HAZARDS CONTROL PROGRESS REPORT NO. 52

January through June 1976

August 15, 1976

Prepared for U.S. Energy Research & Development
Administration under contract No. W-7405-Eng-48

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Printed in the United States of America

Available from

National Technical Information Service

U. S. Department of Commerce

5285 Port Royal Road

Springfield, VA 22161

Price: Printed Copy \$5 Microfilm \$13.00

Page Range	Domestic Price	Page Range	Domestic Price
001-025	\$ 2.50	326-350	10.00
026-050	4.00	351-375	10.50
051-075	4.50	376-400	10.75
076-100	5.00	401-425	11.00
101-125	5.50	426-450	11.25
126-150	6.00	451-475	12.00
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LAWRENCE LIVERMORE LABORATORY
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UCRL-50007-76-1

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Foreword

This report is divided into three major sections. The first section, *Progress Reports*, covers the status of activities undertaken or continuing during the period; additional reports or separate publications will cover the final results of these activities. The second section, *Technical Notes*, contains reports on interesting activities of a more limited scope on which further reporting is not anticipated. The third section lists recent publications.

Readers who are interested in more detail regarding any item may contact the authors of the reports who are listed in the Contents.

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Progress Reports

RADIATION PROTECTION

Further Realistic Torso Phantom Development

Introduction. In previous Hazards Control Progress reports,^{1,2} we discussed the progress on construction of a realistic torso phantom to be used for calibrating detectors designed to measure heavy elements such as ²³⁹Pu *in vivo*. This report covers further work on phantom construction, including reconstruction of the rib cage, preliminary measurements to establish chest contour, final shaping of the major organs, and investigation of tissue-equivalent (T.E.) material.

Organ Shaping. After dissecting the cadaver that was the source of the phantom rib cage, both the organs and the foam mold of the torso cavity developed some distortion. As a result, the lungs, heart and liver had to be reshaped to assure close fit and compatibility. The lungs in particular had shrunk significantly.

We used the reshaped plaster cast of the torso void² to make a plastic female mold that represented the eventual phantom cavity. The plaster lung casts were put in position in the cavity, and plaster was added to fill out each one. In doing so we used the cavity as a mold for the outside surface of the lungs.

After reshaping the lungs, we reshaped the heart to fit in place between the lungs. Dr. Sexton Sutherland, an anatomist at the University of California Medical Center, San Francisco, guided us in that part of the work. After reshaping the heart, we filled the space between the lungs that would normally contain large blood vessels, esophagus, and trachea with silicon rubber (Fig. 1). That was done as a temporary step

Fig. 1. Plaster casts of lungs and heart with silicon rubber filler, in-place in torso cavity.

so that we could reshape the liver cast. In the phantom, the same space will be filled with muscle-equivalent plastic which has a hollow esophagus and depressions for radioactive "lymph nodes."

We reshaped the liver cast by adding plaster to conform to the cavity surface at the front of the torso, and removing material from the back of the liver so that the original volume was retained. In the finished phantom the spleen and kidneys will be imbedded, as cast from the originals, in muscle-equivalent material used to simulate absorption by the intestines and other surrounding tissue.

As a result of reshaping, the lung and heart volumes were changed; however, the liver volume is essentially the same. The new lung volumes are 2180 ml right and 1689 ml left, for a total of 3869 ml compared with a published value for "average adult" of 3915 ml.³ The heart volume was reduced from 865 to 748 ml, compared with an average of 742 ml for 168 adult males.³ Although the purpose of organ reshaping was better fit, we seem to have achieved more appropriate size as well.

Reconstructing the Rib Cage. We dissected and cleaned the trabecular bone of the rib cage. As a result, the bone marrow was removed. A method for replacing the marrow with MIX-D,⁴ a tissue-equivalent wax-based material, had previously been used successfully,² so we used the technique to fill all of the bones in the rib cage. The method simply requires submersing the bones in molten MIX-D in an evacuated chamber. The chamber is brought to atmospheric pressure to force the MIX-D into the bones and then they are removed, cooled, and scraped. After scraping, we removed traces of the wax from most of the rib surface by micro-blasting with CaCO₃ powder to improve the bond of the polyurethane tissue-equivalent plastic after phantom assembly.

The final step was to assemble the rib cage by drilling fine holes in the ribs and vertebrae so they could be pinned together with nylon tennis racket string. We used nylon squid line to maintain rib spacing and add strength to permit handling. The assembled rib cage is shown on the plaster torso cavity cast (Fig. 2).

Torso Contour. An important part of plutonium measurement in the lungs by external counting is determination of the fraction of low-energy x-rays transmitted through the overlying tissue. Differences in human physique can cause very large differences in x-ray transmission. Hazards Control whole-body

Fig. 2. Assembled rib cage on plaster cast of torso cavity.

Fig. 3. Torso cavity and ribs with chest contour in place. Circles on chest contour represent normal detector position.

counter operators have adopted an ultrasonic technique used in medicine for organ imaging to measure the thickness of tissue at selected points between lung and detector.⁵ The chest thickness for a given person is characterized by an exponential average of tissue thickness at 70 to 80 points under the detectors. When this technique is used, the usual range of adult males is 20 to 25 mm, with virtually all males falling in the range 15 to 45 mm.

On the basis of this information, we have decided to construct the basic phantom with a 15-mm chest wall thickness. We will cover the range of physiques by add-on chest plates of T.F. material in four thicknesses up to a maximum chest wall of about 45 mm.

Because the cadaver had a chest wall thickness greater than 20 mm, we will have to reduce the external torso cast appropriately. Therefore, a clear plastic chest plate was cast from the plaster mold and marked with a grid using 25-mm spacing. We drilled holes at the grid points, then aligned the chest plate over the void mold and rib cage (Fig. 3). We mapped the chest wall thickness i.e., plaster-to-plastic-shield separation over the chest and upper abdomen. The chest plate will be transferred to the original torso cast. The cast can then be reduced at the grid points by the difference between the mapped separations and the desired chest wall thickness, as determined from ultrasonic measurement data for a number of workers.

Tissue-Equivalent Materials. For purposes of phantom construction, "tissue equivalence" means that the linear transmission properties of the material for photons with energies greater than 15 keV will simulate those of tissue. The density, effective atomic number (Z) and electron density all contribute to simulation; however, it is not necessary to match each of those properties to achieve functional simulation.

The tissues to be simulated include bone, muscle, adipose, cartilage, and lung material. The human rib cage provides the necessary bone simulation, but the other materials will be simulated with plastic materials. The muscle-equivalent material will make up the greatest portion of the phantom mass. It will be used for the basic phantom, add-on layers of chest tissue over the lungs, and the major organs except for the lungs. Adipose-equivalent material will be present only in a few add-on layers. Cartilage, which has a density approximately 10% greater than muscle, will be used only to connect ribs to sternum.

We have selected polyurethane for use as the basic material for tissue simulation because of its durability and low Z composition which nominally is 9% hydrogen, 66% carbon, 4% nitrogen, and 20% oxygen, with a density of 1.06 g/cm³. For lung-equivalent material, we plan to use foamed polyurethane with a

Fig. 4. Relative transmission, ^{239}Pu L x rays.

	ρ (g·cm ³)
(1) Polyethylene	0.93
(2) Animal fat	0.90
(3) Polyurethane	1.06
(4) Lucite	1.18
(5) Mix-D (LLL)	0.97
(6) Rando muscle	1.03
(7) Temex	1.01
(8) Water	1.00
(9) Polyurethane +4.3% CaCO ₃	1.08
(10) Beef steak	1.03
(11) Frigero T. F. fluid	1.06

density of about 0.25 g·cm⁻². Polyurethane is more transparent to low-energy x rays than muscle, so a higher-Z material must be added to reduce the linear transmission to an appropriate value. Preliminary measurements were reported² with polyurethane containing 3.5 wt% sulfur. Because of the sulfur odor, we have made samples with 4.3% CaCO₃ instead of sulfur. A comparison of the attenuation of polyurethane and a number of other tissue-equivalent materials as well as animal tissues and reference liquids are shown in Fig. 4. Compositions for most of these materials appear in Table 1.

Table 1. Elemental composition of tissue-equivalent materials

Material	Element												
	H	C	N	O	Na	Mg	P	S	Cl	K	Ca	Ti	Fe
Polyethylene ^a	14.4	85.6											
Adipose ^b	12.0	64.0	0.8	22.9	0.051	0.002	0.016	0.073	0.12	0.032	0.002	-	0.002
Polyurethane ^c	9.22	68.9	3.71	18.2									
Lucite ^d	7.57	59.5	32.9										
Mix-D ^d	13.5	77.7	3.4			3.9						1.4	
Rando muscle ^e	8.83	64.4	4.05	20.35					2.24	(Sb - 0.08)			
Temex ^f	9.6	87.0	0.06	0.47				1.53	(Zn - 0.45)			0.33	
Water ^g	11.2			88.8									
Polyurethane ^e + 4.3% CaCO ₃	8.82	66.4	3.55	19.5							1.72		
Muscle ^h	10.2	12.3	3.5	72.9	0.08	0.02	0.2	0.5		0.3	0.007		
Frigero ^h	10.2	12.3	3.5	72.9	0.07	0.02	0.2	0.32	0.68	0.39	0.01		
T. F. fluid													
Cartilage ⁱ	10.4	10.4	2.7	75.2	0.4			0.6	0.3				
Rando (C-15) ^c	5.67	74.2	2.02	18.2		(Sb - 0.08)							

Note: composition of specific sample depends on date of purchase.

^aBy direct calculation.

^bRef. 6.

^cRef. 9.

^dLLL analysis.

^eBy calculation, 60.8% paraffin

30.4% (CH₂)_n, 6.4% MgO, 2.4% TiO₂.

^fRef. 7.

^gRef. 8.

^hRef. 9.

ⁱRef. 10.

^jRef. 3.

Fig. 5. Albedo dosimeters studied.

At this point we need to define more specifically what physical material we intend to use as a reference for simulation. At 15 keV, the linear attenuation of beefsteak and water differ significantly from Frigierio muscle-equivalent fluid. It is unclear which of these best simulates the tissue of the human male chest. Because our beefsteak and water attenuation curves are rather close and because water is universally available as a reproducible absorber, we have chosen to use water as our transmission standard for lean tissue or muscle. The reference for cartilage will then be the

Fig. 6. Response dependence for various source-moderator configurations.

Fig. 7. Four albedo badge designs calibrated with a Cf source in various thicknesses of hydrogen moderation.

absorption by a thickness of water 10% greater than the physical thickness simulated (an equivalent density of $1.10 \text{ g}\cdot\text{cm}^{-2}$). For lung-equivalent material, we will use absorption by a layer of water having a thickness 25% of the physical thickness of lung simulant (i.e., an equivalent density of $0.25 \text{ g}\cdot\text{cm}^{-2}$). We have not yet chosen an adipose reference.

Work Remaining. After reshaping the chest contour we will cast the torso shell, including the rib cage. We will have to develop a technique for making polyurethane foam lungs with the proper density ($\sim 0.25 \text{ g}\cdot\text{cm}^{-2}$) and loaded with microcurie quantities of plutonium. All the organs will be cast in T.E. plastic, and T.E. material will be used to simulate absorption of body tissue other than major organs. We will have to make specific provisions for the esophagus and lymph node spaces.

Acknowledgments. We wish to thank Norm Boyer and Robert Taylor in the plastics shop and Charles Harder in Hazards Control for their support.

Albedo Neutron Dosimeter Development

Work previously reported^{11,12} covered studies of

the response of an albedo-type neutron dosimeter to various neutron source and moderator configurations. This work has been expanded to include several different badge configurations. Figure 5 depicts the badge designs evaluated in this manner. The dosimeter was exposed while in contact with polyethylene water-filled phantoms 166 mm thick, 267 mm wide, and 180 mm high. Figure 5(a) is the design previously reported. Figure 5(b) is a modified version of the dosimeter designed by Hankins while at LASL,¹³ including a TLD-600/700 pair on top of the normal dosimeter package. Figure 5(c) is similar to that developed by Boggs.¹⁴ It consists of a TLD-600/700 pair shielded from incident thermal neutrons and a TLD-600/700 pair unshielded, both with a thermal neutron shield between the TLDs and the reflecting body. These designs were studied primarily in an effort to reduce the dependence of a typical albedo dosimeter response on the separation distance from the reflecting body. The data were reduced as previously reported¹¹ i.e., a comparison of the ratio of incident-to-albedo neutron response (D_1/D_2) with the sum of their response in apparent R per rem ($\Sigma(D_1 \text{ or } D_2) R/\text{rem}$). A summary of that work is plotted in Fig. 6 along with a similar analysis of the modified Hankins badge

from data taken by Hopkins.¹⁵ The dependence of this type of analysis on source-moderator configuration for either badge type can clearly be seen. The sources, moderators, and calibration facility have been described in a previous HCL Progress Report.¹⁶ Figure 7 is a plot of the three dosimeter design (the third design being tried with each cadmium and boron as the neutron shield) results when calibrated with a ²⁵²Cf source in various thicknesses of hydrogen-containing moderator (polyethylene and water).

Adding a thermal neutron shield between the body and the dosimeter greatly decreases the sensitivity of the dosimeter response to separation distances. In the non-shielded case, only a 25-30% decrease was

observed at distances up to 5 cm. However, this was considerably different from the free air exposure situation, even though doubling the shield thickness did not alter badge response when the badge was in contact with the phantom. There was, as can be noted in Fig. 7, a considerable loss in sensitivity to higher energy neutrons due to the additional shield. The difference in shape of the data taken with the Cd shield from that with the B shield is probably due to differences in neutron absorption cross section. The greater difference at higher effective neutron energies is due to the dosimeter's greater dependence on albedo neutrons in that region. Varying the shape of the front shield influenced the data very little.

INDUSTRIAL HYGIENE

Enhanced Filtration Test System (EFTS) Aerosol Generator

The EFTS filter test system has been designed for using various types of aerosols to evaluate enhanced filtration methods. To date, all testing with the EFTS has been done with the modified Wright¹⁷ design nebulizer shown in Fig. 8. Modification of the Wright design consists of changes in critical dimensions of the nebulizer jets and construction materials. The EFTS nebulizers are constructed of hardened brass and stainless steel rather than plastic to allow for operation

at pressures greater than 207 kPa, and to reduce the rate of orifice erosion. A comparison of the critical dimensions of various Wright nebulizers is shown in Table 2.

Table 2. Critical dimensions of various Wright nebulizers (see Fig. 8)

Nebulizer	Critical dimensions (mm)		
	(a)	(b)	(c)
Chalk River (plastic)	0.787	1.499	0.864
Wright No. 3 (plastic)	0.864	1.608	1.115
Wright No. 4 (plastic)	0.848	1.603	1.115
IASI built (plastic)	0.742	1.585	1.156
EFTS (brass)	0.635	1.730	1.240
EFTS (stainless steel)	0.635	1.650	1.245
EFTS (plastic)	0.748	1.603	1.194

Fig. 8. Cutaway drawing of Wright nebulizer.

Table 3 compares the Mass Media Aerodynamic Diameter (MMAD) and geometric standard deviation (σ_g) of three modified Wright-type nebulizers at comparative pressures. Each of the three nebulizer configurations produces a distinct particle size (MMAD) and a given pressure, with a moderate degree of polydispersity. This permits comparative analysis of filter (or process) efficiency as a function of particle size.

Aerosol particle size can be further regulated by varying the input pressure to the nebulizer, and by varying the solution strength (wt% NaCl dissolved in water). Decreasing the nebulizer pressures increases the

Table 3. Particle size^a of the EFTS aerosol nebulizer types

Nebulizer	Jet material	Jet dimensions		Pressure		MMAD	σ_g
		Orifice diam (mm)	Slit wt (mm)				
				(kPa)	(psi)		
#1L ^b	Lucite	1.60	1.19	207	30	0.67	2.1
#1 Gd to #4 Gd	Gold ^c	1.73	1.21	207	30	0.82	2.25
				276	40	0.80	2.1
				345	50	0.76	2.05
#1 Gd to #4 G ^c	Stainless steel	1.65	1.245	207	30	1.2	2.3
				276	40	0.96	2.0
				345	50	0.93	2.06

^aParticle size as measured by six-stage cascade impactor (Ref. 18).

^bMaximum safe operating pressure for Lucite nebulizer jets is 207 kPa (30 psi).

^cGold plate (0.0005 in. thick); over hardened brass.

MMAD, and decreasing the percent of NaCl in solution decreases the MMAD.

The concentration (mg/m³) of NaCl particulate can be regulated by varying the number of nebulizers used. Present testing procedures require maintaining a relatively constant concentration of aerosol at flowrates of 5.9, 11.8, 17.7, and 23.6 l/s. For current filter efficiency studies, one nebulizer is being used for each 5.9 l/s airflow increment.

Figure 9 illustrates the NaCl aerosol concentration as a function of nebulizer pressure at a flow rate of 23.6 l/s for each individual nebulizer, and for combinations of two and four nebulizers.

Other aerosols, such as DOP, methylene blue and uranine, are compatible with the flow characteristics of the 25-l/s EFTS, and are under consideration for future experimentation. For initial investigations, however, the air-generated NaCl aerosol is ideal for real-time analysis (by flame photometer) of filtration methods less than 99.9% efficient.

Generation of Monodisperse Test Aerosols

One of the major goals of the enhanced filtration program is to determine the efficiency of HEPA prefilters as a function of particle size. To make such

measurements, we have acquired a monodisperse aerosol generator with a vibrating orifice.

The generator operates by feeding a liquid solution through a 5-, 10- or 20- μ orifice vibrated at some applied frequency. The frequency of vibration, as well as the solution feed rate and concentration, govern the particle size distribution according to the equations.

Fig. 9. NaCl aerosol concentration versus nebulizer pressure (stainless steel nebulizer jets); 23.6 l/s flowrate; 1% NaCl solution; for various combination of Wright-type nebulizer.

$$D_d = \left(\frac{6Q}{\pi v} \right)^{1/3} \quad (1)$$

and

$$D_p = (C + 1)^{1/3} D_d \quad (2)$$

where

D_d = liquid droplet diameter (cm),

Q = solution feed rate (cm³/min),

v = frequency of vibration (Hz),

D_p = solid particle diameter (cm),

C = volumetric concentration (cm³/cm³), and

f = volumetric fraction of non-volatile impurities in solution (cm³/cm³).

Most particles have naturally acquired electrostatic charges from electron transfer during contact or separation, or from free ion diffusion.¹⁹ To prevent agglomeration and assure monodispersity, these charged particles must be neutralized. The aerosol neutralizer, which is part of the aerosol flow system, contains 10 mCi of ⁸⁵Kr. This neutralizes the particle charge by producing positive and negative air ions which attach themselves to the surface of oppositely charged particles. With the proper residence time in the aerosol neutralizer, the "charge level" is reduced to Boltzman equilibrium.²⁰

We collected the generated aerosol particles on 25-mm-diam Nucleopore polycarbonate membrane filters with a 0.2 μ m pore opening, placed in a millipore filter holder. The aerosol was then sampled at 3.9 l/min to maintain linear flow lines between the aerosol flow system and the collection inlet. This method is referred to as iso-kinetic sampling, and is necessary for particles $\geq 5 \mu$ m. This sampling rate was maintained for particles $\leq 5 \mu$ m as well. We used an automated system which combines a quantimet image analyzer and a light-scattering microscope to count the total number of particles on the filter within several fields of view. We then calculated the total number of particles on the filter, based upon the number of fields observable within the collection area of the filter. On the average, 1×10^6 particles were collected on each filter; five collections were made for each set of experimental conditions. We counted approximately 1×10^3 particles for each sample to determine the average particle diameter, the standard deviation, and the degree of monodispersity. These data were made available via computer graphics, and showed good predictability in particle size generation. We used the number of particles collected from a specific volume of the aerosol to verify the predicted concentration of particles in the flow. The aerosol concentration was found to be 61% of the predicted theoretical value. This was primarily due to wall loss.

We have generated methylene blue and sodium chloride aerosols with a high degree of monodispersity and these results are summarized in Table 4. Particles

Table 4. Operating characteristic of monodisperse aerosol generator

Particle composition	Solution concentration (cm ³ /cm ³)	Frequency (kHz)	Solution feedrate (cm ³ /min)	Orifice size (μ m)	Particle size (μ m)	
					Calculated	Observed
Methylene blue ^a	0.005	67.01	0.15	20	7.1	7.9, 7.9, 7.5 ^c
Methylene blue ^b	0.0001	99.66	0.15	10	1.68	1.6-1.7 ^d (σ = 1.1)
Methylene blue	0.0001	99.72	0.15	10	1.68	2.2 ^c
Sodium chloride	0.005	120.90	0.15	20	5.83	5.6 ^d (σ = 1.04)
Sodium chloride	0.01	60.99	0.15	20	9.22	9.2 ^d (σ = 1.06)
Sodium chloride	0.0025	130.53	0.077	10	3.61	4.03 ^d (σ = 1.05)
Sodium chloride	0.00025	185.50	0.077	10	1.55	1.96 ^d (σ = 1.07)

^a50:50 vol% isopropanol and water. Such a solution had a tendency to plug the filter located between the pump and orifice.

^b50:50 vol% ethanol and water.

^cObtained using Quantimet Particle Counter.

^dObtained from scanning electron microscope.

generated from methylene blue (50:50 isopropanol: H_2O) were solid spheres, as shown in Fig. 10.

Particles generated from crystalline solids such as sodium chloride are more complex²¹ (note Figs. 11-14). Because of the rapid surface evaporation of

Fig. 10. Methylene blue particles 1.5-1.7 μm diameter; $C = 0.0231$.

Fig. 11. Sodium chloride aerosol particle 9.2 μm in diameter; $C = 0.01$.

Fig. 12. Sodium chloride particles 5.6 μm in diameter; $C = 0.005$.

Fig. 13. Sodium chloride particles 4.03 μm in diameter; $C = 0.0025$.

Fig. 14. Sodium chloride particle 1.96 μm in diameter; $C = 0.00025$.

the solvent, the concentration of the solute increases toward the surface of the droplet. Just before complete evaporation, an intercrystalline NaCl lattice is formed that is strong enough to retain the spherical form of the droplet. If the evaporation rate of the droplet were decreased, the solute might have the time necessary to equilibrate with the solvent. Complete evaporation would yield a solid particle rather than a hollow one.

To date, we have generated monodisperse aerosol particulates as small as 1.55 μm . Future work with monodisperse aerosols will include attempts to generate sodium chloride particulates as small as 0.5 μm .

INSTRUMENT DEVELOPMENT

Transmission Anode X-ray Tube Development

A development program for transmission anode x-ray tube (TRAX) has been underway for several years, and has periodically been updated in progress and other reports.²²⁻²⁴ Much of the earlier work was associated with x-ray fluorescence analysis programs; our recent efforts have been aimed at producing high-purity (i.e., nearly monoenergetic) beams of various energies for the Hazards Control calibration facility. This new application has resulted in some significant changes in the design of the tube and the manipulation of the beam. The changes fall into three categories: electro-mechanical design, target (anode) design, and filter design.

Early versions of TRAX suffered from problems with high-voltage breakdown, especially in the high-vacuum feed-through insulator. Here, the requirements for vacuum integrity, mechanical support and alignment, high heat conductivity, and high-voltage insulation led us through a series of in-house designs and modifications of extant commercial insulators. None of these proved satisfactory, so we turned to an outside vendor who collaborated with us on a custom design. Sadly, this insulator, while satisfying most of our requirements, proved unreliable in standing off the 50 kV we normally used on the anode, so a second vendor was contacted and another design, shown in Fig. 15, was developed. The resulting insulator has proven quite satisfactory, and, in fact, operates at 60 kV which is 20% over design requirements. Thermal connection from the anode to the heat exchanger is provided by a 12.7-mm-diam

OFHC copper rod, which is also dimensionally stable enough to provide precise alignment of the anode and the electron gun. Alignment is further aided by making the tube body from a solid block of stainless steel, rather than the welded tubing structure used before.

In TRAX, the design of the anode is crucial to system performance. One parameter of extreme importance is the ability to dissipate the thermal energy deposited in the anode by the electron beam without the temperature exceeding the melting point of the target material. In tubes with target materials such as silver, molybdenum or copper, the combination of good conductivity and/or high melting points allowed anodes to be made of a 0.08- to 0.16-mm foil in good contact with the heat exchanger. Foils of such thickness not only generate characteristic K x rays when struck by energetic electrons, but also provide

Fig. 15. High-voltage insulator. (1) Al_2O_3 ceramic, (2) vacuum flange; (3) seals (4) copper conductor.

Fig. 16. Sputtered target transmission anode x-ray tube (TRAX).

a filter which selectively passes the characteristic radiation while absorbing unwanted bremsstrahlung. If the absorbed bremsstrahlung is above K absorption edge of the foil, additional characteristic radiation is generated.

Demands for other target materials, such as antimony, praseodymium, and neodymium required a different approach. Our latest design, shown in Fig. 16, consists of a massive beryllium substrate, with a thin (typically 5 μm) coating of the desired material sputtered onto it. This layer is thick enough to stop the electrons, but is thin enough to allow excellent heat transfer to the beryllium which is thermally connected to the heat exchanger. A foil, also made of the target element, is placed into the beryllium. This foil, while thermally isolated from the beam spot, provides the same filter/converter properties as did the target foil in the original design.

Two anodes have been manufactured using this technique: one of praseodymium, one of neodymium. In brief, the Pr target worked well at moderate power levels (1-15 W), but the sputtered layer failed at higher levels. The Nd target has proven quite successful, surviving 60-W levels for extended periods with no apparent degradation. I intend to explore other sputtered target materials in the near future.

For calibration uses, the ideal beam would be truly monoenergetic. In TRAX, we can only approach this condition, and we must trade off intensity to do so. The technique involves use of external filters which preferentially absorb undesirable portions of the output spectrum. Using the beam from the Nd target tube as an example, Fig. 17 shows the spectrum without the filter/converter foil in place, while

Fig. 17. Spectrum from tube without internal filter - has only 5- μm Nd layer.

Fig. 18(a) through (e) show the effect of successive stages of external filtration. The spectra were taken with a Si(Li) detector, and have been corrected for detector efficiency. I have arbitrarily defined the "peak-to-total" figure as the fraction of the total photon output lying in a band whose width is 10% of the $K\alpha_1$ energy, and whose upper limit is the $K\alpha_1$ peak — the region between the vertical lines. The intensity is the number of photons in that region relative to the beam with no external filter. Thus, for

a specific application, one can select a filter which provides the most appropriate combination of beam purity and intensity.

Finally, good beam uniformity is desirable. That is, there should be equal intensity in all parts of the beam. To measure this, we exposed film to the beam, digitized the image with a scanning microdensitometer, and plotted the data on a computer. Figure 19 shows a cross section of the beam, averaged over a path 200 μm wide. Figure 20 is an intensity contour map.

Fig. 18. Spectrum from tube with various internal filters.

Fig. 19. Cross-section map of beam intensity.

Fig. 20. Contour map of beam intensity. Contour intervals = 4% change in intensity.

Fig. 21. Isometric map of beam intensity.

looking straight into the beam. The contour lines represent 4% changes in intensity. The blank region in the center, that is, in an area whose intensity is within $\pm 2\%$. Finally, Fig. 21 is an isometric projection of the data.

Future developments under consideration will concentrate on development of higher-Z targets (and hence, higher energy beams). This will entail some redesign to accommodate the necessarily higher accelerating voltages.

A Parameterized Pulse-Height Response Matrix for a ^3He Proportional Counter

Proportional counters and ion chambers filled with ^3He have been used for a number of years to measure line strengths in structured neutron spectra.²⁵⁻²⁷ Much of this work has been devoted to the measurement of structure in delayed neutron energy spectra following fission. Generally, these detectors have not been used to measure continuous neutron energy spectra because of the complexity of the pulse-height response function, and the difficulty in attempting a Monte Carlo calculation of the response matrix. For neutron energies above 1 MeV the pulse-height response function is complicated by (1) the rapidly decreasing (n,p) cross section, (2) ^3He recoils produced by elastic scattering, (3) a continuum caused by events which deposit some of their energy outside the detector active volume, and (4) pulse pile-up due to the large thermal neutron count rate.

In the work reported here, we have used

monoenergetic neutrons to study two ^3He proportional counters. Both detectors are 5.1 cm diam $\times$ 45.7 cm active length with 4 atm ^3He . Detector A has 3 atm argon stopping gas while detector B has 3 atm krypton. Each was irradiated with monoenergetic neutrons in the range 0.05-7 MeV produced by the $^7\text{Li}(p,n)$ or $^3\text{H}(p,n)$ reactions, and pulse-height spectra were obtained. In all cases, we used a dePangher long counter to monitor the neutron fluence during a run. Consequently, the areas of the Gaussian peak due to (n,p) reactions, the ^3He recoil distribution, and the pulse-height continuum are all normalized to unit neutron fluence through the detector. In general, the neutrons were incident perpendicular to the detector axis to minimize the uncertainty in source-detector separation.

To adapt the experimental response measurements for use in unfolding continuous spectra, the experimental data have been parameterized and represented by a semi-empirical model. The pulse-height response was decomposed into three relatively simple components in the following manner: After discarding the pulse-height data in and below the thermal peak and above the thermal peak to an energy two times the full width at half maximum, the data are decomposed into (1) a Gaussian representation of the (n,p) reaction peak, (2) a Gaussian smoothed trapezoidal representation of the ^3He recoil distribution, and (3) an exponential representation of the underlying continuum.

Each column of the ^3He response matrix is normalized to unit neutron fluence, and is expressed as follows:

$$R_{ij} = NT\sigma_{np} \Delta E f_g \left[\frac{e^{-\frac{1}{2}(i-\delta_A)^2}}{\sqrt{2\pi}\delta_A} \right] + \frac{NT\sigma_c \Delta E}{(1-\alpha)^2} \frac{dE}{d\alpha} f_R$$

$$NT\sigma_{np} \Delta E \frac{dE}{d\alpha} e f_{c_0} f_{c_1} (j-1) \quad (3)$$

where

- N = number of ^3He nuclei in detector active volume,
- T = length of data acquisition (s),
- σ_{np} = cross section for ^3He (n,p) reaction (cm^2),
- ΔE = energy bin width (MeV),
- f_g = efficiency function for Gaussian peak,
- δ_A = detector resolution (FWHM) in channels,

σ_s = elastic scattering cross section for ${}^3\text{He}(n,n)$,
 $\alpha = (A - 1/A + 1)^2 = 0.25$,
 E_i = neutron energy for i th bin,
 dE/dx = calibration factor (MeV/channel) in raw pulse-height data,
 f_R = trapezoidal function which represents ${}^3\text{He}$ recoil distribution,
 f_{c_0}, f_{c_1} = parameters which describe the continuum distribution.

The main thrust of the present work has been to determine the parameters which describe the distributions $f_{c_0}, f_{c_1}, f_R, \delta_x$, and the parameters of f_R , all as functions of incident neutron energy. To begin with, the Gaussian component of the response is described by two parameters, f_g and δ_x , which are functions of neutron energy (gas gain is assumed constant). Figure 22 shows the detector resolution (FWHM) as a function of energy deposited in the active volume. The data shown were obtained at a gas gain of 4 for detector A and 14 for detector B. Also shown are polynomials fitted to the data which give an approximate description of the FWHM as a function of energy deposited. Thus

$$\delta_x = g(\delta_0 + \delta_1 E_{\text{dep}}) \quad (4)$$

where g = electronic gain (channels/MeV) = $1/dE/dx$.

The (n,p) full charge collection efficiency, ϵ , is shown in Fig. 23. A reasonably adequate fit to the

Fig. 22. Resolution vs energy deposition for two ${}^3\text{He}$ proportional counters. Detector A: $\text{FWHM}/E_{\text{dep}} = 0.0434 + 4.37 \times 10^{-3} E_{\text{dep}}$. Detector B: $\text{FWHM}/D_{\text{dep}} = 0.062 - 0.0014 D_{\text{dep}}$.

Fig. 23. Absolute (n,p) peak efficiency as a function of energy deposition for two ${}^3\text{He}$ proportional counters. ϵ = Area/No. of n,p reactions, calculated
 $\epsilon = 0.68e^{-0.0587E_{\text{dep}} - 0.096E_{\text{dep}}^3}$.

data is obtained using an exponential function of the form:

$$f_g = e^{(f_0 + f_1 E_{\text{dep}} + f_2 E_{\text{dep}}^2)} \quad (5)$$

where the parameters f_0, f_1, f_2 are characteristic of a particular detector.

Pulse-height response to events in which charge collection is incomplete appears to be well represented by an exponential with parameters f_{c_0} and f_{c_1} whose energy dependence may be represented by a simple polynomial. The polynomial coefficients for the two detectors whose response was measured are as follows:

$$f_{c_0} = \alpha_0 + \alpha_1 E_{\text{dep}} + \alpha_2 E_{\text{dep}}^2,$$

$$f_{c_1} = \beta_0 + \beta_1 E_{\text{dep}}.$$

	α_0	α_1	α_2	β_0	β_1
Detector A:	-0.356	-0.774	0.04	-0.015	0
Detector B:	-0.356	-0.774	0.04	-0.01	-0.002

Fig. 24. Model approximation to ^3He recoil distribution. Parameters C_L , C_H , h_L , h_H determine the area and shape of the distribution.

Finally, the ^3He recoil distribution produced by elastic scattering is represented by a trapezoid which is smoothed by a Gaussian of resolution given by Eq. (4). The trapezoid before smoothing is shown in Fig. 24. Since the charge collected from a ^3He recoil is slightly less than from a $^3\text{H-P}$ pair at the same energy, the detector response is shifted toward lower pulse height. The "knee" of the recoil distribution then extends over pulse heights in the range

$$f_L \{g(1 - \alpha)E_n\} \leq \text{pulse height} \leq f_H \{g(1 - \alpha)E_n\}. \quad (6)$$

Figure 25 shows the experimental data for f_L and f_H as functions of energy, as well as the polynomial fits to the data.

Additionally, the area of the recoil distribution is less than that obtained from the cross section and fluence, and the slope of the distribution is nonzero. At the present time, the data on recoil detection efficiency and slope of the distribution are sparse and uncertain. Trial values have been selected and used to construct a preliminary response matrix. At present, the area of the recoil distribution is assumed to be only 0.3 times that calculated from the neutron fluence and detector cross section, and the slope of the distribution is such that $h_H/h_L = 0.2$.

Using the response functions which are parameterized as described above, a response matrix is calculated for a particular set of pulse-height data and detector. Selected columns of a typical 50×50 response matrix are shown in Fig. 26. This matrix is used to unfold the incident neutron spectrum. Many

Fig. 25. Charge collection efficiency vs neutron energy for ^3He recoil events. Parameters f_L and f_H are related to the charge collection by:

$$C_L = f_L (1 - \alpha)E_n g + X_0$$

$$C_H = f_H (1 - \alpha)E_n g + X_0$$

where g = electronics amplification (channels/MeV)

X_0 = electronics bias

Fig. 26. ^3He response functions for monoenergetic neutrons.

spectra obtained with monoenergetic sources have been unfolded in this way to determine the efficacy in calculating the response matrix according to Eq. (3).

Fig. 27. Spectrum of 567-keV monoenergetic neutrons obtained with ^3He proportional counter.

- a) Pulse-height data
- b) Unfolded spectrum

Fig. 28. Spectrum of 1.2-MeV monoenergetic neutrons obtained with ^3He proportional counter.

- a) Pulse-height data
- b) Unfolded spectrum

Some results of these efforts are shown in Fig. 27-29. Examination of the figures shows that the overall representation of the pulse-height response is a reasonably good approximation above 100-200 keV. But the representation of the "knee" of the recoil distribution is still imperfect. In addition, significant distortion of the response due to pulse pile-up is observed at count rates exceeding a few hundred H_2 .

The greatest difficulty and uncertainty, lies in the recoil component of the response. This component may be suppressed by a factor of 10-20 by using pulse-shape discrimination. In many applications this would be the recommended procedure, and the response matrix would be simplified. We have found,

however, that all three components are suppressed to varying degrees by pulse-shape discrimination. The recoil and continuum responses are very sensitive to the risetime discriminator level. Our interest is largely in field measurements where we have found it difficult or impossible to accurately obtain a risetime discriminator setting which reproduces one used in the laboratory. The response function obtained in the field with pulse-shape discrimination may be seriously different from the one measured in the laboratory and is, itself, difficult to determine. Consequently, field measurements are being made without resorting to pulse-shape discrimination, and efforts are continuing to accurately parameterize the recoil distribution.

Fig. 29. Spectrum of 2.03-MeV monoenergetic neutrons obtained with ^3He proportional counters.

- a) Pulse-height data
- b) Unfolded spectrum

FIRE SAFETY

Heat Release Rate Measurements Using a Radiant Panel Surface Flammability Apparatus

The heat release rate calorimeter, a relatively new testing device, is currently being proposed as a standard fire test method. Essentially, it consists of an enclosure in which a large heat source uniformly exposes a test specimen to a range of radiant flux levels in a specified air flow field. As the specimen burns it releases energy at a rate that depends on: (1) the heat flux absorbed at the surface, (2) the composition of the material, and (3) the thermal properties of the specimen. This energy release is measured either by sensing the average temperature rise in the enclosure exhaust, or by a comparative technique that relates the temperature history caused by exposure of the specimen, to the heat content of a gas burned to duplicate the temperature-time history of the test exposure. Regardless of the technique used, extreme care is exercised in establishing and controlling the fluid- and heat-transfer characteristics of the testing apparatus. Unfortunately, the standard deviation of the resultant data from most devices currently being reported is relatively large. This indicates that the non-uniformity of the testing materials controls the statistical response of the test method. Intuition then tells us that it may be possible to obtain statistically relevant information by conducting more test exposures using simpler testing procedures.

When heat release rate (HR^2) results were identified as useful to the LLL flammability assessment program,

a simpler testing technique was suggested, and is now being evaluated to see whether or not our intuition is correct.*

Essentially, we have modified a flame-spread testing apparatus so that it provides a large-area irradiance field to expose a vertically oriented test specimen to a heat flux.[†] Other modifications include increasing the air flow rate of the exhaust fan and the installation of panels to reduce spurious air currents. The exposure schedule provides for several types of flammability response measurements, including ease of ignition, flame spread rate, and total and peak heat release rates. In the latter measurements, the heat released by the tested materials is sensed by a set of thermocouples, connected in parallel, located in the exhaust stack of

*Currently designed HR^2 calorimeters are conceptually simple devices, as discussed above. However, in their attempts to avoid the complications resulting from the transient behavior of materials suddenly exposed to intense heat sources, the calorimeter designers take great pains to control or define their parameters — both input (heat source, combustion, and excess air flow) and output (heat losses, combustion gas composition and air leaks). Consequently, it is necessary to provide the calorimeter with a well-defined, insulated enclosure, complicated gas controls, and sophisticated analytical instrumentation. It is our contention that the HR^2 from most materials persists for a long enough period at relatively high and constant levels that second-order loss factors are not too important in the assessment of HR^2 .

[†]The Radiant Panel Surface Flammability Test Apparatus (ASTM E-162).

Fig. 30. Test parameters for heat release rate calorimeter.
 a) Time to ignition versus irradiance for gypsum board.
 b) Downward flame-spread rate versus irradiance for gypsum board.
 c) Peak 1-min heat release rate versus irradiance for gypsum board.

the hood. The specimen size is varied (from a maximum of 150 X 450 mm) to insure that all combustion products are accepted by the hood, and that no flames contact the thermopile.

Two materials have been tested to date, and the results are encouraging since they fall within the range of data received from more sophisticated devices. Figure 30 shows respectively the time to ignition (t_i), the downward flame spread rate (d_2), and the heat release rate (HR^2) of standard gypsum wall board exposed to several irradiance levels. These data indicate that t_i increases monotonically as H decreases, and that both d_2 and HR^2 are linearly proportioned to H over the tested irradiance range. The highlighted symbols on the HR^2 curve identify test results from other techniques. Since the LLL data fall within this range, we are initially satisfied that, for this material, the technique is adequate.

Preliminary measurements of the HR^2 of masonite (pressed particle board) have been made at an irradiance of 5.2 W/cm². The average HR^2 of three

tests is 14.3 W·cm⁻²·min⁻¹ which is in the range of results for similar materials found in the literature. Several standard materials have been obtained for test from the National Bureau of Standards and Stanford Research Institute. They are currently stored in an environmental chamber which conditions flammability test specimens to specified levels of moisture content and temperature. When time permits these materials will be tested, and the results will validate or disprove the technique.

Recalibration and Use of High-Flux Heater in NBS/LLL Smoke Chamber

Introduction. In our previous report,²⁸ we described the development, design, calibration, and use of a new high-flux radiant heater in our NBS/LLL Smoke Chamber. Because some questions arose as to the use of a 30°-view angle radiometer, we obtained a 180°-view Gardon foil radiometer, and recalibrated the heater as described herein.

Recalibration. In preparation for this work, the 65-mm-square copper plates were completely refurbished, remeasured, and reweighed. New thermocouple leads were peened into small holes drilled in the backs of the plates rather than being soldered on. We calibrated each voltage setting for the heater as follows:

- We established the setting, and allowed the heater to equilibrate as determined by the reading of the thermocouple located in the center of the heater face.
- We then placed the 30°-view radiometer in front of the heater, and obtained an equilibrated reading.
- We slid the heater in front of the 38-mm-diam circular copper disc and obtained a fast reading for $dV/d\theta$. The same was done successively for square copper plates A and B.
- We placed the 180°-view radiometer in front of the heater and obtained an equilibrated reading.
- Another set of $dV/d\theta$ values was obtained for the three copper standards.
- We then established another voltage setting to the heater and repeated the preceding steps. The maximum setting was 116V/20A, with an indicated heater temperature of 1385°C.

We made two sets of calibrations on two different days. The results are plotted in Fig. 31 showing the millivolt output for the two radiometers against the heat fluxes received by the three copper standards. It is interesting to note the coincidence of the values obtained for copper plate A with the factory calibration of the 180°-view radiometer.

In summary, the maximum heat obtained on the face of a (blackbody) sample is given as follows:

	For circular disc	For square plate
Area (cm^2)	11.40	42.36
Flux (W/cm^2)	13.4 ± 0.1	11.65 ± 0.15

Whereas, as noted in our previous report it is more logical to use the square copper plate (which is the same size and shape as the exposed face of samples tested), most of the NBS-type chambers in the country use the circular disc. This measures the flux on the central circular 11.4 cm^2 area. For this reason, we shall use the circular disc value so that others may compare their results with ours.

Following the calibrations, the two radiometers were successively mounted in rectangular coordinate jigs and the fluxes across the face of both the heater and the exposed sample face area were scanned at several heater settings. A typical set of data is shown in Fig. 32; the results are summarized in Table 5. From the table it can be seen that the ratio of the average flux received over the sample face to that received in

the center is quite uniform for the various heater settings.

Smoke Density of Douglas Fir and White Pine. We used the heater at various heat fluxes, approximately 2.5, 5.0, 7.5, 10.0, and $13.4 \text{ W}/\text{cm}^2$ to determine the smoke-density development of 6.4-mm-thick specimens of preconditioned Douglas fir and white pine. The following series of tests was made:

- Closed chamber without piloted ignition
- Closed chamber with piloted ignition, but with ventilation rates of 3, 6, 12, and 20 air changes per hour, respectively.

The results of these tests are given in Tables 6 and 7 and in Figs. 33 through 40. Tables 5 and 6 (for Douglas fir and white pine respectively) show the values obtained for maximum specific optical density, obscuration time, time to maximum density, and time to autoignition, plus appropriate comments. Figures 33 and 34 for Douglas fir and 35 and 36 for white pine show the specific-optical-density (D_s) time curves for unpiloted and piloted ignition conditions, respectively. Figure 37 (Douglas fir) and Fig. 38 (white pine) show plots of D_m (maximum specific optical density) versus ventilation rates. Figure 39

Fig. 31. Heat flux received by Medtherm radiometers from LLL heater as measured by blackened copper plates.

Fig. 32. Heat flux scan of heater opening and sample face. Note: Heater is set to yield 2.50 W/cm^2 using 30° view radiometer located at center of sample face position as calibrated against square copper plate A.

shows the effect of heat flux on time to autoignition in a closed chamber. Figure 40 shows the effect of ventilation at two heat fluxes on the time to autoignition.

The data and observations made during the tests show several interesting heat- and ventilation-response

phenomena. These are described in the following paragraphs.

The nonventilated density-time curves for both woods are typical of what we have seen in the past for a flux of 2.5 W/cm^2 , with one minor exception. Under piloted ignition the Douglas fir specimens

Table 5. Heat flux scans of heater across sample-face area

Heater flux ^a (W/cm^2)	30° view radiometer ^b		180° view radiometer ^c	
	Average flux across sample face (W/cm^2)	Ratio face/center	Average flux across sample face (W/cm^2)	Ratio face/center
2.5	2.10	0.84	2.25	0.90
5.0	4.30	0.86	4.40	0.90
7.5	6.3	0.84	6.76	0.90
10.0	-	-	8.90	0.89

^aValue given is based on average flux received by 65-mm-square copper plate "A" and as read by radiometer indicated.

^b49 points scanned.

^c5 points scanned: center +4 points near corners.

exhibited variable responses, as shown by the envelope in Fig. 34. This is ascribed to the varying amounts of resin present in this wood.

At the 5-W/cm² flux, nonventilated and without a pilot flame, rapid pyrolysis occurs, yielding about the same smoke density as for the 2.5-W situation. Autoignition tends to occur at 4-5 min of exposure. When this happens pressure builds up in the chamber, causing the pressure relief valve (set at 1000 Pa) to be actuated, and resulting in a dip in the density-time curve.

At the higher heat fluxes (unpiloted and nonventilated), autoignition times are inverse to the radiation. However, pyrolysis rates increase with the increase in heat flux and combustion is inefficient, judging by the dirty orange color of the flames and the persistence in relatively high smoke density values attained. White pine seems to be a little "cleaner" than Douglas fir.

At heat fluxes of 2.5- and 5-W/cm² and under the condition of piloted ignition, combustion seems to keep pace with the pyrolytic generation of particulates,

Table 6. Smoke density data using LLL high-flux heater

Material - solid Douglas fir 6.4 mm thick, vertical grain orientation.

Exposure conditions			Results ^a				Remarks
Radiant flux (W/cm ²) ^b	Pilot flame	Ventilation (air changes/hr)	D _m	T ₁₆ (min)	T _m (min)	T _{ai} (min)	
2.5	No	0	518	1.9	14	-	
5.2	✓	✓	538	0.63	7	4.7	
7.85	✓	✓	282	0.30	5	0.43	
9.9	✓	✓	313	0.22	4	0.25	
13.7	✓	✓	395	0.17	3	0.10	
2.5	✓	3	588	1.32	12.5	-	
✓	✓	6	465	1.59	14	-	
✓	✓	12	381	1.52	12	-	
✓	✓	20	245	1.63	12	-	
5.0	✓	3	>660	0.55	4	2.20	2/3 Auto ignited
✓	✓	6	>660	0.55	5	5.4	2/3 Auto ignited
✓	✓	12	~660	0.59	5	5.4	3/3 Auto ignited
✓	✓	20	189	0.59	2	1.7	2/2 Auto ignited
7.5	✓	3	180	0.28	4	0.52	3/3 Dirty orange flame
✓	✓	6	150	0.30	4	0.52	3/3 Dirty orange flame
✓	✓	12	119	0.29	2	0.49	3/3 Dirty orange flame
✓	✓	20	116	0.31	1	0.54	3/3 Dirty orange flame
10	✓	3	237	0.20	3	0.26	2/2 Dirty orange flame
2.4	Yes	0	198	3.6	14	-	Yellow flame
4.9	✓	0	8	2.9	17	-	Dirty orange flame
7.4	✓	0	165	0.51	5	-	Dirty orange flame
10.0	✓	0	294	0.31	4	-	Dirty orange flame
13.0	✓	0	321	0.21	3	-	Dirty orange flame

^aD_m - Maximum specific optical density.

T₁₆ - Time (min) for D_s = 16 i.e. to visual obscuration.

T_m - Time to reach D_m (min).

T_{ai} - Time (min) at which autoignition occurs.

^b Flux values were corrected as necessary to agree with recent calibrations.

Table 7. Smoke density data using LLL high flux heater

Material - Solid white pine 6.4 mm thick, vertical grain orientation

Exposure conditions			Results ^a				Remarks
Radiant flux (W/cm ²)(b)	Pilot flame	Ventilation (air changes/hr)	D _m	T ₁₆ (min)	T _m (min)	T _{au} (min)	
2.6	No	0	503	1.62	11	-	
5.2	✓	✓	510	0.54	5	3.7	1/3 Autoignited
7.9	✓	✓	191	0.27	3	0.25	4/4 Autoignited
9.9	✓	✓	264	0.19	3	0.15	3/3 Autoignited
13.7	✓	✓	287	0.13	3	0.09	3/3 Autoignited
2.5	✓	3	477	1.22	11	-	
✓	✓	6	426	1.20	10	-	
✓	✓	12	289	1.32	10	-	
✓	✓	20	230	1.42	9	-	
5.0	✓	3	660	0.50	4	~4.0	2/3 Autoignited
✓	✓	6	533	0.49	4	3.9	2/3 Autoignited
✓	✓	12	516	0.47	3	3.8	2/3 Autoignited
✓	✓	20	91	0.45	1	0.9	2/3 Autoignited
7.5	✓	3	171	0.28	5	0.40	3/3 Orange-yellow flame
✓	✓	6	162	0.24	3	0.40	3/3 Orange-yellow flame
✓	✓	12	130	0.75	3	0.27	3/3 Orange-yellow flame
✓	✓	20	86	0.27	3	0.28	3/3 Orange-yellow flame
2.4	Yes	0	71	5.1	12	-	Flare ups
5.2	✓	✓	73	0.5	4.5	-	
7.5	✓	✓	122	0.31	4	-	Dirty yellow-orange flames Several chamber over- pressures
9.9	✓	✓	225	0.22	3	-	
13.0	✓	✓	290	0.15	3	-	

^aD_m = Maximum specific optical densityT₁₆ = Time (min) for D_s = 16% i.e. to visual obscuration.T_m = Time to reach D_m (min).T_{au} = Time (min) at which autoignition occurs.^bFlux values were corrected as necessary to agree with recent calibrations.

judging by the relatively low smoke densities obtained. At the higher heat fluxes, the situation is the same as for the unpiloted cases.

Except for one apparent anomaly - Douglas fir ventilated at 3 air changes per hour - increasing ventilation causes a decrease in maximum smoke density for a heat flux of 2.5 W/cm². At a 5-W heat flux, ventilations in the range of 3 to 12 air changes per hour yield smoke densities for Douglas fir outside the range of our detection apparatus; a similar tendency is observed for white pine, although the densities are measurable. It takes a high ventilation rate, e.g., 20 air changes per hour, to reduce the maximum smoke density to values in the range of 100-200.

At the higher heat fluxes, increasing ventilation plus earlier autoignition seems to combine to produce smoke densities lower than for the 5-W case, although the effect of ventilation is less pronounced than for the 2.5-W case.

In a closed chamber, both woods exhibit very similar times to autoignition as a function of heat flux, at an exposure of 5 W/cm² the times are in the range of 3.5 to 4.5 min. At and above 7.5 W/cm², autoignition is quite rapid (Fig. 39).

At a 5-W exposure, ventilation seems to have little effect until the ventilation reaches 20 air changes per hour when autoignition is enhanced. At a 7.5-W flux, lack of or considerable variations in ventilation apparently have no effect on autoignition time.

Fig. 33. Effect of radiant heat flux on smoke density development of Douglas fir, with no ventilation.

Discussion. A flux of 5 W/cm^2 alone or combined with mild to moderate ventilation rates seems to be a critical value, at least for the woods tested, in the generation of smoke particulates. These appear to be pyrolysates, for the most part liquid tars, which combust inefficiently under the conditions existing in the smoke chamber. It will be interesting to see what the effects are when polymeric materials are exposed to the same bracketing heat, flame, and ventilation parameters.

It also appears that the variable heat-flux and ventilation capabilities may yield a unique approach to evaluating autoignition phenomena for various materials.

Fig. 34. Effect of radiant heat plus pilot flame on smoke density development of Douglas fir with no ventilation.

Fig. 35. Effect of radiant heat plus pilot flame on smoke density development of white pine with no ventilation.

Fig. 37. Effect of ventilation rate and radiant heat flux on maximum specific optical density of Douglas fir.

Fig. 36. Effect of radiant heat plus pilot flame on smoke density development of white pine with no ventilation.

Fig. 38. Effect of ventilation rate and radiant heat flux on maximum specific optical density of white pine.

Fig. 39. Effect of heat flux on exposure time to autoignition for Douglas fir and white pine. Closed chamber - no ventilation.

Fig. 40. Effect of ventilation and radiant heat flux on time to autoignition for Douglas fir and white pine.

The LLL Full-Scale Fire Test Facility - Preliminary Results on Smoke Plugging of HEPA Filters

Introduction. Previously we reported²⁹ that LLL was constructing a full-scale fire test cell to be used in evaluating the adverse effects of unwanted fires on the HEPA filters installed in exhaust ventilation systems in laboratories where toxic or radioactive materials are used. We also detailed the program outline to be followed, and the fuel and smoke loadings to be employed.*

*Smoke loadings are given as D/C (dirty-to-clean) ratios of fuel loadings. A dirty fuel produces large quantities of smoke particulates in a well-ventilated fire, e.g., fire-retarded cellulosic and plastic materials. A clean fuel, does not, e.g., certain plastics and non-fire retarded celluloses.

This report covers a description of the facility, and summarizes the results of preliminary experiments to evaluate the filter smoke-plugging problem.

Description of Test Facility. As shown in Fig. 41, the full-scale fire facility consists of:

- A fire test room 100 m³ in volume, and an associated small control room containing viewports into the test cell; gas sampling and analytical equipment; a control valve and meter for the sprinkling system; and a cathode-ray display tube capable of showing data obtained from the various centers on a 30-s update arrangement.
- High- and low-takeoff exhaust ductwork leading through a damper to the experimental duct system.

Fig. 41. Full-scale fire test facility.

- A bypass air pollution control system (APC).
- A pit for waste water collection and monitoring.
- A computerized data acquisition system.

The fire test cell consists of a rectangular, columnar steel framework onto which a steel skin with a Q-deck exterior is welded. Some 1600 stainless forks are welded to the inside of the steel skin walls and ceiling for holding the insulation. This insulation consists of 38 mm of Kastolite SK-7 with a 985°C rating, followed by 100 mm of Kastolite KS-4V with a 1400°C rating. Kastolite is an alumina-silica mixture

containing a small amount of lime. The floor of the cell, which is sloped to sprinkler drains, consists of a sand bed overlaid with fire brick, and finally overcast with Kastolite. The finished interior dimensions of the cell are 5.9 m long X 4.0 m wide X 4.2 m high.

Figure 42 shows some of the details of the ductwork. The left-hand section is the experimental modularized portion. Figure 43 shows the cell and external equipment. The high-pressure-drop venturi scrubber is used when the test is completed to clear

Fig. 42. Experimental ductwork for fire test facility.

Fig. 43. Fire test cell.

Fig. 44. Air pollution control system.

the room of smoke particulates and any toxic gases. Figure 44 shows a somewhat detailed view of the air pollution control (APC) system. Figure 45 shows the interior of the test cell looking toward the control room, and Fig. 46 shows the data acquisition/reduction system in the computer room. Figure 47 is an interior view of the control room showing the gas analysis instrument rack.

Construction of the facility was finished during the summer of 1975. The next several months were spent in installing the ductwork, APC system, waste drain

tanks and data acquisition system. Following this, some time was spent in installing and connecting the sensors, followed by a period of debugging. We have added more instrumentation during the testing program so that additional data are available from the later burns.

Summary of Tests and Results. A brief description of the tests conducted to date and a summary of the results obtained are given in Table 8. These are reported in detail elsewhere.³⁰

Fig. 45. Test cell interior.

Fig. 46. Data acquisition/reduction system.

Fig. 47. Gas analysis instrument rack.

Discussion. The tests made to date have been preliminary in nature because we have been adding instrumentation and improving our techniques. However, we have observed certain trends in the results:

- Where no attempt is made to increase the exhaust ventilation, the compartment overpressurizes soon after a fire is started.
- In the case of a dirty fire, increasing exhaust ventilation tends to make the filter plug that much sooner.
- If a significantly large "dirty" item in an

otherwise "clean" fuel loading becomes involved in a fire, a large quantity of smoke particulates will be generated.

- The smoke particle size distribution, seems to be tending toward the larger sizes; this suggests that they may be more easily scrubbed out than previously thought.

After a few more check fires, we plan to start testing and evaluating countermeasures. This will involve the use of single and sequential sprays with or without additives and with or without prefiltering techniques.

Table 8. Summary of tests and results on full scale testing of smoke-plugging of HEPA filters

No.	Purpose and Description	Fuel load (kg/m ²)	D/C ratio	Results
1	To check operating/safety characteristics of system, using moderate but dirty fuel load, of laboratory furnishings.	9.7	2.66	Cell overpressurized and heavy smoke particulates were generated in ~3 min. Smoke particulate analysis in duct indicated 50% of weight was of 2 μ m size or larger. Moderate ceiling temperatures throughout test.
2	To determine effect of moderate but clean load of laboratory furnishings. To attempt to depressurize cell by regulating ventilation	10.5	0.57	Clean fire until polyester hood ignited, then very sooty. Increasing ventilation kept room depressurized temporarily, but filter plugged sooner (1-kg of soot). Fuel loss: 9% total; 6.6% clean; 12% dirty.
3	To evaluate efficiency of APC (air pollution control) system, using liquid-fuel pan fire.	-	-	APC system worked well. No data recorded.
4	To determine effects of free burning diesel fire, on combustion rate, smoke-particulate production, and filter plugging.	0.55	-	Smoke dense, moderately heavy, and of fine particulate size: 1000 mg/m ³ ; 50% <1 μ m by weight. Filter plugged at about same time as fire was put out, e.g., 20 min.
5	To evaluate effect of moderate, but very dirty fuel loading; to test use of gas sampling/analytical system.	10.5	5.1	Room pressurized/vision obscured in 1.5 min.; fire decayed at 3 min. (lack of oxygen) for 12 min. when hood burned through, then flared up somewhat. Unburned hydrocarbons reached 3200 ppm, CO reached 3600 ppm; both in about 5 min.
6	To evaluate use of oversize filter, using a two-stage diesel fuel burn. Flow readjusted to normal (250 v/s) between stages.	0.59	-	First-stage fire burned out in 19 min., almost plugging filter. Second stage burned 3 min. when filter ruptured.
7	To determine filter-plugging using well ventilated (500 v/s) clean burning fire, e.g., wood crib.	8.2	0	Fire burned well for 16 min., and was extinguished. Little evidence of filter plugging, higher than usual temperatures noted. CO = 30,000 ppm in duct.
8	Repeat of No. 8 but with normal ventilation - 250 v/s.	8.3	0	Good burning for 5 min., then decay to lower steady state for 14 min. Brief flareup during APC switchover. Cell overpressurized at 2-3 min. Dense, acid, liquid-particulate smoke. CO ~60,000 ppm/20 min. O ₂ ~4.4%/20 min., both in exhaust duct.

DECONTAMINATION

Enhanced Filtration

Introduction. Since the last Progress Report we have focussed our attention on the following areas:

- Construction of a 25 l/s filter test system.
- Exploring the basic theory of electrified filter media.
- Assessing the efficiency increase of various material: when placed in an electric field.
- Verifying the particle size range of methylene blue and sodium chloride particles generated with

a vibrating orifice monodisperse aerosol generator.*

25 l/s Filter Test System. Filters were tested using the apparatus shown in Fig. 48. The system consists of an air mover, a sodium chloride aerosol generation system, prefilter test section and experimental diagnostic equipment.

*See p. 7 of this Progress Report.

Fig. 48. Schematic diagram of filter testing system.

A high-volume air sampler is used to draw filtered air into the system, and the flow is measured by a mass flowmeter. A variable transformer between the sampler and the power source maintains the flow between 2 and 25 l/s.

The aerosol is produced by four air-driven, modified Wright-type nebulizers.* The nebulizer screws into a 500-ml polyethylene bottle containing a solution of dissolved salt. Air pressure supplied to the nebulizer aspirates the solution from the reservoir to the nebulizer where it is atomized into small droplets.

The droplets mix with air in the aging chamber, evaporate, and subsequently form a test mixture of cubical salt particles in a moving air stream. The resultant aerosol characteristics are a function of the airflow, nebulizer air pressure, solution concentration, and the number of nebulizers used.

Particle size characteristics of a 1% sodium chloride solution at a nebulizer pressure of 340 kPa show the aerodynamic mass median diameter to be $0.95 \mu\text{m}$ with a geometric standard deviation of 2.0. The mass concentration using all four nebulizers at 23.6 l/s is 4.5 mg/m^3 .

System diagnostic equipment shown in Fig. 49 includes a flame photometer to determine filter efficiency and an electronic manometer to record pressure differential as a function of time. Relative humidity, temperature, and air flowrate are monitored throughout the experiment.

The dielectrophoretic filtration experiments were conducted in the test section located just in front of

the HEPA filter. Figure 50 shows the duct in the disassembled test assembly expanding to accommodate a 19.1×19.1 -cm-square filter. The filter is held between two perforated stainless steel plates separated with polyethylene spacers. A negative d.c. high-voltage lead is connected to the front screen, and the rear screen is grounded.

Nineteen of the 46 filtering media tested are shown in Table 9. Testing was normally conducted at face velocities of 65 cm/s. However, a glass and polyester filter were tested at face velocities of 16.3, 32.5, 48.8, and 65 cm/s.

The efficiency was measured at voltages ranging from 0-15 kV, and the pressure differential was monitored throughout the testing period. The enhancement coefficient (the ratio of the dielectrophoretic to initial efficiency) was then calculated.

Theory of the Effect of Electric Fields on Filter Media. If a fibrous filter media is placed between two plates in an electric field, the field lines are distorted and appear to emanate radially from the fiber surface. Particles, whether charged or neutral, passing through the filter bed become polarized and are attracted most efficiently to the areas of highest field intensity.³¹ Such a phenomenon has been termed "dielectrophoresis" by Pohl.³²

Rivers³³ has presented a mathematical description of electrified filter theory. Filter efficiency, E_f , can be expressed as

$$E_f = 1 - e^{-\frac{-svQ}{A}} = 1 - e^{-\frac{-svVA}{A}} \quad (7)$$

*Discussed in more detail on p. 6 of this Progress Report.

where

s is the surface area of the fibers,
 v is the particle drift velocity toward the fiber surface,
 Q is the air flowrate,
 A is the face area of the filter, and
 V_a is the air velocity through the filter. Filter efficiency is therefore increased by minimizing the flow of air and maximizing the filter media surface area and draft velocity.

Drift velocity is a resultant of inertial (V_i) and electrical (v_e) characteristics, and can be shown as

$$v = v_i + v_e = v_i + v_p + v_c \quad (8)$$

where v_p and v_c are the polarization and Coulombic contributions.

The polarization component can be expressed as

$$v_p = \frac{[d^2 E^2]}{12\pi\mu (R + d/2)} \quad (9)$$

where

d is the particle diameter,
 E is the average field gradient,
 μ is the viscosity of air, and
 R is the fiber radius.

E can be calculated from

$$E = V/L \quad (10)$$

Equation (9) shows the v_p increased according to the square of the field strength and that the larger particles undergo the strongest attraction toward the fiber.

The contribution of the Coulombic effects can be approximated from

$$v_c = \frac{NqE}{3\pi\mu d} \left[1 + \frac{K^2}{(R + d/2)^2} \right] \quad (11)$$

where N is the number of unbalanced charges/particle and q is the electron charge.

Fig. 49. Filter test system diagnostic equipment.

Fig. 50. Disassembled dielectrophoretic test system.

Whitby¹⁴ estimated the contribution of each effect for one type of filter, and the results are noted in Table 10. Note that at particle sizes below $2\ \mu\text{m}$ the Coulombic component of the drift velocity is the most important. However, at about $5\ \mu\text{m}$, the polarization effects theoretically dominate.

Dielectrophoresis Experiments. The ideal filter would have a relatively high initial efficiency and enhancement coefficient, but a low initial pressure and pressure drop growth. The results shown in the Table 10 indicate that no filter medium is outstanding in all four categories. However, glass (runs 14, 22, 23, 26, 27, 28, 30, and 40), polyester (run 21), and dacron fibers (run 35) are all strong candidates. Woven fibers and reton mats have high initial efficiencies, but their pressure drop is so rapid that they are unsuitable in this type of application.

Figures 51 and 52 show the efficiencies for polyester and glass fiber materials. Here the efficiency

is shown as a function of applied voltage at various face velocities. With no voltage applied, the efficiency, as one might suspect, is greater with increasing face velocities. However, as the voltage between the plates is increased, the inertial component of the efficiency becomes less important and the electrical effects begin to dominate. At $16.3\ \text{cm/s}$ the efficiency of the polyester fiber is increased by a factor of 100, and the glass fiber increases from 29 to $>99\%$ when the voltage is increased from 0 to 12 kV. This behavior has also been observed by Fielding³⁵ and Bogardus³¹ using a 0.3- and $1.0\text{-}\mu\text{m}$ DOP aerosol.

Planned Experiments. The dielectrophoretic approach to filter enhancement appears extremely promising. During the next six months we plan to determine (1) what effects particle charging has on filter efficiency, (2) the fiber characteristics of the ideal filter, and (3) what filter design would best fit into existing systems.

Table 9. Results of filter efficiency tests

Run No.	Fiber material	Initial ΔP (in. H_2O)	Rate of ΔP change (in. H_2O/hr)	Filter efficiency, E_f (%)				Enhancement coefficient [($E_f E_0$)/ E_0]		
				$0(E_0)$	4 kV	8 kV	12 kV	4 kV	8 kV	12 kV
14	Glass	0.055	0.01	14.4	19.7	26.5	30.5	0.37	0.84	1.16
15	Wool polypropy- lene	0.98	0.66	68.7	76.6	80.8	80.3	0.12	0.18	0.17
20	Teflon	2.85	4.6	80.4	-	83.5	-	-	0.039	-
21	Polyester	0.092	0.012	12.3	21.4	29.0	36.8	0.74	1.36	1.99
22	Glass	0.502	0.144	37.1	51.5	64.8	70.8	0.39	0.75	0.91
23	Glass	0.165	0.0084	19.0	26.8	34.8	45.7	0.41	0.90	1.41
26	Glass	0.069	0.005	9.6	15.2	19.4	24.7	0.55	1.02	1.6
27	Glass	0.29	0.13	45.6	62.1	75.6	81.3	0.36	0.66	0.78
28	Glass	0.23	0.070	27.6	39.5	51.5	62.2	0.43	0.87	1.39
29	Glass	0.070	0.010	9.6	12.4	15.3	17.3	0.29	0.59	0.80
30	Glass	0.10	0.046	14.8	17.4	21.0	-	0.18	0.42	-
32	Glass	0.075	0.003	5.5	8.8	12.7	15.4	0.60	1.31	1.83
34	Polyester	0.075	0.004	14.1	15.4	16.8	-	0.015	0.031	-
35	Dacron	0.42	0.05	27.5	28.8	31.6	-	0.05	0.15	-
37	Glass	0.11	0.02	7.4	10.2	11.1	-	0.38	0.50	-
38	Polypropy- lene	1.35	2.6	62.0	62.9	64.6	-	0.01	0.04	-
40	Glass	0.25	0.04	30.6	54.0	70.9	79.0	0.76	1.32	1.58
41	Polyester	2.08	7.7	71.0	72.3	73.2	-	0.018	0.031	-
44	Polyester	0.08	0.01	8.1	12.1	17.1	-	0.49	1.11	-

Table 10. Drift velocities as a function of particle size^a

Particle diameter	Drift velocity		
	Inertial (cm/s)	Polarization ^b (cm/s)	Coulombic ^c (cm/s)
0.5	0.06	0.05	0.76
1	0.09	0.19	0.88
2	0.15	0.74	1.50
5	0.65	4.00	1.18
10	2.29	13.1	1.12
20	2.91	38.7	1.01

^a $R = 9 \times 10^{-4}$ cm, $I = 10.5$ esu, 180×10^{-6} poises
 $V_d = 6.6$ cm/s, $\sigma = 5.88$, and $q = 4.8 \times 10^{-10}$ esu

^b Calculated from Eq. (3)

^c Calculated from Eq. (5)

Fig. 51. Filter efficiency as a function of flow and voltage for a polyester batting.

Fig. 52. Filter efficiency as a function of flow and voltage for a glass fiber filter.

Technical Notes

RADIATION PROTECTION

Calibration of Andersson-Braun Remmeters with Track Etch Detectors

For the past four years, measurements of the environmental neutron dose levels at the LLL site perimeter have been made with ^{235}U track etch detectors encased in Andersson-Braun neutron remmeter moderators. The track etch detector consists of a vapor-plated $2\text{-}\mu\text{Ci}$ ^{235}U foil and a 25-mm diameter disk of 8- μ -thick polycarbonate. The polycarbonate is stretched over a lucite ring and held in place with a steel clamp ring. This steel ring also encircles the ^{235}U foil, keeping the foil and the polycarbonate face to face. Fission products from neutron-induced fission in the ^{235}U foil produce damage tracks in the polycarbonate film. These tracks are enlarged by etching in 60°C 6N NaOH for 1.5 hr. and are then counted by the spark technique³⁶ (Fig. 53).

In spark counting, a high voltage (500-800 V d.c.) is applied across the etched polycarbonate foil (B), the enlarged damage tracks providing an air gap or break-down path between conductors A and C. When a spark occurs, the potential across A and C drops to some value below the break-down potential, causing the spark to cease. The thin film of aluminum on conductor A is burned away in the vicinity of the spark, insuring that the track will not be counted repeatedly. The voltage then rises to the break-down point, and a spark occurs through another track. This cycle continues until there are no more tracks overlaid by the aluminum conductor. The sensitivity of this system is approximately 35 counts/mrem, with tracks resulting from spontaneous fission accounting for 20 to 30 counts for a 6-month period. The limit of

Fig. 53. Schematic of spark etch detector.

detection is therefore less than 1 mrem, providing a detector is sensitive enough for low neutron doses found at the fencelines.

In the latter part of 1975, we noted that the dose at the fenceline south of Building 212, as indicated by the track etch detector, was 100 mrem for a given period of time, while measurements by a health physicist indicated that the actual dose for that time period was 140 mrem. We suspected that this may have been due to the existence of a saturation point somewhere above the range of doses normally encountered. To determine response of track etch detectors with increasing dose levels, we exposed 33 foils for various times to a plated ^{252}Cf source, which would correspond to remmeter doses from zero to approximately 200 mrem. The response of the etch detector was clearly nonlinear (Fig. 54).

The decrease in response of spark foils with increasing dose is due to a combination of possible events. A spark burns a hole in the aluminumized conductor A on the order of 130-155 μ in diameter. No other tracks which are overlaid by this void can then be counted. Since the foil area exposed for counting is 2.84 cm^2 , this permits a maximum number of 1.4×10^4 spark holes, or a dose equivalent of approximately 400 mrem. Because the track spacing in the foil is random, the response error increases throughout the experimental range, beginning at low doses. In addition, the spark deteriorates the

Fig. 54. Spark etch detector response.

Fig. 55. Spark etch track.

polycarbonate around the original track, causing adjacent tracks to be perforated and counted with the subject track as one event. Finally, different angles of penetration of fission products result in overlying tracks at high dose rates (Fig. 55). These tracks can all be perforated by one spark and thus be counted as one event, or the adjacent tracks can remain intact and not be counted simply because the initial spark has removed the aluminum conductor overlying them. The contribution of these factors increases with total dose. However, the last two phenomena play a relatively minor role.

Optical counting of fission tracks in thick polycarbonate is a convenient method of augmenting spark counting techniques for dose levels in our range of interest. This method requires only a 25-mm-diam disk of 40-mil polycarbonate placed next to the previously mentioned ^{235}U foil. Damage tracks are produced in the same manner as in spark foils, and use of the Andersson-Braun moderator results in the same relative energy response characteristics. The polycarbonate disk is etched in 60°C 6N sodium chloride for 2 hr, and the tracks are counted optically using a transmission light microscope. Track/density is used to determine a dose equivalent.

Determining the proper ratio of tracks/cm² to mrem in thick polycarbonate was done as follows:

1. Using the low scatter facility in Building 255, a series of thick polycarbonate etch track detectors housed in Andersson-Braun moderators was exposed to the following spectra:
 - (1) Unmoderated ^{252}Cf
 - (2) ^{252}Cf moderated by 10 cm of polyethylene
 - (3) ^{252}Cf moderated by 20 cm of aluminum
 - (4) Unmoderated PuBe
 - (5) PuBe moderated by 10 cm of polyethylene
 - (6) PuBe moderated by 20 cm of aluminum
 These exposures were made 1 m from the source tube for time equivalent to a total dose equivalent to 500 mrem.
2. The samples were then etched and counted with a transmission light microscope at 400 X (Table 11). The optimum etching time for track counting was determined to be 2 hr.
3. Having determined optimum etching time, additional exposures were made using the same six spectra for times equivalent to 250 mrem.
4. These samples were etched and counted, yielding the results shown in Table 12.

Table 11. Etch track detector results for various Cf and PuBe spectra at 500 mrem

Spectrum	Counts/cm ²	Counts/cm ² -mrem
(1)	11 354	22.7
(2)	12 008	24.0
(3)	8 596	17.2
(4)	11 766	23.5
(5)	11 052	22.1
(6)	8 798	17.6

Table 12. Etch track detector results for various Cf and PuBe spectra at 250 mrem

Spectrum	Counts/cm ²	Counts/cm ² -mrem
(1)	5516	22.1
(2)	5640	22.6
(3)	4410	17.6
(4)	5657	22.6
(5)	5475	21.9
(6)	4459	17.9

Some explanation should accompany these data, since the counts/cm²-mrem are noticeably different in some cases. The Andersson-Braun moderator over-responds in the range from 25 to 200 keV, while the response falls off rapidly above 7 MeV.³⁷ The value for counts/cm²-mrem for the three spectra from a specific source (i.e., Cf or PuBe) is then a function of that particular spectral distribution. To obtain an overall calibration number for tracks/cm²-mrem in thick polycarbonate, the values for all spectra at both dose levels were averaged, yielding 21.0 tracks/cm²-mrem.

Particularly noteworthy is the agreement between values for a given spectrum at 250 and 500 mrem. This suggests a very linear response throughout the dose range of interest. This is expected because the optical counting method exhibits none of the characteristics which cause large response deviations in spark counting. Primarily, optical counting of each track eliminates the problem of undetected tracks due to effects such as those produced by the size difference between a track and its corresponding spark hole in the aluminum. Secondly, overlying tracks can be optically separated by adjusting the microscope focus, and thus be recorded as separate events. Finally, since there is no high-voltage spark present to damage adjacent tracks, a sample can be recounted as indefinite number of times.

Summary. The optical counting method of environmental neutron dose measurement has many advantages over the spark counting method. The thick polycarbonate foils are easier to prepare and etch, and there is less chance of damage due to handling. Overall sensitivity is greater with the optically counted etch detector, and the response is more linear than with the type used in spark counting. However, due to the long and tedious counting procedures, the optically counted etch detector has been reserved for use only in areas where the total dose normally exceeds 50 mrem for the exposure period. Selective use of a combination of both detectors provides both increased sensitivity and response in areas of high doses, and rapid dose measurement in low dose areas.

The Neutron Spectrum at the LLL Site Boundary

As part of LLL's program to monitor personnel radiation exposure, dosimeters are placed at the site boundary. Among these monitors are neutron dosimeters placed at the site boundary near the high-intensity 14-MeV neutron source (ICT). For some time we have been interested in measuring the neutron energy spectrum at the position of the dosimeters to (1) provide a proper calibration of the dosimeter

Fig. 56. Pulse-height data: (a) NE 213 detector, (b) ³He proportional counter, and (c) hydrogen proportional counter.

response, and (2) to provide physics data for the design of shielding in the event additional shielding is desired.

One previous spectrum measurement was made³⁸ but was incomplete because of equipment failures. A second measurement has now been made and is reported below.

During the week of 17 May 1976, the ICT was operated at maximum intensity (60 mA) for a total of about 60 hr. Neutron energy spectrum measurements were made on the fence line, at the point nearest the 14-MeV neutron source. Three detectors were used to obtain pulse-height data: (1) a 3.8 cm diam X 15 cm active length hydrogen proportional counter filled to 4 atm pressure; (2) a 5.1 cm diam X 45.7 cm active length ³He proportioned counter containing 4 atm ³He and 3 atm argon; and (3) a 11.4 cm diam by 5.1 cm deep NE213 liquid scintillator.

Pulse-height data were obtained by using the electronics configuration described in an earlier Progress Report.³⁹ Single parameter data were stored in a portable pulse-height analyzer and reduced by using the unfolding code NUTSPEC.⁴⁰ Results of the measurements are preliminary since the sensitivity of the proportional counter was minimally adequate for the existing neutron intensity, and statistics were poor. This is clear from the raw pulse-height data shown in Fig. 56.

The reduced neutron spectral data are shown in Fig. 57. Error estimates shown in the figure include only the statistical and fitting errors. Systematic errors are not included. Some assurance that the systematic

Fig. 57. Unfolded neutron spectrum at site boundary near ICT.

errors are not large is obtained from spectral measurements of monoenergetic neutron sources and PuBe or ²⁵²Cf sources which are in good agreement with calculations and measurements reported in the literature.

It is anticipated that another measurement will be made with one or more large proportional counters filled with hydrogen or methane. With greater sensitivity and better statistics, the data may be reported with greater confidence.

Light Particle Track Registration for Fast Neutron Personnel Dosimetry

Introduction. The three most prevalent detection methods used for personnel neutron dosimetry are: (1) NTA film (used in the photographic process to detect neutron-induced recoil tracks); (2) TLD albedo (detecting neutrons scattered from the body with a thermoluminescent dosimeter); and (3) fission track detectors (using a dielectric track recorder to detect neutron-induced fission from fissionable radiators).⁴¹ None of these can be considered a wide-range, general-purpose dosimeter because of serious deficiencies in the detection process or detector characteristics. Such problems include fading (NTA), gamma sensitivity (NTA and to a much lesser degree, albedo), inappropriate energy response (albedo), poor sensitivity for low-energy neutrons (NTA and most fission track detectors), and the need for radioactive material in personnel dosimeters (fission track detectors).

In recent years, we have studied the possible use of plastics such as cellulose nitrate or polycarbonate for detection of recoil particles or fast neutron induced (n, α) reactions. An attractive feature of light particle registration is that the reactions we would detect are responsible for the greatest part of the fast neutron dose in biological materials. Such a detector might be good enough to replace existing dosimeters, or more likely would be added to an existing dosimeter such as the TLD albedo to enhance its dependability and versatility.

One of the greatest obstacles to the use of light particle registration is that conventional track counting requires laborious, manual counting with a high-power (~400X) optical microscope. Our emphasis has been to find and adapt a process which improves track visibility. We have recently studied two methods for track contrast enhancement.

For many years, Kodak Pathe[®] has produced a special film which consists of a 10-μ layer of cellulose nitrate⁴² impregnated with deep red dye, which is coated on one side of a sheet of polyester. The neutron-induced particle tracks can be chemically

Fig. 58. Electrochemically etched tracks in polycarbonate (70x).

etched and enlarged so that they perforate the red layer and thus are readily visible with a low-power microscope ($\sim 35\times$), particularly when a green or blue filter is used over the incident light.

The second process involves electrochemical etching⁴³⁻⁴⁵ to enlarge neutron damage tracks in polycarbonate so that they are also readily visible with a low-power microscope (Fig. 58). Simply stated, electrochemical etching involves enlarging the track by applying an a.c. potential across the sample during etching in a caustic solution.

Energy Response. Important to the use of these detectors is their energy response and sensitivity. As part of a recent series of monoenergetic neutron calibrations at the LLL Cyclograph,^{46,47} we placed samples of a polycarbonate (LR115) and conventional cellulose nitrate (Kodak Pathe' CA8015) on an arc 100 mm from the target point. Using the reactions shown in Table 13, we were able to determine the neutron energy response of the detectors from 0.27 to 5.7 MeV and from 12.6 to 18.9 MeV. The neutron fluence was measured with a de Pangher long counter

Table 13. Cyclograph neutron reaction summary

Reaction	Particle energy	Neutron energies
${}^3\text{H}(p,n){}^3\text{He}$	2.3 MeV	0.27-1.51 MeV
	3.5	0.61-2.72
	6.5	1.48-5.73
${}^3\text{H}(d,n){}^4\text{He}$	2.5 MeV	12.6-18.9

and indium foils, using the ${}^{115}\text{In}(n,n'){}^{115\text{m}}\text{In}$ reaction. The long counter was 2 m from the target, while the indium foils were placed at the location of each sample (100 mm from the target).

After irradiation, the LR115 polycarbonate samples were etched using the conditions shown in Table 14. We have found that the neutron response of the electrochemically etched polycarbonate (E.C.E.P.) samples are enhanced significantly by exposure to UV light before being etched. The UV exposure enhances the photo-oxidation process⁴⁸ which increases the reactivity of the etching process in the damage track. One reason the UV exposure is effective with our samples is that, to reduce background caused by amplification of surface abrasions, we buy polycarbonate which is covered with thin polyethylene film. This film may retard the natural photo-oxidation.

After etching, the tracks have the size distribution shown in Fig. 59. The distributions were measured with a commercial optical particle size analysis system. Data for the CA8015 cellulose nitrate were obtained several years ago,⁴⁹ so specific analysis was not done on the current samples. The size distributions give

Table 14. Track detector processing parameters

Cellulose nitrate: CA8015	- 10% NaOH
	40°C
	3 hr
LR115	- 10% NaOH
	60°C
	2.75 hr
Polycarbonate: 1 hr exposure to UV (301, 312, 333 and 365 nm)	
Etch - 28% KOH,	
22°C	
800 V, 2000 Hz	
5 hr

Fig. 59. Track size distribution in light particle detectors.

some idea of the relative visibility of the two track enhancement techniques.

The measured responses of the detectors are shown in Fig. 60. Note that the track density represents total

tracks on both sides of a 1-cm^2 E.C.E.P. sample area. The scatter in the data for the CA8015 film is so bad that we could not include it. Therefore, the solid lines are shown only for rough comparison with the

Fig. 60. Energy response of light particle track detectors.

enhanced track techniques. The magnitude of estimated errors in counting and fluence measurement are shown by the four error bars. Note also that the E.C.E.P. response in the region from 5.7 to 12.6 MeV can be interpolated easily (dashed line), but for LR115 it is not clear, without further investigation, what happens.

Background. Surface abrasions cause artifacts which contribute to the background for both E.C.E.P. and LR115 techniques. We ignore tracks in the E.C.E.P. samples which lie on a scratch or do not look like normal neutron-induced tracks. Examples of abnormal tracks are given elsewhere.⁴⁴ In practice, we have found the working background in our E.C.E.P. material to range from 1.0 to 4.0 tracks cm^{-2} with a mean of about 2.5 tracks cm^{-2} . The LR115 background is about 10 times as great, with the same relative range. It is quite convenient to count as much as 10 cm^2 to obtain a given data point with E.C.E.P. samples, but the LR115 samples are more tedious to count, so only an area of 2.4 cm^2 can be counted in the same amount of time.

Sensitivity. In both techniques, there are factors which affect the track background and thus cause the variations to be somewhat nonstatistical. These include variations due to abrasion and, in the case of LR115, variations in film thickness. These variations made it difficult to accurately determine sensitivity; however, experience tells us that an induced track density equal to the normal background is necessary for detection. If we use that criterion, together with the energy response data just presented and accepted neutron-to-dose-equivalent conversion factors,⁵⁰ Fig. 61 gives us an estimate of the dose equivalent sensitivity as a function of energy.

Dependence on Orientation. We have found that with distributed neutron spectra such as that from ^{252}Cf , there are more tracks on the neutron incident side of the detector than on the exit side. A tentative explanation is that the etching process is most active at the tip of a track closest to the sample center. For incident neutrons, the associated charged particles have their greatest value of LET near the center of the

Fig. 61. Estimated sensitivity of light particle track detectors.

Fig. 62. Ratio of incident to exit side track densities in neutron irradiated electrochemically etched polycarbonate.

sample, causing more etchable damage where the etching process is most active.

The energy response of this directionality is shown in Fig. 62. Note that the deviation of R from unity is prominent only near the low-energy response threshold. However, in distributed spectra, there are enough neutrons in this energy range to be useful for determining orientation. This information would, of course, be valuable for assessing the effect of body interaction on dosimeter response, and could be quite helpful for making a more accurate dose estimate.

Discussion. The two neutron track detection methods considered are only sensitive to neutrons with energies greater than 1.5 MeV. As a result, neither can

be considered useful as a single, general-purpose dosimeter. However, the fast neutron sensitivity (<50 mrem), flat energy response, and orientation dependance of the E.C.E.P. process make it attractive for use in conjunction with an albedo dosimeter. It would be difficult to automate the etching and readout of the E.C.E.P. technique; however, in conjunction with an automated TLD albedo system, the E.C.E.P. need only be etched and counted if the albedo dosimeter (typically quite sensitive) showed a positive response.

Acknowledgments. The authors wish to thank Art Biermann and Chuck Shettevold for the track size analyses.

INSTRUMENT DEVELOPMENT

Monitoring System for Detecting Environmental Levels of ^{41}Ar

The Environmental Evaluation Group of the Hazards Control Department must measure the concentration of any of our atmospheric effluents which might reach the project boundaries or beyond. One such effluent is ^{41}Ar , produced by neutron activation of atmospheric ^{40}Ar during normal operation of the LPTR. We were asked to design an instrument to make such measurements. The requirements were: Sub-MPC sensitivity, portability, and, since several installations might eventually be required, low cost was desirable.

^{41}Ar undergoes beta decay with a 1.83-hr half-life, and emits both a 1.198-MeV (E_{max}) beta and a 1.293-MeV gamma. To measure concentration by gamma counting would require either energy discrimination and/or heavy shielding, both of which

tend to be costly. Beta counting eliminates both of these requirements. However, one must be sure that only ^{41}Ar betas are being counted. Fortunately, ^{41}Ar is the only gaseous, energetic beta-emitting species found in our reactor effluent with a lifetime long enough to allow transport to the site boundary.* Consequently, if a monitoring system excludes radon daughters (which tend to be associated with filterable particulates) and provides shielding against external beta radiation, any beta counts recorded will be due to ^{41}Ar . A detector with high beta sensitivity and low gamma sensitivity, such as a Geiger-Mueller tube, will

* ^{85}Kr produced by activation does exist but in such minuscule quantities as to be negligible. ^{85}Kr produced as a fission product is contained in the fuel elements, and is not part of the effluent.

Fig. 63. External view of ^{41}Ar monitoring system.

Fig. 64. Detector assembly removed from 55-gal drum.

provide some discrimination against external gamma background.

We have designed and built an instrument that meets these requirements, utilizing ideas published by Jones.⁵¹ It consists of a steel 55-gal drum with two 1B85 G-M tubes mounted axially at the center. These are supplied with high voltage by an external supply, and the signal is picked off via a blocking capacitor and fed directly to the negative input of a scaler/timer. A small rotary-vane pump pulls air through a membrane filter with a $0.8\text{-}\mu\text{m}$ pore size at about 0.5 l/s . Figure 63 shows an external view of the system, and Fig. 64 shows the detector assembly removed from the drums.

The instrument, while bulky, is not heavy, and can easily be carried. It requires 110 V a.c. (unregulated) which may be supplied by a portable generator for field use. The electronic package requires only two NIM-widths and is normally housed in a small, portable NIM-Bin. The prototype is not weatherproof; however, the feature could easily be added if required.

The instrument was calibrated against a special ion chamber whose response to ^{41}Ar was well documented.⁵² Air from the main exhaust duct of the LPTR building which contained considerable ^{41}Ar was pumped into the drum. Then a closed loop was set

up, circulating the drum's contents through the ion chamber and back into the drum. Count rates in the G-M tubes were corrected for dead-time losses, and then related to activity in the ion chamber. This was repeated at various concentrations. A final calibration factor was finally derived from these data:

$$\frac{\mu\text{Ci } ^{41}\text{Ar}}{\text{cm}^3} = \frac{\text{net counts/min}}{5.5 \times 10^8}$$

Backgrounds in normal environments are $70\text{--}80\text{ counts/min}$. This gives a minimum detectable concentration (MDC) (at the 95% confidence level) of $\sim 1.5 \times 10^{-8}\text{ }\mu\text{Ci/cm}^3$, in 10 min counting time. The ^{41}Ar MPC for uncontrolled areas is $4 \times 10^{-8}\text{ }\mu\text{Ci/cm}^3$ so in 10 min , we can detect levels of about 40% of the MPC. Extending the counting time to 100 min drops the MDC to $\sim 5 \times 10^{-9}\text{ }\mu\text{Ci/cm}^3$, or just over $1/10\text{ MPC}$. Since the response time of the system is on the order of 30 min (to equilibrium), these counting times are not excessively long. Thus, we seem to have met the design requirements. Field testing, scheduled for the near future, will be necessary to determine reliability, but the extreme simplicity of the system offers hope for long life.

Publications

T. Jordan Powell *An Unusual Problem in Tritium Dosimetry: Tritium in Glass*. Lawrence Livermore Laboratory, Rept. UCRL-77674 (presented at the Twenty-First Annual Meeting of the Health Physics Society, San Francisco, California, June 27-July 2, 1976).

Glass microballoons having a diameter in the range of 30 to 100 μm and a wall thickness of about 7 μm are used as targets for laser fusion experiments. The microballoons are filled with a deuterium-tritium gas mixture by diffusing the gas through the wall of the microballoon at high temperature and pressure. A typical microballoon might contain several microcuries of tritium after loading.

Hazards analyses indicate that these microballoons will present little hazard if they remain intact. If a microballoon is broken, a slight hazard will be present for a short time due to the release of the contained tritium. However, a long-term respiratory hazard may exist because of tritium contained in interstitial spaces in the glass fragments.

We have examined the glass fragments of a number of broken microballoons, and many are in the respirable size range. Further, we have confirmed that tritium is trapped in these fragments and retained for long periods of time. The fragments are relatively insoluble in lung fluids and may be considered to behave in the lungs as an ICRP Class Y aerosol. In this case, the dosimetry of the contained tritium differs radically from that of tritium in more common forms. For example, an incident that deposits 100 μCi of tritium in this form in the pulmonary lungs will deliver a radiation dose of about 20 rad to the lungs over the course of several years. Contrasted with this dose, 100 μCi of tritium inhaled as water will deliver a dose of only 0.007 rad to the lungs (and whole body).

W. J. Silver, C. L. Lindeman, J. W. Meadows, E. H. Willes and D. R. McIntyre, *Environmental Monitoring at the Lawrence Livermore Laboratory 1975 Annual Report*, Lawrence Livermore Laboratory, Rept. UCRL-50027-75 (1976).

In 1975, the average annual gross beta activity on air filters was about half of that observed during 1974. There were corresponding decreases in specific fission product radionuclides, reflecting the reduction in fallout of nuclear debris from the large-yield Chinese atmospheric nuclear explosion in 1973. Airborne ^{238}U concentrations at Site 300 were higher than those at Laboratory perimeters because of the use of "depleted" uranium (a byproduct of ^{235}U

enrichment) at the Site. These uranium concentrations were well below the standards set by ERDA.

Soil samples collected in the off-site vicinity of the Laboratory and at Site 300 were analyzed for plutonium. There were negligible changes from the levels previously reported.

Water samples collected within the Livermore Valley and Site 300 exhibited normal background gross beta and tritium activities.

Vegetation samples collected in areas generally downwind from the Laboratory contained tritium activities generally higher than those collected in areas where the Laboratory's contribution should be minimal. Tritium levels in Site 300 vegetation were essentially unchanged from those observed in 1974.

The median annual gamma radiation dose rate at the Laboratory perimeter was 62 mrem compared with 59 mrem as the median background dose rate in the off-site vicinity. Operation of a 14-MeV neutron generator near the south Site perimeter continues to be a source of elevated radiation.

Both Laboratory perimeter and Site 300 annual average airborne beryllium concentrations were less than 0.002% of the appropriate standard.

Releases of heavy metals to the Livermore sanitary sewer system conformed to the discharge regulations of the city of Livermore.

No Laboratory effluent resulted in an estimated radiation dose to the nearest resident exceeding 1 mrem. Assessment of the radiation doses to an individual from the environmental activities listed in this report demonstrates that the dose contribution from Laboratory operations in 1975 was very small compared with the approximately 100-mrem/year dose locally received from natural sources.

P. H. Gudiksen, T. R. Crites, and W. L. Robison, *External Dose Estimates for Future Bikini Atoll Inhabitants*, Lawrence Livermore Laboratory, Rept. UCRL-51879 Rev. 1 (1976).

To evaluate the potential radiation doses that may be received by the returning Bikinians, we surveyed the residual radioactivity on Bikini and Eneu Islands in June of 1975. An integral part of the survey included measurements of gamma-ray exposure rates which are used to estimate external gamma-ray doses. The survey showed that on Bikini Island the rates are highly variable: values near the shores are generally of the order of 10 to 20 $\mu\text{R/h}$, while those within the interior average about 40 $\mu\text{R/h}$ with a range of roughly 30 to 100 $\mu\text{R/h}$. Eneu Island, however, is characterized by more or less uniformly distributed

gamma radiation levels of less than 10 $\mu\text{R}/\text{h}$ over the entire island.

These data, in conjunction with population statistics and expected life styles, allowed us to estimate the potential external gamma-ray doses associated with proposed housing locations along the lagoon road and within the interior portions of Bikini Island as well as along the lagoon side of Eneu Island. As expected, living on Eneu Island results in the lowest doses: 0.12 rem during the first year and 2.9 rem during 30 years. The highest values, 0.28 rem during the first year and 5.9 rem over 30 years, may potentially be received by inhabitants living within the interior of Bikini Island. Other options under consideration produce intermediate values.

C. L. Lindeken, J. H. White, A. J. Toy and C. W. Sunbeck, *Ambient Environmental Radiation Monitoring at the Lawrence Livermore Laboratory*, Lawrence Livermore Laboratory, Rept. UCRL-77106 (1975) (presented at the Health Physics Society Ninth Midyear Topical Symposium on Operational Health Physics, Denver, Colorado, February 9-12, 1976).

Thermoluminescence dosimetry is the principal means of measuring ambient gamma radiation at the Lawrence Livermore Laboratory. These dosimeters are used at 12 perimeter locations and 41 locations in the off-site vicinity of the Laboratory, and are exchanged quarterly. Control dosimeters are stored in a 75-mm-thick lead shield located out-of-doors to duplicate temperature cycling of field dosimeters. Effect of dosimeter response to radiation in the shield is determined each quarter. Calibration irradiations are made midway through the exposure cycle to compensate for signal fading. Terrestrial exposure rates calculated from the activities of naturally occurring uranium, thorium, and potassium in Livermore Valley soils vary from 3 to 7 $\mu\text{R}/\text{hr}$. Local inferred exposure rates from cosmic radiation are approximately 4 $\mu\text{R}/\text{hr}$. TLD measurements are in good agreement with these data. Off-site and site perimeter data are compared, and differences related to Laboratory operations are discussed.

Paul H. Gudiksen and Thomas R. Crites, *A Comparison of Gamma-Ray Exposure Rate Measurements at Bikini Atoll*, Lawrence Livermore Laboratory, Rept. UCRL-77532 (1975) (presented at the Health Physics Society Ninth Midyear Topical Symposium on Operational Health Physics, Denver, Colorado, February 9-12, 1976).

A radiological survey of Bikini and Eneu Islands of the Bikini Atoll was conducted during June 1975 to assess the potential radiation doses that may be received by the returning Bikinians. Bikini Atoll was

one of the U.S. nuclear weapons testing sites in the Pacific. An integral part of the survey included measurements of the gamma-ray exposure rates at 1 m above the ground with portable NaI instruments at nearly 2700 locations on the two islands. For comparison purposes, similar measurements were made with a pressurized ion chamber at approximately 200 locations, and with LiF and $\text{CaF}_2:\text{Dy}$ thermoluminescent dosimeters (TLDs) at 80 locations. The results indicate that the NaI scintillators overresponded because of their nonlinear energy characteristics. The responses of the LiF dosimeters and the pressurized ion chamber agreed to within 13%. Attenuation studies with LiF TLDs indicated that roughly 25% of the total free air exposure rate at 1 m was due to beta radiation.

Dennis R. Slaughter, *A Simple Method to Unfold NE 213 and Stilbene Neutron Spectral Data*, Lawrence Livermore Laboratory, Rept. UCRL-77849 (1976) (presented at the RSIC Seminar-Workshop on Radiation Energy Spectra Unfolding, ORNL-RSIC-40, Oak Ridge, Tenn. (April 12-13, 1976)).

A very simple method is proposed wherein recoil proton scintillation data are reduced using the derivative unfolding technique. The approximations described require minimal calculation or storage and may be readily implemented on a mini computer. A digital filter is incorporated which yields a smoothed approximation to the slope of the pulse-height distribution, with error estimates. In order to accurately represent the scintillator light output without introducing discontinuities in the unfolded spectrum, the light output data are fitted using B-splines whose first and second derivatives are continuous.

D. E. Hankins, *Evaluation of the Fast Neutron Dose Equivalent Using the Thermal Neutron Response of LiF TLD Material*, Lawrence Livermore Laboratory, Rept. UCRL-77678 (1976) (presented at the Twenty-First Annual Meeting of the Health Physics Society, San Francisco, California, June 27-July 2, 1976).

A procedure was developed to evaluate the fast neutron dose equivalence from the thermal neutron readings of LiF TLDs located in personnel badges. The "percent thermal" (dose rate of the thermal neutrons divided by the total neutron dose rate) is determined using a survey procedure which is fast and easy to make under field conditions. The percent thermal must remain fairly constant for this procedure to be useful. Extensive surveys were made at LLL facilities and, although large differences were found between various facilities, within each one the percent thermal remains

power at the time, the need for solutions to these problems will intensify.

Richard V. Griffith and John C. Fisher, Light Particle Track Registration for Fast Neutron Personnel Dosimetry. Lawrence Livermore Laboratory, Rept. UCRL-77661 (1976) (presented at the Twenty-first Annual Meeting of the Health Physics Society, San Francisco, California, June 27-July 2, 1976).

A study has been made of the neutron energy response and background levels of three track-etch materials used for the registration of recoil nuclei. The ^{252}Cf isotope source was used to produce monoenergetic neutrons in the range 0.1 to 19 MeV. We determine the relative sensitivity of conventional cellulose nitrate as well as two materials which make use of track-etch enhancement techniques: Kodak Pathé FR115 cellulose nitrate¹ with red dye contrast enhancement² and polycarbonate film with acetic anhydride etching.³ We also made measurements of the response of the FR115 and polycarbonate materials to ^{252}Cf fission neutrons with a variety of moderators. Although the sensitivities of the track-etched materials are not as high as that for conventional cellulose nitrate, overall optically, the track-etches are much lower and the limits of detection are below 0.5 rem. For both FR115 and polycarbonate, care must be taken in handling and etching to prevent unacceptable background variations.

Both the FR115 and polycarbonate, with acetic anhydride etching, can be useful for fast neutron dosimetry. An attractive approach would be to use one of them in combination with a TLD albedo dosimeter to take advantage of the ease and sensitivity of the albedo technique at low doses and the energy response of the track registration at levels above a few hundred μrem .

I. A. Gibson and D. S. Muntan, Simplified Scheme for Radiometric Plume Calculations. Lawrence Livermore Laboratory, Rept. UCRL-77092 (1976) (presented at the Ninth Midyear Topical Symposium of the Health Physics Society, Denver, Colorado, February 13-17, 1976).

A simplified mathematical scheme to estimate external whole-body gamma radiation exposure rates from gaseous radioactive plumes was developed for the Rio Blanco Gas Field Nuclear Simulation Experiment. The method enables one to calculate swiftly, in the field, downwind exposure rates knowing the meteorological conditions and gamma radiation

exposure rates measured by detectors positioned near the plume source. The method is straightforward and easy to use under field conditions without the help of mini-computers. It is applicable to a wide range of radioactive plume situations. It should be noted that the Rio Blanco experiment was detonated on May 17, 1975, and no seep or release of radioactive material occurred.

Norman J. Alvares, Ignition Inhibitors for Cellulose Materials. Lawrence Livermore Laboratory, Rept. UCRL-78026 (1976) (presented at the 172nd American Chemical Society Meeting, San Francisco, California, August 29-September 2, 1976).

By exposing samples to various irradiance levels from a calibrated thermal radiation source, the ignition responses of blackened alphacellulose and cotton cloth with and without fire-retardant additives were compared. Samples treated with retardant compounds which showed the most promise were then isothermally pyrolyzed *in vitro* so that comparisons between the pyrolysis rates of the samples could be obtained. This was done to gain further insight into the mechanisms of ignition and degradation in attempts to arrive at a better understanding of flammability.

Alphacellulose samples containing a mixture of boric acid, borax, and ammonium dihydrogen phosphate could not be ignited by irradiance up to $40 \text{ cal cm}^{-2} \text{ sec}^{-1}$ (15 W cm^{-2}). At higher irradiances the specimens ignited, but flaming lasted only until the flammable gases were depleted. Cotton cloth containing a polymeric retardant with the designation THRC + MM was found to be ignition-resistant to irradiances below $70 \text{ cal cm}^{-2} \text{ sec}^{-1}$ (29 W cm^{-2}). Comparison of the pyrolysis rates of the retardant-treated mechanism is qualitatively the same.

Similar ignition-response measurements were also made with specimens exposed to ionizing radiation. We observed that gamma radiation results in ignition retardance of cellulose, while irradiation by neutrons does not.

N. J. Alvares, Trends Between Residential and Industrial Fire Protection for Upright Public Safety. Lawrence Livermore Laboratory, Rept. UCRL-77774 (1976) (presented at the Fifth Environmental Symposium, SRI, Menlo Park, California, May 12-13, 1976).

In 1976, the losses in the United States totaled about 0.25% of the GNP, or 4.4 billion dollars. Statistics on distribution of the types show that 50% involve residential dwellings, 10% industrial, institutional, and educational buildings, 21% are due to transportation-related factors, and the remaining

¹ Barber, *Proc. 1966 Meet. Soc.*, 13, 530 (1970).

² M. S. Gibson, *Health Phys.*, 27, 530 (1976).

38% include forest, grassland, and rubbish fires. These statistics show that industrial and public facilities account for almost 50% of the financial loss statistics, while residential and transportation losses amount to 36 and 11% of the total respectively. More than 60% of fire fatalities are attributed to building fires, and of these, almost 90% occur in private residences.

This brief survey reveals that a relatively small number of fires are responsible for the major dollar losses, and the major loss of life in fires results from residential fires, where the number of fatalities per fire are relatively small.

Can technology be applied to reduce either the financial disaster incurred during industrial fires, or the life loss in residential fires? The evidence indicates that residential fire mortality will not be significantly reduced by technical solutions. However, there is also the potential for large life loss in industrial fires, and these could be reduced significantly by technical solutions. Therefore, increased efforts to secure the optimum amount of industrial fire protection could certainly reduce our financial losses, and possibly protect more lives.

James R. Gaskill, *Coordination of Fire Terminology in ASTM*, Lawrence Livermore Laboratory, Rept. UCRL-78327 (1976) (presented at the 14th USERDA Air Cleaning Conference, Sun Valley, Idaho, August 2-3, 1976).

Legal and technical challenges as to the meaning of terms used in the field of fire technology have led to the establishment of a broad-based, coordinating group within ASTM. Their purpose is to ensure that definitions and terms related to ASTM fire test standards are technically correct and cannot be used in an inappropriate or misleading manner. The mechanics of such coordination are described and lists of terms under different stages of review are given.

Thomas M. Distler and Carla M. Wong, *Pesticide Residue Analysis of Storm-Drain Waters, 1975*, Lawrence Livermore Laboratory, Rept. UCID-17079 (1976) March 12, 1976.

Runoff-water samples for the first, third, and fourth quarters of 1975 were analyzed for pesticide residues at LLL and independently by the LFE Environmental Analysis Laboratories. For the compounds analyzed, upper limits to possible contamination were placed conservatively at the low parts-per-billion level. In addition, soil samples were also analyzed.

Future work will continue to include quarterly sampling and will be broadened in scope to include quantitative analysis of a larger number of compounds.

A study of recovery efficiency is planned. Because of the high backgrounds on soil samples together with the uncertainties introduced by the cleanup procedures, there is little hope of evaluating the distribution of a complex mixture of pesticides among the aqueous and solid phases in a drainage sample. No further sampling of soil from the stream bed is therefore contemplated.

Gary O. Nelson and A. Nicholas Correia, *Respirator Cartridge Efficiency Studies: VIII. Summary and Conclusions*, Lawrence Livermore Laboratory, Rept. UCRL-77654 (1976).

The service life of organic vapor cartridges has been determined at various flow, concentration, temperature, humidity and solvent vapor conditions. A simplified empirical expression to estimate service life has been derived which compares favorably with experimental values. This paper has been submitted and accepted by the American Industrial Hygiene Association Journal.

Gary O. Nelson, Charles P. Richards and Robert D. Taylor, *Air Filtration Enhancement Using Electronic Techniques*, Lawrence Livermore Laboratory, Rept. UCRL-77852 (1976) (presented at the 14th USERDA Air Cleaning Conference, Sun Valley, Idaho, August 2-3, 1976).

High efficiency particulate air (HEPA) filters are commonly used at radioactive handling facilities to remove airborne contaminants. If filter service life could be increased, the following benefits could be realized: (1) less handling of contaminated filters, (2) reduction of the radioactive waste inventory, and (3) reduction in the number of filters required. The major thrust of our research effort is to significantly improve the effectiveness of the low efficiency prefilter which precedes the HEPA filter using electronic techniques.

We are currently considering two approaches - particle agglomeration and dielectrophoresis. Particle agglomeration focuses on coalescing the small particles into larger ones in order to remove them more efficiently by the prefilter. Dielectrophoresis involves placing an electric field across the nonconductive prefilter which causes particle and prefilter polarization. This condition greatly increases the prefilter effectiveness.

Our testing medium currently consists of a 0.8 μ m mass median aerodynamic diameter sodium chloride aerosol with a geometric standard deviation of 2.1 flowing at 25 l/s. The system can accommodate up to four Wright-type nebulizers and can produce a salt concentration of 1-10 g/l.

G. O. Nelson, C. P. Richards, R. D. Taylor and A. H. Biermann, *Enhanced Filtration - October-December 1975 Progress Report*, Lawrence Livermore Laboratory, Rept. UCID-16949-75-4 (1976).

A 25-l/s aerosol generation system has been built. The aerodynamic mass median diameter of the particle is 0.64 with a standard deviation of 2.1. Preliminary experiments using electrostatic precipitator-type particle agglomerators are shown.

E. E. Volk and R. D. Partidge, *Decontaminating Gloved Boxes*, Lawrence Livermore Laboratory, Rept. UCRL-77657 (1976) (submitted to Health Physics).

Gloved boxes used in work with transuranium nuclides generally become contaminated at levels >10 nCi/g, requiring their emplacement in interim isolation (i.e., up to 20 yr) at an ERDA-controlled site. Because such glove boxes are frequently too large to be packaged in DOT- and NRC-approved shipping

containers, they must be modified (e.g., by decontamination to levels >10 nCi/g) so that they need not be sent to interim storage. A new, relatively efficient method of decontaminating a glove box involves (1) spraying the internal surfaces of the box with a solution of 30% nitric acid and 1% hydrofluoric acid, (2) allowing the box to stand for at least 1 hr to enable the acid to dissolve the nuclide particles wedged in microscopic crevices, (3) withdrawing the spent acid solution by suction, (4) rinsing the box two or three times with water, and (5) removing the remaining visible water with absorbent paper. The concentration of contaminant remaining in the air-dried box is determined with an alpha survey meter probe. If the level averages >10 nCi/cm², the acid wash is repeated. If the level is <10 nCi/cm², the box is filled with other radioactive waste and packaged as low-specific-activity waste for shipment to a commercial burial site.

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