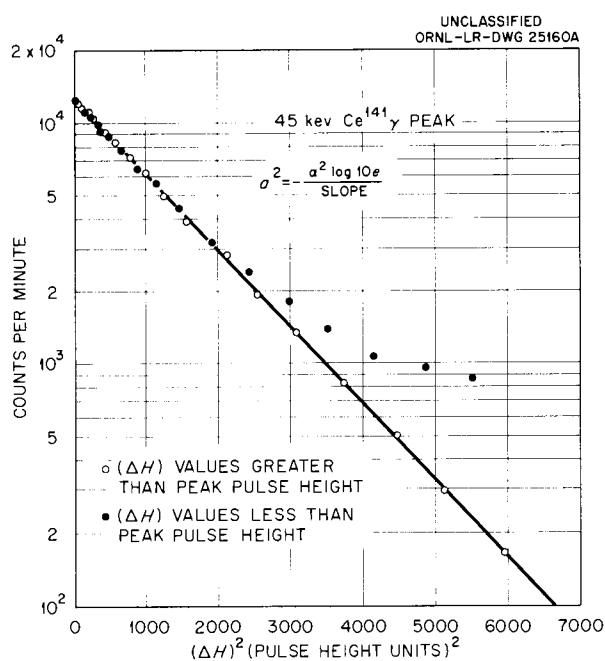


Fig. 93. Gaussian Representation of Data from a Scintillation Spectrometer for a Monoenergetic Gamma Line.

expected response curves for these lines are plotted with the relative intensities as given by Compton and Allison, the overlap shown in Fig. 95 is obtained. The individual lines as seen by the spectrometer are shown; above them is shown the result of adding the intensities of the two α doublet components, and at the top is the still broader curve due to the sum of the intensities of all four lines. If a curve for log count rate vs square of deviation is plotted as discussed above, the slope would be too flat because the curve is too broad, and the resolution constant calculated from this would be too small. This error was corrected in two ways. For the x rays from radioactive isotopes, the intensity is low; so it is not feasible to try to filter out some of the lines. Hence a correction factor was calculated by making plots similar to those in Fig. 95, and from them, points were taken for the plot of log count rate vs square of deviation. The correction factor is the ratio of the calculated breadth of a monoenergetic line to that of the computed composite line, and a plot of these factors vs energy of the line is given in Fig. 96. Multiplying this factor by the observed width

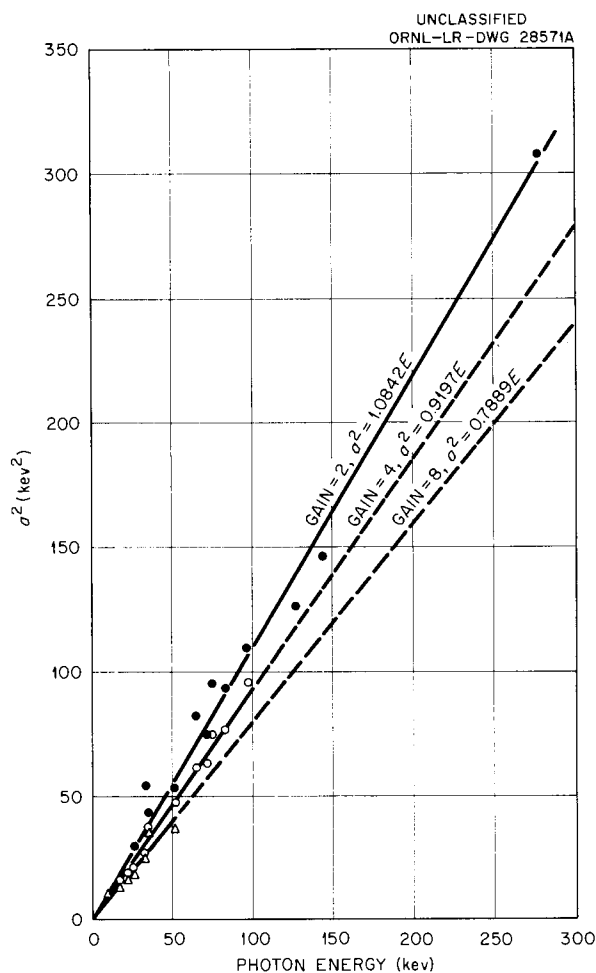


Fig. 94. Resolution Constant vs Energy for a Scintillation Spectrometer. Different window widths were used for the three gain settings, giving three curves instead of one.

parameter a^2 of a K series x-ray peak gives the expected breadth parameter for a monoenergetic line at this energy. In addition, it may be noted that whenever an unknown line is observed whose width parameter as seen by the spectrometer is greater than the predicted one, we may be sure that the line is actually multiple, even though the components may be far too close together to be resolved by the instrument.

The sharp break in the mass absorption coefficient curve at the K absorption edge permits experimental narrowing of the K series x-ray peak. The K beta lines may lie just above the absorption edge of a properly chosen filtering element

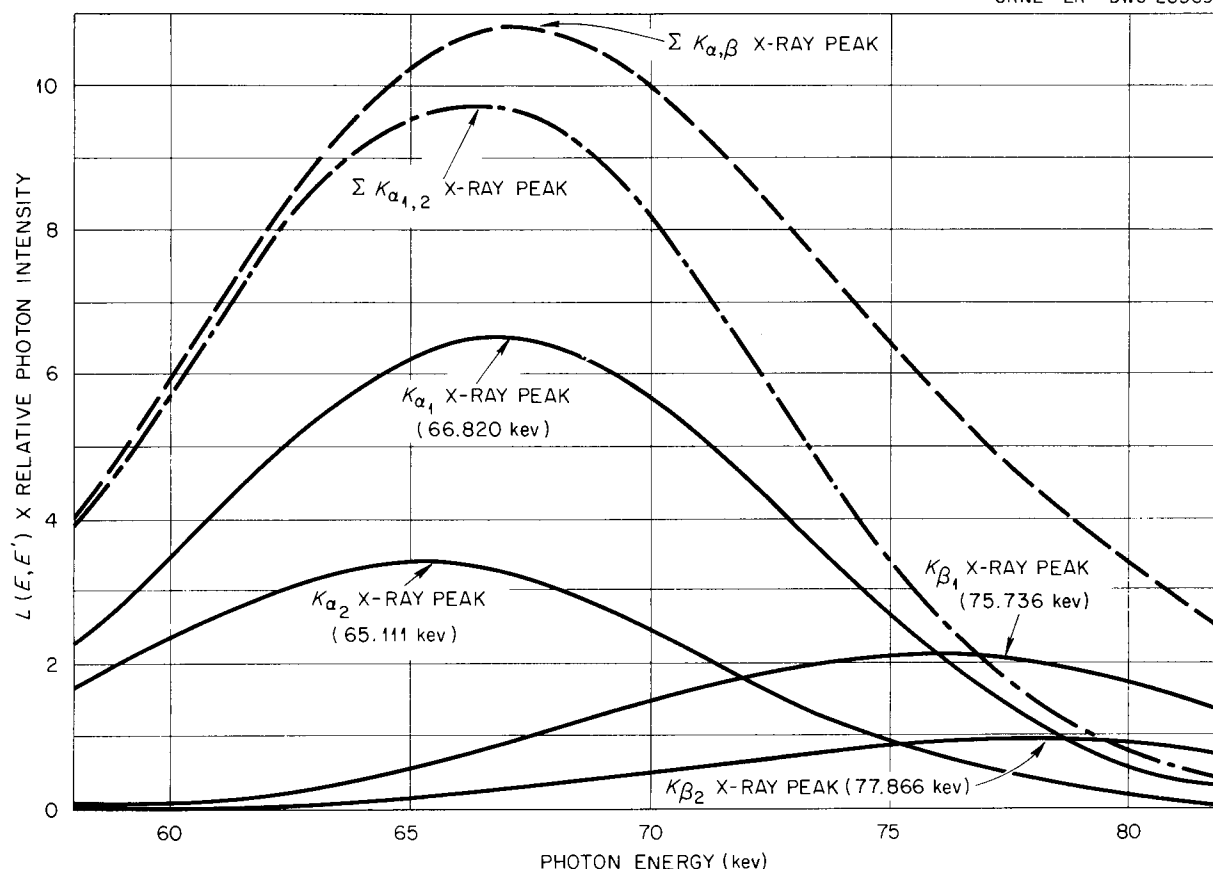
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Fig. 95. Calculated Response Curves of the Scintillation Spectrometer to the Various Series-K Lines of Platinum, and to Combinations of the Lines.

and be strongly absorbed, while the K alpha lines lie below and are only slightly absorbed.

Figure 94 shows a plot of the resolution parameter a^2 as a function of energy. The points were obtained by all three methods: from gamma rays, from corrected breadths of K series peaks of isotopic x rays, and from filtered K x rays. The three lines are for three different gain settings of the amplifier, for which the relative window breadth of the analyzer varied. Table 40 shows the sources used to obtain the two calibration curves of pulse height vs energy and of resolution parameter vs energy. These techniques can considerably extend the usefulness of the scintillation spectrometer both for measurements in the low-energy region and for the detection of multiplicity in unresolved spectral lines.

The correction of the bremsstrahlung spectra for spectrometer resolution has been made⁴⁴⁻⁴⁷ and

the method of matrix inversion was used, but such a procedure requires the use of a large digital computer and an appropriate code to handle the problem. We have used a simpler approximate method suitable for use with a desk calculator — a method which was suggested by Freedman *et al.*⁴⁸ As these authors pointed out, the measured spectrum $M(E)$ is related to the true spectrum $N(E)$ by the equation

$$(7) \quad M(E) = \int_0^{E'_{\max}} N(E') L(E, E') dE',$$

or

$$(8) \quad M(E) \cong \sum_i N(E'_i) L(E, E'_i) \Delta E'_i.$$

⁴⁷L. A. Beach, R. B. Theus, and W. R. Faust, NRL-4277 (Dec. 1, 1953).

⁴⁸M. S. Freedman *et al.*, *Rev. Sci. Instr.* **27**, 716 (1956).

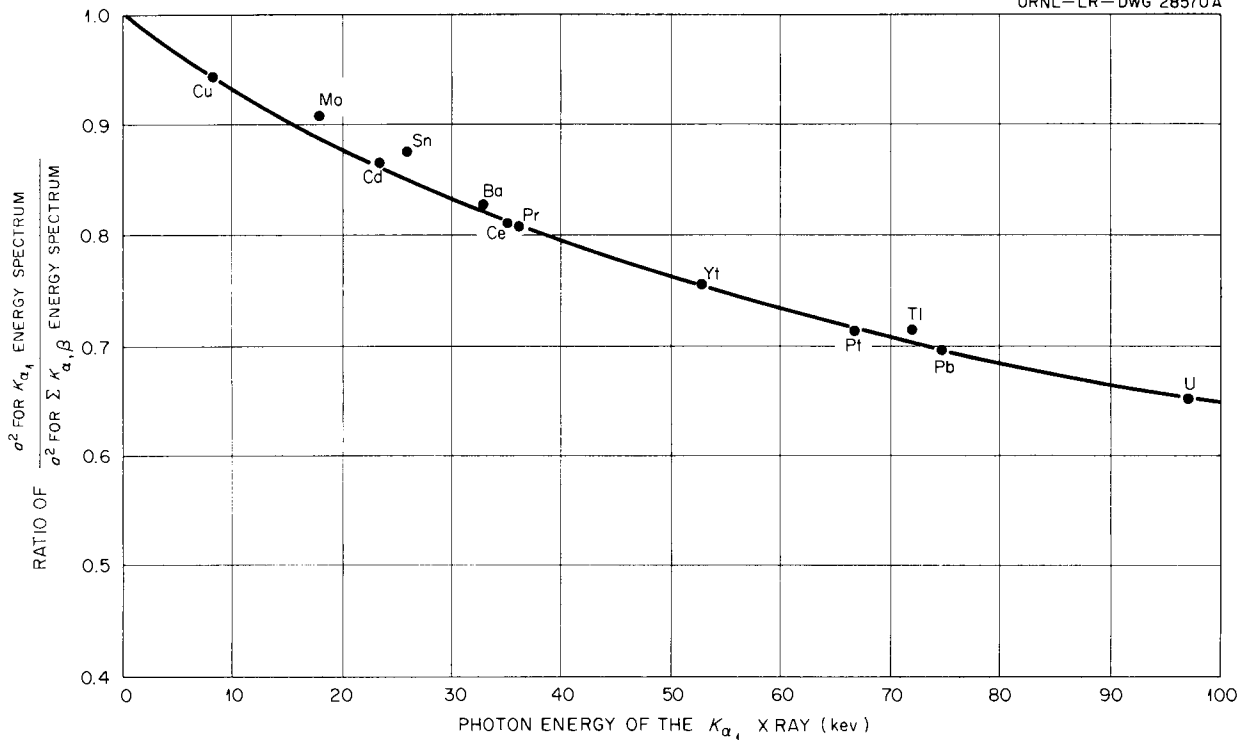
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Fig. 96. Correction Factor To Give the Resolution Constant (σ^2) of the Scintillation Spectrometer for a Monoenergetic X-Ray Line from That Observed for the Whole K-Series.

In practice the integral equation (7) is solved by an iterative method as suggested in the reference. The measured spectrum $M(E)$ is used in order to start the iterative procedure, and a new spectrum $M_1(E)$ is generated:

$$(9) \quad M_1(E) = \sum_i M(E_i) L(E, E'_i) \Delta E_i.$$

This new spectrum will, in general, be even more distorted by the Gaussian smearing function than the measured spectrum will be. As a first approximation the true spectrum $N_1(E)$ may be assumed to differ from the measured spectrum $M(E)$ by the same amount as $M_1(E)$ differs from $M(E)$ and in the opposite direction; that is, if $M_1(E) > M(E)$, then $N_1(E) < M(E)$. Thus

$$(10) \quad \begin{aligned} N_1(E) &= M(E) - [M_1(E) - M(E)] \\ &= 2M(E) - M_1(E). \end{aligned}$$

The approximate spectrum $N_1(E)$ is then substituted into Eq. 8, and a new smeared spectrum

$M_2(E)$ is generated. This new spectrum is used as above to generate a second approximation to the true spectrum. In general,

$$(11) \quad N_i = N_{i-1} - (M_i - M).$$

In the problems considered by Freedman *et al.* and in those considered here, the convergence was rapid, with only a few iterations being required for determining a spectrum $N(E)$ which, when substituted into Eq. 8, yielded a spectrum differing by 5% or less from the measured spectrum at all energies.

Figure 97 shows the original measured spectrum $M(E)$ and the spectra resulting from the first few iterative approximations.

After the corrections for the Gaussian distribution had been made by the iteration method, the spectrum was still distorted by (1) the iodine escape peak, which is prominent at low energies, and (2) the finite probability that an x ray may pass through the crystal without interacting, which is important at the high energies. The distortion

Table 40. Sources Used for Calibrating the Scintillation Spectrometer

Gamma Rays	K X Rays from Radioactive Isotopes*	Fluorescent K X Rays		Energy (kev)
		Radiator	Filter	
		Cu		8.07
		Mo		17.75
		Cd		23.36
		Sn		25.60
		Ba		32.65
		Ce	Ba	34.6
	Pr			36.2
	Yb			52.6
		Pt	W	66.2
	Tl			72.3
		Pb	Hg	74.3
Tm^{170}		U	Th	97.2
Os^{191}				125.9
Ce^{141}				145
Hg^{203}				279

* Daughter element.

caused by the escape of the characteristic K radiation of iodine from the crystal was removed from the iterated experimental spectrum by calculating the number of pulses gained and lost at each energy by this process. If $P(E)$ is the true number of photons of energy E striking the crystal, and $N(E)$ is the number of pulses observed corresponding to energy E , then

$$(12) \quad N(E) = P(E) - F(E) N(E) + F(E + K) P(E + K),$$

where $F(E)$ is the "escape ratio" calculated by Axel,⁴⁹ the ratio of the number of iodine K x-ray photons leaving the crystal to the number of incident photons of energy E ; K is the average energy of the iodine K x rays, 28.4 kev. Equation 12 may be solved for $P(E)$ under the assumption

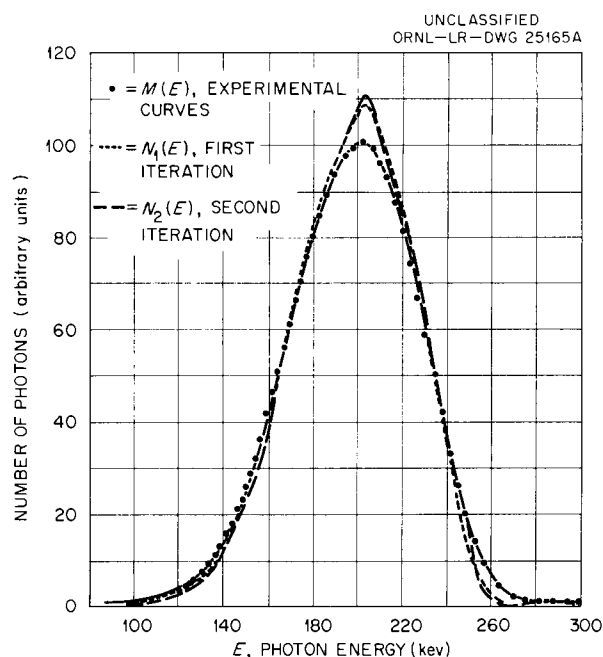


Fig. 97. The Effect of the First and Second Iterations on the Experimental Data for the 250-kev X-Ray Spectrum.

that $F(E + K) P(E + K) \ll N(E + K)$, which gives

$$(13) \quad P(E) = \frac{N(E) - \frac{F(E + K) \cdot N(E + K)}{1 - F(E + K)}}{1 - F(E)}.$$

Axel's ratios were recalculated according to more recent values⁵⁰ for the absorption coefficients of sodium iodide, the fluorescent yield of iodine, and the probability that a photoelectric interaction with iodine will eject a K electron.

To correct for the incomplete absorption of the x rays in the crystal, the corrected $P(E)$ is divided by the absorption of the crystal, which is calculated from its thickness and Grodstein's coefficients.

The Compton continuum has been neglected, since it is only prominent at energies greater than those used in this experiment. In the experimental 250-kev spectrum, it was only 2% of the primary photoelectric peak.

Figures 98 through 101 show the comparison of the theoretical with the corrected experimental spectra calculated by the method just outlined for

⁴⁹P. Axel, *Rev. Sci. Instr.* 25, 391 (1954).

⁵⁰G. W. Grodstein, *Natl. Bur. Standards Circ. No. 583* (1957).

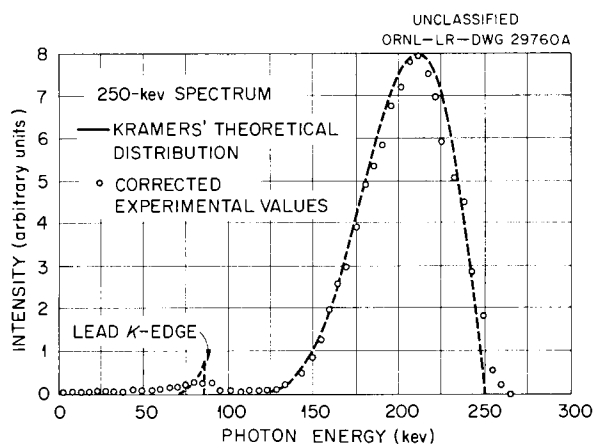


Fig. 98. Comparison of Corrected Experimental Values with the Theoretical Kramers' Distribution; 250-kev Spectrum.

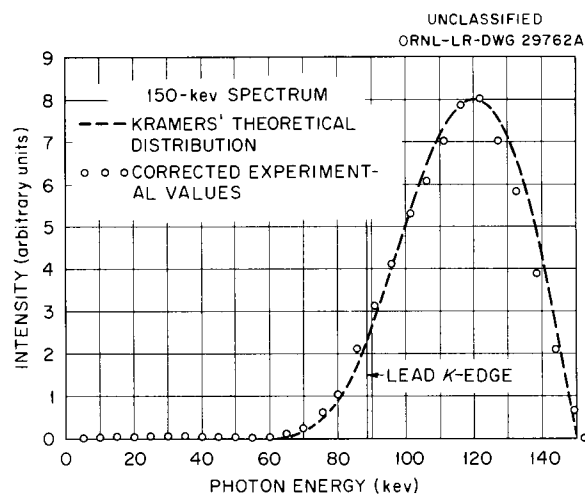


Fig. 100. Comparison of Corrected Experimental Values with the Theoretical Kramers' Distribution; 150-kev Spectrum.

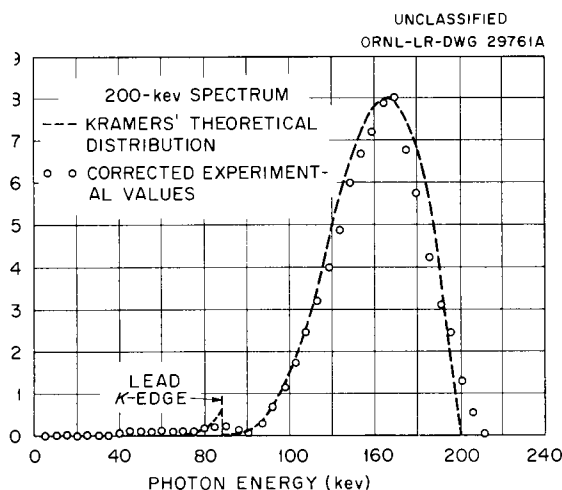


Fig. 99. Comparison of Corrected Experimental Values with the Theoretical Kramers' Distribution; 200-kev Spectrum.

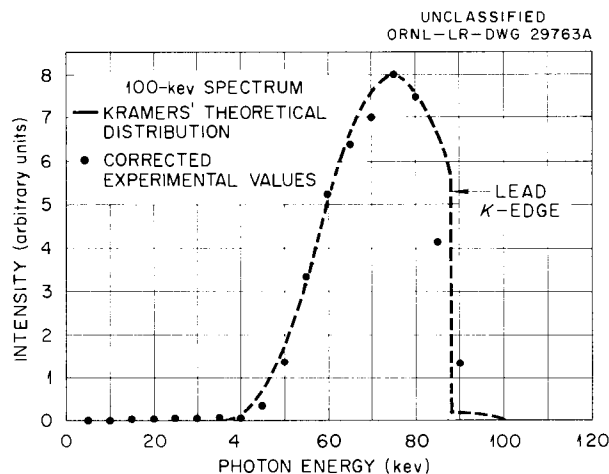


Fig. 101. Comparison of Corrected Experimental Values with the Theoretical Kramers' Distribution; 100-kev Spectrum.

250-, 200-, 150-, and 100-kv spectra excited by the constant-potential machine. The most probable and the average energies for each spectrum were obtained as well.

As a comparison with these values, the effective energy of each spectrum was obtained by the absorption method. The effective energy is defined as the energy of a monochromatic x-ray beam which would have the same absorption coefficient as the given spectrum, for an incremental thickness of standard absorber. That is, the

absorption coefficient of the given spectrum is calculated from the slope of the tangent to the curve of log intensity vs filter thickness, with the tangent being drawn at the point corresponding to the filter thickness which was used as a component of the total filter.

The results at 250-kv constant potential may be considered as typical. Absorption runs were made with tin and copper filters, with the results shown in Fig. 102. An air ionization chamber of Bureau of Standards design was used as a detector. The

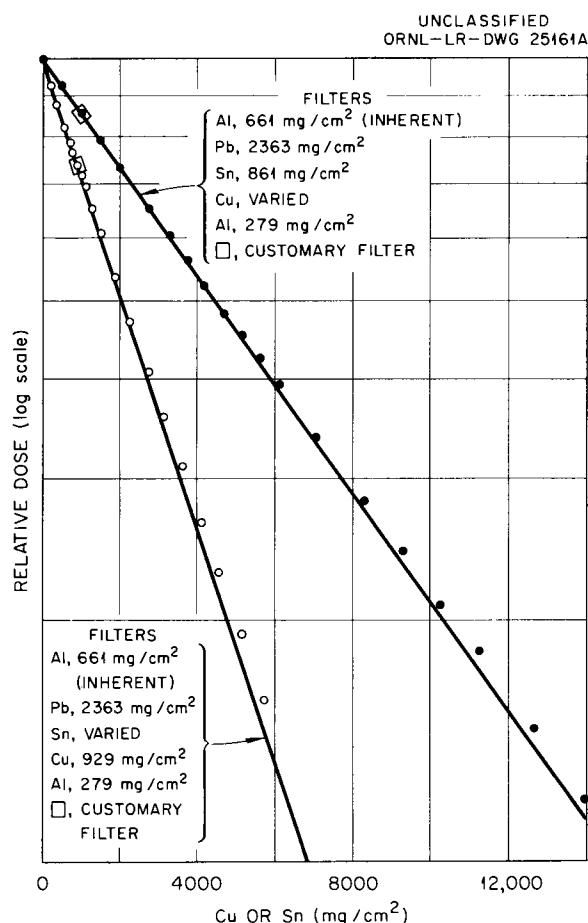


Fig. 102. Constant Potential (250 kv) Absorption Curves Obtained by Varying the Thickness of Cu or Sn on the Customary Filter. The filters were always placed on the x-ray beam in the order shown with the Pb closest to the target.

filters were analyzed chemically for higher Z elements, and none were found in sufficient percentage to affect the absorption curves. The effective energies were calculated by least squares from the data for the tin and copper filters.

The absorption curves are somewhat affected by the backing material for the detector. Additional absorption data were, therefore, taken with a Victoreen condenser r-meter as the detector, backed by a stack of half-inch-thick sheets of Lucite, Al, Cu, Sn, and Pb, in that order, and by various combinations of these. An additional run was made with a narrow x-ray beam directed out an open window in the laboratory, so that the x-ray beam struck nothing but detector and air.

The effective values from these curves differed by about 5% among themselves.

The filters used⁵¹ (similar to those used by Ehrlich and Fitch) are given in Table 41, and the final calibration data for the x-ray beams are shown in Table 42. The estimates of the average and the effective x-ray energy for a given setting fall within a range of about 3%.

Measurement of Electron Flux in Media Bombarded by X Rays

Although the ionization per unit volume and the energy deposited per unit mass by radiation are useful indices of radiation exposure, it is well known that a more detailed knowledge of the interaction of the radiation with the absorber is required before biological damage may be estimated. In fact, the differences in biological damage found for the same energy deposition from the different radiations has led to the introduction of a factor called the "relative biological effectiveness," which is characteristic of each radiation. Platzman⁵² has pointed out that a study of the low-energy electrons liberated in a medium by the passage of ionizing radiation may be an important preliminary to a more thorough understanding of the mechanism of biological damage.

McKay,⁵³ in his review article on secondary emission, has mentioned some of the many experimental difficulties found in the application of conventional electron spectrometers to low-energy spectral measurements. In order to avoid these difficulties, a stopping-potential type of energy analysis was applied to the electrons ejected by x rays from the inner surfaces of two parallel plates. The region between the plates was kept at high vacuum and constituted a cavity in an effectively infinite medium which made the experimental situation especially amenable to theoretical interpretation.

The experiment consisted in measuring the current which flowed in the vacuum chamber as a function of the voltage applied to the chamber. It became apparent that the chamber was, in fact, a spectrometer and that the spectral distribution of the electron flux could be obtained from the

⁵¹See M. Ehrlich and S. H. Fitch, *Nucleonics* 9(3), 5 (1951).

⁵²R. L. Platzman, *Radiation Research* 2, 1 (1955).

⁵³K. G. McKay, *Advances in Electronics and Electron Phys.* 1, 65 (1948).

Table 41. Health Physics 250-kv Constant-Potential X-Ray Machine

Kilovoltage Setting	Inherent Filtration Equivalent*		Added Filters							
			Pb		Sn		Cu		Al	
	mm	mg/cm ²	mm	mg/cm ²	mm	mg/cm ²	mm	mg/cm ²	mm	mg/cm ²
250	2.45	661	2.08	2363	1.18	861	1.07	959	1.03	279
200	2.45	661	0.63	716	2.55	1863	1.04	929	1.03	279
150	2.45	661			1.19	913	3.12	2790	1.03	279
100	2.45	661	0.48	548					1.03	279

*Determined from absorption curves with no added filtration; data given by Glasser *et al.*, *Physical Foundations of Radiology*, 2d ed. p 245-49, Hoeber, New York, 1952.

Table 42. Comparison of Average, Most Probable, and Effective Energies

Approximate X-Ray Tube Voltage (kv)	Average Energy (kev)		Most Probable Energy (kev)		Effective Energy of Absorbers (kev)				Profile Width* (kev)	
	Corrected Experimental Spectrum	Kramers' Theoretical Distribution	Corrected Experimental Spectrum	Kramers' Theoretical Distribution	Pb	Sn	Cu	Al	Corrected Experimental Spectrum	Kramers' Theoretical Distribution
100	70.5	72.3	75	75			69.0	68.0	14.2	15.3
150	115.2	116.4	123	120			115.5		21.4	22.5
200	159.7	159.5	170	165			158.7		24.7	27.4
250	201.5	201.6	212	215	194.8	196.5	200.9		30.3	32

*Expressed as half width at half-maximum intensity.

current-voltage curve if the angular distribution from the plate surfaces were known. The experimental situation is shown in Fig. 103. It is apparent that all electrons which are ejected from the upper negative plate will reach the positive plate, while, on the contrary, a fraction f of those emitted from the lower plate which have an energy V' exceeding the potential V across the plates will fail to traverse the cavity. Of course, all electrons with $V' < V$ also will fail to cross from the lower to the upper plate. Those electrons which fail to reach the upper plate will return to the lower plate and thus constitute no current in the external circuit. However, their counterparts from the upper plate will still make the crossing, and being uncompensated by electrons crossing from the lower plate will produce an

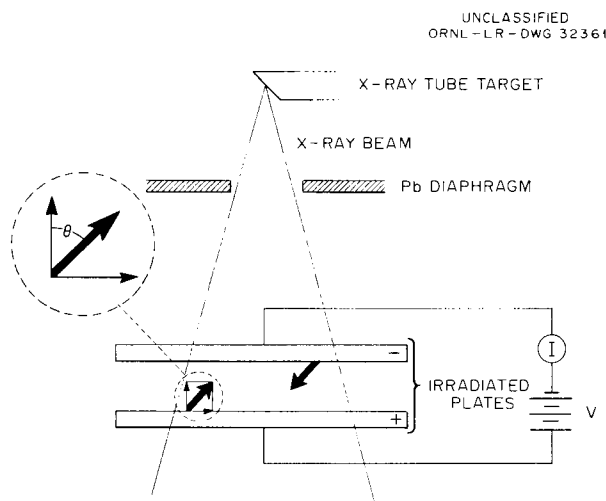


Fig. 103. Schematic of Apparatus for Determining the Flux of Electrons in Irradiated Matter.

external current I , given by the following equation:

$$(1) \quad I = eAR \left[\int_0^V N(V') dV' + \int_V^\infty f(V, V') N(V') dV' \right],$$

where e is the electronic charge in esu, A is the plate area in cm^2 , R is the dose rate in roentgens per second, and $N(V')$ is the flux of electrons emerging from either plate with an energy between V' and $V' + dV'$ in $\text{cm}^2 \cdot \text{r}^{-1} \cdot \text{v}^{-1}$. Clearly, if the electrons are all emitted normal to the plates, $f(V, V') = 0$. If the angular distribution is isotropic,

$$(2) \quad f = \alpha \int_{\cos^{-1}\sqrt{V/V'}}^{\pi/2} 2\pi \sin \theta d\theta = 2\pi\alpha \frac{\sqrt{V}}{\sqrt{V'}} = \frac{\sqrt{V}}{\sqrt{V'}},$$

where α is a normalization constant and is equal to $(2\pi)^{-1}$ for an angular distribution normalized to unity in the 2π solid angle outside the plate.

If the angular distribution follows a cosine law,

$$(3) \quad f = \alpha \int_{\cos^{-1}\sqrt{V/V'}}^{\pi/2} 2\pi \cos \theta \sin \theta d\theta = \pi\alpha \frac{V}{V'} = \frac{V}{V'},$$

where α is again a normalization constant which is now equal to $(\pi)^{-1}$. The factor $f(V, V')$ may now be represented by $(V/V')^n$, where n takes on the values ∞ , $1/2$, and 1 for the three cases mentioned above. Thus

$$(4) \quad I = eAR \left[\int_0^V N(V') dV' + \int_V^\infty \left(\frac{V}{V'} \right)^n N(V') dV' \right],$$

and this equation may be differentiated with respect to V to yield

$$\frac{dI}{dV} = eAR \left[N(V) + nV^{n-1} \int_V^\infty (V')^{-n} N(V') dV' - \frac{V^n N(V)}{V^n} \right],$$

$$\frac{d^2I}{dV^2} = eAR \left[n(n-1) V^{n-2} \int_V^\infty (V')^{-n} N(V') dV' - nV^{n-1} \frac{N(V)}{V^n} \right].$$

The solution for $N(V)$ is then

$$(5) \quad N(V) = \frac{1}{eAR} \left[\left(\frac{n-1}{n} \right) \frac{dI}{dV} - \frac{V}{n} \frac{d^2I}{dV^2} \right].$$

For straight-ahead emission, $n = \infty$ and

$$(6) \quad N(V) = \frac{1}{eAR} \frac{dI}{dV}.$$

For the isotropic case, $n = 1/2$ and

$$(7) \quad N(V) = \frac{1}{eAR} \left[-\frac{dI}{dV} - 2V \frac{d^2I}{dV^2} \right].$$

Finally, the cosine angular distribution with $n = 1$ yields

$$(8) \quad N(V) = \frac{1}{eAR} \left[-V \frac{d^2I}{dV^2} \right].$$

The differences in these results indicate that considerable care must be used in specifying the angular distribution before an evaluation of the current-voltage curve is made. In order to avoid having to assume any of the simplified distributions, the problem may be altered slightly. The distribution inside the plates may be assumed to be isotropic, and the electrons may be followed as they progress through the surface barrier and cross the cavity. If all kinetic energies are measured from the bottom of the conduction band, then the flux of electrons in the material (as opposed to those leaving the surface) which have a kinetic energy E , equal to the sum of the Fermi energy, the photoelectric work-function, and the applied potential V , may be shown⁵⁴ to be

$$(9) \quad N(E_F + \phi + V) = \frac{1}{eAR} \left[-\frac{dI}{dV} - 2(E_F + \phi + V) \frac{d^2I}{dV^2} \right].$$

⁵⁴D. L. Nelson et al., *Measurement of Electron Flux in Media Bombarded by X-Rays*, ORNL-2521 (July 25, 1958).

A typical current-voltage curve is shown in Fig. 104 for chemically cleaned, hydrogen-annealed plates. It is apparent that the flux is predominantly in the low-energy region. The first- and second-derivative curves for these plates are given in Figs. 105 and 106. The yield, obtained from the saturation portion of the first curve, was $0.0063 \text{ esu} \cdot \text{cm}^{-2} \cdot \text{r}^{-1}$, and for aluminum and graphite plates, 0.0011 and 0.00024, respectively.

The energy distribution of the flux obtained from Figs. 105 and 106 and from Eq. 9 is shown in Fig. 107. The line represents an attempt to fit

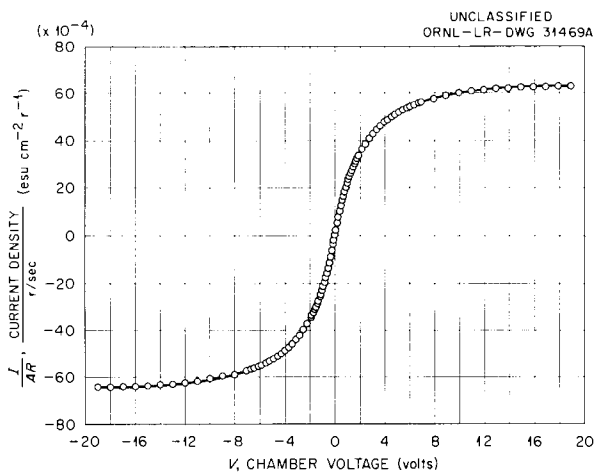


Fig. 104. Current-Voltage Curve for Hydrogen-Annealed Copper Plates.

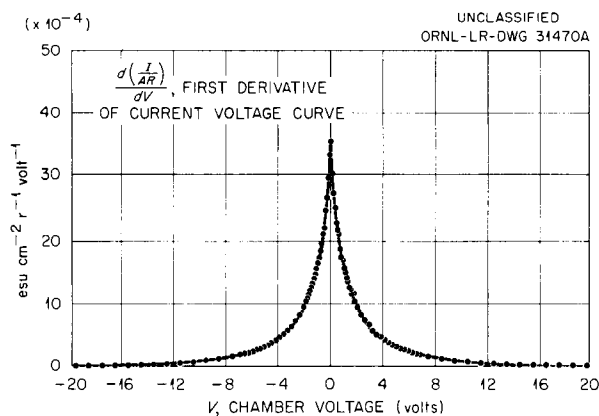


Fig. 105. First Derivative of the Curve of Fig. 104.

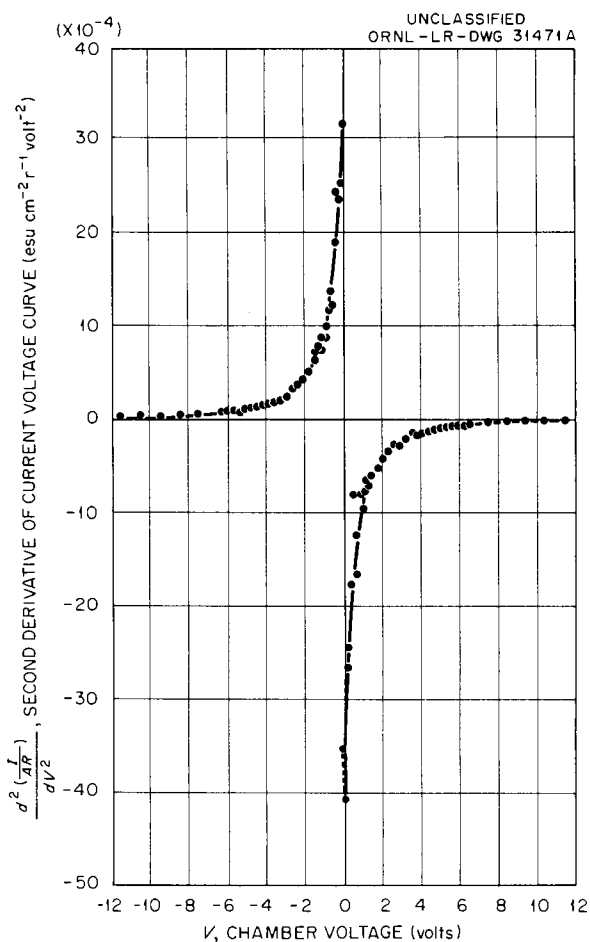


Fig. 106. Second Derivative of the Curve of Fig. 104.

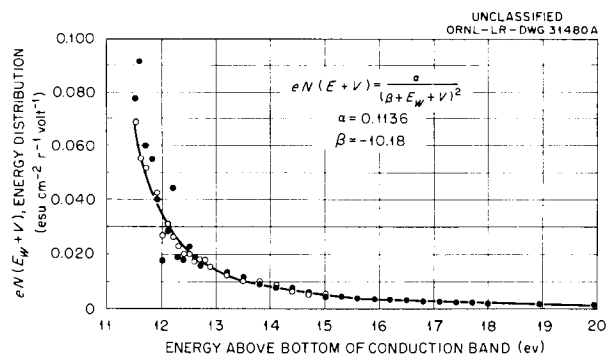


Fig. 107. Energy Distribution of Electron Flux in Irradiated Copper. Points are experimental; curve is a plot of the function shown, with empirical parameters α and β .

the data with a function of the form

$$(10) \quad eN(E_w + V) = \frac{\alpha}{(\beta + E_w + V)^2} \text{ esu} \cdot \text{cm}^{-2} \cdot \text{r}^{-1} \cdot \text{v}^{-1},$$

where $E_w = E_F + \phi$. Spectra for aluminum and graphite are shown in Figs. 108 and 109 with the constants α and β , which give a fit to Eq. 10. The empirical parameter β is about equal to $1 - E_w$ for all three materials studied. Thus the

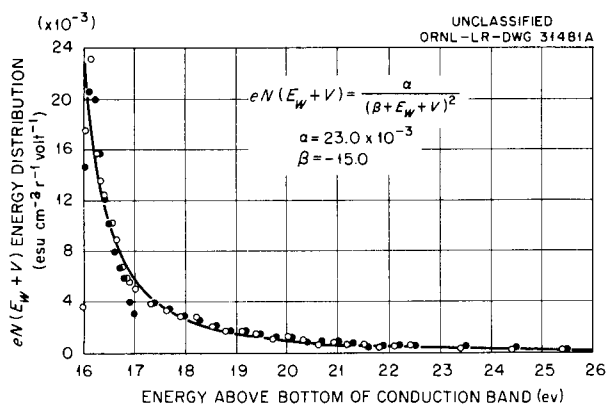


Fig. 108. Energy Distribution of Electron Flux in Irradiated Aluminum.

flux of electrons of energy between E and $E + dE$ is given by

$$(11) \quad \phi(E) = \frac{\alpha}{[E - (E_w - 1)]^2} \text{ esu} \cdot \text{cm}^{-2} \cdot \text{r}^{-1} \cdot \text{v}^{-1},$$

where α has the values 0.11, 0.023, and 0.0021 $\text{esu} \cdot \text{v} \cdot \text{cm}^{-2} \cdot \text{r}^{-1}$ for copper, aluminum, and graphite, respectively.

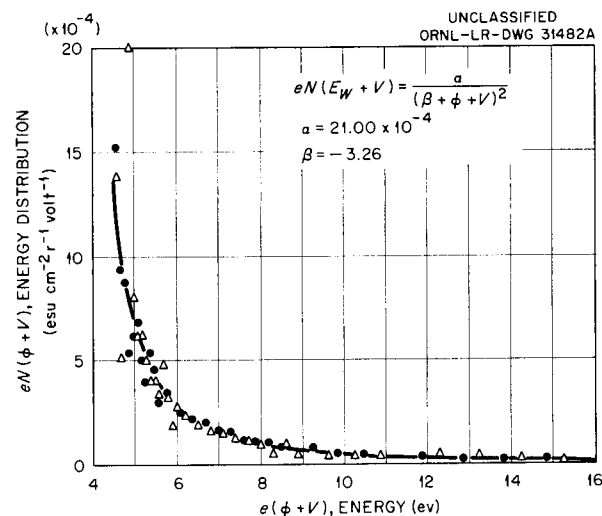


Fig. 109. Energy Distribution of Electron Flux in Irradiated Graphite.

INTERNAL DOSIMETRY

E. G. Struxness

W. S. Snyder

B. R. Fish

APPLIED RADIOBIOLOGY

B. R. Fish

Inhalation Studies

G. W. Royster, Jr.

M. B. Edwards

W. E. Lotz

G. J. Dodson

B. R. Fish

J. L. Thompson

Single Acute Inhalation of U_3O_8 Aerosol. — Eighteen dogs were exposed to single inhalations of a U^{235} oxide aerosol. Groups of three animals each were sacrificed 1, 24, 48, 96, 192,

and 384 hr after the completion of the 1-hr exposure. The aerosol consisted of U_3O_8 particles bound into chainlike aggregates which had an average mean particle size of 0.33μ , while the average standard deviation was 0.13 (Table 43). A description of the exposure method and apparatus has been given in previous progress reports.^{1,2}

¹S. R. Bernard *et al.*, *H-P Semiann. Prog. Rep.* July 31, 1956, ORNL-2151, p 1.

²S. R. Bernard *et al.*, *H-P Ann. Prog. Rep.* July 31, 1957, ORNL-2384, p 1.

Table 43. Disappearance of Enriched Uranium from the Lungs of Dogs After Inhalation of a U_3O_8 Aerosol

Time of Sacrifice After Inhalation (hr)	Dog No.	Total Exposure (d/min)	Mean Particle Size (μ)	Percentage of Total Exposure Remaining in Lungs at Sacrifice
1	49	39,326		12.6
	50	26,644		18.7
	101	26,803	0.3142 ± 0.2840	24.2
	102	144,340	0.7178 ± 0.6538	18.7
				$\bar{X} = 18.6$
24	103	24,704	0.3374 ± 0.3556	19.5
	104	42,561		15.0
	105	84,379		17.3
				$\bar{X} = 17.3$
48	106	51,813		9.8
	107	38,542	0.4926 ± 0.5548	15.5
	117	24,625		8.04
				$\bar{X} = 11.1$
96	115	92,877	0.3527 ± 0.3102	14.7
	116	34,459		15.1
				$\bar{X} = 14.9$
192	108	98,368		12.2
	113	66,486	0.3465 ± 0.3239	16.1
	114	139,129	0.4224 ± 0.3996	8.03
				$\bar{X} = 12.4$
384	111	60,473	0.4232 ± 0.3514	9.94
	112	103,495	0.4317 ± 0.3803	13.7
	110	110,403	0.3298 ± 0.3050	9.4
				$\bar{X} = 11.01$
			$0.3294^* \pm 0.1299^{**}$	

*Weighted mean particle size.

**Weighted standard deviation.

A measured volume of air containing a known concentration of the aerosol defined the dose to which the animals were exposed. Lung burden, in terms of percentage of the exposed dose retained, may be described by $19.2 t^{-0.0846}$ for t in hours (see Fig. 110). During this initial phase of the study, the effective half life was 648 hr.

Acute Inhalation of Uranium Compounds by Man. — Three separate uranium inhalation incidents have been followed by analyses of the excretion of the personnel involved in the incidents. A description of the efforts made to evaluate the magnitude of the exposures can serve to illustrate the methods currently available.

One incident involved the inhalation of a relatively insoluble uranium oxide. Over a period of about two weeks, an employee was exposed to the fume produced in grinding and polishing a uranium-metal part. Urine specimens indicated an excretion of 0.05 mg/day at the time the employee was removed from exposure. The first estimate of the lung burden was obtained by assuming that of the amount which reached the bloodstream from the lung, about two-thirds appeared in the urine directly and that this quantity dominated any slower elimination from other body organs. The initial rate of elimination from the lung to the bloodstream was not known, but the effective half life was taken to lie between 12 and 120 days. The lung burden was then estimated from the relation

$$\begin{aligned}\text{excretion rate} &= 2/3 \times 0.693/12 \times \text{lung burden} \\ &= 0.05 \text{ mg/day}\end{aligned}$$

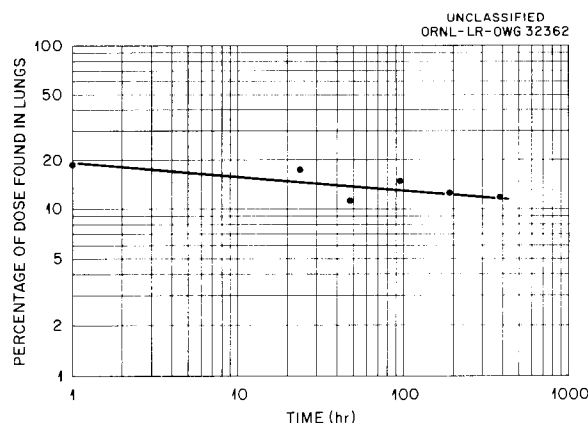


Fig. 110. Lung Retention of Uranium by Dogs After Acute Inhalation of Uranium Metal Fumes (U_3O_8).

and was found to be about 1.3 mg (13 mg when the 120-day half life was used). This relation provides an estimate for only that portion of the lung burden that had direct communication with the bloodstream. The portion eliminated through the GI tract is a highly sensitive function of particle size, but was estimated in this case to have been a fourth of the total lung burden at cessation of exposure.

Subsequent air samples made during a mockup of the operation were analyzed for composition by electron diffraction. Particle-size distribution was estimated by sampling with a modified cascade impactor. The particles, predominantly U_3O_8 , had a mass median diameter of 1.9μ and a geometric standard deviation of 1.8. Air-activity levels around the operation were highly variable and depended upon sampling position and operating technique. The maximum activity was 0.3 mg/m^3 . If a breathing rate of $10 \text{ m}^3/\text{shift}$ and an alveolar retention of 20% are assumed, it would have been possible for personnel to accumulate as much as 6 mg in the alveoli over the known period of exposure.

The excretion-rate data for the first 100 days postexposure may be fitted with the expression $0.0433e^{-0.0347t} + 0.0067e^{-0.00231t}$ (mg/day). If it is assumed that the total initial exposure to alveolar lung will eventually be recovered from the urine, then by integrating this form it may be estimated that the initial lung burden was 4.1 mg. The above expression predicted a total excretion for the first 140 days to be 2.04 mg. During the same period, measurements in an in vivo counter operated at the Argonne National Laboratory by Marinelli and Miller indicated a 2.07-mg reduction in body burden. Also, the accumulated urine excretion determined by the Y-12 Bioassay Laboratory showed the total to be 2.00 mg over the first 140 days. By using the value 4.1 mg and by extrapolating the urinary excretion rate curve back to the time the exposure ceased, it can be shown that the ratio of lung burden to initial daily excretion rate was about 80. Such a multiplication factor might be used for obtaining an estimate of the alveolar burden.

Two other acute exposures involved single inhalations of uranyl nitrate, chiefly as an airborne mist. It is recognized that the effective half life of uranyl nitrate in the lung lies between a few hours and a day. Because uranyl nitrate is rapidly absorbed in lung fluids, the excretion

pattern, after the first few days, for a lung exposure to this compound is similar to that shown by an intravenous injection.

Urine specimens from the person involved in the lowest-dose incident were first obtained 153 hr after exposure. At the time about 0.0155 mg of uranium was being excreted per day. That excretion level was comparable to the rate of excretion seen by Bassett *et al.*³ in a study employing single intravenous injections of uranyl nitrate in human subjects. Bassett's data for the excretion of uranium from six humans can be approximated by the power function $57.2t^{-1.8}\%$ /hr. This equation predicts that at 153 hr the urinary excretion per day should represent 0.16% of the initial dose. Interpreting this acute inhalation on the same pattern as a single injection yielded an estimated 9.7 mg for the initial dose, based on the first urine sample. Similar calculations based on the results of subsequent urine specimens provided estimates ranging down to 1 mg. Urinary excretion of uranium had reached a very low level (about 0.0001 mg/day) three months after the exposure, and by that time approximately 0.5 mg had been excreted. Evaluation of urinary uranium from an inhalation on the injection basis leads to overestimation of the initial exposure. For that reason the best approximation to the initial dose was taken to be between 0.5 and 1.0 mg.

The case involving the highest gravimetric dose was first sampled 29 hr after the incident. Urinary excretion was found to be about 22 mg/day at that time. This excretion rate was somewhat higher than the levels reported by Bernard and Struxness⁴ from a study of the distribution and excretion of uranium in man. In that study, patient VI received the largest gravimetric dose, and the excretion rate was represented by the expression $33.2t^{-1.36}\%$ of the injected dose per hour. By use of the first spot urine sample (a single void) and the excretion function above, the inhaled dose was calculated to have been 270 mg. Subsequent urine specimens were composited over succeeding 24-hr periods and yielded an estimate of 500 mg for the initial exposure.

³S. H. Bassett *et al.*, *The Excretion of Hexavalent Uranium Following Intravenous Administration. II. Studies on Human Subjects*, UR-37 (June 25, 1948).

⁴S. R. Bernard and E. G. Struxness, *A Study of the Distribution and Excretion of Uranium in Man. An Interim Report*, ORNL-2304 (June 4, 1957).

An Aerosol Impinger-Precipitator. — The initial deposition pattern, hence the fate, of an inhaled aerosol is a sensitive function of particle-size distribution. Many other physiological factors such as solubility and rates of phagocytosis are also influenced by particle size. In an experimental study of chronic inhalation, the aerosol size distribution indices are essential control factors. An airborne aerosol is very difficult to characterize as to particle-size distribution. A sampling device may tend to change the apparent size of certain particles by breaking up large aggregates or, oppositely, by promoting the agglomeration of smaller particles into chains or aggregates. In addition, a sampler may tend to discriminate against certain sizes of particles or to segregate the deposit so that representative fields are not available for optical counting.

Particle sizes of physiological significance lie in a band that overlaps the lower limit of resolution for light microscopes. Therefore it is necessary to use the electron microscope in order to more adequately cover the size range of importance. The sample may be collected on some primary medium, such as a molecular filter, and then may be transferred to an electron microscope target. It is more convenient, however, for the sample to be collected directly on the microscope target grid. Baum⁵ has described a sampling device which combines the collection properties of direct impaction for large particles with the collection properties of electrostatic precipitation for smaller ones, the deposit being collected on a microscope target. A sampler has been built at the Laboratory, and, although the collection mechanisms are similar to those of Baum's instrument, there are some differences that may prove to be useful.

Figure 111 shows the major features of the developmental model of the sampler. A stream of air is directed against the microscope target by a modified 16-gage hypodermic needle. The larger particles will be collected by impaction on the grid, but many of the small particles will be deflected and will follow the air stream. An electric field is established between the orifice at the tip of the needle and the target so that these small particles may be collected more efficiently.

⁵J. W. Baum, *Electrical Precipitator for Aerosol Collection on Electron Microscope Screens*, HW-39129 (Sept. 22, 1955).

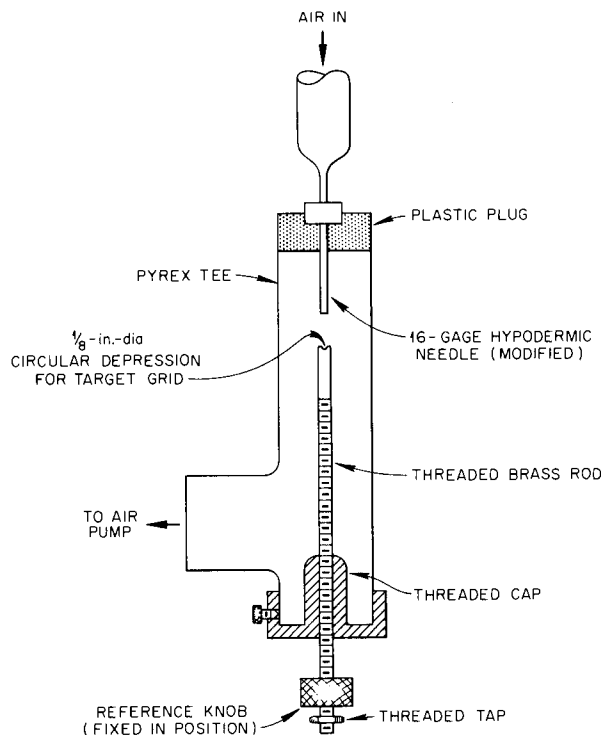
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Fig. 111. Developmental Model of an Aerosol Sampler Designed to Collect Directly on an Electron-Microscope Grid.

Electrostatic precipitation depends upon two essentially different stages. The first stage produces an electrostatic charge on individual particles, while the second stage provides an electric field which promotes their collection. Ordinarily, both functions are performed in one stage by using very high voltages. With a view toward a possible future application as a portable air sampler, there is an advantage in using as low a collecting voltage as would be consistent with high efficiency. It is expected that frictional forces in the tube and the orifice will impart some charge to the particles. However, if adequate efficiencies are not obtained, then some expedient, such as passing the aerosol through a layer of ionized air, may be used. The ionization may be done in a manner similar to that described by Saunders and Quinn.⁶

⁶B. G. Saunders and R. L. Quinn, *Electrostatic Precipitator for Measuring Particle-Size Distribution in Aerosols*, ORNL-1656 (Jan. 19, 1954).

Distribution and Excretion Studies

J. R. Muir

N. L. Gillum

B. R. Fish

K. R. Lape

G. W. Royster, Jr.

Distribution and Excretion of Uranium. — It has been suggested by Tannenbaum⁷ and Bernard and Struxness⁴ that the amount of uranium injected may affect its distribution and excretion. A pilot experiment was undertaken in order to evaluate such an effect and to establish more definitely the factors involved in the deposition of soluble uranium compounds in the animal body, particularly in the kidneys.

Mongrel dogs received intravenous injections of uranyl nitrate and were serially sacrificed at periods of two, four, eight, etc., weeks. The dogs were injected on a body-weight basis at one of three dose levels: 10 μg of U^{235} per kg, 100 $\mu\text{g}/\text{kg}$, and 1 mg/kg. The higher dose levels were selected because they approximate the doses received by the Boston patients.⁸

The radiochemical analysis of the tissues and body fluids obtained from 11 dogs at sacrifice has begun. All the urine was collected for two days following injection, and then 24-hr samples were obtained once weekly until sacrifice. The urine specimens were analyzed radiochemically by use of a method described by Royster.⁹ Figure 112 presents this data, the points for each dose level being connected by straight-line segments and each point representing the average excretion rate of from one to four animals. It is seen that the percentage excretion rate varied inversely with gravimetric dose.

Electrodeposition of Urinary Uranium. — This laboratory has for several years used a modification of the Cohen and Hull method for the electrodeposition of uranium and has been successful in assaying a large number of urine samples with reasonable precision. It has been noted that a recovery of $(92.3 \pm 3)\%$ could be obtained from pure aqueous solutions of uranium but that the recovery was lower and considerably more variable from wet-ashed urines. The lowered recovery of urinary

⁷A. Tannenbaum and H. Silverstone, *Natl. Nuclear Energy Ser. Div. IV* 23, 59 (1951).

⁸S. R. Bernard et al., *H-P Semiann. Prog. Rep.* July 31, 1955, ORNL-1942, p 1.

⁹G. W. Royster, Jr., *Electrodeposition of Uranium from Urine* (to be published).

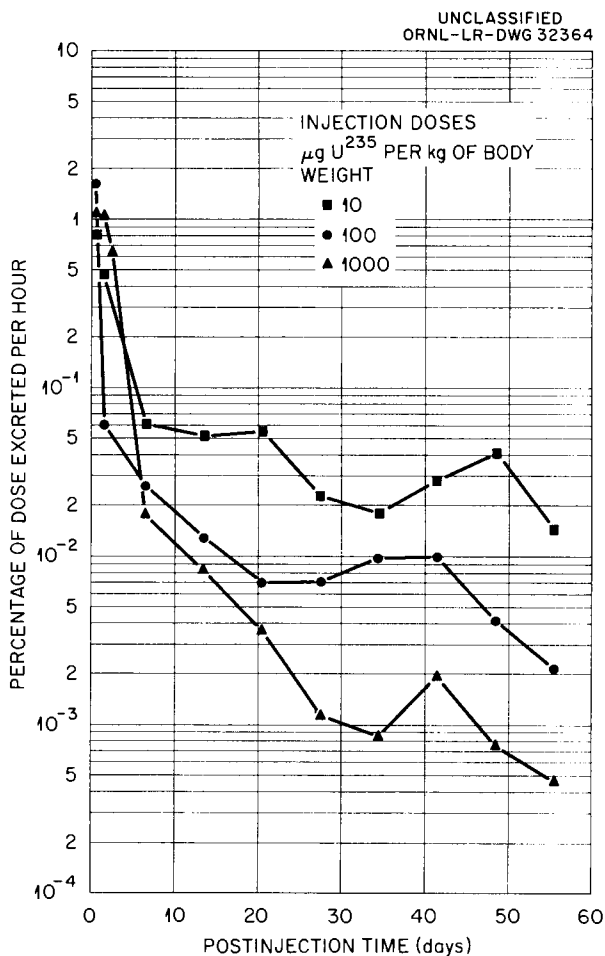


Fig. 112. Urinary Excretion of Uranium by Twelve Mongrel Dogs, Following Intravenous Injection.

uranium was generally attributed to a codeposition of inorganic salts which leads to loss of alpha activity by absorption.

In an effort to determine the composition of these undesirable deposits, a series of samples was plated after adding the major cations normally found in urine to aqueous solutions of uranyl nitrate. The cations - Ca, Mg, Na, and K - were tested separately to determine the effect of each. The results of these tests are shown in Fig. 113. The plates to which calcium and magnesium were added showed a decreasing recovery as the cation concentration increased. A white deposit was visible on all plates to which calcium and magnesium were added. Sodium and potassium showed

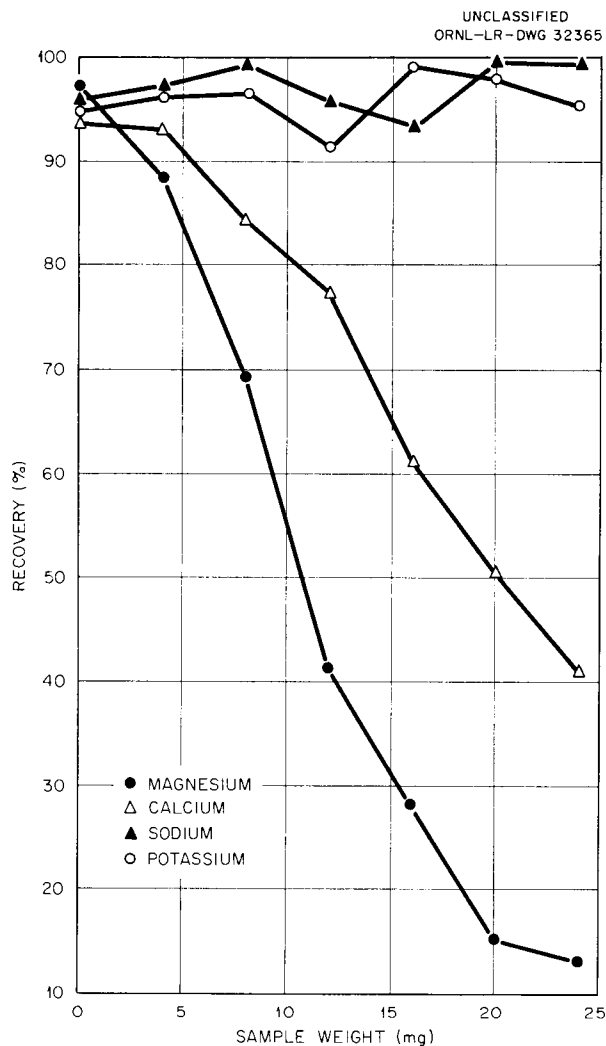


Fig. 113. Electrodeposition of Major Cations Found in Urine.

no white deposits on the plates, and the recovery was not affected.

Sodium carbonate, which is one of the stronger complexers of uranium, was added to urine solutions to hold the uranium in solution while the calcium was removed as the insoluble carbonate. Magnesium is removed as the phosphate. Removal of calcium and magnesium from the digested urine solutions was very effective in eliminating the extraneous salt deposits on the films. Recoveries from 33 different urines tested averaged $(89.5 \pm 4)\%$.

Ingestion Studies and Whole Body Counting

W. E. Lotz

J. W. Hollis, Jr.

F. W. Sanders

Experimental Body Burden Counter. — The first model of a device for measuring the body burden of U^{235} in a small live animal was described in ORNL-2384.¹⁰ Initial tests with a Po-Be source showed that the background due to the source was too high. Changes in the apparatus which were made in order to reduce the source background included the use of an Sb-Be neutron source and the addition of layers of paraffin and boral (see Figs. 114 and 115).

A phantom, which consisted of U^{235} in aqueous solution contained in a polyethylene bottle about the size and shape of a mouse, was used to test the sensitivity of the device. The best response obtained was a $(28.7 \pm 1.9)\%$ increase in the background count rate for a phantom containing $1 \mu\text{C}$ of U^{235} . In order for the device to be useful, the sensitivity would have to be increased by a factor of 10^3 over the best results obtained.

It was calculated that during a 5-min counting period, the dose received by the phantom consisted of 1.16 mrad of neutron and gamma radiation from the source, 42.3 mrad from the fissioning nuclei, and 1260 mrad from the uranium alpha radiation.

¹⁰S. R. Bernard *et al.*, *H-P Ann. Prog. Rep. July 31, 1957*, ORNL-2384, p 1.

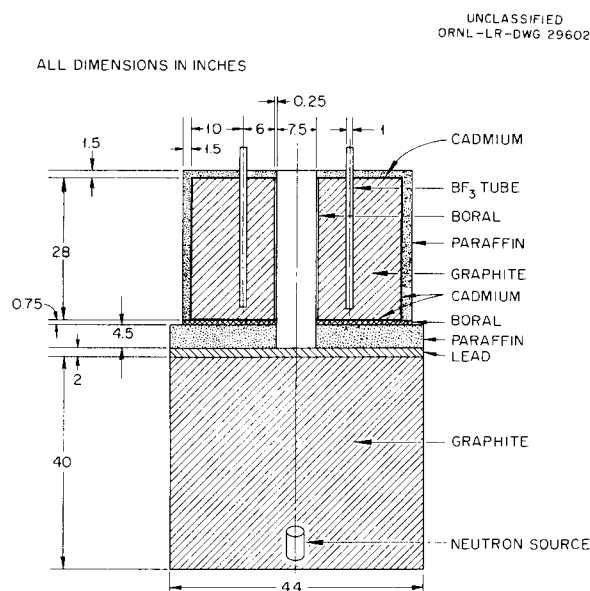


Fig. 114. Diagram of Body-Burden Counter.

Thus the additional dose due to the counting procedure was only 3.5% of the dose produced by the contained uranium.

Construction and testing of this device were undertaken to study the feasibility of measuring the body burden of U^{235} in man by this method. Although the results indicated that the additional dose to the subject could be held to a low level, a great improvement in sensitivity is necessary before the instrument can be of much value.

Distribution of $Mn^{52,54}$ and Cs^{137} in Mice. — Eight groups of five young female mice, whose weights ranged from 20 to 25 g, were used for determining the tissue distribution of $Mn^{52,54}$ after 2, 4, 8, 16, 32, 50, 64, and 77 days of continuous ad libitum feeding of the radionuclide in the drinking water ($0.1 \mu\text{C}$ of $Mn^{52,54}$ per milliliter). In the case of Cs^{137} , 50 mice were evenly divided into five groups and fed the isotope in the drinking water ($1.0 \mu\text{C}$ of Cs^{137} per milliliter) for 2, 16, 32, 64, 94, and 259 days. The concentrations in tissues after various periods of ad libitum feeding are given in Tables 44 and 45.

INTERNAL DOSE ESTIMATIONS

W. S. Snyder

Internal Dose Handbook Revision

K. Z. Morgan

M. J. Cook

W. S. Snyder

M. R. Ford

In the 1957 annual report,¹¹ the proposed changes in the revisions of the national¹² and international¹³ handbooks on internal dose were outlined. During the past year the two committees have agreed to combine these publications and to issue jointly a single internal dose report. This has necessitated a complete revision of the text and many alterations of calculations in order to conform to the joint requirements. The preliminary version has been sent to the members of both internal dose committees and to the editorial committee of the ICRP (International Commission on Radiological Protection).

Maximum permissible concentration (MPC) values have been calculated for insoluble material and

¹¹M. J. Cook *et al.*, *H-P Ann. Prog. Rep. July 31, 1957*, ORNL-2384, p 39-41.

¹²U.S. National Bureau of Standards, *Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water*, Handbook 52, GPO, Washington, 1953.

¹³International Congress of Radiology, "Recommendations of International Commission of Radiological Protection," *Brit. J. Radiol. Suppl.* 6, 23-29 (1955).

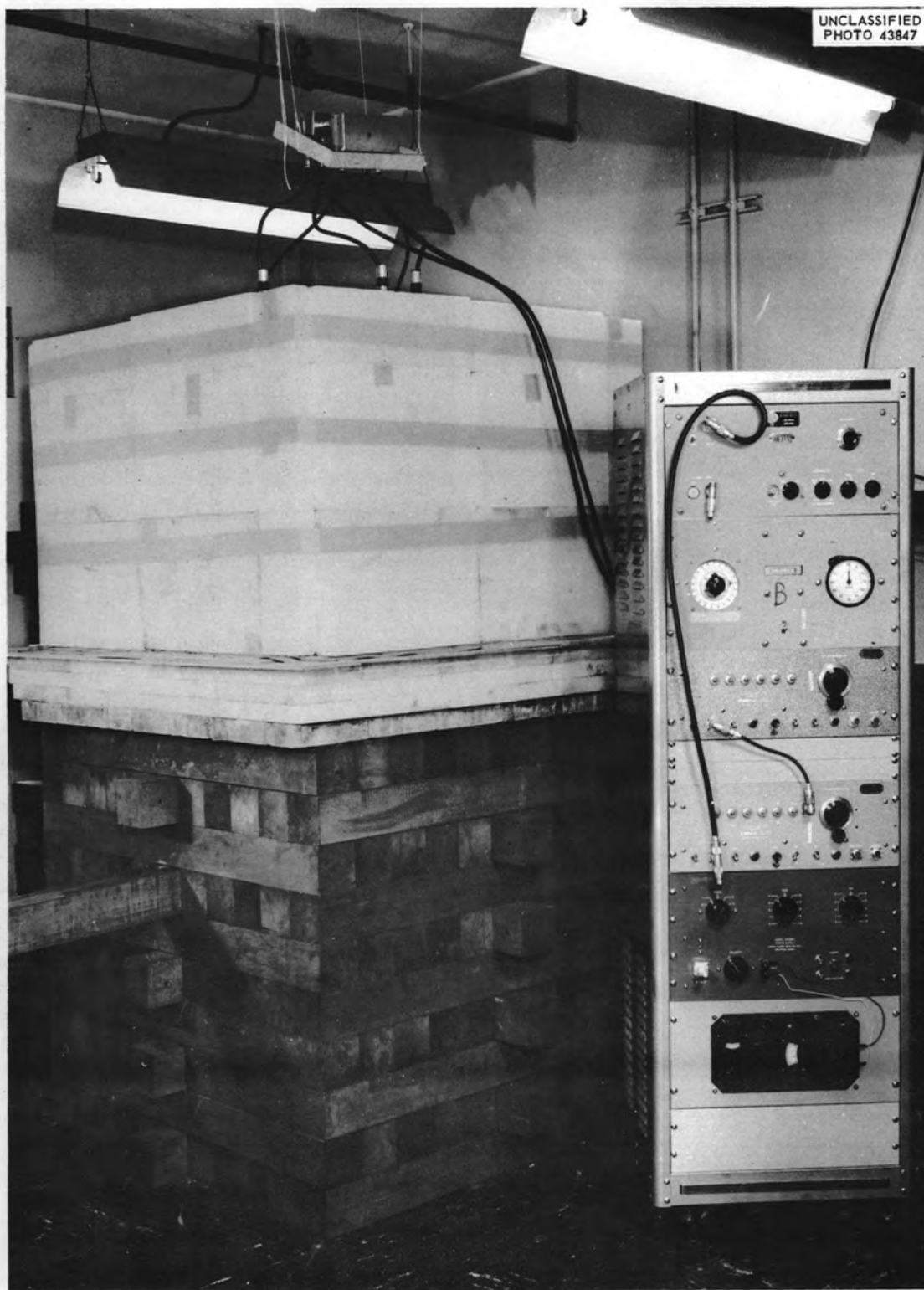


Fig. 115. Photograph of Body-Burden Counter.

Table 44. Concentration of Mn^{52,54} in Various Organs of the Mouse After Continuous ad Libitum Feeding in the Drinking Water
(per cent daily intake per gram of tissue)

Organ	Days of Continuous Feeding							
	2	4	8	16	32	50	64	77
Liver	0.23	0.39	0.48	0.60	0.39	0.48	0.72	0.55
Kidney	0.11	0.33	0.50	0.68	0.58	0.74	0.88	0.70
Muscle	0.22	0.38	1.06	0.74	1.00	1.23	1.16	0.61
Pancreas	0.22	0.54	0.57	0.92	0.66	0.81	0.89	0.82
Femur	0.28	0.49	0.33	0.38	1.07	0.48	0.44	0.37
Ovary	0.25	1.14	0.32	0.30	0.42	1.16	0.48	0.31
Ovarian tract	0.11	0.41	0.30	0.41	0.28	0.34	0.30	0.30
Lung	0.15	0.21	0.38	0.40	0.24	0.34	0.50	0.19
Spleen	0.08	0.36	0.31	0.16	0.27	0.34	0.54	0.45
Fat	0.06	0.33	0.04	0.12	0.08	0.10	0.07	0.15
Heart	0.02	0.14	0.26	0.35	0.33	0.62	0.58	0.54
GI tract and contents								
Large intestine	11.46	16.46	12.70	9.36	13.24	1.51	9.52	9.41
Cecum	17.44	15.08	10.09	9.93	8.25	1.22	9.55	10.14
Small intestine	2.61	1.84	2.21	1.05	0.97	0.59	0.97	2.54
Stomach	3.78	2.45	2.09	2.09	2.20	0.57	1.10	2.17

have been included in the revision. These additions practically double the number of estimated MPC values previously discussed.

A third alteration has been the addition of 13 more radionuclides, making a total of 240, which may be compared with the 70 radionuclides in Handbook 52 (ref 12) or the 86 in the ICRP handbook¹³ (see Fig. 116).

The fourth major addition has been the compilation of a bibliography (about 1500 references) from which the biological data were taken.

Inorganic Analysis of Human Tissue

M. J. Cook E. L. Grove¹⁴
I. H. Tipton¹⁵ K. K. McDaniel

During the past year, human tissue has been analyzed by emission spectroscopy, flame photom-

etry, neutron activation, and wet chemistry. The emission spectroscopy has been done under sub-contract No. 380 by the Physics Department of the University of Tennessee. The methods of collection, preparation, and spectrographic analysis have been described previously.¹⁶ A fourth memorandum has been issued which gives the data from tissue received from Baltimore, Maryland.¹⁷ The analyses of the tissues received from Seattle and

¹⁴Consultant, University of Alabama.

¹⁵Consultant, University of Tennessee.

¹⁶I. H. Tipton *et al.*, *Methods of Collection, Preparation and Spectrographic Analysis of Human Tissues*, ORNL CF-57-2-2 (Feb. 28, 1957).

¹⁷I. H. Tipton *et al.*, *Spectrographic Analysis of Normal Human Tissue from Baltimore, Maryland*, ORNL CF-57-11-33 (Nov. 11, 1957).

Table 45. Concentration of Cs^{137} in Various Organs of the Mouse After Continuous ad Libitum Feeding in the Drinking Water
(per cent daily intake per gram of tissue)

Organ	Days of Continuous Feeding					
	2	16	32	64	94	259
Muscle	8.27	63.07	53.31	66.43	58.37	67.25
Ovary	11.54	64.84	18.21	47.88	35.77	18.84
Pancreas	16.95	28.64	20.99	39.96	22.14	
Heart	12.99	32.43		36.39	21.61	20.37
Femur	6.58	29.30	19.84	31.78	25.41	30.71
Spleen	10.13	26.34	19.91	31.76	20.54	26.65
Liver	7.53	17.24	12.95	20.98	36.82	14.90
Lung	9.01	18.19		25.71	13.22	15.90
Brain	2.58	18.30		20.50	9.99	12.32
Kidney	12.43	30.29	21.75	30.78	16.60	22.96
Fat	1.02	3.36	2.30	5.54	3.21	
GI tract and contents						
Stomach	2.33	10.14	7.44		17.46	11.43
Large intestine	3.16	12.46	12.96		12.38	17.18
Small intestine	13.62	57.85	44.44		25.49	19.37
Cecum	2.85	7.57	4.52		13.33	13.61

Tacoma have been finished, and a report will be issued soon.¹⁸

Flame photometric analysis for sodium and potassium in the samples analyzed spectrographically is authorized under subcontract No. 1084 and was performed by the School of Chemistry of the University of Alabama. The summary of the results is presented in Table 46. A correlative study indicates that there is no appreciable loss of sodium and potassium in the ashing process.

For neutron activation analysis, samples of liver, kidney, spleen, lung, bone, ovary, and testis were dried and sent to the Analytical Chemistry Division, ORNL. The results are presented in Table 47.

¹⁸J. H. Tipton *et al.*, *Spectrographic Analysis of Normal Human Tissue from Seattle and Tacoma, Washington*, ORNL Central Files Memo (to be published).

Calcium in bone was determined by wet chemistry at the College of Home Economics, University of Tennessee, and the results are presented below:

Age (years)	Per cent in Ash
10-19	34.8
20-29	34.8
30-39	33.7
40-49	34.0

From these four methods for tissue analysis, a table of the chemical composition of human tissue was prepared and will be issued in *Report of Committees on Permissible Dose for Internal Radiation - 1958 Revision*. The information is presented as Table 48.

In addition to the analyses of tissues from the United States, the analysis of tissues from foreign countries is being continued.

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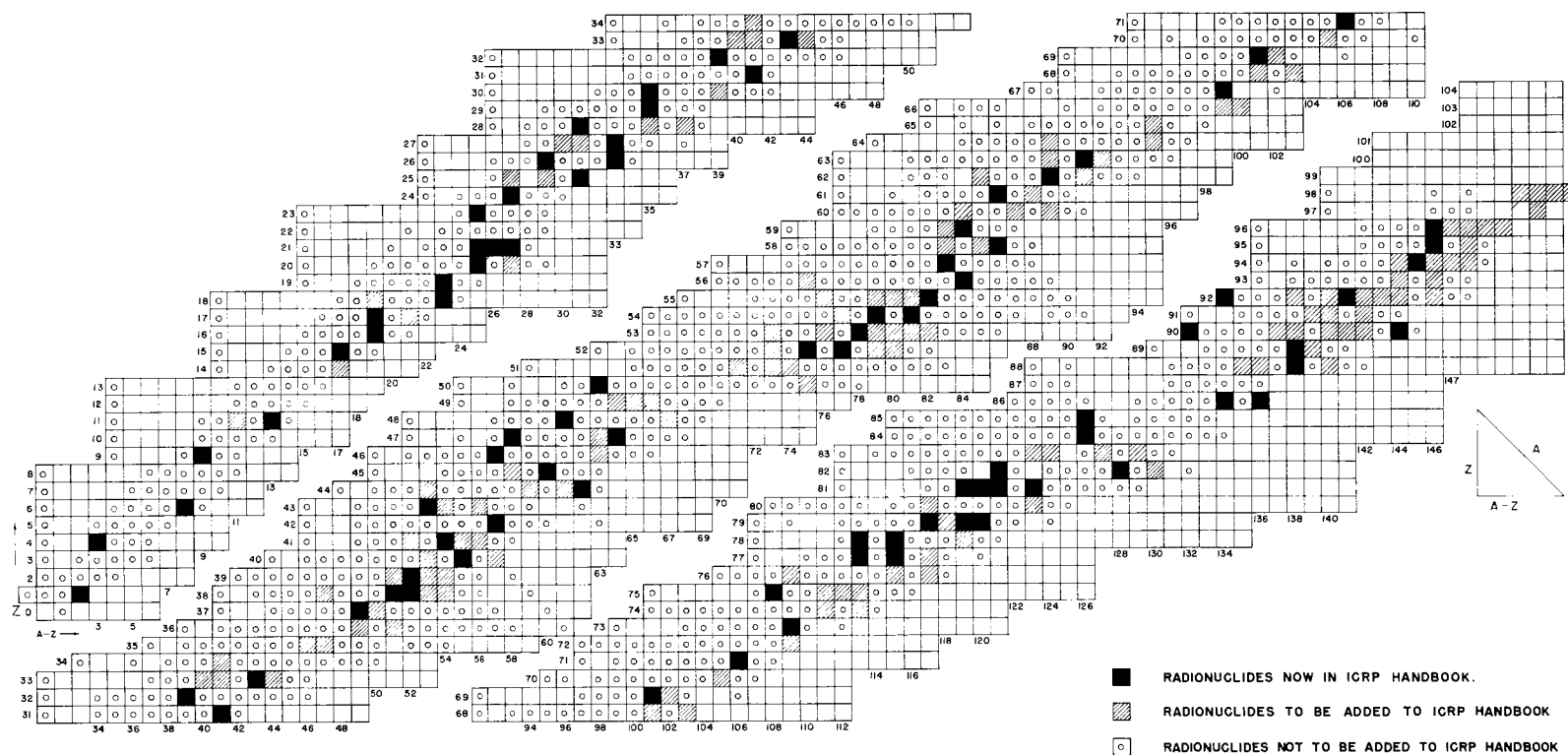


Fig. 116. Radionuclides in Revised Internal-Dose Handbook.

PERIOD ENDING JULY 31, 1958

Table 46. Sodium and Potassium in Ashed Human Tissue As Determined by Flame Photometry

Tissue	Element	Analysis (%)			
		Denver ^a	Dallas ^b	Baltimore ^c	Miami ^d
Aorta	Na	14.5	18.2	10.3	17.1
	K	8.76	9.94	6.14	11.8
Brain	Na	10.8	9.96	11.1	9.70
	K	19.1	17.5	18.0	16.4
Esophagus	Na	20.6	20.2	20.6	20.2
	K	19.3	19.1	18.7	18.4
Heart	Na	11.9	11.9	12.4	13.5
	K	25.4	25.2	25.0	24.0
Large intestine	Na	15.1	13.9		17.8
	K	20.4	18.2		17.5
Sigmoid colon	Na			15.3	
	K			21.4	
Small intestine	Na	14.2	15.6		15.4
	K	22.2	19.0		18.4
Jejunum	Na			15.9	
	K			19.6	
Kidney	Na	18.9	18.4	20.8	19.0
	K	16.2	17.8	17.7	18.2
Liver	Na	11.3	9.36	9.04	9.70
	K	19.5	17.2	17.6	17.4
Lung	Na	18.4	19.1	17.2	19.0
	K	17.2	17.4	17.9	18.8
Muscle	Na	5.90	5.32	7.12	7.18
	K	30.6	28.2	31.1	28.6
Pancreas	Na	10.7	8.98	11.8	12.0
	K	21.8	20.4	21.0	19.4
Prostate	Na	15.3	16.6		
	K	17.6	17.6		
Spleen	Na	8.15	8.49	6.4	9.9
	K	24.6	23.7	24.0	22.3
Stomach	Na	16.1	16.6	16.6	18.2
	K	18.8	18.6	19.0	18.7
Testis	Na	18.2	19.7	18.0	19.9
	K	17.4	19.2	18.2	19.9
Thyroid	Na	19.6			
	K	13.2			
Trachea	Na		20.7	16.0	
	K		9.99	9.0	
Urinary bladder	Na	17.6	16.9	19.7	19.7
	K	20.0	17.4	17.7	18.6

Table 46 (continued)

Tissue	Element	Analysis (%)			
		Denver ^a	Dallas ^b	Baltimore ^c	Miami ^d
Uterus	Na		19.2		20.4
	K		17.8		18.5
Diaphragm	Na		12.8	12.0	15.0
	K		23.8	25.0	23.8
Larynx	Na			9.53	9.75
	K			6.64	5.66
Skin	Na			22.6	
	K			9.07	
Omentum	Na				15.9
	K				14.4

^aI. H. Tipton *et al.*, *Progress Report: Spectrographic Analysis of Tissues for Trace Elements. July 1, 1955 Through December 31, 1955*, ORNL CF-56-3-60 (March 12, 1956).

^bI. H. Tipton *et al.*, *Spectrographic Analysis of Normal Human Tissue from Dallas, Texas*, ORNL CF-57-2-3 (Feb. 28, 1957).

^cI. H. Tipton *et al.*, *Spectrographic Analysis of Normal Human Tissue from Baltimore, Maryland*, ORNL CF-57-11-33 (Nov. 11, 1957).

^dI. H. Tipton *et al.*, *Spectrographic Analysis of Normal Human Tissue from Miami, Florida*, ORNL CF-57-2-4 (Feb. 28, 1957).

Table 47. Elements in Human Tissue as Determined by Neutron Activation

Tissue	Elements ($\mu\text{g/g}$, dry tissue)											
	As	Br	Co	Cs	Cu	K	Mn	Na	Sb	Se	Te	Zr
Bone	0.016	0.39	0.83	8.6	0.7	2.1	0.4	378	0.26		428	
Kidney	0.091	1.3	0.64	1.1	20	8.9	4.5	848	0.82	2.37	199	105
Liver	0.28	0.47	0.74	1.4	46	18.8	4.0	518	0.45		2500	
Lung	0.36	2.4	0.26	1.6	9	10.6	1.5	945	0.53		56.3	
Ovary	0.07	1.8	0.08	0.71	5.0	7.5	0.9	984	0.15		79.3	
Spleen	0.055	0.61	0.37	2.3	6.0	15.1	0.5	548	0.44		96.4	
Testis	0.18	1.4	0.2	1.1	4.5	8.6	0.7	974	0.2		177	

Table 48. Elements in the Body Organs of Standard Man*
($\mu\text{g/g}$ of wet tissue)

Tissue	Elements																					
	Ag	Al	As	Au	B	Ba	Be	Bi	Br	Ca	Cd	Cl	Co	Cr	Cs	Cu	F	Fe	Ga	I	K	Lo
Adrenal (6)**	0.016	0.62	<1.4	<0.16	<0.11	0.025	<0.01	<0.05		44	<0.4		<0.04	0.07	<0.7	1.1		43	<0.005		1100	<0.4
Aorta (75)	0.015	0.8	<2.8	<0.2	<0.1	0.15	<0.03	<0.15		970	<0.6		<0.07	0.06	<1.4	1.3		46	<0.009		1370	<0.9
Blood		0.16 ^(S)								50		320 ^(R)				1 ^(S)	0.3 ^(S)	0.5 ^(S)			11	
Bone (99)	<0.1	<0.44	0.01 ^(N)	<2.5	<3	1.63			0.3 ^(N)	10 ^{+6(E)}	N.D.	1700 ^(E)	0.6 ^(N)	<0.49	6 ^(N)	0.6	127 ^(E)	115		0.3 ^(E)	880	
Brain (94)	0.045	0.29	<4	<0.13	<0.14	<0.01	<0.04	<0.05		110	<0.9	1300 ^(E)	<0.05	<0.01	<0.03	5.8	0.5 ^(E)	48	<0.01	0.1 ^(E)	3300	<1.3
Breast (7)	<0.002	0.4	<2.5	<0.09	<0.08	0.015	<0.03	<0.03		64	<0.2		<0.02	0.03		0.36		4.4	<0.003		310	<0.3
Gastrointestinal tract												600								0.4 ^(E)		
Esophagus (39)	<0.004	0.82	<2.1	<0.08	<0.07	0.05	<0.02	<0.03		120	<0.5		<0.03	0.05	<0.01	1.3		34	<0.007		1510	<0.7
Stomach (88)	<0.006	0.52	<1.7	<0.07	<0.14	0.038	<0.02	<0.5		115	<0.43		<0.04	0.03	<0.01	1.7		29	<0.006		1520	<0.6
Duodenum (51)	<0.006	0.75	<1.7	<0.07	<0.06	0.038	<0.02	<0.12		86	<0.56		<0.04	0.03		2.3		39	<0.006		1520	<0.6
Jejunum (66)	<0.004	0.57	<2.1	<0.13	<0.12	0.04	<0.02	<0.5		76	<0.6		<0.04	0.03	<0.02	2		33	<0.007		1600	<0.7
Ileum (68)	<0.005	2	<0.9	<0.1	<0.04	0.088	<0.01	<0.04		135	<0.26		<0.04	0.04		1.9		28	<0.003		1230	<0.3
Cecum (31)	<0.009	1.26	<1.2	<0.09	<0.05	0.12	<0.01	<0.1		155	<0.32		<0.26	0.047		1.4		23	<0.004		1060	<0.4
Sigmoid colon (72)	<0.007	1.05	<0.9	<0.05	<0.04	0.09	<0.01	<0.04		115	<0.26		<0.1	0.05	<0.01	1.5		21	<0.003		1320	<0.3
Rectum (33)	<0.004	0.74	<0.9	<0.03	<0.03	0.054	<0.01	<0.07		140	<0.24		<0.06	0.04		1.2		28	<0.003		1580	<0.3
Heart (96)	<0.006	0.3	<2.6	<0.09	<0.09	0.01	<0.03	<0.04		53	<0.6	1250 ^(E)	<0.04	0.02	<0.02	3.51	0.5 ^(E)	50	<0.009	0.9 ^(E)	2500	<0.1
Kidney (102)	<0.01	0.41	0.03 ^(N)	<0.12	<0.12	0.027	<0.03	<0.02	0.4 ^(N)	115	32	2100 ^(E)	0.2 ^(N)	0.03	0.34 ^(N)	2.9	0.8 ^(E)	76	<0.01	0.2 ^(E)	2030	<1
Larynx (31)	<0.015	0.61	<4	<0.13	<0.13	0.215	<0.04	<0.08		1920	<0.9		<0.05	0.07	<0.03	1.1		28	<0.013		1530	<1.3
Liver (102)	0.018	0.71	0.1 ^(N)	<0.12	<0.14	<0.013	<0.03	<0.07	0.2 ^(N)	70	2.44	1250 ^(E)	0.3 ^(N)	0.02	0.6 ^(N)	8.6		185	<0.011	1.1 ^(E)	3100	<1.1
Lung (102)	<0.008	24	0.09 ^(N)	<0.08	<0.09	0.183	<0.02	<0.04	0.6 ^(N)	130	0.7	2550 ^(E)	0.06 ^(N)	0.2	0.4 ^(N)	1.3	0.6 ^(E)	300	<0.011		2120	<0.8
Muscle												600 ^(E)					0.4 ^(E)			0.9 ^(E)		
Diaphragm (42)	<0.004	0.35	<1.8	<0.06	<0.06	<0.03	<0.018	<0.05		74	<0.43		<0.13	0.04	<0.3	1.35		44	<0.006		2600	<0.6
Pectoral (21)	<0.005	0.20	<2.7	<0.09	<0.09	<0.011	<0.03	<0.04		37	<0.6		<0.04	0.03		0.71		28	<0.003		2830	<0.9
Psoas (86)	<0.007	0.34	<3.0	<0.10	<0.10	<0.014	<0.03	<0.05		49	<0.7		<0.10	0.03		0.9		36	<0.01		3330	<1
Omentum (35)	0.002	1.26	<0.26	<0.01	<0.01	0.026	<0.003	<0.12		40	<0.09		<0.01	0.04	<0.002	0.34		28	<0.001		400	<0.09
Ovary (9)	<0.005	0.7	0.01 ^(N)	<0.07	<0.09	0.04	<0.03	<0.04	0.33 ^(N)	220	<0.6		0.02 ^(N)	0.14	0.13 ^(N)	1.06		35	<0.009	0.9 ^(E)	1350	<0.9
Pancreas (95)	<0.007	0.47	<2.4	<0.11	<0.11	0.026	<0.02	<0.06		120	1.2	1600 ^(E)	<0.05	0.04	0.04	1.7		39	<0.008	0.5 ^(E)	2800	<0.8
Prostate (27)	0.021	0.89	<2.9	<0.2	<0.15	0.05	<0.03	<0.04		320	<0.76		<0.07	0.05	<0.02	1.4		30	<0.01		2200	<1.0
Skin (18)	0.013	2.8	<1.2	<0.05	<0.12	0.161	<0.01	<0.03		150	<0.28	2600 ^(E)	<0.02	0.33	<0.01	0.90		16	<0.005	1 ^(E)	800	<0.4
Spleen (97)	<0.006	1.1	0.02 ^(N)	<0.1	<0.12	<0.022	<0.03	<0.052	0.16 ^(N)	84	<0.74	1550 ^(E)	0.1 ^(N)	0.02	0.6 ^(N)	1.22	0.3 ^(E)	330	<0.01		3500	<1.1
Testis (53)	<0.005	0.5	0.04 ^(N)	<0.11	<0.09	0.011	<0.03	<0.08	0.33 ^(N)	100	<0.6	2330 ^(E)	0.05 ^(N)	0.03	0.3 ^(N)	1		27	<0.009		2100	<0.9
Thymus (2)	0.002	0.23	<1.2	<0.04	<0.04	0.009	<0.01	<0.02		63	<0.28		<0.02	0.01		0.42		19	<0.004		540	<0.4
Thyroid (11)	<0.04	1.2	<3.1	<0.1	<0.1	0.26	<0.03	<0.1		260	<0.8	1700 ^(E)	<0.08	0.03	<0.02	1.1		66	<0.01	350 ^(E)	1100	<1
Tongue (3)	<0.005	0.62	<3.2	<0.11	<0.11	0.04	<0.03	<4.4		175	<0.75		<0.04	0.06		1.43		33	<0.011		2800	<1.1
Trachea (35)	<0.02	1.85	<3.4	<0.47	<0.13	0.113	<0.03	<0.19		950	<0.8		<0.05	0.07	<0.02	1.2		47	<0.011		2200	<1.1
Urinary bladder (70)	<0.003	0.61	<1.4	<0.1	<0.11	0.03	<0.01	<0.02		130	<0.38		<0.02	0.04	<0.01	0.95		25	<0.005		1730	<0.5
Uterus (17)	<0.005	0.44	<2.6	<0.09	<0.09	0.05	<0.03	<0.04		185	<0.61		<0.07	0.24	<0.02	0.95		27	<0.009		1730	<0.9
Vagina (7)	<0.005	0.3	<2.6	<0.09	<0.09	0.03	<0.03	<0.04		200	<0.58		<0.14	0.07		0.90		32	<0.009		1500	<0.9

*Unless otherwise indicated, all these values are taken from the studies of Tipton *et al.* (see refs a, b, c, and d, Table 46).

**Number in parentheses after the name of the tissue indicates the number of samples of that tissue analyzed.

(N) Determined by neutron activation.

(F) Determined by flame photometry.

(E) M. Everett, *Medical Biochemistry*, 2d ed., Paul B. Hoeber, Inc., New York, 1946.

(B) E. J. King and T. H. Belt, "The Physiological and Pathological Aspects of Silica," *Physiol. Revs.* 18, 329-635 (1938).

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(H) J. B. Hursh and A. A. Gates, "Body Radium Content of Individuals with No Known Occupational Exposure," *Nucleonics* 7(1), 46-59 (1950); R. F. Palmer and F. B. Queen, "Normal Abundance of Radium in Cadavers from the Pacific Northwest," *Am. J. Roentgenol. Radium Therapy Nuclear Med.* 79, 521-529 (1958).

Table 48 (continued)

Tissue	Elements																					
	Li	Mg	Mn	Mo	Na	Nb	Ni	P	Pb	Re	Rb	Ru	Sb	Si	Sn	Sr	Te	Ti	Tl	V	Zn	Zr
Adrenal (6)		50	0.19	0.57		<0.45	0.28	1000	0.13			<0.05	<0.9	125 ^(B)	0.19	0.02		<0.13	<0.05	<0.005	8.4	<0.5
Aorta (75)	0.02	230	0.12	<0.09	2400 ^(F)	<0.9	<0.15	1100	2.51		9.9	<0.09	<2.8		0.41	0.43		<0.17	<0.09	<0.01	26	<0.9
Blood	0.02 ^(S)	43	0.14 ^(S)		1750 ^(E)			380	0.3 ^(S)					2.5 ^(S)	0.23 ^(S)						9.3 ^(S)	
Bone (99)	0.003	980	<0.1	<1.5	1300 ^(F)	<2		8 × 10 ⁴ (E)	6.6	1.7 × 10 ⁻⁸ (H)	10		0.2 ^(N)		<1.3	15	321 ^(N)	<1		<1.5	66	
Brain (94)	<0.007	140	0.26	<0.1	1700 ^(F)	<1.3	<0.1	3300	<0.4		28	<0.13	<4.0	26 ^(B)	<0.1	0.04		<0.15	<0.13	<0.01	13	<1.3
Breast (7)		20	0.03	<0.03		<0.3	<0.07	140	0.04			<0.04	<2.5		0.26	0.06		<0.05	<0.04	<0.004	2.8	<0.4
Gastrointestinal tract														41 ^(B)								
Esophagus (39)	0.013	120	0.12	<0.08	1900 ^(F)	<0.7	<0.09	850	0.16		7.2	<0.07	<2.1		0.8	0.1		<0.12	<0.07	<0.007	22	<0.7
Stomach (88)	0.009	100	0.31	<0.06	1300 ^(F)	<0.6	<0.06	1160	0.19		8.6	<0.06	<1.7		0.35	0.12		<0.07	<0.06	<0.012	18	<0.6
Duodenum (51)	0.009	100	0.47	<0.06	1700 ^(F)	<0.6	<0.08	1230	0.4		8.6	<0.06	<1.7		0.54	0.12		<0.08	<0.06	<0.007	20	<0.6
Jejunum (66)	0.009	120	0.61	<0.07		<0.7	<0.06	1460	0.32		23	<0.07	<2.1		0.52	0.15		<0.08	<0.07	<0.008	20	<0.7
Ileum (68)		150	0.63	<0.05	1600 ^(F)	<0.3	<0.13	1000	0.28			<0.03	<0.9		1.13	0.22		<0.06	<0.03	<0.018	21	<0.3
Cecum (31)		200	0.93	<0.04		<0.4	0.24	680	0.28			<0.04	<1.2		2.2	0.31		<0.06	<0.04	<0.015	22	<0.4
Sigmoid colon (72)	0.01	175	0.45	<0.03		<0.3	0.15	800	0.18		5.9	<0.03	<0.9		0.6	0.25		<0.1	<0.03	<0.047	18	<0.3
Rectum (33)		180	0.43	<0.03	1500 ^(F)	<0.3	0.14	760	0.49			<0.03	<0.9		0.94	0.19		<0.08	<0.03	<0.037	26	<0.3
Heart (96)	0.008	160	0.19	<0.09	1300 ^(F)	<0.9	<0.12	1600	<0.2		15.1	<0.09	<2.6	25 ^(B)	<0.15	0.05		<0.09	<0.09	<0.009	27	<0.9
Kidney (102)	0.008	130	0.85	0.40	2200 ^(F)	<1	<0.09	1700	1.24		10.7	<0.1	0.3 ^(N)	21 ^(B)	0.28	0.08	62 ^(N)	<0.14	<0.1	<0.011	48	<1
Larynx (31)	0.03	300	0.19	<0.13	2700 ^(F)	<1.3	<0.17	1700	2.2		12.6	<0.1	<4		0.18	1.0		<0.17	<0.13	<0.013	29	<1.3
Liver (102)	<0.006	165	1.3	1.13	1400 ^(F)	<1.1	<0.11	2700	2.		63	<0.1	0.2 ^(N)	24 ^(B)	0.4	0.03	1075 ^(N)	<0.14	<0.11	<0.014	46	<1
Lung (102)	<0.03	95	0.18	<0.08	1900 ^(F)	<0.8	<0.27	1110	0.68		7.3	<0.08	0.1 ^(N)	130 ^(B)	0.74	0.12	13 ^(N)	2.8	<0.08	<0.071	14	<0.8
Muscle														12 ^(B)								
Diaphragm (42)		140	0.13	<0.06	1600 ^(F)	<0.6	<0.08	1200	0.17			<0.06	<1.8		<0.09	0.06		<0.07	<0.06	<0.006	43	<0.6
Pectoral (21)		145	0.05	<0.09	1900 ^(F)	<0.9	<0.11	1440	0.19			<0.09	<2.7		<0.21	0.02		<0.10	<0.09	<0.009	46	<0.9
Psoas (86)	<0.005	190	0.05	<0.1		<1	<0.11	1700	0.14		31	<0.1	<3.0		<0.11	0.03		<0.10	<0.1	<0.01	51	<1
Omentum (35)	0.005	22	0.05	<0.01	350 ^(F)	<0.09	0.04	170	0.14		2.0	<0.01	<0.26		0.03	0.04		0.07	<0.01	<0.002	3.3	<0.09
Ovary (9)	0.011	86	0.17	<0.09		<0.9	<0.1	940	0.13		4.5	<0.09	0.03 ^(N)	18 ^(B)	1.15	0.25	14 ^(N)	<0.1	<0.09	<0.009	13	<0.9
Pancreas (95)	<0.005	165	1.16	<0.08	1400 ^(F)	<0.8	<0.09	2500	0.72		62	<0.08	<2.4	34 ^(B)	<0.27	0.07		<0.11	<0.08	<0.008	27	<0.8
Prostate (27)	0.013	175	0.22	<0.1	2000 ^(F)	<1	<0.1	1100	0.91		6.9	<0.1	<2.9	70 ^(B)	0.31	0.24		<0.16	<0.1	<0.01	87	<1
Skin (18)	0.021	73	0.2	<0.05	920 ^(F)	<0.4	0.44	310	0.55		2.0	<0.04	<1.2	55 ^(B)	0.28	0.1		0.61	<0.04	<0.008	6	<0.4
Spleen (97)	<0.006	130	0.11	<0.11	1200 ^(F)	<1.1	<0.1	2200	0.67		43	<0.11	0.1 ^(N)	34 ^(B)	0.24	0.05	260 ^(N)	<0.18	<0.11	<0.013	19	<1.1
Testis (53)	0.009	115	0.14	<0.09	2100 ^(F)	<0.9	<0.09	1400	0.15		12	<0.09	0.05 ^(N)	31 ^(B)	0.21	0.06	42 ^(N)	<0.1	<0.08	<0.009	15	<0.8
Thymus (2)		27	0.04	<0.04		<0.4	<0.04	370	0.07			<0.04	<1.2	260 ^(B)	0.05	0.05		<0.06	<0.04	<0.004	4.2	<0.4
Thyroid (11)	0.023	73	0.23	<0.1	2200 ^(F)	<1.0	<0.11	520	0.14		4.2	<0.1	<3.1	45 ^(B)	0.28	0.13		<0.49	<0.1	<0.01	27	<1
Tongue (3)		175	0.23	<0.11	2000 ^(F)	<1.1	0.24	1300	<0.06			<0.11	<3.2		0.14	0.07		<0.11	<0.11	<0.011	29	<1.1
Trachea (35)	0.044	260	0.18	<0.11	3000 ^(F)	<1.1	0.18	880	1.1		21	<0.11	<3.4		0.51	0.48		<0.26	<0.11	<0.01	15	<1.1
Urinary bladder (70)	0.014	130	0.14	<0.05	1600 ^(F)	<0.5	<0.08	740	0.17		6.6	<0.05	<1.4	19 ^(B)	0.25	0.15		<0.09	<0.05	<0.006	22	<0.5
Uterus (17)	0.004	110	0.10	<0.09	2000 ^(F)	<0.9	<0.23	1000	0.19		5.6	<0.09	<2.6		0.1	0.15		<0.1	<0.09	<0.009	20	<0.9
Vagina (7)		85	0.09	<0.09	2000 ^(F)	<0.9	<0.13	6100	0.16			<0.09	<2.6		0.2	0.17		<0.1	<0.09	<0.009	16	<0.9

EDUCATION, TRAINING, AND CONSULTATION

E. E. Anderson

AEC FELLOWSHIP PROGRAM

E. E. Anderson M. F. Fair

The present group of 27 AEC Fellows in Radiological Physics, one Air Force officer, and one civilian from the Watertown Arsenal completed their year of graduate study at Vanderbilt University in June 1958. The records for the year indicate that 70% of the group showed an average of "B" or better. Four of the group reported to Idaho Falls for their applied health physics training, and the remainder are now at the Laboratory. One Air force officer, who took his academic work at the University of Rochester, is taking his applied health physics training with the group at the Laboratory. Twelve of the AEC Fellows have been granted a six-month extension of their Fellowship and will work on research problems in the Health Physics Division at ORNL to complete the requirements for the M.S. degree. The AEC Fellows for the 1958-59 program were selected in March, and the group of 35 will enroll at Vanderbilt University in September.

OTHER ACTIVITIES

E. E. Anderson M. F. Fair L. C. Emerson

The authors conducted a five-week course in health physics at the Centre d'Etudes Nucleaires, Mol, Belgium, from September 28 to October 31,

1957. This was the second international course to be held in Europe, the first having been given in Stockholm, Sweden, in 1955. The course in Belgium was attended by 27 students from 19 countries and was under the joint sponsorship of the World Health Organization, the Belgian government, and the USAEC.

A 10-hr health physics lecture series was presented for the Chemical Technology Division. A special short course in health physics fundamentals was presented from January 20 to March 14, 1958, in an effort to fill an immediate need for personnel who have some understanding of the nature, scope, and magnitude of health physics problems. Students were selected in a manner that rendered maximum value to the AEC, with applications from the AEC and its major contractors receiving primary consideration. All applicants were actively engaged in health physics, industrial hygiene, or related fields and were on the payroll of the sponsoring organization. Ten persons were enrolled in the course. Lectures and discussion periods on particular phases of health physics were given for

1. The ORNL Orientation Program,
2. MIT Practice School,
3. ORINS Radioisotope Techniques Course,
4. ORINS UT-AEC Military Veterinary Radiological Health Course.

APPLIED HEALTH PHYSICS

J. C. Hart

A. D. Warden

APPLIED INSTRUMENTATION AND
MONITORING SECTIONS

D. M. Davis

Area Monitoring

H. H. Abee

Radioactive Particulate Fallout. — Fallout data from the Laboratory's 18 monitoring stations are shown in Figs. 117–120. The effect of weapons testing during the summer months is evidenced by a significant rise in activity collected at stations 11–17 and at the one remote station, located at Berea, Kentucky.

An abnormal increase in fallout was recorded at ORNL (stations 1–10) during October. Analysis showed the activity to consist of Pa^{233} affixed to Fe_2O_3 (rust) particles. Later, the source of the activity was traced to a cell ventilation duct which serviced one of the chemical processing installations. The chemical separations process was immediately discontinued, allowing time for corrective measures which included replacement of the duct and the installation of a particle-filter system.

Surface Stream Surveys. — A cooperative program with TVA, designed to determine the quantity of naturally occurring radioactive elements in certain TVA lake waters, has been undertaken. Composite samples of lake water were collected over three-month intervals by TVA personnel at seven major dam sites. All samples were analyzed at ORNL for uranium, thorium, radium, and potassium, with analysis being performed on both liquid and suspended solid phases of the samples. Results of analyses are being tabulated for later distribution.

The data from the 1954, 1955, and 1956 surveys of bottom sediment contamination in the Clinch and Tennessee Rivers have been cataloged. The surveys were made at predetermined intervals along the Clinch and Tennessee Rivers below the outfall of White Oak Creek and included gamma measurements at the surface of the bottom sediment as well as radiochemical analyses for fission product content. It is significant to note

that fission product activity in the silt increased from the summer of 1954 to the summer of 1956 by factors as high as 3.6 in the Clinch River and 2.5 in the Tennessee. The major radioactive constituents found fixed in the silt were cesium, cerium, and cobalt, with smaller amounts of strontium, yttrium, trivalent rare earths, and ruthenium.

Routine day-to-day radioactive-liquid-waste-monitoring results with respect to fission product contamination in the Clinch River downstream from ORNL are shown in Figs. 121–123. It is significant to note that at no time did the calculated weekly average concentration of radioactivity in the Clinch River exceed the MPC, for population-at-large, for drinking water.

Personnel Monitoring

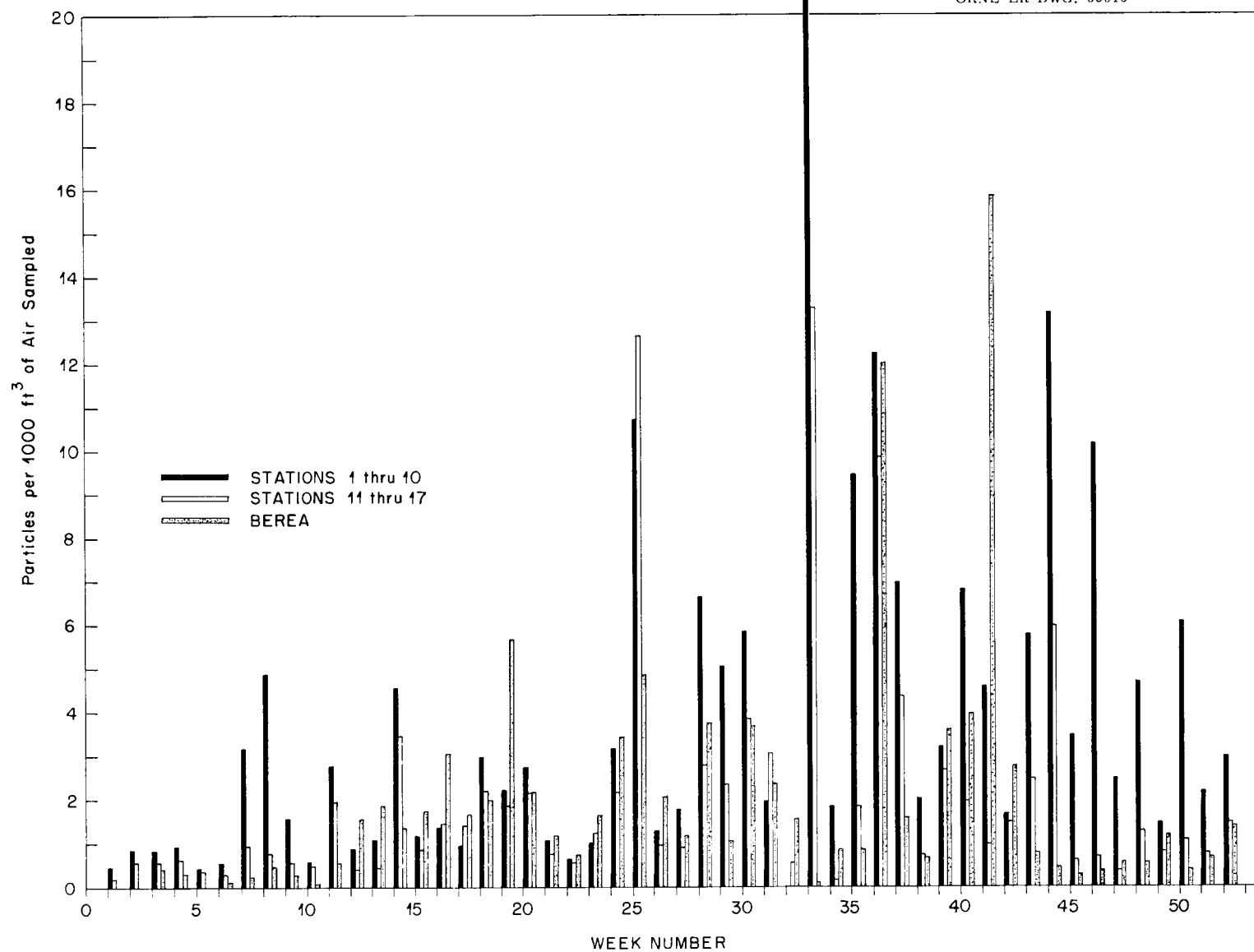
J. C. Ledbetter

Film Meters. — A new badge-film meter for ORNL employees was designed and placed in service. The new design incorporates four basic features as follows: the meter doubles as a security identification device and meter; it is relatively small and compact and weighs only 31 g; a special film emulsion is provided for gamma measurement up to 1000 rem; neutron dosimetry above 10 rad is accomplished through neutron activation of the elemental sulfur and gold components of the badge.

Assays and Instruments

E. D. Gupton

Monitoring of Airborne Plutonium. — Due to the low MPC values for airborne plutonium, hazardous quantities may be masked by the variable quantities of naturally occurring alpha emitters normally present in air. A continuous air sampler, designed to overcome this difficulty, has been assembled and tested. The principle of operation depends upon the relatively constant alpha-to-beta ratio produced by natural emitters. The device

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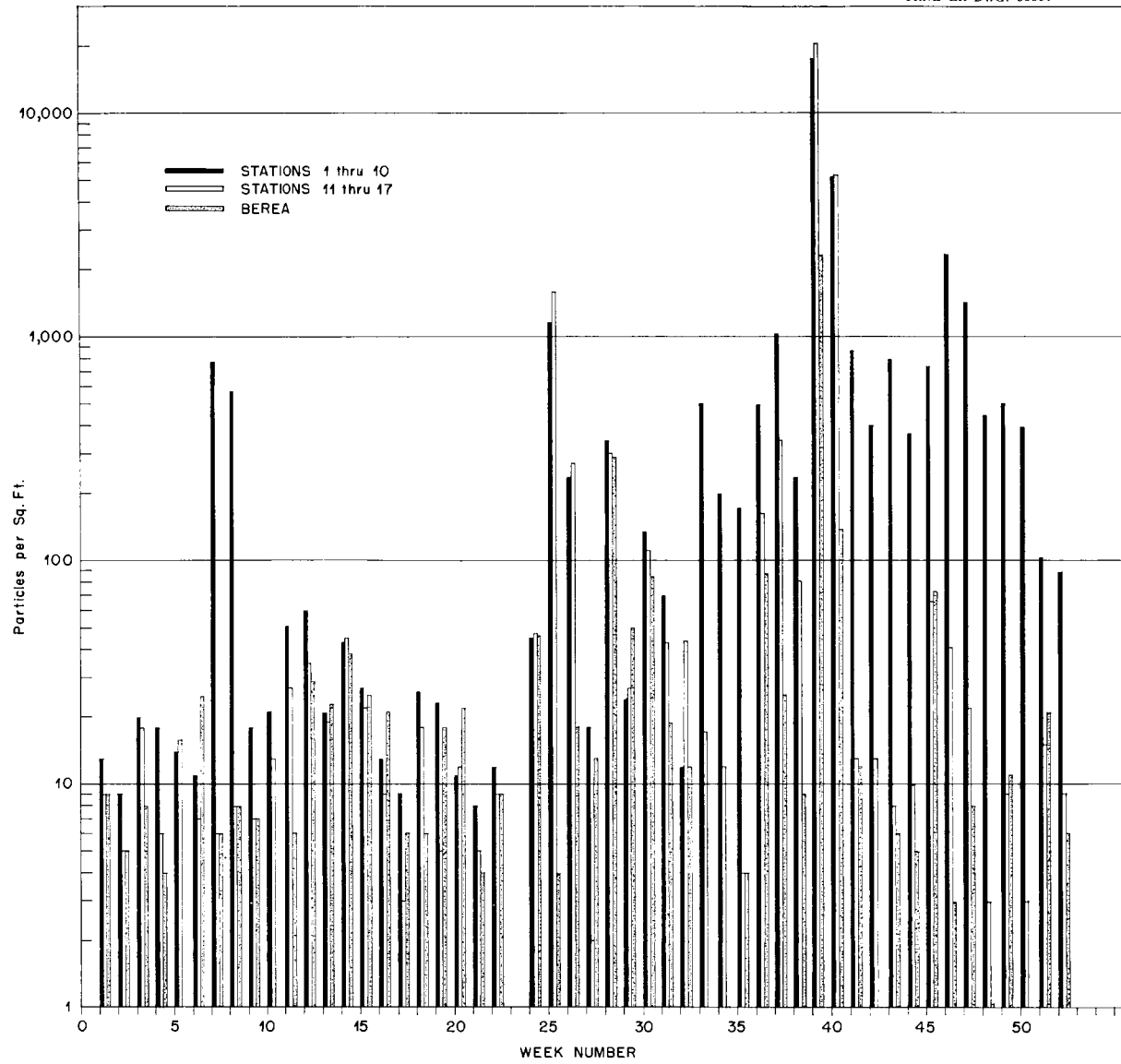


Fig. 118. Particle Fallout, 1957.

PERIOD ENDING JULY 31, 1958

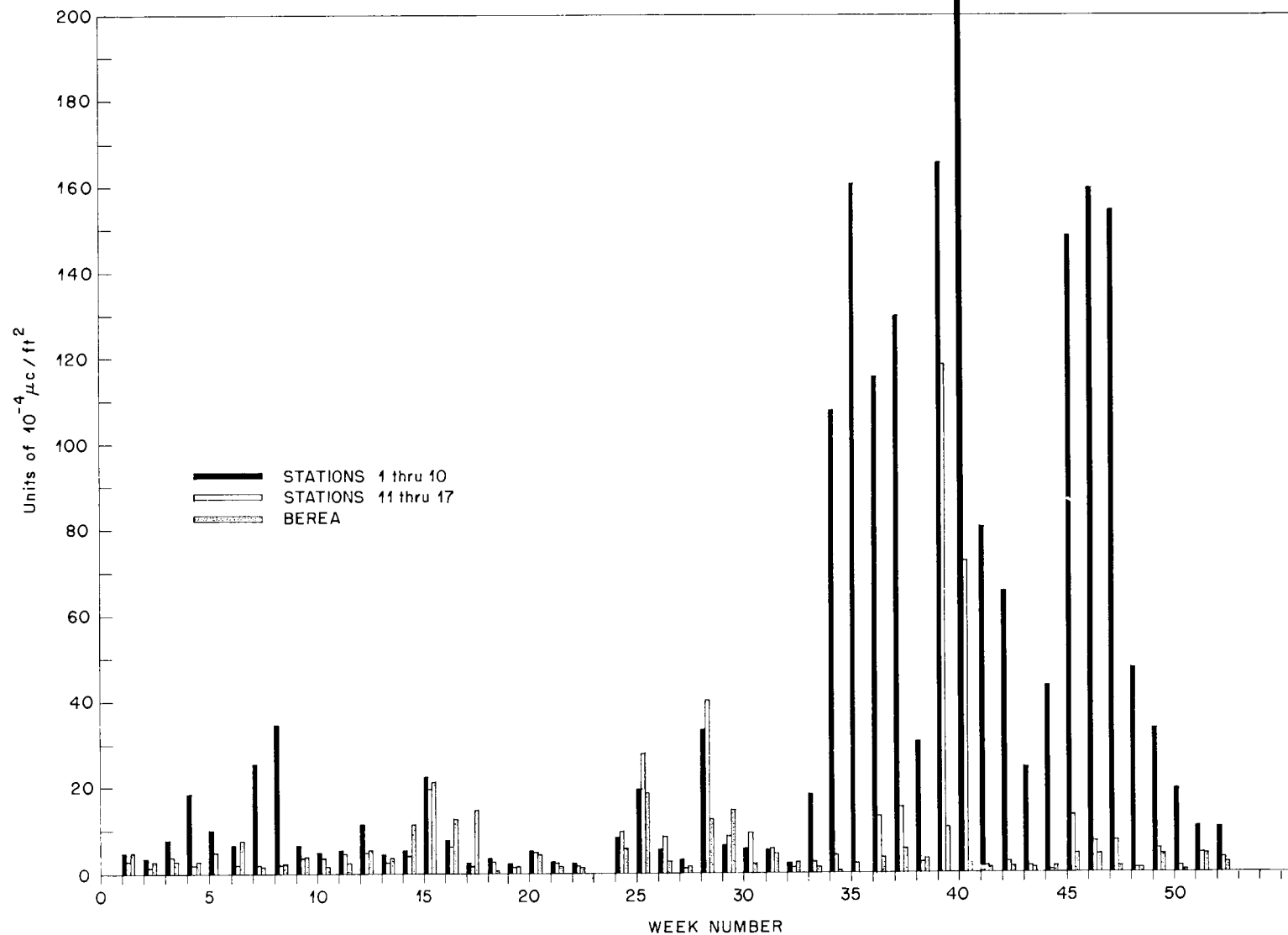


Fig. 119. Radioactive-Particle Fallout, 1957.

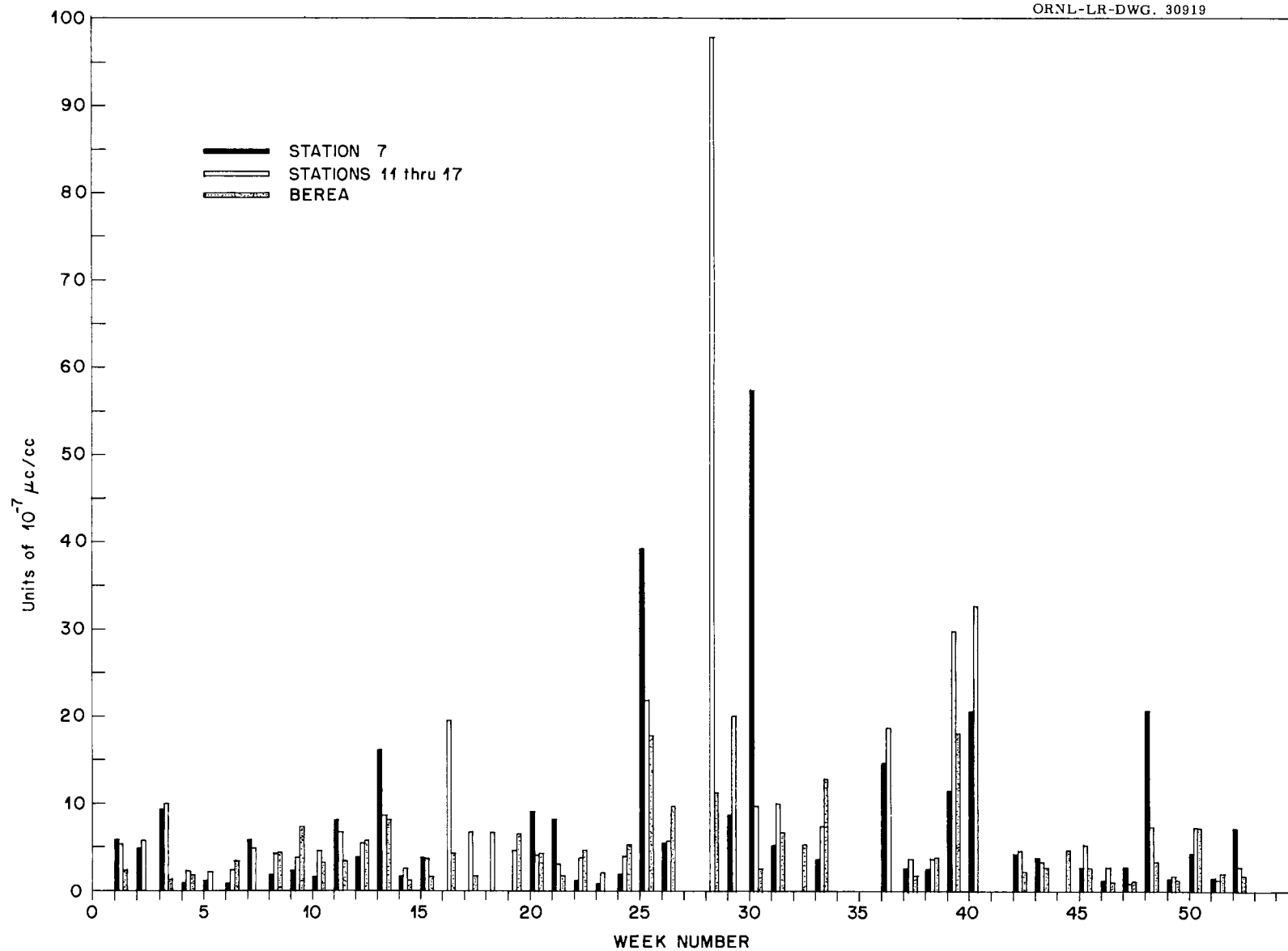


Fig. 120. Radioactivity in Rain Water, 1957. Where no data is shown, no rainfall occurred.

PERIOD ENDING JULY 31, 1958

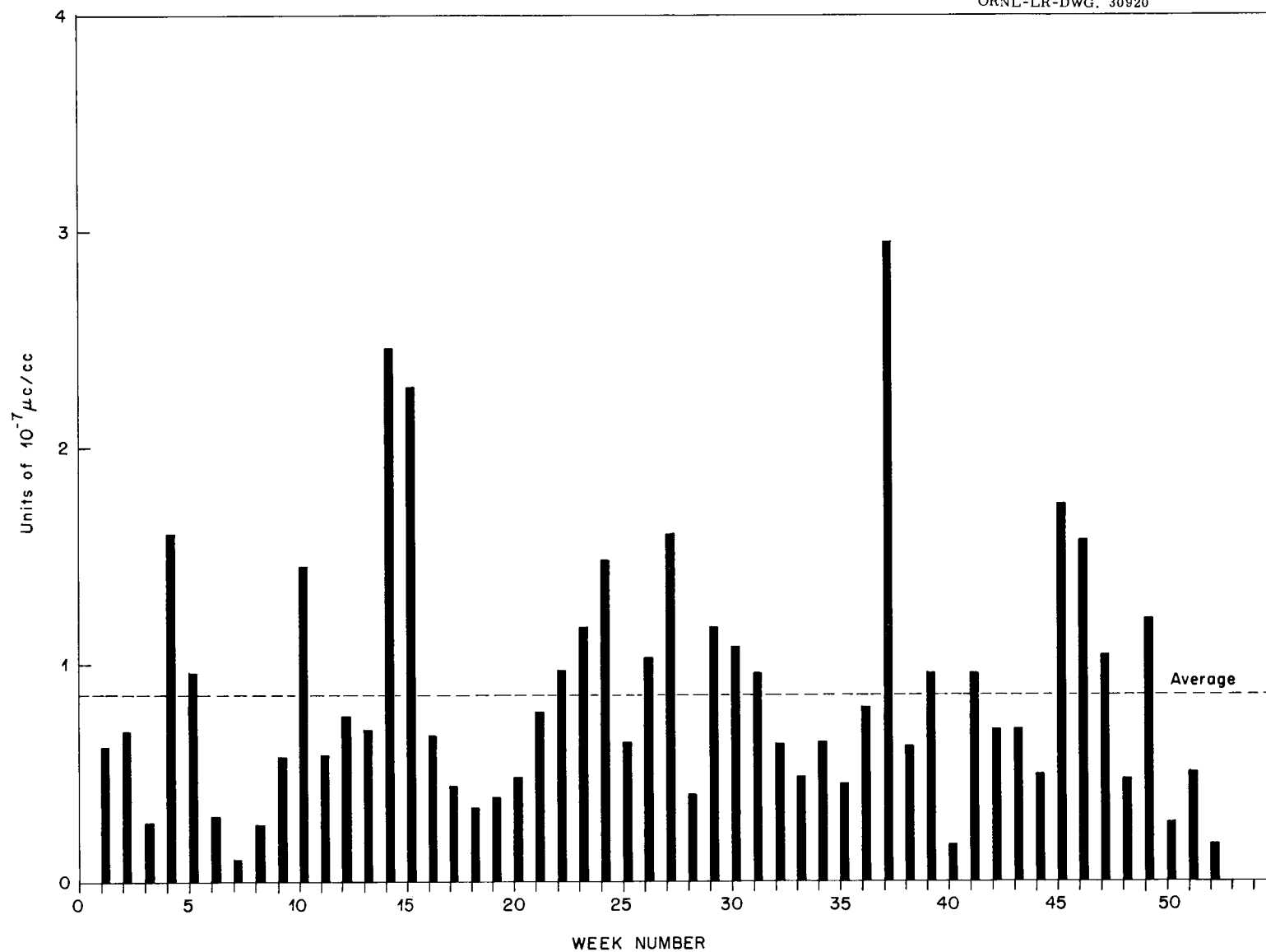


Fig. 121. Weekly Average Concentration of Radioactivity in the Clinch River, 1957.

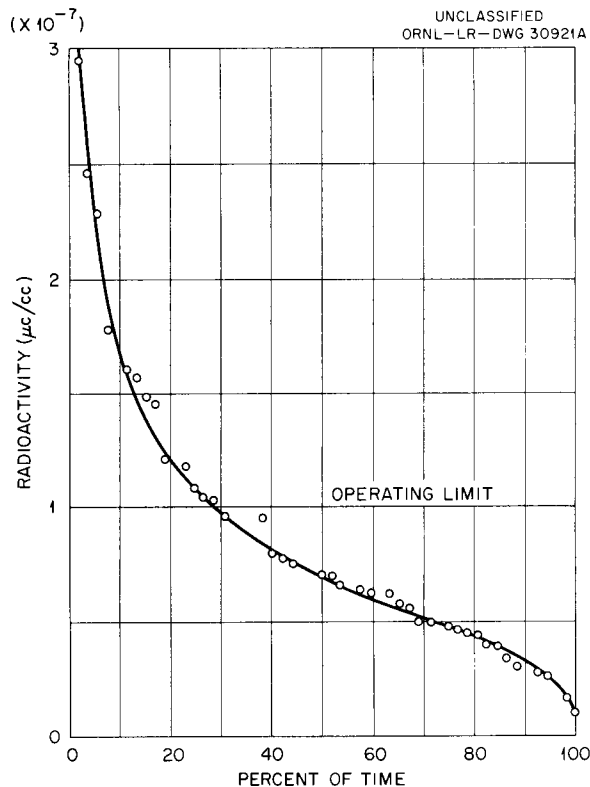
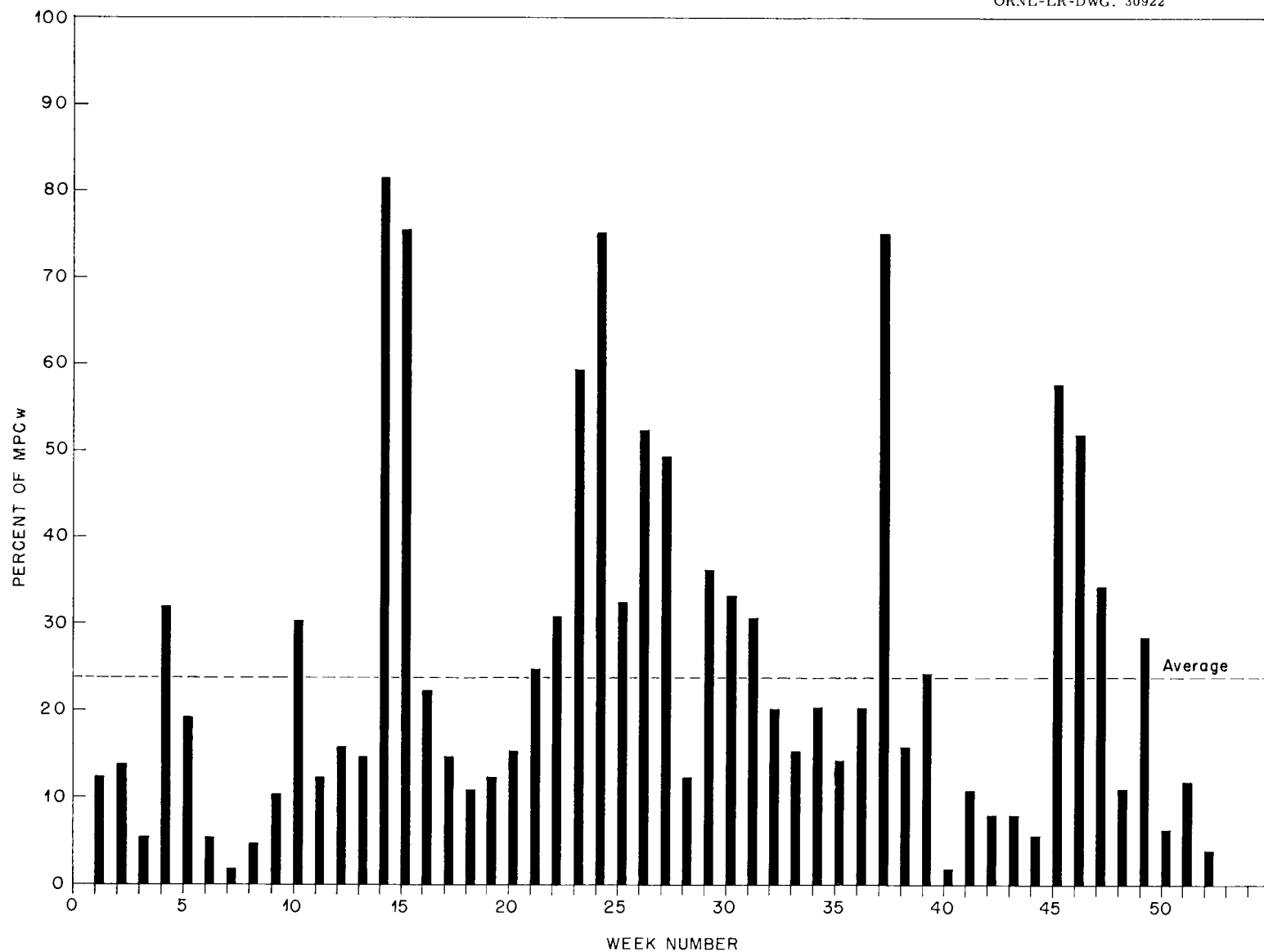


Fig. 122. Calculated Probable Average Concentration of Radioactivity in the Clinch River, 1957.

continuously samples air and gives a simultaneous read-out for both alpha and beta activity. By means of a dual-channel rate meter, the alpha and beta counting rates due to background are adjusted to cancel each other so that a zero net counting rate is obtained for background counts. Deviation from zero indicates a change in the ratio of alpha-to-beta activity, and a change of significant magnitude causes an audible alarm and sets off a series of warning lights. A read-out recorder provides the data necessary for interpretation.

Gonad Dose from Diagnostic X Ray. — Reported estimates of the average gonad dose to age 30 for the general population vary widely. In order to obtain a better estimate for the gonad dose to age 30, a cooperative program with the ORNL Health Department involving ORNL employees was undertaken. The study showed that dose varies greatly with the techniques used in the administration of the x rays. Of significance was the conclusion that doses received from medical x rays can be greatly reduced by simple planning for the use of proper filtration and collimation of the x-ray beam. At present, a second program is under way to determine the average gonad dose received per year for a typical cross section of Oak Ridge residents. Data collected for approximately 5000 residents, in all age groups, are being studied.



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PAPERS

S. I. Auerbach

Ecological Studies of Radioactive Waste Disposal, First Annual Texas Conference on the Utilization of Atomic Energy, The Texas A&M College System, March 30–April 2, 1958, College Station, Texas.

R. D. Birkhoff

Interpretation of Experiments Utilizing a Stopping Potential, Southeastern Section Meeting of the American Physical Society, April 10–12, 1958, Huntsville, Alabama.

R. D. Birkhoff, D. R. Nelson, H. H. Hubbell, Jr., and R. H. Ritchie

Measurement of Electron Flux in Media Bombarded by X-Rays, Health Physics Society Third Annual Meeting, June 9–11, 1958, Berkeley, California.

R. D. Birkhoff and R. H. Ritchie

Finite Source Considerations for the Spherical Condenser Spectrometer, American Physical Society Meeting, May 1–3, 1958, Washington, D. C.

L. C. Emerson

Preliminary Efforts Towards the Establishment of Surface Contamination Standards, Health Physics Society Third Annual Meeting, June 9–11, 1958, Berkeley, California.

H. H. Hubbell, Jr., F. N. W. Noll, and J. S. Cheka

Comparison of Theoretical and Experimental Filtered X-Ray Spectra, Radiological Society of North America, November 17–22, 1957, Chicago, Illinois.

A. P. Hull (presented by H. H. Hubbell, Jr.)

A Small Personnel Dosimeter for Beta and Gamma Radiation, Health Physics Society Third Annual Meeting, June 9–11, 1958, Berkeley, California.

R. J. Morton

Community Health Problems Due to Peace Time Uses of Radiation, Radiation Protection Symposium, American Public Health Association Annual Meeting, October 11–15, 1957, St. Louis, Missouri.

Expanding Scope and Concepts of Environmental Health, First General Session, Annual Health Conference, June 9–12, 1958, Rochester, New York.

R. H. Ritchie and G. S. Hurst

Penetration of Weapons Radiation, Health Physics Society Third Annual Meeting, June 9–11, 1958, Berkeley, California.

W. S. Snyder

Calculation of Radiation Dose, Health Physics Society Third Annual Meeting, June 9–11, 1958, Berkeley, California.

J. C. Villforth, H. H. Hubbell, Jr., and R. D. Birkhoff (presented by H. H. Hubbell, Jr.)

Calibration of a Scintillation Spectrometer at Low Energies, Southeastern Section Meeting of the American Physical Society, April 10–12, 1958, Huntsville, Alabama.

E. B. Wagner and G. S. Hurst

Advances in the Standard Proportional Counter Method of Fast Neutron Dosimetry, Fourth Annual Meeting of Professional Group on Nuclear Science of the Institute of Radio Engineers, October 30–November 1, 1957, New York, New York.

LECTURES

E. E. Anderson

Nuclear Energy Fundamentals, University of North Dakota, April 15, 1958, Grand Forks.

Radiation Hazards and Current Practices in Radiation Protection, Knoxville College, March 24, 1958, Knoxville, Tennessee.

Four lectures at Iowa State Teachers College, Cedar Falls, April 17, 1958: *Biological Effects of Ionizing Radiation*; *Nuclear Energy Fundamentals*; *Radiation Hazards and Current Practices in Radiation Protection*; *Radioactive Waste Disposal*.

Radiation Hazards and Current Practices in Radiation Protection, University of Tennessee, December 4, 1957, Memphis.

Radiation Hazards and Current Practices in Radiation Protection, University of Texas, December 11, 1957, Houston.

Health Physics, Veterinary Radiological Health Course, sponsored by UT-AEC Experiment Station, March 24 and June 27, 1958, Oak Ridge, Tennessee.

Principles of Health Physics, Oak Ridge Institute of Nuclear Studies, January 17 and May 29, 1958, Oak Ridge, Tennessee.

E. E. Anderson and M. F. Fair

Health Physics Fundamentals, series of lectures from January 20 to March 14, 1958, ORNL, Oak Ridge, Tennessee.

Health Physics Lecture Series, Chemical Technology Division, ORNL, Oak Ridge, Tennessee (10 hour course, November and December 1957).

S. I. Auerbach

The Role of Biological Sciences in Health Physics, Symposium on Education and Training for Health Physics, December 3, 1957, Oak Ridge, Tennessee

Environmental Surveillance, Environmental Health Aspects of Nuclear Reactor Operations Course, Robert A. Taft Sanitary Engineering Center, January 31, 1958, Cincinnati, Ohio.

Long Range Objectives in Radioecology, Bio-Medical Program Directors Meeting Savannah River Plant, February 17, 1958, Aiken, South Carolina.

Ecology and Radiation Ecology, Health Physics Radiological Physics Course, ORNL, March 12, 1958, Oak Ridge, Tennessee.

Ecological Studies in Radioactive Waste Areas, presented to the following groups:

1. ORSORT Students, ORNL, January 28, 1958, Oak Ridge, Tennessee.
2. Zoology Department, University of Georgia, January 20, 1958, Athens.
3. Beta Kappa Chi Scientific Society, Central State College, February 4, 1958, Wilberforce, Ohio.
4. Zoology Department, South Illinois University, February 5, 1958, Carbondale.
5. Medical Branch, The University of Texas, April 2, 1958, Galveston.
6. Beta Beta Beta Biological Society, Berea College, May 2, 1958, Berea, Kentucky.
7. Science Club, Mississippi State College, May 20, 1958, State College.

R. D. Birkhoff

The Response of Anthracene to Monoenergetic Electrons, Health Physics Seminar, ORNL, October 2, 1957, Oak Ridge, Tennessee.

K. K. Bohnsack

Soil Arthropod Studies in the Vicinity of the ORNL Radioactive Waste Pits, Health Physics Division Seminar, ORNL, August 1957, Oak Ridge, Tennessee.

HEALTH PHYSICS ANNUAL PROGRESS REPORT

K. E. Cowser

Waste Handling Treatment and Disposal at ORNL, presented to the following groups:

1. U. S. Public Health Service, Program on Environmental Health Aspects of Nuclear Reactor Operations, December 12, 1957, Cincinnati, Ohio.
2. Oak Ridge Institute of Nuclear Studies, 16th and 17th Session of Veterinary Radiological Health Course, March 17 and 23, 1958, Oak Ridge, Tennessee.
3. American Petroleum Institute Committee on Waste Disposal, January 23, 1958, Oak Ridge, Tennessee.

D. A. Crossley, Jr.

Fission Products in Insect Populations on Vegetation, Health Physics Division Seminar, ORNL, October 30, 1957, Oak Ridge, Tennessee

L. C. Emerson

Health Physics, Centre D'Études pour les Applications de l'Énergie Nucleaire, October 1-31, 1957, Mol, Belgium.

Health Physics, Shielding Reactor Safety, presented to the following groups:

1. TVA Engineers Association, December 20, 1957, Huntsville, Alabama.
2. TVA Engineers Association, March 21, 1958, Rogersville, Tennessee.
3. TVA Engineers Association, April 11, 1958, Nashville, Tennessee.
4. TVA Engineers Association, April 12, 1958, Paducah, Kentucky.

Radiation Units, American Petroleum Institute Committee on Radioactive Waste, January 22, 1958, Oak Ridge, Tennessee.

Health Physics Lecture Series AEC Short Course in Health Physics Fundamentals, ORNL, March 3-5, 1958, Oak Ridge, Tennessee

Radiation Hazards Associated with Self-Sintering, Applied Health Physics Seminar, ORNL, March 7, 1958, Oak Ridge, Tennessee.

Reactor Safety, Vanderbilt University, March 23 and 25, 1958, Nashville, Tennessee.

Waste Disposal, Vanderbilt University, April 21-22, 1958, Nashville, Tennessee.

Course in Health Physics Fundamentals, personnel in Health Physics Division, ORNL, 1957-1958, Oak Ridge, Tennessee.

M. F. Fair

Health Physics, Veterinary Radiological Health Course, sponsored by UT-AEC Experiment Station, March 17, 1958, Oak Ridge, Tennessee.

Principles of Health Physics, Oak Ridge Institute of Nuclear Studies, September 20, 1957 and May 8, 1958, Oak Ridge, Tennessee.

J. C. Howell

A Bird Census of Melton Valley, Health Physics Division Seminar, ORNL, August 14, 1957, Oak Ridge, Tennessee.

H. H. Hubbell, Jr.

What the Health Physicist Should Know About X-Rays, Health Physics Seminar, ORNL, November 13, 1957, Oak Ridge, Tennessee.

A. P. Hull

Some Designs, Past and Present, in Beta Dosimetry, Health Physics Seminar, ORNL, March 5, 1958, Oak Ridge, Tennessee.

J. B. Lackey

Ecological Studies on Clinch River Plankton, Health Physics Division Seminar, ORNL, September 11, 1957, Oak Ridge, Tennessee.

R. J. Morton

Preparation for Public Health Responsibilities in Radiation Protection, Monthly Student-Faculty Seminar, School of Public Health, University of North Carolina, January 6, 1958, Chapel Hill.

Radioactive Waste Disposal Research, ORSORT Students, ORNL, April 14, 1958, Oak Ridge, Tennessee.

Environmental Considerations in the Location of Nuclear Power Facilities, Luncheon Meeting of Sanitation Section, New York Public Health Association, Annual Health Conference, June 9-12, 1958, Rochester, New York.

D. Nelson

Energy Distribution of Electrons in Irradiated Media, Health Physics Seminar, ORNL, February 5, 1958, Oak Ridge, Tennessee.

R. H. Ritchie

Application of Plumbbob Data to Ichiban Dosimetry, Health Physics Seminar, ORNL, December 4, 1957, Oak Ridge, Tennessee.

C. C. Sartain

Current Trends in the Application of Nuclear Devices, Health Physics Seminar, ORNL, September 18, 1957, Oak Ridge, Tennessee.

W. S. Snyder

Sampling Procedures to Determine Acute Radiation Effects in Hiroshima and Nagasaki, Health Physics Seminar, ORNL, November 7, 1957, Oak Ridge, Tennessee.

Approximate Solutions of Fredholm Integral Equations, University of Tennessee Mathematics Colloquium, February 26, 1958, Knoxville.

W. S. Snyder and J. A. Auxier

Theoretical and Experimental Aspects of Reciprocity, Health Physics Seminar, July 30, 1958, Oak Ridge, Tennessee.

E. B. Wagner

Advances in Fast Neutron Dosimetry, Health Physics Seminar, October 16, 1958, Oak Ridge, Tennessee.

L. G. Williams and H. D. Swanson

Uptake of Cesium-137 by Algae of Clinch River and its Tributaries, Health Physics Division Seminar, ORNL, August 21, 1957, Oak Ridge, Tennessee.

H. P. Yockey

Science and Information Theory, Instrumentation and Controls Division Seminar, ORNL, September 9, 1957, Oak Ridge, Tennessee.

Information Theory, Aging and Radiation Damage, Health Physics Division Seminar, ORNL, September 25, 1957, Oak Ridge, Tennessee.

Information Theory in Biology, Electronuclear Research Division Seminar, ORNL, October 1, 1957, Oak Ridge, Tennessee.

The Concept of Information Theory in Biology, Biological Seminar, Medical College of Virginia, November 6, 1957, Richmond.

The Concept of Information Theory in Biology, Mathematics Colloquium, University of Tennessee, November 13, 1957, Knoxville.

Application of Information Theory to Biological Radiation Damage, Applied Nuclear Physics Division Seminar, April 9, 1958, Oak Ridge, Tennessee.

Information Theory and the Biophysics of Aging and Radiation Damage, Physics Division Seminar, Vanderbilt University, May 16, 1958, Nashville, Tennessee.

Information Theory and Statistical Mechanics, Theoretical Physics Seminar, June 2, 1958, Oak Ridge, Tennessee.

Progress reports previously issued by the Health Physics Division are as follows:

ORNL-1596	Period Ending July 31, 1953
ORNL-1684	Period Ending January 31, 1954
ORNL-1763	Period Ending July 31, 1954
ORNL-1860	Period Ending January 31, 1955
ORNL-1942	Period Ending July 31, 1955
ORNL-2049	Period Ending January 31, 1956
ORNL-2151	Period Ending July 31, 1956
ORNL-2384	Period Ending July 31, 1957

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