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LAWRENCE LIVERMORE NATIONAL LABORATORY
University of California · Livermore, California · 94550

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MASTER

Foreword

This report, which covers the period from October 1981 to September 1982, 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 may contact the authors of the reports.

Contents

PROGRESS REPORTS

Industrial Hygiene

Field Testing of Carbon Adsorber Beds	2

Instrument Development

A Real-Time Monitor for Tritium in Waste Water	5

Environmental Protection

Adding an On-Line X-Ray Fluorescence Analyzer for Metals to the LLNL Sewer Monitoring System	11

Radiation Protection

Progress in Multi-Target TRAX Development	13
Development of a Passive Personnel Neutron Dosimeter/Spectrometer—DOSPEC	17

Fire Safety

Effectiveness of Standard and Alcohol-Resistant Aqueous-Film-Forming Foam (AFFF) on Alcohol Fires	25

TECHNICAL NOTES

Industrial Hygiene

Determination of Explosives Using High-Performance Liquid Chromatography	29
Self-Contained-Breathing-Apparatus Protection at High Work Rates	33

Radiation Protection

Neutron and Gamma-Ray Measurements at the LANL SHEBA Critical Assembly	36
Additional Calibrations of the PTB Beta Sources	45
Probability of Interaction and Energy Deposition of Neutron-Induced Recoil Protons in a Cell Plasma Membrane	47
Calibration of the PNR-4 Neutron Remmeter for Weapons-Like Spectra	50
A Single-Calculation Comparison of MORSE76L and ANISN	52
Effective Absorption Thickness of the ⁷ Li TLD for Beta Particles	56

Fire Safety

Evaluation of the Fire Properties and Potential Safety Hazard of Building 431's Exterior Wall Panels	59

PUBLICATION ABSTRACTS	67

Progress Reports

Field Testing of Carbon Adsorber Beds

P. M. Siewarengen, J. S. Johnson, R. A. da Roza, and C. A. Harder

Activated carbon adsorbers are used in air cleaning systems at LLNL to remove hazardous gases or vapors. The Safety Science Group has been working on an in-place field test procedure to measure the residual adsorption capacity (RAC) of large-bed activated carbon adsorbers. The effects of humidity and organic vapor load on testing for RAC have been studied. A parallel-flow sampling manifold for an in-place field test is near completion and will soon be installed; laboratory experiments have been performed that will guide testing of the cartridges from the sampling manifold. Finally, a portable field test instrument is being prepared for testing carbon beds at LLNL.

Experimental

Previous work has indicated that a chromatographic theory can be used to provide a measurement of the residual adsorption capacity (RAC) of carbon adsorbers.^{1,2} Recent work done in the Safety Science Group at LLNL has also shown that pulse testing with methane or ethane gas can indicate the RAC of a carbon bed.³ In this case, activated carbon cartridges (cylindrical, 2 in. in diameter by 4 in. long) were loaded with varying amounts of toluene. The carbon was then tested with a 5-ml pulse of ethane. Typical data are shown in Table 1. As the RAC decreases, the retention time (RT) for ethane on the adsorber also decreases; RT of the test gas is thus proportional to RAC. When data from a series of such experiments are compiled, analytical-type curves can be derived. Such an analysis is shown in Fig. 1.

Although adsorber testing with a weakly adsorbed gas like ethane can provide information on the condition of the bed, our work has also shown a limitation of this type of test. From Fig. 1, it is apparent that above 50% relative humidity (RH), a pulse test will provide no information about the RAC of an adsorber bed. The humidity of the air in the system containing the adsorber bed is thus a critical parameter. It should be noted that in these laboratory experiments, the carbon was subjected to a 24-h equilibration period in the humidity conditions chosen for testing. The purpose of the equilibration was to maintain the amount of water on the carbon and in the air stream as nearly constant as possible during testing.

Evidence to show that the amount of water on carbon has an effect on the RT of the test gas is shown in Table 2. The data presented were obtained under experimental conditions identical to those shown in Table 1. The first column shows the RH conditions during equilibration and testing. The second column shows the RT observed for ethane at the start of the test, before any toluene

Table 1. Results of pulse-testing^a a carbon cartridge^b with ethane gas.

Exposure time (h)	RT (min)	RAC (%)
0	2.50	100
0.40	1.70	90
0.72	1.50	83
1.02	1.25	76
1.40	1.00	67
1.52	0.85	64
1.74	0.71	58
2.10	0.49	50
2.31	0.35	45
2.50	0.22	40
3.04	0.14	27

^a Flow velocity of ethane = 8.2 cm/s.

^b Sutcliffe Speakman 2089C carbon loaded with 3000 ppm toluene in air flow of 10 liters/min (air at 40% RH, 25°C).

Figure 1. RAC of carbon cartridges pulse-tested with ethane, at various relative humidities of air.

was loaded on the carbon. The RT for ethane decreases as the RH increases. Eventually, RT decreases to the condition described above, where no information on RAC is provided.

Two other phenomena pertaining to carbon were revealed during this investigation. First, at RH values greater than 50%, the RAC of the carbon is still quite high. As shown in the fourth column of Table 2, nearly the same mass of toluene can be removed from the air stream at high RH conditions as at low. Adsorbed water can apparently affect the carbon surface so that the weakly adsorbed ethane is barely retained, causing the pulse test to fail. The more strongly adsorbed toluene, however, is still retained by carbon even in the presence of significant quantities of water.

A second important phenomenon is desorption. A carbon cartridge completely saturated with

toluene was left in the flow system and exposed to humid air containing no toluene; the downstream air was monitored and the toluene was observed to desorb from the carbon. With time, the RAC of the carbon increased and returned to a level near that of the new material. This was measured by a steady increase in the RT for ethane (pulse tested) as desorption proceeded. In the case of carbon tested at humidities of 40% or lower (where the ethane pulse test is a good indicator of RAC), the RT values returned to their original (preloading) values. In the higher-RH cases, the carbon was found to have renewed its capacity for toluene. When we again challenged the bed with an air flow containing 3000 ppm toluene, the carbon cleaned the air as it did before saturation.

Table 2. Effect of relative humidity on the RT of ethane.

% RH	RT of ethane at start of test (min)	Weight of carbon in adsorber bed (g)	Weight of toluene (g)	Weight of water after 24-h equilibration (g)
<3	16.4	109.4	35.5	—
30	8.0	111.9	26.3	8.3
40	2.5	113.7	24.8	14.6
50	0.31	109.8	21.8	33.9
60	0.16	111.9	21.8	34.9
70	0.13	110.8	24.0	36.3

Discussion

The desorption of toluene that we observed raised several questions. First, what are the general trends of desorption on compounds of interest to industrial hygienists at LLNL? Second, what desorption trends are observed when chemicals are compared by physical constants such as boiling point, vapor pressure, molecular size, and functional groups (polarity) on the molecule? Finally, can a "retentivity index" for organic molecules be derived that would be of functional value to the industrial hygiene community?

Future Work

An instrument is currently being modified that will allow in-place field testing of existing adsorber beds. In contrast to the fixed-manifold system for in-place testing, this approach employs a portable instrument. A gas chromatograph is being fitted with a pump and a gas-sampling valve (GSV); the GSV has two sampling loops that al-

low alternate measurement of the air stream above and below the carbon bed. Testing can then be conducted according to American Association for Contamination Control Standard CS-8. By this standard, a commercial freon is released into the air stream upstream from the carbon bed; the challenge concentration must be at least 10,000 times greater than the minimum sensitivity of the detection system. At least three upstream and three downstream determinations are then made at intervals of one minute, and there must be no downstream indication of the challenge gas. We expect the modified gas chromatograph to allow us to carry out this type of testing.

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INSTRUMENT DEVELOPMENT

A Real-Time Monitor for Tritium in Waste Water

C. J. Huntzinger, D. E. Hankins, J. L. Cate, Jr., M. Dreicer, and A. A. Odell

We are developing an automatic on-line tritium monitoring system for a combined industrial and sanitary waste water stream. It will allow early detection and mitigation of either chronic or acute tritium spills to the sewer. Two major tasks were identified: to provide a means of obtaining relatively pure and representative samples, and to develop a reliable low-level tritium detector. This article reviews the highlights of our progress during the past year in solving these two tasks.

History

Since it was formed 30 years ago, LLNL has maintained a constant environmental surveillance program to ensure that concentrations of potentially hazardous materials in its effluents are well below existing guidelines. One of these effluents is the combined flow of sanitary and industrial waste water. Initially, sampling consisted of taking monthly sludge samples from the Livermore Water Reclamation Plant and a 24-h composite sample as the waste water left the Laboratory boundary. About 13 years ago the waste water monitoring capabilities were improved vastly with the addition of on-line low- and high-energy-photon detectors and a pH monitor.¹ We are continuing to improve our monitoring capabilities by adding an on-line x-ray fluorescence analyzer (XRFA) for metals (described in the next article), and by developing this tritium waste water monitor.

System Specifications

The monitoring system has two subunits: sample collection and radiation detection. It must be:

- Sensitive enough to detect 1-20 pCi HTO/ml (2.2-44.1 dpm HTO/ml).
- Economical to operate on a continuous basis.
- Rugged enough to operate at a remote environmental sampling station.
- Easy to maintain on a routine basis.
- Simple for a casual user to operate.

- Capable of being connected to the existing microcomputer alarm system.
- Capable of obtaining a representative and particulate-free liquid sample.

Investigations

Sample Collection

Osborne and his colleagues at Atomic Energy of Canada, Ltd., have vast experience collecting tritiated water vapor with bubblers² and their studies prove that this approach to collecting a clean sample works. In our application, a dry gas would be bubbled through a representative waste water stream, leaving behind suspended particulates and dissolved chemicals. The vapor along with any HTO present would be collected from the gas stream. We constructed an experimental bubbler (Fig. 1). The collected sample vapor remained clean even when the starting solution had various combinations of chemicals, such as potassium permanganate, food colorings, light motor oil, and detergents. We also found we could transfer sufficient sample vapor for our counting requirements at room temperature.

Two major difficulties became apparent. It would take a large number of vapor flow meters, relative humidity indicators, level sensors, and valves to accurately control the rate of vapor transfer. There was also a question as to how we would introduce the vapor into the liquid scintillator cocktail. About this time a new plastic product called Nafion, developed by Du Pont to dry process-gas streams, came to our attention. It is a

Figure 1. Experimental HTO vapor bubbler.

semipermeable membrane, similar to solid sulfuric acid, that is permeable to water only. It appeared that we could solve these two problems by passing the liquid scintillator through the center of the Nafion tubing, with the vapor sample around the exterior of the tubing, and have the vapor diffuse directly into the scintillator. We obtained three samples of tubing, ranging in size from 0.625-mm i.d. by 0.875-mm o.d. to 2.75-mm i.d. by 3.125-mm o.d. Our studies showed that tritiated water vapor diffused into the liquid scintil-

lation cocktail, but the largest concentration ratio we measured between scintillator and starting sample water was 0.0078, which was disappointingly low—the goal being to achieve the largest decimal up to a maximum of one, which would represent the same concentration of HTO in the scintillator as in the sample water.

At this point we decided that the benefits of a simpler sampling method would more than compensate for a decrease in counting efficiency due to chemicals in the purified sample. Recently, a

new type of filter media called cross-flow filters or tangential flow filters has been developed, primarily to concentrate biological cell colonies. They differ from conventional "dead-end" filters in that the sample stream continuously washes the filter face, greatly extending its useful life.

The Acroflux capsule cross-flow filter made by Gelman seemed to be the leading candidate because of its modular design, low cost, and ease of disposal. We acquired a 1.2- μm pore size unit and installed it at the monitoring station for a trial run. It was downstream from the macerator, a mechanical device that reduces all particulates to a size of 100 μm or less. The sample waste water stream was flowing at approximately 3 gal/min. Unfortunately, the capsule operated for approximately 10 min before its flow was occluded with a fibrous matrix. This matrix resisted attempts to clean it by backwashing.

The principles of cross-flow filtration are relatively straightforward so we decided to design our own prototype unit. After numerous modifications, we settled on the configuration shown in Fig. 2. The waste water stream flows down the XRFA chute over the face of the 49-mm-diam filter disk. The disk is placed on the expanded metal filter support, sealed to the back of the chute with an O-ring, and held in place with a screw-on retainer ring.

A variety of pore sizes of filter disks have been tried, ranging in size from 10 μm down to 0.5 μm . (For reference, 1.2 μm is considered general filtration and 0.2 μm is considered sterilization.) The most important quality seems to be the macroscopic surface texture. Smooth-surface filters have lasted approximately 10 times longer than rough-surface filters of comparable pore size. A 1- μm Teflon filter disk, wetted with ethanol, has lasted the longest—approximately 10 days at a filtrate flow rate of 0.23 ml/min.

Radiation Detection

Numerous methods are available for the detection of tritiated water but few meet our sensitivity requirement of 1–20 pCi HTO/ml. We considered these methods:

- Sample enrichment followed by a less sensitive detection method.
- Conversion of sample to a counting gas for internal proportional counting.
- Mixing sample vapor with a counting gas for internal proportional counting.
- Counting sample vapor through a large-volume ion chamber.
- Passing condensed sample through a solid scintillator array.
- Continuous batch processing of condensed sample in a liquid scintillation counter.

Figure 2. Prototype cross-flow filtration unit mounted in XRFA monitor flume.

- Passing condensed sample through a liquid-scintillation-counter flow cell.

The first two methods were discarded because they involve complicated chemical processes not easily done on a continuous basis and the enrichment process would take days to complete. The next two methods involving counting the sample vapor in the gaseous phase were discarded because they did not have adequate sensitivity. For example, at 70°F and 80% relative humidity, 10 pCi HTO/ml of liquid (a middle value for our sensitivity requirement) corresponds to 0.15×10^{-15} Ci HTO/ml of vapor. This is a small fraction of one disintegration per minute per milliliter of vapor. It could not be detected because the background count rate is large and there are potential electrical instabilities and loss of sensitivity as the vapor content of the counting gas increases. The last three methods utilize scintillation counting. A solid scintillator array was preferred because of its simplicity, but it was discarded because it has low sensitivity due to the short range of tritium betas (6 μ m in water), possible chemical reactions between the waste water and fluor, and chronic loss of sensitivity due to biofouling (we have loosely defined biofouling as the unwanted growth of microscopic, opaque colonies of micro-

organisms). Liquid scintillation counting in a continuous batch mode (counting one discrete vial while another is being collected) would have provided the greatest sensitivity, but requires a prohibitively elaborate system of flowmeters, valves, level sensors, and moving parts.

Liquid scintillation counting with a continuous flow cell appeared to be an optimum compromise between sensitivity and simplicity. The condensed sample stream would be mixed with a precise amount of liquid scintillation cocktail before being pumped through a flow cell detector. The major drawback to this method was the expense of using liquid scintillation cocktail continuously. We decided to design the detector to limit cocktail consumption to 1 gal per week (0.4 ml/min), which seemed to be the best compromise between sensitivity and cocktail expense.

There are a number of simple liquid scintillation flow cell detectors on the market. Most of them were designed to monitor high-specific-activity effluents from High Performance Liquid Chromatography (HPLC) columns. We were able to acquire a Radiomatic Instruments and Chemical Co., Inc., Model HP on long-term loan (see Fig. 3). The detector element is a thin-wall Teflon tube coiled flat and sandwiched between two

Figure 3. Radiomatic Instruments and Chemical Co. Model HP radioactive flow detector (top cover open).

Figure 4. Scintillation flow cell.

matched photomultiplier (PM) tubes. The usable cell volume is 2 ml (see Fig. 4).

We characterized the performance of the unit as follows:

- The optimum ratio of scintillator to sample was between 3.5/1 and 4/1, depending on the scintillator used. At ratios of 3/1 and below, the mixture went into an unstable two-phase state and at ratios of 1.3/1 and below it became a viscous gel.

- The maximum counting efficiency for tritium was measured at 22.6%.

- The minimum detectable activity (MDA) was calculated using the method of *Tritium Measurement Techniques*.³ It was 34 pCi/ml with "known" (stable) background and 47 pCi/ml with "unknown" (unstable) background.

- The two available electronic windows were used to establish a quench correction curve by the internal channel ratio (ICR) method. As the quench of the solution increases the entire spectrum is shifted down in energy. The ratio of the count rate in the two overlapping windows gives an indication of how far the spectrum has been shifted. Using a set of standard quench solutions, it is fairly easy to develop a curve that shows the amount of quench present for any unknown channel ratio.

We believe one of the largest potential maintenance problems is biofouling within the tritium monitor tubing and scintillation tubing. To avoid this we recently installed an Ultra Dynamics

Model 500 ultraviolet flow sterilizer. While the dissolved chemicals and turbidity in the waste water have prevented us from achieving the 16,000- $\mu\text{W-s}/\text{cm}^2$ dose recommended by the U.S. Public Health Service for sterilization of drinking water supplies, we have observed a reduction in the particulate hangup within the XRFA chute and associated tubing. An additional benefit has been a reduction in the olfactory index of the 24-h composite sample.

Future Investigations

In the near future, we plan to operate the model HP flow cell detector in conjunction with the cross-flow filter disk. Initially this will be for short-term attended runs, and if successful for long-term unattended runs. When these runs are completed and we are satisfied the combined system will meet our expectations, we will begin to design and construct our own flow cell detector. It will probably include the following design features:

- A flat, spiral-wound coil of thin-wall Teflon tubing sandwiched between the two matched PM tubes, similar to the flow cell developed by Schram and Lombaert.⁴ The light-transmission properties of the cell will be improved by filling all voids with water or silicon oil, coupling the cell to the PM tubes with optical grease, and placing reflectors around the perimeter of the cell.

- The two PM tubes will be used in the coincidence counting mode.

- The flow cell and PM tubes will be placed in a layered metal shield.

- An external standard source will be used as an indicator for quench correction. The external source should provide statistically more significant results than the internal channel ratios (ICR) method. The source will be actuated by a simple mechanical system automatically for a 1-min count to determine the quench present, then removed so the sample alone can be counted.

- Standard NIM high-voltage and signal-processing modules.

- Electronic connection to a multichannel analyzer and the alarm system LSI-11. They will be used to calculate the necessary quench correction using the External Spectral Index of Sample method (ESIS), in which the spectrum is divided between numerous adjacent electronic windows rather than two large overlapping windows as in the ICR method. The ESIS spectrum is similar to what might be seen with a multichannel analyzer. The counts in each channel are used to calculate the first moment of the spectra. A set of known quench standards is used to develop a curve that relates the first moment of a sample to the necessary quench correction. This method is more sensitive than the external standard channel ratio method because the numerous small channels allow small changes of quench to be detected more readily.

- A single-pump motor with two heads to deliver the sample and liquid scintillator streams.

- Combination of the two streams in a mixing tee or packed-bed reactor. This will avoid the need for a mechanical stirring cell.

The tritium monitor may be utilized to detect other dissolved isotopes in the waste water. Occasionally, a Lab employee has a nuclear medicine scan off site; if he returns to work later that day, there is a good chance he will void some of the remaining diagnostic isotope. The high- and low-energy-photon detectors at the monitoring station easily detect trace quantities of these diagnostic isotopes. The tritium monitor might allow us to rapidly determine if a high photon count rate was caused by a nuclear medicine diagnostic isotope; we would add additional electronic windows that would be set specifically to monitor for the low-energy photons or beta particles from common diagnostic isotopes.

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ENVIRONMENTAL PROTECTION

Adding an On-Line X-Ray Fluorescence Analyzer for Metals to the LLNL Sewer Monitoring System

J. L. Cate, D. W. Rueppel, and C. J. Huntzinger

About 13 years ago, we built a monitoring system for the LLNL sanitary sewage effluent. This system monitored for radioactive materials and excursions in pH and has, with some modernization, served the lab faithfully.^{1,2} We later recognized a need to monitor certain toxic materials—namely, chromium, nickel, copper, and zinc—not so much for environmental reasons, but to protect the microbial digesters at the Livermore Water Reclamation Plant against upset caused by these elements. Over the last several years we have developed a system based on isotope-excited x-ray fluorescence analysis that can detect these four elements at or below our effluent concentration limits; it also monitors the levels of manganese, iron, cobalt, arsenic, bromine, mercury, and lead.

Components

The system consists of an exciter/detector unit and a data processing unit. The exciter (a 20-100-mCi ^{109}Cd source) and detector (a xenon-filled proportional counter) are arranged in a carefully optimized geometry over a sampling flume (Fig. 1). A continuous sewage sample passes through a macerator, which reduces solids to particles <100 μm in diameter, and then flows as a thin sheet down the flume where it is irradiated by the

source. Characteristic x rays from trace elements in the sewage are detected by the proportional counter and the signals routed to the data processing unit. This device, a DEC LSI-11/2 microcomputer, acts both as a pulse-height analyzer generating a 256-channel x-rays spectrum and as a data reduction computer. In this latter role, it contains software which converts any detected x-ray intensities to elemental concentrations, compares these to preset limits and, if required, generates an alarm signal (see Table 1).

Figure 1. Cross section of the exciter/detector geometry.

Table 1. LLNL maximum permissible metal discharge limits and XREA alarm levels.

Single metal ^a	LLNL limit (ppm)	XREA alarm level (ppm) ^b	
		500-s response time	5000-s response time
Cr	100	119	37
Cu	10	16	5
Zn	50	14	4
Ni	10	21	7

^a Combinations: total concentration, to be less than 100 ppm.

^b The alarm levels vary somewhat, depending on the age of the exciting source. These are typical values.

Status

The computer has also assumed from the original sewer monitor the functions of monitoring radiation, pH, and flow. The system has undergone extensive testing and debugging while operating in parallel with the old monitor and is now ready to be connected to the alarm system and assume the whole monitoring responsibility. (The system has been described in much greater detail in other publications.¹⁻⁵)

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RADIATION PROTECTION

Progress in Multi-Target TRAX Development

J. L. Cate and C. J. Huntzinger

The transmission anode x-ray tube (TRAX) is a device for producing nearly monoenergetic x rays by self-filtration in the target. We have developed a series of such tubes for use in our dosimetry and calibration laboratory, which have been described earlier in this report series.¹⁻⁸ Successful operation of the last of this series—a uranium target tube—culminated our effort to provide the widest possible range of x-ray energies. However, since the last several iterations in design development had been modifications of one existing system, we now had only the uranium, neodymium, silver, and copper tubes in operation. Thus, we were faced with the prospect of building additional devices to round out our inventory of necessary targets. This approach not only required physical redundancy of vacuum systems, electron guns, power supplies, etc., but meant disruption of experimental setups in order to change energies. We had long wanted to develop a tube with multiple targets: this seemed an opportune time—both experimentally and fiscally—to do so.

Design Criteria

We established several basic design criteria. First, we should have at least three target materials: tungsten, gold and uranium. Second, there should be little physical disruption of an experiment in progress associated with a change of energy. Third, the new design should incorporate as much of the existing hardware as possible.

Target Selection

Under these constraints, we considered means of selection of the desired target. Two general schemes exist: either leave the targets stationary and somehow selectively move the electron beam to strike the desired material, or have a stationary electron beam and move the desired target in front of it.

In the first category, an elegant solution is to have a single electron gun, and to steer the electron beam to the desired target by electrostatic or magnetic deflection as in a cathode ray tube. A less sophisticated approach might be to simply have three electron guns and turn on only the appropriate one for the desired target. Several drawbacks to such systems were identified. They

would utilize little of the existing hardware, one required development of a technology with which we had no experience (electron ballistics control), and, most importantly, they inherently had the targets in different physical locations when used. Thus, the beam for each energy would emanate from a different spot, presenting problems in positioning and aligning dosimeters and instruments for calibration. The ultimate users of the facility viewed this as highly undesirable.

The use of moving targets with a fixed beam avoids the last problem and we concentrated our efforts in this area.

Target Array

Again, two approaches can be identified. In one, the targets are arranged along the circumference of a circle, and the holder is rotated to select the desired material. The other configuration is a linear array of targets, requiring a linear motion for selection.

The rotary selection arrangement has some advantages and has been used by other workers in the field.⁹ However, the previous work involved lower-atomic-number targets, allowing

much lower accelerating voltages. This, in turn, allows one to have the targets at ground potential. We were committed to a "double-ended" design where the electron gun is floated at roughly half the required voltage below ground, and the target is at about the same voltage above ground. A rotary target holder design would require both being insulated to hold off 60-80 kV, and providing a means of dissipating some 50-100 W of heat, all while allowing the assembly to be rotated from outside the vacuum system. These formidable engineering problems led us to abandon this initially attractive method in favor of a linear array.

The design problems associated with the linear array again centered around providing vacuum sealing for the moving target array while maintaining a heat-dissipation path and high-voltage insulation. Our solutions to these problems utilized a balanced pair of stainless steel bellows to provide sealing and an adaptation of our old insulating and heat-rejection methods. The arrangement is shown in Fig. 1. A 3/4-in.-diam copper rod has the three foil targets brazed into it. One end of the rod penetrates into a heat exchanger via a Macor insulator and one of the bellows seals. The heat exchanger is filled with a 'fluorocarbon liquid' which has a low ($\sim 80^{\circ}\text{C}$) boiling point and excellent insulative properties, thus allowing the necessary heat rejection while maintaining the necessary voltage standoff. High-voltage connection is made by a simple sliding contact between the end of the HV cable and the copper anode support. The HV cable enters the fluid-filled chamber through a common plastic tube fitting. The other end of the rod is supported by another Macor insulator which, via the other bellows seals (at ground potential) reaches the outside of the x-ray tube. There, three precision detents and a knob permit accurate manual selection of the desired target. The electron gun assem-

bly, exit window, and main housing remain unchanged from the previous design. The tube is actively pumped by an ion pump through a port not shown in the diagram.

Several interesting engineering problems arose during construction. Perhaps the most unusual one involved the brazing of the three target foils and the bellows to the target rod. Each joint needed a special brazing alloy and technique; the orchestration of the conflicting requirements was skillfully handled by D. Berger of the Vacuum Services Laboratory and is the subject of another paper.¹⁰ Another problem concerned the alignment of the target foils with the electron beams. We developed an alignment fixture that keyed on the conical collimators machined for each target. Then we epoxied the final location point (the joint between the anode support insulator and its bellows) after assembling the anode inside the tube body and inserting the alignment rod.

Testing

Initial vacuum testing was successful and we proceeded to high-voltage testing. To sum up a frustrating process, we found that while the insulators would hold off the necessary 140 kV under static conditions, with any usable beam current the arcing rate became excessive. Several changes to the internal insulators followed with some improvement noted, but no real success. We have now added junction shields to the insulators, following criteria developed by Finke.¹¹ These shields reduce the electric field strength at the junction between insulator and charged conductor. These highly stressed junctions have been identified as potent sources of field emission electrons, and may thus lead to electrical breakdown. We are now readying the tube for further testing. Other facets of the tube's operation have been successful; it appears that when we can overcome the arcing problem, the tube will be operational.

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* 3M Co. FC-85 Fluorinert Electronic Liquid.

Figure 1. Diagram of the transmission anode x-ray tube.

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Development of a Passive Personnel Neutron Dosimeter/Spectrometer - DOSPEC

R. V. Griffith and T. A. McMahon

Each of the two most widely used personnel neutron dosimeters—NTA (neutron track type A) film and TLD (thermoluminescence dosimeter) albedo detectors—has significant practical limitations. NTA film is notorious for fading, particularly under adverse environmental conditions (high temperature and humidity). Moreover, since its practical detection limit is about 50 mrem it has a marginal sensitivity, and evaluation requires tedious optical counting with a high-magnification microscope. Albedo dosimeters do not have these limitations, but their energy response is so poor that it is almost the inverse of the neutron-fluence-to-dose-equivalent conversion factor curve (Fig. 1). As a result, neither of these, nor the radioactive fission track detectors, can be considered a generally satisfactory personnel neutron dosimeter. We have developed a prototype composite dosimeter that uses track detectors to augment an albedo detector.

Introduction

This composite dosimeter combines the advantages of the albedo detector (excellent sensitivity and ease of automation) with the fast neutron energy response of plastic etch track detectors. Three track detectors with different fast neutron response (CR-39, polycarbonate, and LR-115 cellulose nitrate) are included in the cadmium box used for the albedo dosimeter. The response of these detectors depends on the inherent properties of the plastic, and does not involve a fission foil radiator. Moreover, because the TLD is used as a screening tool, only the track foils from those dosimeters that appear to have been exposed to 50 mrem or more need to be etched and counted.

We have developed two versions of a dosimeter that embody the albedo detector-track foil concept. DOSPEC I includes a TLD component, plus CR-39, polycarbonate, and LR-115 (Fig. 2). In practice, one or more of the track foils can be and usually is omitted from the badge. If, for example, the contribution from neutrons above 10 MeV is small, the LR-115 is not used. The second version—DOSPEC II—is a combined albedo-CR-39 dosimeter that uses only track detection. The albedo response is provided by a ^6Li or ^{10}B (n, α) radiator. The shape of the etch-track albedo energy response is the same as the TLD albedo response. However, DOSPEC II is gamma-insensitive and the n, α radiator concentration and processing parameters are adjusted so that the overall albedo

Figure 1. Comparison of the response of a Hankins-type albedo dosimeter with the ICRP dose-equivalent conversion.

Figure 2. Components of DOSPEC I dosimeter/spectrometer.

plus CR-39 fast neutron response matches the fluence-to-dose-equivalent response as closely as possible.

DOSPEC I

The energy response of the track detectors depends to some degree on the processing parameters. Although, for example, CR-39 is capable of detecting recoil protons while polycarbonate is not, the specific response function shape depends on a number of factors:

- Thickness and composition of the covering or radiator.
- Chemical vs electrochemical etching (ECE).
- Pre-etch treatment.
- Temperature and concentration of etchant.

- Voltage gradient and frequency of ECE.
- Specific composition of plastics.

The conditions used for processing are presented in Table 1. (The TLDs in DOSPEC I are evaluated using conventional thermoluminescence dosimetry techniques.) The etch-track components are counted semi-automatically using a commercial bacterial colony counter coupled by a TV camera to an optical microscope (Fig. 3). The colony counter provides a simple pattern recognition and counting capability. Therefore, as long as the etch-track images meet the size and contrast requirements of the colony counter, counting is done efficiently and reproducibly. The colony counter can evaluate either dark tracks on a light background or light tracks on a dark background. The format is switch selectable, so we can count either the black electrochemical-etch tracks in CR-39 and polycarbonate, or the light holes in the

Table 1. DOSPEC component processing parameters.

	CR-39	Polycarbonate	LR-115
Radiators	0.06 mm CH ₂	0.025 mm CH ₂	0.013 mm cellulose nitrate
Pre-treatment	3-hour chemical etch at 60°C in 6N KOH	1-hour exposure to UV	None
Etch parameter	5-hour ECE of incident side at 22°C in 5N KOH; 31.5 kV/cm, 2 kHz	5-hour ECE of both sides at 22°C in 5N KOH; 31.5 kV/cm, 2 kHz	3.5-hour chemical etch at 60°C in 10% NaOH

red LR-115 film. A blue filter is used when counting LR-115 to enhance contrast. The colony counter can also be interfaced easily to a small computer for a greater degree of dosimetry system automation.

The relative energy responses of the four dosimeter components are shown in Fig. 4. These data are used as the basis of a four-group matrix that can be used to unfold a crude spectrum from the dosimeter component responses. We have assigned lower energy bounds of thermal, 0.1 MeV, 1.5 MeV, and 5.0 MeV to the energy bins used in this matrix. A spectrum-weighted average has been calculated for each detector and each bin to

create the final matrix (actually, three matrices have been created for different detector selections; see Fig. 5). We have found, for example, that the use of LR-115 has little practical value for the operational conditions at our Laboratory. Therefore, the dosimeters as issued normally include only the other components.

We have incorporated these matrices into a processing code on a programmable calculator (Hewlett-Packard 41CV, with printer). The code performs the calculations by following a simple sequence of steps:

1. Specify plot option, yes or no.
2. Specify matrix dimension.
3. Specify neutron source used to normalize response of dosimeter components (none, Cf or PuBe).

Figure 3. Commercial bacterial colony counter and microscope used for measurement of track densities in DOSPEC etch track detectors.

Figure 4. Relative neutron responses for the four DOSPEC I components. (Albedo response is in relative light units, so the absolute magnitude cannot be compared with etch-track responses in tracks per neutron.)

2 × 2				
	<u>Albedo</u>	<u>CR-39</u>		
Thermal to 0.1 MeV	X *			
0.1 to 18 MeV	X	X		
3 × 3				
	<u>Albedo</u>	<u>CR-39</u>	<u>Polycarbonate</u>	
Thermal to 0.1 MeV	X			
0.1 to 1.5 MeV	X	X		
1.5 to 18 MeV	X	X	X	
4 × 4				
	<u>Albedo</u>	<u>CR-39</u>	<u>Polycarbonate</u>	<u>LR-115</u>
Thermal to 0.1 MeV	X			
0.1 to 1.5 MeV	X	X		
1.5 to 5.0 MeV	X	X	X	X
5.0 to 18 MeV	X	X	X	X

*X represents non-zero detector response for indicated energy bin

Figure 5. Matrices for various detector selections in DOSPEC I, used for determining spectra and calculating dose equivalents.

4. Input responses of dosimeter components for calibration dosimeters, if Cf or PuBe, to normalize matrix responses.
5. Enter dosimeter identification.
6. Calculate fluence in each energy bin.
7. If fluence in any bin is negative, recalculate using modified matrix.
8. If fluence remains negative, set to zero and calculate remaining fluences.
9. Use bin fluence data to calculate bin dose equivalent values.
10. Print spectrum and dose equivalent summary.
11. Plot, if option requested.
12. Repeat steps 5 through 11.

A sample of the output is presented in Table 2.

In April of this year, we submitted both DOSPEC I and DOSPEC II to the CEC-ORNL Personnel Dosimetry intercomparison study. Dosimeters were sent to the Oak Ridge National Laboratory. Some were exposed to distributed-

energy neutrons from the Health Physics Research Reactor (HPRR), while others were sent to the PTB and GSF laboratories in West Germany for mono-energetic exposures (0.57, 1.2, 5.3, and 15.0 MeV) plus irradiation by ²⁵²Cf in a 15-cm-radius D₂O-filled sphere. The dose-equivalent evaluations for both DOSPEC I and II are presented in Table 3, along with the reference values. The DOSPEC I spectral results are presented as fractions normalized to a total fluence in Table 4. LR-115 was not evaluated for these dosimeters, so there are only three energy bins shown.

DOSPEC II

The key to the success of the DOSPEC II concept is obtaining a balanced response so that the fast neutron response of the CR-39 plus the track density contributed by the n,α radiator albedo response results in a full energy response that matches the fluence-to-dose-equivalent conver-

Table 2. Example of the output spectrum and dose equivalent.

DOSPEC	RESULTS	RELATIVE FLUENCE
DO YOU WANT PLOTS? ENTER 1=YES, 0=NO	THERM TO .1 MEV FN -	.001 .01 .1 1.
1. ***	7.20+06 ***	THERM ----- ----- -----
COMPONENTS?	REM	
4. ***	1.12-02 ***	
CALIBRATION SOURCE USED?	.1 MEV TO 1.5MEV	
NONE=0, CF=1 OR PUBE=2	FN -	
	5.84+06 ***	.1 MEV ----- ----- -----
1. ***	REM	
ENTER CF252 ALBDO RES/DE	1.49-01 ***	
754,000. ***	1.5MEV TO 5. MEV	1.5MEV ----- ----- -----
ENTER CF252 CR39 RES/DE	FN -	
1,739. ***	1.20+07 ***	
ENTER CF252 P.C. RES/DE	REM	5. MEV ----- ----- -----
63. ***	4.45-01 ***	
ENTER CF252 LR115 RES/DE	ABOVE 5. MEV	
10. ***	FN -	
	2.22+06 ***	
DOSPEC 10?	REM	
	9.36-02 ***	RELATIVE DOSE EQUIV.
T. MC MAHON	TOTAL- FN -	0.0 1.0
	2.72+07 ***	THERM ----- ----- -----
INPUT ALBDO DATA	TOTAL- REM	
3.01+05 ***	6.99-01 ***	
INPUT CR39 DATA		.1 MEV ----- ----- -----
1.36+03 ***		
INPUT P.C. DATA		1.5MEV ----- ----- -----
5.34+01 ***		
INPUT LR115 DATA		5. MEV ----- ----- -----
8.04+00 ***		

Table 3. CEC-ORNL personnel neutron dosimetry intercomparison summary.

Spectrum	Reference dose equivalent (mSv)	DOSPEC I dose equivalent (mSv)	DOSPEC II dose equivalent	
			(mSv) ^a	(mSv) ^b
Bare HPRR	0.62	0.67	1.20	0.75
	11.13	9.30	12.10	11.6
HPRR with 13 cm steel	0.64	0.51	0.97	0.52
	10.99	8.01	8.02	7.57
HPRR with 20 cm concrete	0.48	0.43	0.82	0.37
	9.43	7.38	10.70	10.2
HPRR with 12 cm lucite	0.59	1.32	1.25	0.80
	10.96	11.80	13.00	12.6
0.57 MeV	0.70	0.98	-	-
	8.37	6.68	-	-
	0.81	-	1.63	1.18
	8.37	-	9.46	9.01
1.2 MeV	6.00	6.56	-	-
	1.50	2.17	-	-
	7.01	-	9.24	8.79
	2.50	-	4.29	3.84
5.3 MeV	6.50	7.33	-	-
	3.99	5.10	-	-
	9.49	-	13.4	13.0
	2.99	-	5.55	5.10
15.0 MeV	13.13	18.2	14.40	14.0
	1.05	1.31	-	-
	0.89	-	0.93	0.48
²⁵² Cf in 15 cm D ₂ O	10.97	11.6	11.9	11.4
	1.20	1.91	-	-
	3.61	-	4.02	3.57

^a Values reported for the CEC-ORNL intercomparison.

^b Values adjusted for error in background.

sion factor curve as closely as possible. A mathematical synthesis of the response is illustrated in Fig. 6. Two of the key variables are concentration of ⁶Li and/or ¹⁰B in the n,α radiator, and the degree of etching performed. The n,α track response is quite sensitive to the etch parameters, primarily because of the short range of the alphas, and other reaction products.

For uniformity in dosimeter processing, we began by testing the etch parameters used for CR-39 in DOSPEC I (with the protective polyethylene uncovered to permit registration of the alphas). We tested two commercially available materials as radiators—CN85 (a plastic sheet covered with natural lithium tetraborate) and BN1 (an ester-based plastic coating with natural boron nitride). The

Table 4. Results of the spectral evaluation of DOSPEC I in the CEC-ORNL intercomparison.

Spectrum	Energy bin fraction (n/cm ²)		
	Thermal to 0.1 MeV	0.1 to 1.5 MeV	Above 1.5 MeV
Bare HPRR	0.295	0.459	0.246
HPRR with 13 cm steel	0.478	0.388	0.132
HPRR with 20 cm concrete	0.718	0.181	0.101
HPRR with 12 cm lucite	0.686	0.193	0.126
0.57 MeV	0.000	1.000	0.000
1.2 MeV	0.000	0.952	0.048
5.3 MeV	0.00	0.319	0.682
15.0 MeV	0.102	0.000	0.898
²⁵² Cf in 15 cm D ₂ O	0.807	0.117	0.076

Figure 6. Calculated optimum response function for DOSPEC II.

BN1 has turned out to be superior, which is fortunate because CN85 is no longer commercially available.

Four pieces of CR-39 and four BN1 radiators are used in the version of the dosimeter tested at Oak Ridge (Fig. 7). The components are arranged as two pairs of radiator-detector combinations. In each pair, the radiators are sandwiched between two pieces of CR-39. The sensitive surfaces of the CR-39 face inward in each pair. An additional benefit accrues from this configuration—orientation dependence. The neutron direction can be determined because the upstream track detector in each pair has a higher track density than those on

the downstream side. This can be very useful for establishing body-orientation corrections.

The DOSPEC II results from the CEC-ORNL intercomparison are presented in Table 3. A problem developed in interpretation of the low dose (<100 mrem) dosimeters. Because of a limited supply of BN1 radiators, we used CN85 for the background controls. However, the backgrounds turned out to be significantly elevated—to the point that they indicated doses on the order of 100 mrem. Whether this was due to alpha contamination of the lithium tetraborate or other causes, these backgrounds were unusable. On the other hand, doses evaluated using DOSPEC II CR-39 background values were uniformly about 45 mrem higher than the reference dose-equivalent values (Table 3, col. 4). If we assume this value should be added to the background, the DOSPEC II results are uniformly quite close to the reference values and, in general, are even better than the DOSPEC I results.

Summary

The tests of the prototype are encouraging and demonstrate the validity of the basic premises of the DOSPEC concepts. Additional work is needed to refine the evaluation process and test the dosimeters more extensively. More effort needs to be directed at proper correction for orientation effects. Finally, the CR-39 used does not represent dosimeter-grade material; such plastic is now becoming available and, after proper test and evaluation, should contribute to improved dosimeter performance.

Figure 7. DOSPEC II components.

FIRE SAFETY

Effectiveness of Standard and Alcohol-Resistant Aqueous-Film-Forming Foam (AFFF) on Alcohol Fires

N. J. Alvares, H. W. Ford, and S. J. Priante

In late 1980, LLNL obtained reserves of gasohol as a means of fuel conservation for laboratory vehicles. To ensure that the Lab Fire Department (FD) had proper resources to combat potential gasohol spill fires, the Fire Science Group was authorized to test extinguishants in the FD inventory with regard to their effectiveness in extinguishing gasohol pool fires and their stability and endurance when layered over nonburning gasohol. The extinguishants tested were high-expansion foam, standard 6% AFFF, and alcohol-resistant 6% AFFF; both experimental and commercially available aeration nozzles were used to apply them. The tests indicated that no advantage is gained by using alcohol-resistant AFFF for either extinguishing or inerting.

Introduction

Fires on spills of flammable fluid are dynamically controlled by physical properties of the fluid, heat transfer, and atmospheric pressure. During fires, these processes are coupled such that the burning rate (rate of fuel consumption) is dominated by heat feedback from the flame. Essentially, burning rate is equal to rate of fuel evaporation (R_e). When burning is steady, energy feedback from fire zone ($\dot{Q}_f$) minus energy sinks and losses to surroundings ($\dot{Q}_L$) divided by the latent heat (L) equals energy required for fuel evaporation or

$$R_e = \frac{\dot{Q}_f - \dot{Q}_L}{L}$$

To modify R_e , extinguishing agents must tamper with $\dot{Q}_f$ or $\dot{Q}_L$; i.e., agents can block $\dot{Q}_f$, increase $\dot{Q}_L$, and thus frustrate fuel evaporation. The action of foams and films during extinguishment operations interferes with fuel vapor release by three correlating mechanisms:

- Blocking radiative and convective heat transfer from flame to fuel surface (reducing $\dot{Q}_f$).
- Cooling the fuel and adjacent exposed surfaces (increasing $\dot{Q}_L$).
- Providing a barrier atop the fuel to prevent vapor release (R_e).

For an agent to be effective it must be delivered faster than the fire can consume it by evaporation and losses; e.g., AFFF is applied most effectively if it is ejected as a low-expansion foam so that it will float on rather than penetrate the fuel surface. Flame and hot surfaces destroy bubble membranes, releasing water which sinks through fuel or evaporates in fire. Therefore, both foam properties and application procedures are critical to the effectiveness of extinguishing operations. Successful extinguishment requires that agent application rate (A_r) be greater than the theoretical amount of agent capable of covering the surface area of the fuel spill (A_L) because of agent destruction (D) by fire and heat:

$$A_r - A_L = \int_0^{t_p} D dt$$

where t_p is endurance time of agent layer. The function t_p is sensitive to thermal and environmental conditions and, with foams, interfacial effects. For example, application of foam to petroleum hydrocarbon is effective because water within foam membranes is immiscible in the petroleum fluid. However, in gasohol (10% ethyl alcohol in gasoline) the affinity of the alcohol for water can result in the destruction of foam membranes and thus reduce or negate the effectiveness of the agent.

Tests

Several tests were conducted to assess the stability and endurance of alcohol-resistant AFFF on gasohol pool fires contained in pans 3 ft and 6 ft in diam (7.07-ft² and 28.3-ft² areas). As a control, standard (non-alcohol-resistant AFFF) was applied in parallel tests.

Initial tests involved application of foam to nonburning pools of gasohol. High-expansion foam provided no sealing protection on these pools; therefore we conducted no further tests of this extinguishant. For both alcohol-resistant and standard AFFF, the endurance of the foam layer was greater than 15 minutes. No differences in appearance were noted during the application or testing periods.

Figure 1 shows typical results of the extinguishment tests (in No. 9—small experimental nozzle applying aerated alcohol-resistant AFFF to a fully developed 6-ft-diam pool of burning gasohol). Table 1 summarizes the times until control and extinguishment of these fires. (Control time is

defined as the time when approximately 90% of the fire is extinguished.) To ensure that the fire attained steady-state burning we allowed it to burn for 1 min before initiating extinguishing procedures. During this period, the pan edges became very hot and destroyed the foam, resulting in substantial periods between control time and complete extinguishment. The last photo in Fig. 1 shows flamelets that persisted during this period.

Conclusions

LLNL no longer stocks gasohol as fuel for vehicles. Therefore, the practical utility of this report is moot. However, it is interesting that both standard and alcohol-resistant AFFF appeared to be nearly equally effective in extinguishing gasohol fires. If there is a significant price differential between the two formulations, our recommendation based on these test results would be to use the cheaper of the two.

Table 1. Control and extinguishment time for standard and alcohol-resistant 6% AFFF applied to gasohol fires.

Run No.	Extinguishment time (s)	Nozzle size	Pan size	Type of AFFF	Control time (s)
1	17	Small*	Small	Standard	—
2	29	Small	Small	Standard	—
3	23	Small	Small	Standard	—
4	15	Small	Small	Alcohol-resistant	—
5	15	Small	Small	Alcohol-resistant	—
6	17	Small	Small	Alcohol-resistant	—
7	163	Small	Large	Alcohol-resistant	—
8	49	Small	Large	Alcohol-resistant	—
9	61	Small	Large	Alcohol-resistant	25
10	130	Large	Large	Standard	32
11	90	Large	Large	Standard	35
12	120	Large	Large	Standard	49
13	73	Large	Large	Alcohol-resistant	45
14	90	Small	Large	Standard	35
15	100	Small	Large	Standard	35
16	60	Large	Large	Standard	15
17	86	Large	Large	Standard	25

* Expansion ratios:

Small nozzle, standard AFFF 60:1

Small nozzle, alcohol-resistant AFFF 75:1

Large nozzle, standard AFFF 10:1

Large nozzle, alcohol-resistant AFFF 10:1

Figure 1. Alcohol-resistant AFFF applied to burning gasohol.

Technical Notes

INDUSTRIAL HYGIENE

Determination of Explosives Using High-Performance Liquid Chromatography

R. D. Szidon

We have used high-performance liquid chromatography to measure trace levels of the explosives HMX, RDX, PETN, and TATB distributed as dust in working environments. As little as 1-3 μ g of individual explosives on a filter paper swipe can be detected. Each explosive can be measured without interference from the others by appropriate selection of chromatographic conditions.

Introduction

Industrial hygienists, toxicologists, and environmental scientists are increasingly interested in the analysis of explosives, because of mounting evidence that many explosives are mutagenic and/or carcinogenic in laboratory animals and that many aromatic nitro derivatives possess biological properties detrimental to animals and humans.¹

High-performance liquid chromatography (HPLC), coupled with a sensitive and selective detector, can be used for the analysis of trace amounts of both volatile and nonvolatile materials and is nondestructive in all cases where explosives have been studied by this approach. HPLC is conducted near ambient temperature. This is preferable to medium- or high-temperature gas chromatography, since most explosives will not survive the thermal conditions required for their determination as gases.

We have used HPLC with ultraviolet detection to determine microgram amounts of 1,3,5,7-tetranitro-1,3,5,7-tetrazacyclooctane (HMX), 1,3,5-trinitro-1,3,5-triazacyclohexane (RDX), pentaerythritol tetranitrate (PETN), and 2,4,6-trinitro-1,3,5-benzenetriamine (TATB) collected on filter paper swipes.

Results

The equipment used in this study consisted of a Hewlett-Packard 1084B liquid chromatograph

with a variable-wavelength ultraviolet detector, a variable-volume injector, and an automatic sampling system. Filter paper swipes were immersed in a suitable solvent and agitated in an ultrasonic cleaner to dissolve the explosives. Acetonitrile was used to dissolve HMX, RDX, and PETN, while dimethylformamide was used to dissolve TATB.

HMX and RDX were determined simultaneously by elution from a bonded-phase RP-18 LiChrosorb column (25 cm $\times$ 4.6 mm) with a 60% water-40% methyl alcohol mobile phase.² The ultraviolet detector was set at 230 nm for highest sensitivity. A typical chromatogram is shown in Fig. 1. Calibration curves obtained from standard solutions of HMX and RDX are given in Fig. 2. Detection limits are on the order of 1 μ g for HMX and 2 μ g for RDX.

PETN cannot be determined under these conditions because the eluting solvent, methyl alcohol, absorbs ultraviolet radiation at 200 nm, the optimum wavelength for the determination of PETN. No attempt was made to determine TATB with this system, because TATB is not soluble in the eluting solution.

PETN was determined by elution from the RP-18 LiChrosorb column by using 40% water-60% acetonitrile as the solvent as shown in Fig. 3. The ultraviolet detector was set at 200 nm. This system also provides separation of PETN from HMX and RDX. The relationship between ultraviolet response and quantity of PETN is shown in Fig. 4; the detection limit is approximately 2 μ g.

TATB can be eluted from a Chromosorb LC-9 NH₂ column with a mixture of 1.5%

Figure 1. HPLC separation of HMX and RDX (column, RP-18 LiChrosorb; mobile phase, 60% water-40% methyl alcohol; UV detector, 230 nm).

Figure 2. Calibration curves for HMX and RDX by HPLC.

dimethylformamide, 43% toluene, and 55.5% heptane.¹ Ultraviolet absorption at 355 nm is most sensitive for detection. A typical chromatogram of TATB in this system is shown in Fig. 5. A calibration curve is shown in Fig. 6; the detection limit for TATB is 3 µg.

Summary

HPLC with ultraviolet detection is useful for the rapid determination of trace quantities of the explosives HMX, RDX, PETN, and TATB. By proper selection of columns, eluting solvents, and detection wavelengths, each explosive can be measured without interference from the others.

Such analyses are useful in the monitoring of working environments for these potentially harmful substances.

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Figure 3. HPLC chromatogram of PETN (column, RP-18 LiChrosorb; mobile phase, 40% water-60% acetonitrile; UV detector, 200 nm).

Figure 4. Calibration curve for PETN by HPLC.

Figure 5. HPLC chromatogram of TATB (column, Chromosorb LC-9 NH₂; mobile phase, 1.5% dimethylformamide-43% toluene-55.5% heptane; UV detector, 355 nm).

Figure 6. Calibration curve for TATB by HPLC.

Self-Contained-Breathing-Apparatus Protection at High Work Rates

J. W. Stengel

More than 60 years ago when the U. S. Bureau of Mines was created, a schedule for testing and approving breathing apparatus was developed for the mining industry. Their need was for long-duration (2- to 4-hour) apparatus for mine rescue. Out of that background evolved the national approval and certification testing of apparatus used throughout all industry; this is the testing done by the National Institute of Occupational Safety and Health (NIOSH). The technical specifications in current testing are still biased toward the needs of the mining industry—for example, the moderate ventilation rate used is appropriate for long durations. We conducted tests that showed that the appropriateness of using a moderate work rate in testing equipment that is intended for short-duration work of high intensity (such as firefighting) is highly questionable.

Standards

The maximum work rate of the machine testing and the average human subject testing conducted by NIOSH corresponds to a minute volume (ventilation rate) of 40 standard liters per minute (SLPM).¹ By contrast, a NIOSH-funded study² determined that a mean minute volume of 58 SLPM was achieved by actual firefighters at work (see Fig. 1). Failure to attain the necessary protection from a breathing apparatus in a fire atmosphere (immediately dangerous to life and health) can easily mean loss of control of the fire and resulting property loss, injury, or fatality. These are serious consequences which are beyond standard NIOSH laboratory testing. Yet these problems are faced daily, even hourly, in municipal fire departments throughout the United States.

A very basic problem for firefighters working at high ventilation rates is loss of protection. At some point (work level) and for one of many possible reasons (e.g., improper fit or adjustment, growth of beard, sweaty facial skin, loss of proper shape of facepiece, etc.), a leakage of the facepiece will occur. The point at which this occurs is considered to be the limit of protection provided by that SCBA (self-contained breathing apparatus). The most protective of all SCBA designs is that which maintains a positive facepiece pressure throughout the entire breathing cycle. This design is called "positive-pressure," because when a leakage occurs in the facepiece, it will occur outward. "Pressure-demand" apparatus will allow toxic atmosphere to flow into the facepiece during leakage. For that reason, positive-pressure SCBA is mandatory for the fire service. This position is

Figure 1. Minute volume of firefighters at work.

supported by the Occupational Safety and Health Administration, the National Fire Protection Association, and the American National Standards Institute.³⁻⁵

Experiments

The ability of a firefighter's SCBA to maintain positive facepiece pressure is directly related to the work rate. A review of the data from the NIOSH study² indicated that approximately 98% of firefighters would be capable of sustaining a high work rate corresponding to a minute ventilation of 100 SLPM. Other studies have indicated that some firefighters are capable of much higher work rates when tested under laboratory conditions.⁶ Figure 1 illustrates that fewer than 10% of firefighters are adequately served by the 40-SLPM ventilation rate used by NIOSH. The median work rate would occur at approximately 60 SLPM, and a 100-SLPM ventilation rate would protect greater than 95%.

Therefore, preliminary tests were undertaken to determine whether the NIOSH-approved SCBA found in use in the fire service today are capable of maintaining positive facepiece pressure

at 100 SLPM. NIOSH-approved SCBA were measured using the experimental set-up depicted in Fig. 2. Results of testing are depicted in Fig. 3. Data from machine testing show that seven of 12 approved breathing apparatus are capable of maintaining positive pressure at a high work rate. Additionally, two apparatus tested from other countries were also able to pass this test. Maximum breathing resistances were also noted. Maximum resistance taken with the more important absolute value of the magnitude of the pressure change is a good indicator of perceived discomfort due to breathing resistance. The ideal product would have a high positive pressure to ensure protection, and a low peak resistance for comfort.

Conclusions

Added protection for the wearer can be gained by increasing the design work rates of breathing apparatus to maintain a minimum of 0.5 in. H₂O positive facepiece pressure when subjected to a work rate corresponding to 100 SLPM. One-third of all apparatus tested can already meet this criterion. Another one-third may be able to meet it by minor adjustments or modification.

Figure 2. Diagram of SCBA testing apparatus.

RADIATION PROTECTION

Neutron and Gamma-Ray Measurements at the LANL SHEBA Critical Assembly

D. E. Hankins, R. V. Griffith, J. H. Thorngate, and D. W. Rueppel

We recently performed some measurements on the SHEBA critical assembly at the Los Alamos National Laboratory to determine the leakage spectra and dose rates of neutrons and gamma rays at various distances from the assembly and through shields of steel and concrete.

SHEBA

The SHEBA critical assembly is a clean-geometry critical assembly employing fuel of 4.8% enriched uranium as the fluoride. It was constructed during 1980 to evaluate accidental-criticality-alarm detectors for enrichment plants, to provide radiation spectra and intensity measurements to benchmark calculations on a low-enrichment solution system, and to provide radiation fields to calibrate personnel dosimetry. The reactor vessel is a simple stainless steel cylindrical tank with a wall 6.35 mm (1/4 in.) thick. A single safety rod along the axis provides shutdown without sacrificing cylindrical symmetry. Control is effected by varying the solution level through calibrated metering valves. To facilitate measurements out to 300 m, SHEBA is housed in a thin metal shed outside of Kiva I at the Los Alamos Critical Assemblies Laboratory.

Measurements

Measurements were made at distances from 4.5 to 69 m from the assembly. A 5-1/4-in.-thick steel shield was positioned at 4 or 12 m from the assembly and concrete shields 8 and 16 in. thick were placed at 12 and 4 m, respectively.

Neutron doses were determined using the 9-in.-sphere PNR-4 remmeter and from the readings of TLDs (^6Li and ^7Li) placed inside the 9-in. sphere. Thermal neutron doses were measured using the BF_3 tube of the PNR-4 instrument and with pairs of ^6Li and ^7Li TLDs. We used albedo neutron dosimeters of the "Hankins" type and ap-

plied the 9- to 3-in.-sphere ratio technique to evaluate the albedo readings. Gamma-ray dose measurements were made using TLDs located in air and on the phantoms. Two types of phantoms were used; the first was the head and torso section from a REMAB phantom (containing bones) and the second was the LLNL water-filled torso phantom. To provide input data to evaluate a solution criticality accident, saline solutions of Na_2CO_3 (sodium carbonate) were placed in both phantoms and the activity of the ^{24}Na was determined. Sulfur pellets 1-1/2 in. in diameter were exposed on the phantoms and the ^{32}P activity was used to determine the fast neutron (>2.9 MeV) component of the neutron fluence. The ratio of the gamma dose in air to the gamma dose on the phantom and the ratio of γ to n was calculated.

Neutron spectrum measurements were made with the NE-213 scintillator, and with multispheres over the full range from thermal to 20 MeV. A 0.5-in. $\times$ 0.5-in. ^6LiI scintillator was used in the multispheres. We obtained spectrum-weighted dose-equivalent values from the NE-213 and multisphere measurements for comparison with other techniques. Above 0.4 MeV, neutron energy spectra were measured with a 2-in. $\times$ 2-in. NE-213 scintillator used with a pulse-shape discriminator to suppress gamma-ray response. Measurements were made at two detector gains, covering the energy ranges 0.3 to 2 MeV, and 1 to 12 MeV. A multisphere system composed of a 0.5-in. $\times$ 0.5-in. ^6LiI detector with a cadmium shield and 3-, 5-, 8-, 10-, and 12-in.-diameter spheres of polyethylene was used to measure the entire neutron energy spectra, but with poorer resolution than the NE-213 detector.

Figure 1. SHEBA neutron spectra obtained with the NE-213 detector at 5 m.

Neutron Dose

In order to take full advantage of the NE-213 resolution and the multisphere energy range, we began reducing the neutron data by unfolding the NE-213 response, using a program that differentiates the proton recoil response and corrects for detector characteristics such as nonlinear light output and multiple scattering. Figure 1 shows the neutron spectra obtained with the NE-213 detector at 5 m from SHEBA with and without the shields. The NE-213 results were also an input parameter for the evaluation of the multisphere responses used to calculate the spectra below 0.3 MeV. Figure 2 presents a comparison of the full-range composite neutron spectra thus obtained at 5 m.

The results obtained in this study are given in Tables 1 through 4. They have been normalized to an ion chamber reading (used to determine the reactor power) of 0.1×10^{-9} A (~ 0.154 W) for most of the dose measurements and to 0.5×10^{-7} A for the activations of Na and sulfur. All calibrations were to a bare ^{252}Cf neutron source or a ^{137}Cs gamma-ray source in the LLNL calibration facility.

Figure 3 shows the neutron dose rate as determined by the 9-in. sphere. Two points are shown at many of the distances—one obtained with TLDs placed inside the sphere, and the sec-

ond with the BF_3 tube. The decrease in the neutron dose rate for the unshielded (bare) assembly follows the inverse square relationship very closely. The concrete shield is a very effective shield, reducing the neutron dose rate at 5 m by almost an order of magnitude. The slope of the curve in Fig. 3 for the concrete shield is less than the inverse square curve. This is caused by neutron leakage around the shield as the distance from the shield is increased.

With the steel shield the decrease in dose rate is more rapid than inverse square. The rapid drop behind the steel shield must be caused by the energy dependence of the 9-in. sphere, which overresponds to low-energy neutrons (by a factor of ~ 7 at the 25-keV iron cross section window). At larger distances from the shield, the number of these 25-keV energy neutrons is decreased by scatter and air absorption, causing the bend in curve shown in Fig. 3. At the larger distances the leakage around the shield causes the curve to have a slope that is less than inverse square. However, even at 69 m the effect of the shield in reducing the neutron dose rate is still apparent. The rapid drop in neutron dose rate at small distances behind the steel shield is not seen in the gamma dose rate, which is discussed later.

There is reasonably good agreement between the rem-meter, NE-213 scintillator, and multisphere results (see Tables 1 and 3). The NE-213

Figure 2. Comparison of SHEBA neutron spectra at 5 m.

scintillator results are slightly lower for the steel- and concrete-shielded data and the multisphere results are low at 14 m. The multisphere results are also lower than the remmeter results at 69 m. The NE-213 scintillator has a lower energy threshold of 400 keV and its readings were expected to be low when compared to an instrument sensitive to all neutron energies.

The ratio of the 9- to 3-in. spheres given in Table 1 decreases as the distance from the bare assembly increases, indicating that a gradual softening of the spectrum is occurring. (High 9- to 3-in. ratios are obtained from fast spectra and low ratios from low-energy spectra.) The results behind the steel shield indicate that the spectrum is softening very rapidly as the distance from the shield is increased. At the larger distances the 9- to 3-in. ratios appear to be merging at a ratio of around 0.3 but data at greater distances were not obtained. It is reasonable to assume the ratios would be the same at greater distances.

The thermal neutron dose rates measured with the bare probe from the PNR-4 instrument and with TLDs are plotted as a function of distance from the assembly in Fig. 4. There is an unexplained difference in the instrument and TLD readings; the TLDs are higher for the bare and steel-shielded assembly and lower behind the concrete shield. The thermal neutron dose rates from the bare assembly and behind the concrete

Figure 3. Neutron dose rates determined with the 9-in.-sphere remmeter as a function of distance from the assembly. Measurements were made with (1) the BF_3 probe and (2) the ^6Li and ^7Li TLDs in the sphere.

Table 1. Neutron dose rates obtained with the 9-in.-sphere remmeter and the thermal neutron results obtained at the SHEBA critical assembly.

Shield and shield distance	Distance from assembly (in)	Dose rate, 9-in. sphere (mrem/h)		Ratio of 9- to 3-in. spheres		Dose rate, thermal neutron (mrem/h)				% thermal neutrons			
						In air		On phantom		In air		On phantom	
						Instrument	TLD	REMAB phantom TLD	LLNL phantom TLD	Instrument	TLD	REMAB phantom TLD	LLNL phantom TLD
		Instrument	TLDs	Instrument	TLDs								
Steel at 4 m	4.5	34.5		0.68		0.067				0.19			
	5.0	24.2	24.8	0.56	0.61	0.069	0.088	0.19	0.29	0.30	0.36	0.78	1.04
	7.0	8.80	9.44	0.45	0.43	0.070	0.090		0.19	0.81	0.95		1.96
	9.0	5.35	4.72	0.42	0.32	0.056	0.067		0.14	1.06	1.42		2.99
	14	3.09		0.32		0.049				1.60			
	35	0.58		0.31		0.012				2.03			
Bare	5	57.2	66.4	0.59	0.61	0.35	0.51	1.06	1.24	0.61	0.77	1.59	1.86
	7	30.9	38.3	0.59	0.66	0.20	0.22		0.68	0.66	0.58		1.79
	9	17.1		0.56		0.11				0.66			
	14	7.64	8.59	0.57	0.59	0.037	0.068		0.20	0.75	0.80		2.32
	35	1.43	1.66	0.46		0.014				1.01			
	69	0.37	0.38	0.38		0.005				1.22			
Steel at 12 m	14	3.56	2.82	0.39		0.045				1.27			
	35	0.80		0.28		0.016				1.98			
	69	0.29		0.34		0.0047				1.61			
8 in. of concrete at 12 m	14	2.69	2.42	0.33		0.056				2.09			
16 in. of concrete at 4 m	5	8.73	8.55	0.34	0.40	0.21	0.14	0.28	0.32	2.40	2.21	3.26	3.79
	7	6.53	5.71	0.30	0.30	0.14	0.11		0.23	2.09	1.95		4.08
	9	2.92 low	5.22	0.38	0.35	0.042 low	0.082		0.17	1.48	1.57		3.22

Table 2. Albedo neutron dosimeter and gamma-ray measurements made at the SHEBA critical assembly.

Shield and shield distance	Distance from assembly (m)	Albedo neutron dosimeters						Gamma ray							
		Calibration factor using TLDs in 9-in.-sphere doses		Dose rate using inst 9/3 ratio (mrem/h)		Dose rate using TLD 9/3 ratio (mrem/h)		TLDs in air (mR/h)	TLDs on phantom		Personnel TLD badges on phantom		Ratio of TLD on phantom to in air		Ratio of gamma (in air) to neutron (9-in.-sphere TLD)
		REMAB phantom	LLNL phantom	REMAB phantom	LLNL phantom	REMAB phantom	LLNL phantom		REMAB (mR/h)	LLNL (mR/h)	REMAB (mR/h)	LLNL (mR/h)	REMAB	LLNL	
Steel at 4 m	4.5														
	5.0	0.55	0.61	17.8	19.8	19.5	21.7	0.42	0.58	0.61	0.60	0.64	1.38	1.44	0.017
	7.0		0.96		9.26		9.49	0.35		0.46		0.43		1.29	0.037
	9.0		1.37		5.29		3.85	0.21		0.30		0.32		1.43	0.044
	14														
35															
Bare	5	0.87	0.86	78.4	77.7	81.7	81.0	5.42	6.32	6.05	6.54	5.71	1.17	1.12	0.082
	7		0.79		32.8		47.6	2.47		3.31		3.20		1.34	0.064
	9														
	14		0.96		10.9		11.2	0.61		0.81		0.77		1.34	0.071
	35														
69															
Steel at 12 m	14														
	35														
	69														
8 in. of concrete at 12 m	14														
16 in. of concrete at 4 m	5	0.96	0.99	6.15	5.54	7.40	7.69	0.69	0.81	0.86	0.84	0.85	1.18	1.25	0.081
	7		1.57		7.66		5.85	0.42		0.67		0.62		1.57	0.074
	9		1.29		6.75		5.22	0.29		0.42		0.38		1.44	0.056

Table 3. Results obtained with the multisphere neutron instrument and the NE-213 detector at the SHEBA critical assembly.

Shield and shield distance	Distance from assembly (m)	Multisphere					Total kerma (rads/run) ^a	NE-213 dose rate (mrem/h)
		Total flux (n/cm ² ·s ⁻¹)	Dose equivalent rate (mrem/h)	Kerma rate (ergs/g·h ⁻¹)	Element 57 dose rate (rads/h)	Average energy (MeV)		
Steel at 4 m	4.5							
	5.0	401	23.5	0.194	0.00251	0.532	0.317	20.9
	7.0						(0.121) ^b	
	9.0						(0.0603) ^b	
	14							
	35							
Bare	5	1107	68.9	0.557	0.00753	0.694	0.921	65.2
	7						(0.531) ^b	
	9							
	14	148	7.04	0.0570	0.000808	0.489	0.0942	9.2
	35							
	69							
Steel at 12 m	14							
	35							
	69	7.18	0.212	0.00167	0.0000278	0.255	0.00276	
8 in. of concrete at 12 m	14							
16 in. of concrete at 4 m	5	210	7.34	0.0570	0.000923	0.350	0.186	5.9
	7						(0.124) ^b	
	9						(0.114) ^b	

^a Bare assembly and steel shield exposures were 20 min; concrete shield exposure was 40 min.

^b Calculated on the basis of 9-in.-sphere (TLD) results at these distances.

Table 4. Sulfur and sodium activation results obtained from the SHEBA critical assembly (normalized to 0.5×10^{-7} A).

Shield and shield distance	Distance from assembly (m)	Sulfur fluence (n/cm ²)		Sodium activity (pCi/mg)		Sodium activation/ rad (kerma) (pCi/mg)		Ratio of sulfur fluence to sodium activation	
		REMAB phantom	LLNL phantom	REMAB phantom	LLNL phantom	REMAB phantom	LLNL phantom	REMAB	LLNL
Steel at 4 m	4.5								
	5.0	7.0×10^6	6.1×10^6	3.45	3.50	10.9	11.0	2.03×10^{11}	1.74×10^{12}
	7.0		3.0×10^6		1.59		13.1		1.88×10^{11}
	9.0		2.3×10^6		1.15		19.1		2.00×10^{12}
	14								
	35								
Bare	5	7.1×10^7	5.8×10^7	9.85	9.88	10.7	10.7	7.21×10^{12}	5.87×10^{12}
	7		3.3×10^7		5.81		10.9		5.68×10^{12}
	9								
	14		1.1×10^7		1.15		12.2		9.56×10^{12}
	35								
	69								
Steel at 12 m	14								
	35								
	69								
8 in. of concrete at 12 m									
16 in. of concrete at 4 m	5	7.6×10^6	7.6×10^6	3.53	3.30	19.0	17.7	2.15×10^{12}	2.30×10^{12}
	7		5.2×10^6		2.92		23.5		1.79×10^{12}
	9		2.7×10^6		2.28		20.0		1.18×10^{12}

Figure 4. Thermal neutron dose rates as a function of distance from the assembly. Measurements were made with (1) the bare BF_3 probe from the PNR-4 remmeter and (2) ^6Li and ^7Li TLDs.

are almost equal. The curve for the steel-shielded assembly peaks at a distance of about 6 m from the assembly. At the larger distances all the curves appear to merge.

When the TLDs are placed on a phantom the thermal neutron dose is increased by fast neutrons moderated to thermal in the phantom and by incident thermal neutrons reflected by the phantom. The readings of TLDs exposed on a phantom increased by a factor of ~ 3 for the bare assembly and a factor of ~ 2 for the concrete- and steel-shielded assembly. There is a difference in the thermal response from the different phantoms; the Remab phantom has slightly lower readings than the LLNL phantom. The percentage of the total neutron dose (determined by the 9-in. sphere) that is contributed by thermal neutrons is given in Table 1.

Figure 5 shows the results obtained from the albedo neutron dosimeters plotted as a function of the 9- to 3-in. ratio. The ratios used in this figure were those obtained using TLDs in the spheres, which differed only slightly from the instrument results. The calibration factors were obtained by dividing the albedo dosimeter readings (^6Li minus ^7Li) by the neutron dose determined using the TLDs placed inside the 9-in. sphere. The line drawn on the figure is the standard curve used to evaluate albedo neutron dosimeters based on the 9- to 3-in.-sphere technique. The points shown on Fig. 5 fall close to the curve, indicating that the 9- to 3-in.-sphere technique can be used accurately with this assembly shielded or unshielded.

The dose rates obtained from the albedo neutron dosimeters are given in Table 2. Two evaluations were made with each albedo reading, one with the 9- to 3-in. ratio determined using TLDs inside the 9- and 3-in. spheres and the second with the 9- to 3-in. ratio determined using the BF_3 probe in the spheres. We also used both the

Figure 5. Albedo neutron dosimeter calibration factors as a function of the ratio of the 9- to the 3-in. sphere.

Remab and LLNL-type phantoms at the 5-m distance, giving us four dose determinations at these points. The curve shown in Fig. 5 was used to determine the doses. A bare ^{252}Cf source was used for calibration of the albedo dosimeters. When the results are compared with the neutron doses shown in Fig. 3, we find agreement within about 25%. In Fig. 5, points that lie above the standard curve cause the neutron dose evaluation to be high, and points below the curve cause the doses to be low. This is shown in Table 2, where the bare assembly results are high and the steel- and concrete-shielded results are slightly low. No significant differences in the results obtained with the Remab or LLNL phantoms were found, indicating that either phantom can be used in future studies.

Gamma-Ray Dose

The gamma-ray dose rates measured by TLDs exposed in air as a function of distance from the assembly are shown in Fig. 6. The dose rates measured by TLDs on the phantom are higher than the dose rates in air (averaging about 30 to 35%; see Table 4). This increase is caused by neutron capture in the phantom, which produces gamma rays. Comparing Fig. 4 with Fig. 3, we find the relative positions of the curves for the bare and concrete-shielded assembly to be the same, but the curve for the steel-shielded assembly is lower on Fig. 6 and has a different shape. The steel is obviously providing appreciable gamma-ray shielding.

Figure 6. Gamma-ray dose rates measured in air as a function of distance from the assembly.

Sulfur and Sodium Activation

The fast neutron fluences (>2.9 MeV) determined by using the sulfur pellets are given in Table 4. There is no significant difference in the results obtained with the sulfur pellets located on either of the two types of phantoms.

The sodium activation of the saline solution in the phantoms is shown in Fig. 7. The curves for

Figure 7. Activation of the Na solution in two types of phantoms as a function of distance from the assembly.

Figure 8. Curves used to determine the dose received in a criticality accident, based on the activation of the blood and hair (sulfur fluence). The new points obtained in this study are indicated.

the bare and steel-shielded assembly have about the same shapes as shown in Fig. 3 for the total neutron dose, but the slope for the steel-shielded curve in Fig. 7 is different and follows closely an inverse square relationship. The concrete-shielded results in Fig. 7 are higher than their relative position in Fig. 3. The relative positions of the sodium activation curves in Fig. 7 are not the same as those for either the thermal neutron dose rates shown in Fig. 4 or the fast neutron dose rates shown in Fig. 3, indicating that sodium activation is from a composite of all neutron energies.

The results from the sodium activation and sulfur fluence were plotted on the curve used to determine criticality-accident doses by using only blood and hair activations.* The results are shown in Fig. 8. For this assembly all the points lie near

the top of the figure, indicating that there is considerable sodium activation per rad of fast neutron dose, which makes detection of small doses possible. The sulfur activation is low, however, because the number of >2.9-MeV-energy neutrons leaking from this assembly is less than for metal assemblies. This would make detection of the sulfur activation in the hair difficult for low doses and the evaluation of these low doses correspondingly less accurate, if the hair and blood activation method were used. The concrete-shielded points lie beyond the curve and the previously published curve can now be extended to include these results. The bare assembly points are above the curve but within the scatter of data previously obtained. The 14-m bare point is questionable and could be from counting statistics of the blood and sulfur samples. If this point is correct, it would be interesting in a future study to determine what is happening at larger distances. The steel-shielded points lie between the curves as expected, except for the 9-m point which fell on the upper curve.

* D. E. Hankins, "Dosimetry of Criticality Accidents Using Activations of the Blood and Hair," *Health Physics* 38, 529 (1980)

Additional Calibrations of the PTB Beta Sources

D. E. Hankins

The Hazards Control Calibration Facility has a set of the secondary standard beta-ray sources calibrated by the West German equivalent to our National Bureau of Standards. The sources are usually referred to as the "PTB" sources (the abbreviation for the Physikalisch-Technische Bundesanstalt at Braunschweig, Germany).^{*} We have expanded their usefulness to us by making additional calibrations to cover a wider range of dose rates.

Beta Energies

The set of sources consists of two ^{90}Sr sources ($E_{\max}$ 2.270 MeV), a ^{204}Tl ($E_{\max}$ 0.766 MeV), and a ^{147}Pm source ($E_{\max}$ 0.224 MeV). (By convention, a $^{90}\text{Sr} - ^{90}\text{Y}$ source is referred to as ^{90}Sr and the energy understood to be 2.270 MeV. In fact, the Sr and Y are in equilibrium and the 2.270 MeV is the energy of the Y beta.) These sources were selected to give beta spectra of low, medium, and high energies. The sources came to us with limited, but extremely accurate, calibrations. Only one calibration point is given for the ^{204}Tl , the ^{147}Pm , and the small ^{90}Sr source, and three calibration points at various distances are given for the large ^{90}Sr source. The large ^{90}Sr source is used unfiltered, bare, but the other sources are used with filters or "flatteners" which are used to flatten the beam from the source over an area where a reasonably sized probe would be exposed fairly uniformly.

The maximum beta energy for the sources is not the theoretical values given above (taken from nuclear tables). The spectra from the PTB sources of ^{90}Sr and ^{204}Tl are lower than the theoretical value, which is caused in part by absorbers of 50 and 20 mg/cm², respectively, used to seal the source. In addition, air attenuation, probe window thickness, and the effect of the flatteners all reduce the beta energies.

The ^{147}Pm source has a contaminant of ^{146}Pm (beta $E_{\max}$ 0.78 keV, 4.4-y T_{1/2}) which confuses the beta energy determination. Although this contaminant is small, it results in the observed effective energy for the flattened source at 20-cm dis-

tance being slightly higher than the $E_{\max}$ of ^{147}Pm . This source is sealed with a 5-mg/cm² cover and at 20 cm of air (~25.9 mg/cm²) very few of the ^{147}Pm betas from the source are detected.

Thus the beta energies from the sources (1) are lower than those given in the tables except for the ^{147}Pm source, (2) vary detectably as a function of distance from the source, and (3) are lower if the flatteners are used.

Measurements

In using the sources, we found that the calibration points given by the PTB were too limited to cover the wide ranges of dose rates we desired. We decided to expand the usefulness of these sources by making measurements at several distances without the flatteners using our extrapolation chamber¹ (purchased from Monitor Radiation Corp., Santa Barbara, CA). One advantage was that we would have information with the small ^{90}Sr source with the same energy spectra as the large ^{90}Sr (at the same distance) at dose rates that are different by approximately a factor of 25.

The dose rates obtained with the extrapolation chamber are given in Table 1. The effect of the flatteners on the dose rates is to reduce the dose rates from ^{90}Sr and ^{204}Tl by 36% and for ^{147}Pm by a factor of four. Note also the large decrease in the Pm dose rate when the detector is moved from 11 to 20 cm (a factor of ~11). This rapid drop in dose rate makes accurate measurements difficult since small measurement errors

^{*} Actually, the sources were made in England by Amersham, calibrated at the PTB, and sold by the German firm Buchler.

¹ See W. J. Price, *Nuclear Radiation Detection*, 2nd Ed. (McGraw-Hill Book Co., New York, NY, 1958).

Table 1. Dose rates (rads/h)^a at various distances from the PTB sources, measured with an extrapolation chamber.

Source	Distance from source (cm)				
	11	20	30	50	100
Large ⁹⁰ Sr, no flattener	176			8.87	2.03
Small ⁹⁰ Sr, flattener no flattener	6.69	0.606	0.947	0.333	0.0764
²⁰⁴ Tl, flattener no flattener	0.910		0.0677 0.1064	0.0290	
¹⁴⁷ Pm, flattener no flattener	3.24	0.0729 0.287			

^a Measured on 3/17/82.

become very important. The decrease in dose rate of the ⁹⁰Sr source with distance is slightly more rapid than inverse square. The ²⁰⁴Tl source decrease is significantly more rapid than calculated by using the inverse square rule.

We compared our results with the PTB calibration points in Table 2. Our results were generally in good agreement with the PTB values except for the ¹⁴⁷Pm source, where we are 16.5% high. This could be error from distance measurement or other factors. The PTB values are probably more accurate than our values and should be used when possible.

Conversions

To convert the air dose rates in Tables 1 and 2 into tissue rads, conversion factors of 1.111, 1.139,

and 1.150 are given by the PTB for ⁹⁰Sr, ²⁰⁴Tl, and ¹⁴⁷Pm, respectively. These factors are given only for the calibration points they measured and there is no indication as to how much these would vary at other locations. I doubt they change by more than a percent or two, and using these values for the other points should not result in a significant error.

To determine the depth dose at 7 mg/cm (dead layer of skin thickness) the PTB gives attenuation factors that are different for each measurement point. These values vary from 1.070 to 1.041 for ⁹⁰Sr. The values for ²⁰⁴Tl and ¹⁴⁷Pm are 0.955 and 0.20, respectively. The values >1.0 for ⁹⁰Sr are caused by the buildup factor's being greater than absorption, but for ²⁰⁴Tl and ¹⁴⁷Pm the absorption in the skin exceeds the buildup factor and the dose at a depth of 7 mg/cm² is lower, particularly for ¹⁴⁷Pm.

Table 2. Comparison of measured vs reported dose rates in air (rads/h) from the PTB beta sources.

Source	Distance (cm)	Measured value	PTB reported value ^a	% Difference
Large ⁹⁰ Sr	11	176	175	+0.6
	30		23.9	
	50	8.87	8.54	+3.9
Small ⁹⁰ Sr, flattener	30	0.606	0.591	+2.5
²⁰⁴ Tl, flattener	30	0.0677	0.0720	-6.0
¹⁴⁷ Pm, flattener	20	0.0729	0.0626	+16.5

^a Corrected for decay to 3/17/82.

Probability of Interaction and Energy Deposition of Neutron-Induced Recoil Protons in a Cell Plasma Membrane

C. T. Prevo and T. Straume

The germ cells (oocytes) in female mice are known to be unusually sensitive to killing by radiation.¹ Fifty percent are killed by 7 rads of gamma rays. It is of interest both radiobiologically and for radiation protection purposes to identify the radiosensitive target in these cells. This has recently been studied by Straume² who presents evidence that the target is the oocyte's plasma membrane. In this paper we describe calculations that validate those made by Straume. His calculations use published microdosimetric data³ (event frequency) obtained for a small tissue sphere (mouse-oocyte dimensions) to convert to a tissue shell with the dimensions of the oocyte's plasma membrane.

Calculations

One can calculate the probability of recoil protons depositing energy in the cell membrane because of neutron interactions in the cell if one assumes a spherical model of the cell and restricts the calculation to protons of one energy.

The spherical cell is shown in Fig. 1. The probability that a recoil proton will deposit energy in the membrane because of a neutron interaction in the cell is given by

$$P = \frac{P_1 V_1 + P_2 V_2 + P_3 V_3 + P_4 V_4}{V_{\text{tot}}} \quad (1)$$

where P_1 , P_2 , P_3 , and P_4 are the probabilities that interactions in the volumes V_1 , V_2 , V_3 , and V_4 will result in energy deposition in the membrane. V_1 is the innermost sphere of radius $R_1 - R$ and for which $P_1 = 0$, since protons originating more than a distance R away from the membrane cannot deposit energy in the membrane. Protons originating in the membrane (V_3) will all deposit energy in the membrane and thus $P_3 = 1$. The situation in V_2 and V_4 , which are within range of the membrane, is somewhat more complicated. For these regions the probability that the proton will deposit energy in the membrane depends upon the point position at which the proton is created. For a proton produced at a distance r from the origin and for which $R_1 - R \leq r \leq R_1$, the probability is given by

$$\frac{\Omega}{4\pi} = \sin^2 \frac{\alpha}{2} = \frac{1 - \cos \alpha}{2}$$

where α is shown in Fig. 2. From Fig. 2 we can see that

$$\begin{aligned} \cos \alpha &= \frac{R_1 - r - Z}{R} = \frac{R_1 - r - R_1(1 - \cos \theta)}{R} \\ &= \frac{R_1 \cos \theta - r}{R} \end{aligned}$$

Figure 1. Spherical model of cell. R_1 and R_2 are the inner and outer radii of the membrane. R is the range of a recoil proton.

Figure 2. Geometric relations for cell for Eq. (2).

and since

$$\cos \theta = \frac{r^2 + R_1^2 - R^2}{2rR}$$

then

$$\cos \alpha = \frac{r^2 + R_1^2 - R^2}{2rR} - \frac{r}{R}$$

and

$$\begin{aligned} P_2 &= \frac{\Omega}{4\pi} = \frac{1}{2} \left(1 + \frac{r}{2R_1} - \frac{R_1^2 - R^2}{2rR_1} \right) \\ P_2 &= \frac{\int P_2 dV}{\int dV} \\ &= \frac{1}{2} \frac{\int_{R_1-R}^{R_1} r^2 dr + \int_{R_1-R}^{R_1} \frac{r^3 dr}{2R_1} - \frac{R_1^2 - R^2}{2R_1} \int_{R_1-R}^{R_1} r dr}{\int_{R_1-R}^{R_1} r^2 dr} \\ &= \frac{1}{2} \left[1 + \frac{3}{8R_1} \frac{[R_1^2 - (R_1 - R)^4]}{R_1^3 - (R_1 - R)^3} \right. \\ &\quad \left. - \frac{3(R_1^2 - R^2)}{4R_1} \frac{[R_1^2 - (R_1 - R)^2]}{R_1^3 - (R_1 - R)^3} \right] \end{aligned} \quad (2)$$

For a proton produced at a distance r from the origin and for which $R_2 \leq r \leq R_2 + R$, the probability is given by

$$\frac{\Omega}{4\pi} = \sin^2 \frac{\alpha}{2} = \frac{1 - \cos \alpha}{2}$$

where α is shown in Fig. 3. From Fig. 3 we see that

$$\begin{aligned} \cos \alpha &= \frac{x}{R} = \frac{r - R_2 \cos \theta}{R} = \frac{r}{R} - \frac{r^2 + R_2^2 - R^2}{2rR} \\ P_3 &= \frac{1}{2} \left(1 - \frac{r}{R} + \frac{r^2 + R_2^2 - R^2}{2rR} \right) \\ &= \frac{1}{2} \left(1 - \frac{r}{2R} + \frac{R_2^2 - R^2}{2rR} \right) \\ &= \frac{1}{2} \frac{\int_{R_2}^{R_2+R} r^2 dr + \int_{R_2}^{R_2+R} \frac{r^3 dr}{2R} - \frac{R_2^2 - R^2}{2R} \int_{R_2}^{R_2+R} r dr}{\int_{R_2}^{R_2+R} r^2 dr} \\ &= \frac{1}{2} \left[1 - \frac{3[(R_2 + R)^4 - R_2^4]}{8R[(R_2 + R)^3 - R_2^3]} \right. \\ &\quad \left. + \frac{3(R_2^2 - R^2)[(R_2 + R)^2 - R_2^2]}{4R[(R_2 + R)^3 - R_2^3]} \right] \end{aligned} \quad (3)$$

Results

A Monte Carlo code was written which also calculates the probabilities for energy deposition for (n,p) reactions inside and outside of a spherical cell as well as the average energy deposition in the plasma membrane. The results for proton energies from 0.043 to 0.430 MeV are given in Table 1. The proton ranges were obtained by using log-log interpolation of the values given for water in ICRU Report 16. The results are based on ten batches of 10^5 protons each. The results are in good agreement both with those reported by Straume² for 0.215-MeV protons and with calculations based on Eqs. (2) and (3).

Figure 3. Geometric relations for cell for Eq. (3).

Table 1. Proton energy (E) deposition and energy deposition probabilities (P) in the cell membrane.

E (MeV)	Range (μm)	E_{dep} (MeV)	P_{inside}		P_{outside}	
			Calc	MC ^a	Calc	MC
0.043	0.663	0.00098 ± 0.00006	0.278	0.279	0.225	0.224
0.086	1.116	0.00159 ± 0.00011	0.300	0.302	0.209	0.208
0.129	1.597	0.00227 ± 0.00014	0.327	0.328	0.194	0.193
0.172	2.089	0.00276 ± 0.00011	0.359	0.357	0.182	0.178
0.215	2.643	0.00302 ± 0.00015	0.393	0.394	0.167	0.164
0.258	3.350	0.00328 ± 0.00015	0.445	0.446	0.152	0.147
0.301	4.094	0.00329 ± 0.00016	0.510	0.508	0.138	0.131
0.344	4.870	0.00337 ± 0.00014	0.577	0.579	0.124	0.116
0.387	5.676	0.00328 ± 0.00018	0.654	0.657	0.109	0.103
0.430	6.509	0.00310 ± 0.00017	0.736	0.733	0.099	0.091

^a Monte Carlo.

References

1. R. L. Dobson and T. C. Kwan, "The Tritium RBE at Low-Level Exposure—Variations with Dose, Dose Rate, and Exposure Duration," *Curr. Top. Radiat. Res. Q.* **12**, 44–62 (1979).
2. T. Straume, "Biological Effectiveness of Neutron Irradiation on Animals and Man," PhD thesis, University of California, Davis, CA (1982).
3. H. H. Rossi, "The Role of Microdosimetry in Radiobiology," *Radiat. Environ. Biophys.* **127**, 29–40 (1979).

Calibration of the PNR-4 Neutron Remmeter for Weapons-Like Spectra

C. T. Prevost

There is an error in measuring neutron dose rates with a neutron remmeter because of the differences in the spectral shapes of the field being measured and the calibration source. Errors as large as 30% can result if a PuBe neutron source was used for calibration. We performed a set of calculations to demonstrate that for the type of neutron sources measured in LLNL's Intrinsic Radiation Facility, ^{252}Cf is preferable to either Am-Li or PuBe neutron sources for calibration.

The theoretical calibration factor—(counts/min)/(mrem/h)—for operational and calibration spectra were calculated using the codes DOSE and DOSEM, which were written for the CDC-7600. DOSE calculates the theoretical spectrum-weighted response using a TARTNP calculated response function, shown in Fig. 1 and given in Table 1. TARTNP is a Monte Carlo coupled neutron-photon transport code.¹ DOSEM does the same calculation as DOSE, except that 30 group MORSE-calculated spectra are used. MORSE is also a Monte Carlo coupled neutron-photon transport code.²

The calibration factors and spectral correction factors are shown in Table 2. It was assumed in each case that the instrument had been calibrated with ^{252}Cf .

Ideally, one would like the calibration factors to be small. A dose variance of

$$\sigma^2 = \sum_{i=1}^{30} (DW_i - DC_i)^2$$

where

DW_i = the dose rate of the i^{th} group of a MORSE-calculated neutron spectrum

and

DC_i = the dose rate of the i^{th} group of the calibration source,

Figure 1. PNR-4 response and tissue dose response as a function of energy.

Table 1. PNR-4 response calculated by TARTNP.

Energy (MeV)	Response (counts/n)
2.5×10^{-8}	2.95×10^{-5}
1.0×10^{-6}	3.57×10^{-5}
1.0×10^{-5}	5.81×10^{-5}
1.0×10^{-4}	7.50×10^{-5}
1.0×10^{-3}	9.32×10^{-5}
3.0×10^{-2}	1.53×10^{-4}
5.0×10^{-2}	2.07×10^{-4}
7.0×10^{-2}	2.24×10^{-4}
1.0	2.78×10^{-4}
2.0	3.06×10^{-4}
2.2	3.27×10^{-4}
3.0	2.82×10^{-4}
4.0	2.72×10^{-4}
6.0	2.51×10^{-4}
8.0	2.04×10^{-4}
10.0	1.73×10^{-4}
14.0	1.5×10^{-4}

was used to compare Am-Li, ^{252}Cf , and PuBe as calibration sources for the W48, W68, and W84. The results are shown in Table 3. For the three cases calculated, the variance was minimum for the ^{252}Cf . This indicates that the dose spectrum for the W48, W68, and W84 more nearly matches that of ^{252}Cf than that of Am-Li or PuBe.

Table 2. Theoretical calibration factors for PNR-4 for various neutron spectra.

Source	Calibration factor	Spectral correction factor ^d
Am-Li	62.6	0.848
^{252}Cf	53.1	1.00
Pu ^b	51.8	1.025
PuBe ^c	44.9	1.183
PuBe ^b	43.2	1.229
W48	55.5	0.957
W68	55.6	0.955
W84	58.7	0.950

^a Neutron dose results based on instrument calibration with ^{252}Cf need to be multiplied by this factor.

^b 30 group spectra were used by Intrinsic Radiation as input for MORSE.

^c NBS spectrum.

Table 3. Dose variances of weapon dose spectra for various calibration sources.

Weapon	Am-Li	^{252}Cf	PuBe
W48	1795	82.7	833.8
W68	1900	128	772
W84	147	36	1600

References

1. E. F. Plechaty and J. R. Kimlinger, "TARTNP: A Coupled Neutron-Photon Monte Carlo Transport Code," Lawrence Livermore National Laboratory, Livermore, CA, UCRL-50400, Vol. 14 (1976).
2. T. P. Wilcox, "MORSE-L, A Special Version of the MORSE Program Designed to Solve Neutron, Gamma and Coupled Neutron-Gamma Penetration Problems," memorandum (Sept. 1, 1972).

A Single-Calculation Comparison of MORSE76L and ANISN

C. F. Prevost and J. C. Fisher

Previously, ANISN was used to calculate the dose rates at 1 m for ^{252}Cf in various moderators in cell A of the calibration facility.¹ The cell is 12.2 m long, 9.14 m wide, and 7.32 m from basement to ceiling. Since ANISN is a one-dimensional discrete-ordinates code, it cannot be used to address issues involving asymmetry in the geometry such as the hole in the moderator and the walls of the cell. MORSE is a two-dimensional Monte Carlo radiation-transport code and is well suited for accounting for the effects due to asymmetry.

The first issue raised was: how well do MORSE and ANISN agree in their determination of the dose rates for the same problem? ^{252}Cf in a 15-cm D_2O sphere was picked for the comparison. The same energy group structure, cross sections, geometry, and isotopic compositions were used for both problems. The isotopic composition of the concrete is given in Table 1. The 30 group spectra are given in columns three and four of Table 2 and are shown in Figs. 1-3. Running the MORSE problem with 10 batches of 10^4 neutrons each produced a 1.0% standard deviation on the dose rate at 1 m. This dose rate is 1.9% greater than predicted by ANISN. This is really excellent agreement between the two codes.

Table 1. Isotopic composition of the concrete walls.

Isotope	Atoms/barn-cm
H	0.01386
C	0.01200
O	0.04582
Na	0.00100
Mg	0.00011
Al	0.00174
Si	0.01063
K	0.00046
Ca	0.00152
Fe	0.00035

The second issue raised was: what percent of the flux and dose is below the cadmium cutoff? Using 0.4 eV as the cutoff energy, MORSE predicts that 27.9% of the flux and 4.40% of the dose is below the cadmium cutoff, compared to 29.3% and 4.83% predicted by ANISN. We did an additional calculation to determine what percent of the dose and flux density at 1 m is due to return from the room. This was accomplished by rerunning the problem with ^{252}Cf in the 15-cm D_2O sphere; however, this time the room air and walls were replaced with a vacuum. The difference between the two runs gave that portion due to room return.

The results were a 5.6% return for the dose and 19.9% return on the flux density. Table 2 gives the vacuum-differential spectrum as well as the fluence-to-dose-equivalent conversion factors used for all the calculations. These rem/n-cm^{-2} values are those used in the original ANISN calculations.²

References

1. R. V. Griffith et al., "Multi-technique Characterization of Neutron Fields from Moderated ^{252}Cf and $^{238}\text{PuBe}$ Sources," *National and International Standardization of Radiation Dosimetry*, Vol. II, IAEA-SM-222/14.
2. *Protection Against Neutron Radiation*, NCRP Report No. 38 (1971).

Table 2. The differential spectra for ^{252}Cf in 15-cm D_2O sphere ($\text{n}/\text{cm}^2\text{-MeV}$).

Group	E Top	ANISN	MORSE	MORSE - Vacuum	rem/n-cm ²
1	18.000 MeV	0	0	0	4.278E-08 ^a
2	15.612	0	0	0	4.241E-08
3	13.541	8.524E-10	8.690E-10	8.893E-10	4.209E-08
4	11.745	2.722E-09	2.809E-09	2.803E-09	4.146E-08
5	10.187	7.571E-09	7.933E-09	7.445E-08	4.985E-08
6	8.835	1.606E-08	1.686E-08	1.717E-08	4.085E-08
7	7.663	2.884E-08	3.034E-08	3.009E-08	4.085E-08
8	6.647	4.967E-08	5.342E-08	5.139E-08	4.085E-08
9	5.765	7.793E-08	7.991E-08	7.927E-08	4.085E-08
10	5.000	1.162E-07	1.194E-07	1.219E-07	4.085E-08
11	4.205	1.469E-07	1.551E-07	1.545E-07	4.005E-08
12	3.536	2.017E-07	2.099E-07	2.105E-07	4.026E-08
13	2.973	3.032E-07	3.109E-07	3.121E-07	3.968E-08
14	2.500	3.470E-07	3.551E-07	3.566E-07	3.968E-08
15	1.988	3.013E-07	3.165E-07	3.091E-07	3.885E-08
16	1.581	2.946E-07	3.120E-07	2.892E-07	3.779E-08
17	1.257	2.514E-07	2.679E-07	2.465E-07	3.429E-08
18	1.000	3.026E-07	3.361E-07	2.882E-07	3.053E-08
19	0.841	4.355E-07	4.658E-07	4.256E-07	2.769E-08
20	0.707	5.054E-07	5.476E-07	5.099E-07	2.395E-08
21	0.595	5.457E-07	5.848E-07	5.539E-07	2.973E-08
22	0.500	7.473E-07	8.020E-07	7.241E-07	1.502E-08
23	0.224	2.498E-06	2.592E-06	2.469E-06	8.170E-08
24	0.100	1.198E-05	1.207E-05	1.139E-05	2.572E-09
25	12.61 keV	8.983E-05	8.943E-05	8.053E-05	1.002E-09
26	1.59	6.144E-04	6.001E-04	5.343E-04	1.068E-09
27	200 eV	5.305E-03	5.229E-03	4.591E-03	1.157E-09
28	25.2	3.430E-02	3.383E-02	2.815E-02	1.218E-09
29	3.18	2.397E-01	2.352E-01	3.772E + 00	1.068E-09
30	0.40	7.300E - 00	6.851E - 00	3.772E - 00	1.068E-09
	0.0001				

^a E-08 denotes $\times 10^{-8}$, etc.

Figure 1. ANISN differential spectra for ^{252}Cf in 15-cm D_2O sphere in air.

Figure 2. MORSE differential spectra for ^{252}Cf in 15-cm D_2O sphere in vacuum.

Figure 3. MORSE differential spectra for ^{252}Cf in 15-cm D_2O sphere in air.

Effective Absorption Thickness of the ^7Li TLD for Beta Particles

D. E. Hankins

To evaluate the reading of a thermoluminescence dosimeter (TLD) exposed to beta particles, a correction must be made for the absorption of the beta particles in the TLD material. This absorption results in the TLD's being irradiated nonuniformly and, for very-low-energy beta, the front surface of the TLD may be highly exposed and the back part may not be exposed at all. Calibrations of TLDs are usually made to ^{137}Cs gamma rays which irradiate the entire TLD uniformly. The correction for beta absorption requires that the thickness of the TLD and the energy of the beta be known. In place of the beta energy, one can use the absorption curve results from aluminum if they are available. In both cases, however, the effective thickness of the TLD relative to aluminum, which is the standard for beta studies, is required. The "Beta Particle Range Energy Curve" given in the *Radiological Health Handbook** is for aluminum (although this isn't stated in the book and the curve is often misused for other materials). We determined the effective thickness of our TLDs relative to aluminum.

Introduction

The thickness of materials used for beta shielding is usually stated in mg/cm^2 rather than in cm, in., mils, etc. This is because beta particles are absorbed very nearly the same for a given thickness in mg/cm^2 regardless of the type of material. However, there are fairly significant differences at low-Z materials, such as lithium, and reasonably large deviations from aluminum are found.

At LLNL we use 35-mil-thick ^7Li in our personnel badges. The TLDs are $1/8 \times 1/8$ in. and weigh ~ 0.238 g each. The absorption thickness for 1 cm^2 of the TLD material is calculated to be $236\text{ mg}/\text{cm}^2$. This is considerably thicker than the value we determined below using absorption measurements.

Measurements

We compared the absorption of the ^7Li TLD material with that of aluminum foils using two ^{90}Sr ($E_{\text{max}} 2.27\text{ MeV}$) and a ^{204}Tl ($E_{\text{max}} 0.766\text{ MeV}$)

beta sources. We used the Eberline RO-7 for the large ^{90}Sr source at 11 and 30 cm, and the pancake probe for the small ^{90}Sr and the ^{204}Tl sources. The small ^{90}Sr and ^{204}Tl sources were used with beam flatteners (see the article on "Additional Calibrations of the PTB Beta Source" in this report). We found no significant variation that could be attributed to differences in beta source energies or the use of different types of instruments.

Because the probe size of the RO-7 and the pancake probe is large compared with the size of the TLD array that we were going to use (see below), we used a small hole ($\sim 5/16$ -in.-diam) drilled in a $1/4$ -in.-thick Al disk placed over the front of the probe of the two instruments. A series of 10-mil Al foils was used to obtain an absorption curve for aluminum, which is plotted in Fig. 1.

Twenty-five of the TLDs were positioned in a 5×5 array with their edges touching to form a $5/8 \times 5/8$ in. block of TLDs, which was held together by wrapping with 1-mil Al foil. Three of these TLD blocks were prepared and measurements made with each to ensure that no leakage between chips was occurring. The relative reading of the instruments was determined with the TLDs placed over the hole. The readings are indicated in Table 1 and by the boxes in Fig. 1.

* Bureau of Radiological Health, *Radiological Health Handbook*, U.S. Dept. of HEW, Public Health Service, Rockville, MD.

Figure 1. Effective absorption thickness of ^7Li TLDs (35-mil).

Results

The effective thickness of the TLD material and two layers of Al foil (9.6 mg/cm^2) was found to be between 180 and 190 mg/cm^2 when the ^{90}Sr sources were used, and 160 mg/cm^2 when the ^{204}Tl source was used. The count rates with the

^{204}Tl source were small and the results correspondingly less accurate than the ^{90}Sr results. I recommend that a value of 175 mg/cm^2 be used as the effective thickness of our ^7Li TLD material for betas when Al is used as the reference material. This value applies only to the TLD material; other absorbers in front of the TLD must be considered separately.

Table 1. Relative beta reading of instruments shielded by one thickness of ^7Li TLD material (35-mil), and the effective thickness (compared with aluminum).

Instrument	Source	Distance (cm)	Relative instrument reading	Effective absorption thickness ^a (mg/cm ²)
RO-7	^{90}Sr (large)	30	0.386	180
RO-7	^{90}Sr (large)	11	0.416	190
Pancake	^{90}Sr (small) with flattener	30	0.375	185
Pancake	^{204}Tl with flattener	30	0.015	160

^a includes 9.6 mg/cm² of Al foil.

These results indicate that ^7Li TLDs absorb less of the beta energy than do comparable thicknesses (in mg/cm²) of aluminum. When calculating the correction for the energy absorbed in a TLD to determine the reading expected from a TLD or vice versa, the effective absorption thickness should be used. If the calculations are based on absorption curves from the *Radiological Health Handbook* or from measurements of beta absorption in aluminum, the effective TLD thickness of 175 mg/cm² must be used. An example of this cor-

rection was a measurement we recently made of the beta- and gamma-dose rates of the Godiva IV Critical Assembly at LANL. We used TLDs to measure the dose rates and corrected their readings for self-absorption using the information from an absorption curve obtained at the Assembly with aluminum foils. If we had used the actual rather than the effective Al thickness, the final evaluated beta-dose rate would have been significantly greater than the actual dose rate.

FIRE SAFETY

Evaluation of the Fire Properties and Potential Safety Hazard of Building 431's Exterior Wall Panels

H. K. Hasegawa, K. Staggs, S. J. Priante, and J. A. Sharry

We have performed tests on samples of exterior wall panels from Bldg. 431 at LLNL to determine the fire properties—the potential flamespread and resultant smoke generation—of the coating material (a felt and asphalt composite). These tests will also be used to assess the potential hazard to life if the panels are exposed to a fire. This information will help the Fire Protection Engineering Group to evaluate the proposed expansion plans for Bldg. 431 and to determine appropriate countermeasures to protect life and property.

Introduction

The coating material has been applied to corrugated steel panels. We considered two fire exposure scenarios: (1) an "outside-in" exposure that would subject the exterior side of the panel (coated side) to flame, and (2) an "inside-out" fire that would expose the interior side (uncoated side) to flame. These two exposures would enable us to evaluate the fire and smoke response of the coating as a result of direct flame impingement and also of indirect heating. Our goals were:

- Define an appropriate and realistic exposure fire.
- Perform two large-scale fire experiments and monitor the following:
  1. Time to ignition.
  2. Rate and extent of flamespread.
  3. Smoke generation (qualitative).
  4. Combustion-gas composition and rate of acid ion generation.
  5. O_2 , CO , CO_2 , and hydrocarbon concentrations.
  6. Mass loss rate of burning coating.
  7. Temperatures—
    - a. Both exposed- and unexposed-face temperatures on panels.
    - b. Throughout the test cell.
  8. Thermal fluxes.

Survey of Fire-Exposure Studies

We performed the following survey to ensure that we defined an appropriate and realistic fire

exposure to evaluate the fire response of the panel coating material. Since energy conservation has gained increasing importance, fire research activity dealing with exterior walls has rapidly increased. The primary reason for this stepped-up activity was the appearance of a number of exterior wall systems intended to reduce thermal transfer by using various thicknesses of foamed plastics. Many of these foamed plastics presented a considerable fire hazard if not protected by an appropriate fire-resistant coating or barrier.

One of the primary scenarios investigated for exterior fire spread was flames extending out of a fully involved compartment window and impinging on the exterior wall finish above the window. A number of research organizations performed room-burnout experiments to characterize this thermal assault through a window on the exterior of the enclosure. Table 1 compares the results of experiments at the Building Research Institute (BRI) in Japan, Underwriters Laboratories (UL), and Stanford Research Institute (SRI), International. BRI and UL used wood cribs in similar-size compartments for the various fuel loads listed. The SRI results are taken from experiments using a fuel oil burner to produce the various fire exposures. Actually, the SRI apparatus was designed to simulate fire intensities developed in both real-life situations and exemplar compartment burns (e.g., those by BRI and UL) to evaluate the fire performance of traditional exterior wall treatments.

Figures 1 and 2 show isotherms of flames extending out of the windows of the experimental compartment fires. They illustrate the difference

Figure 1. Study of convection currents created by fires of large area (from Ref. 1).

in flame characteristics as a result of varying room size, fuel loading, and window opening factor $(\frac{A_w}{A_t})^*$. The flame in Fig. 1a had a fuel loading of 41 kg of wood per m^2 of floor area, or 12 kg/m^2 of total surface bounding the room with a window opening factor of $0.028 m^{1/2}$ (167 cm high $\times$ 91 cm wide). Figure 1b corresponds to a fire loading of 40 kg of wood per m^2 of floor area, or 10 kg/m^2 of total surface area and a window opening factor of $0.060 m^{1/2}$ (1 m high $\times$ 3 m wide). Figure 2 summarizes the results of 30 tests in which the fuel load was varied from 24.4 kg/m^2 (5 lb/ft^2) of floor to 97.6 kg/m^2 (20 lb/ft^2) and window opening sizes from 0.61 m wide by 1.82 m high to 1.82 m wide by 2.44 m high.

* A = total window area of room, H = height of window.
 A_t = total area of the surfaces bounding the room.

Exposure Fire Calibration

Excluding the one for 20 lb/ft^2 and 1.0 gpm, the experimental fuel loads and resultant temperature profiles in Table 1 are realistic fire exposures for exterior walls. Therefore, simulating selected temperatures from the table with our burner would provide an ignition source with enough energy to produce a fairly severe fire exposure with a flame height short enough (~ 1 m) to monitor flame spread due to the burning of the coating material.

On the basis of the temperature ranges in Table 1 at the 1-ft. and 4-ft heights, we opted for average temperatures of between 400°C and 600°C and between 200°C and 400°C, respectively. We used a 0.89-m $\times$ 1.98-m sheet of 1.27-cm gypsum wallboard as the calibration panel to characterize the exposure fire. In order to find a proper gas-flow setting for the desired fire, a

Figure 2. The projection of flames from burning buildings (from Ref. 2).

multi-nozzled burner was ignited for a short duration to produce a flame pattern on the panel which would indicate where to locate thermocouples and heat-flux gauges.

The calibration procedure was to ignite the burner and adjust the natural gas flow until a satisfactory flame height was established, at which

point the gas flow was noted. Then the various temperatures and fluxes produced by this gas-flow rate were monitored and recorded. This procedure was repeated until we achieved the combination of the flame height (~ 1 m) and temperatures at the 1-ft and 4-ft heights we defined for the ignition source. We were able to time

Table 1. Comparison of room-burnout experiments.

	Fuel load							
	BRI ^a (wood)		UL ^b (wood)		SRI ^c (gasoline)			
	8.5 lb/ft ²	8.2 lb/ft ²	5 lb/ft ²	20 lb/ft ²	0.25 gpm	0.5 gpm	0.75 gpm	1.0 gpm
Total flame extension above window ^d (ft)	5	5.75	5		2	4	6	8
Temperature (°C)								
1 ft above window	500	500	540	960	270	620	680	790
4 ft above window	300	400	400	870	100	310	560	750
8 ft above window	200	200	340	705	125	210	260	310

^a Building Research Institute, Japan.

^b Underwriters Laboratories.

^c Stanford Research Institute.

^d Above top edge of window.

our gas flow to produce a source fire with a flame height about 1 m and temperatures of approximately 500°C at 1 ft and 250°C at 4 ft. These levels fit the criteria we abstracted from Table 1 fairly well.

Experimental Setup

The experimental apparatus shown in Fig. 3 was designed for the performance of large-scale vertical cable burns but was well suited for this experimental program. As it turned out, we were provided with panel samples which were coated on both sides; therefore, we could evaluate the panel response to both fire scenarios (outside-in and inside-out) with the same experiment. Figure 4 shows the test panel configuration along with the location of both exposed- and unexposed-face thermocouples and the burner. Because the actual wall panel was 2 ft wide, we added a 2-ft-2-in.-wide (0.66-m) corrugated aluminum panel on each side (see Fig. 4) to prevent flames from wrapping around the edges of the test specimen.

The panel configuration was then suspended from the test frame by a cable which was attached to a load cell to monitor dynamic mass loss from the burning of the panel coating. In addition to the above, we monitored and recorded the following:

- Temperatures—
  1. Throughout the test cell.
  2. Inlet and exhaust air temperatures.
  3. Exposed- and unexposed-face temperatures on panel.

- Ventilation rate.
- Heat fluxes (calorimeters) at several locations, approximately 30 cm from the panel.
- Gas analyzers—
  1. Oxygen depletion.
  2. % Carbon dioxide.
  3. % Carbon monoxide.
  4. % Hydrocarbons.
  5. Combustion-gas acid ion concentration.
- Videotape and 35-mm photographs.

Results

The natural gas flow to the burner was set to produce the exposure fire defined in the calibration burns, and the burner ignited. The panel was continually exposed to the ignition source for 660 s (11 min) with very little, if any, fire contribution from the coating material. Figure 5 shows the experiment at the point of greatest flameheight and Fig. 6 illustrates the post-test condition of the panel. The char pattern on the panel is primarily an imprint of the exposure fire, indicating that the material did not contribute to flamespread. The non-burning of the coating is strongly substantiated by Fig. 7, which is a composite plot of the exposed-face temperatures. Note that, once the exposure flame stabilizes, the temperatures flatten out and remain constant for the duration of the test, which indicates that no combustion of the coating occurred. The back-face temperatures were about 45% of the exposed-face temperatures (on the average). Finally, most sensors monitoring

Figure 3. Photograph of experimental apparatus.

Figure 4. Building 431 test panel, showing location of thermocouples (TCs).

Figure 5. Photograph of latter stage of experiment.

Figure 6. Post-test char pattern on panel.

test-cell air temperatures recorded only a minor rise above ambient conditions. Other significant data were as follows:

- In correspondence with the temperature data, incident heat fluxes were very low.
- At a forced ventilation rate of 550 liters/s there was no detectable oxygen depletion.
- Similarly, there was no mass loss recorded by the load cell.
- Carbon monoxide, carbon dioxide, and hydrocarbon readings were at pre-test levels.

Furthermore, analysis of the combustion products for acid components such as HCl and formaldehyde rendered negative results.

Analysis of the Coating Material

Thermogravimetric analysis (TGA) was performed on a portion of the sample to determine the % volatiles, initial degradation temperature, gross degradation temperature, and % ash remaining. A 4.52-mg sample was placed in the TGA apparatus in an air atmosphere, and temperature was programmed at 20°C/min from room temperature to 800°C. Results were as follows:

% volatiles	~0.5% at 125°C
Onset of degradation	260°C
Onset of gross degradation	390°C
% ash	60% at 750°C

A sample was prepared for GC-MS analysis by running an approximately 20-mg sample in the

TGA apparatus up to the final degradation temperature (800°C), cooling the unit to room temperature, and washing down the furnace tube assembly with 10 ml of methylene chloride to remove the pyrolyzed product. This sample-collection procedure was repeated because there was little evidence of pyrolysis products on the furnace tube after the first run.

The methylene chloride-sample mixture was then concentrated down to low volume (~0.5 ml), and a 4- μ l aliquot was injected onto the GC column for component separation. The GC conditions were as follows:

Column	25-ft fused-silica methyl silicon fluid capillary
Starting temperature	50°C
Temperature ramp	10°C/min
Final temperature	230°C

The separated components then went directly into the mass spectrometer, where each compound underwent fragmentation by high-energy-electron impact. The specific chemical patterns of each component produced by the fragmentation were then stored on a computer disk.

The stored information on each compound was then compared against approximately 40,000 compounds on a library disk and the 10 best "hits" from the library search were printed out. The "hits" were then subjected to an additional computer program which compares individual mass peak numbers between the sample and the library "hits." Table 2 lists compounds in the panel insulation identified using this method.

Figure 7. Exposed face temperatures.

Table 2. Analysis of degradation products.

Degradation products	Flash point (°C)	Ignition point (°C)	Boiling point (°C)	Water soluble ?	Extinguishing method	NFPA hazard identification		
						Health	Flammability	Reactivity
2,2-diethyl-1,3-propanediol	102	—	160	Yes	Alcohol foam	2	1	0
Phthalic acid	168	—	289	—	—	0	—	0
Phthalic anhydride	152	570	284	No	H ₂ O or foam	2 ^a	1 ^b	0
1-eicosanol	—	—	—	—	—	—	—	—
Nonadecanol	—	—	—	—	—	—	—	—
2L, 4D-dimethyl heptanol	—	—	—	—	—	—	—	—
Octyl acetate	71	268	199	No	—	2	2	0
Beta helol-nitro octane	—	—	—	—	—	—	—	—
1,2-epoxy-2,4-dimethyl pentane	—	—	—	—	—	—	—	—

^a Chemical is hazardous when inhaled, but self-contained apparatus will protect against inhalation.

^b Chemical must be preheated to ignite. Water may cause frothing if it gets below the surface of the liquid and turns to steam.

Conclusions

On the basis of the preceding test methods and the sample provided us, we concluded that the coating on the panels from Bldg. 431 would not contribute to the fire or generate any significant quantities of smoke. Because of this negative result we attempted to ignite the coating by cutting holes through the panel with an acetylene torch. The material did ignite into small flamelets around the holes, but would extinguish shortly after removal of the torch.

It should be noted that the preceding results were from the specific panel supplied us. Vari-

ables such as coating-mix variations, coating depth, etc. could influence the flammability of the panels.

References

1. S. Yokoi, "Study of Convection Currents Created by Fires of Large Areas," *International Symposium on the Use of Models in Fire Research, Washington, 1961*, pp. 130-147.
2. L. G. Seigel, "The Projection of Flames from Burning Buildings," *Fire Technology* 5 (February 1969).

Publication Abstracts

G. R. Endres, J. M. Aldrich, L. W. Brackenbush, L. G. Faust, R. V. Griffith, and D. E. Hankins, *Neutron Dosimetry at Commercial Nuclear Plants*, Nuclear Regulatory Commission, NUREG/CR-1769 (1981); Battelle Pacific Northwest Laboratories, Richland, WA, PNL-3585 (1981).

A series of measurements of neutron energy spectra, neutron dose-equivalent rates, and personnel neutron dosimeter responses taken at six commercial nuclear power plants are discussed in detail. Results of neutron spectral measurement indicate average or effective neutron energies below 100 keV at all locations studied inside containment of pressurized water reactor (PWR) power plants. The neutron energies were somewhat higher near containment penetrations at a boiling water reactor (BWR) power plant. Neutron spectra were obtained using multispheres and ^3He proportional counter spectrometer systems. Graphs of all unfolded spectra from both measurement systems are presented.

Dose-equivalent rates were determined, where possible, both from the multisphere spectra and from event-size spectra measured with a tissue-equivalent proportional counter (TEPC). There is generally good agreement between the two techniques (within about 20%), although quality factors Q derived from the two types of spectra are quite different.

Results obtained from neutron track emulsion type A (NTA film), polycarbonate track etch and CR-39 plastic, and thermoluminescence (TLD albedo) personnel dosimeters are presented. In the neutron spectra found in reactor containment, the NTA film and polycarbonate track etch dosimeters failed to respond above the levels needed for positive indication of dose. The CR-39 track etch dosimeters showed a low response (50% or less) when calibrated with ^{252}Cf neutron sources. Thermoluminescence-albedo dosimeters showed a very high response but require spectral corrections for proper interpretation.

D. Hankins, *Beta-Energy-Response Determination of the RO-7 Survey Instrument*, Lawrence Livermore National Laboratory, Livermore, CA, UCID-19484 (1982).

We obtained an RO-7 instrument from the Eberline Instrument Corporation to use in a study of the beta dose rates from the Godiva IV Critical Assembly at Los Alamos. We have performed a limited evaluation of the instrument and have determined its energy dependence for beta particles.

D. Hankins, *Evaluation of Neutron Dose Received by Personnel at the LLNL*, Lawrence Livermore National Laboratory, Livermore, CA, UCID-19385 (1982).

This report was prepared to document the techniques being used to evaluate the neutron exposures received by personnel at LLNL. Two types of evaluations are discussed, covering the use of the routine personnel dosimeter and of the albedo neutron dosimeter. Included in the report are field survey results which were used to determine the calibration factors being applied to the dosimeter readings. Calibration procedures are discussed and recommendations are made on calibration and evaluation procedures.

D. Hankins, "Neutron and Gamma-Ray Dose Measurements at the Los Alamos Critical Assembly—SHEBA," presented at ANS meeting, Nov. 1981, San Francisco, CA; *Nuclear News* 24, No. 12 (1981). (Abstract only.)

D. Hankins, *Personnel Neutron Dosimetry*, presented at INPO Radiological Protection Seminar, April 5, 1982, Houston, TX (Institute of Nuclear Power Operations, 1820 Water Place, Atlanta, GA); Lawrence Livermore National Laboratory, Livermore, CA, UCRL-88131 (1982).

D. Hankins, "Personnel Neutron Dosimetry Studies at the Lawrence Livermore National Laboratory," *Eighth DOE Workshop on Personnel Neutron Dosimetry*, June 18-19, 1981, Louisville, KY, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-86170 (1981); Battelle Pacific Northwest Laboratories, Richland, WA, PNL-SA-9950 (1981).

M. W. Nathans, *Duties and Responsibilities of Employers and Employees under OSH Acts and the Industrial Hygienist*, presented at Annual American Industrial Hygiene Conference, Cincinnati, OH, June 6-11, 1982.

The Occupational Health and Safety Act imposes, inter alia, upon employers the duty to comply with standards promulgated under the Act, and to furnish employment and workplaces free from recognized hazards that may cause death or serious harm to employees (29 U. S. C. 654). Employees have a duty to follow the law and the rules as they apply to them. Similar clauses are found in many state statutes.

Court and OSHRA decisions concerning these "duty clauses" affect the mutual relationship between an employer and the industrial hygienist or safety engineer, because they indicate the role of these professionals in the statutory scheme.

The primary issues of interest are the meaning of "recognized hazard" in the general duty clause, and what is required of an employer to enable him to avoid or defend a citation successfully. Following the standard of knowledge in the industry is not necessarily sufficient; an employer maybe held to a higher standard. An employer must discover and eliminate feasibly preventable hazards. The standard is the precautions a conscientious safety expert would take. These include the prevention of *foreseeable* employee misconduct by adequate safety training and enforcement of the work rules.

We have found no case law dealing specifically with an employer's remedies against an expert who makes a mistake which results in the employer's being cited. Such remedies would depend on whether the expert is an independent contractor or an employee. If the latter, the employer has probably only one recourse: fire the "culprit." If the former, an action for negligence may be taken, in which the employer may seek to recover all his provable damages approximately (i.e., legally) caused by the substandard performance of his consultant. The employer would have to prove, however, that the expert has failed to meet the standard of expertise that prevails in the community. An action on the contract for breach of warranty may be taken only if the contract contains an express warranty.

C. T. Prevo, Neutron and Gamma Radiation Transport Problems Solved Using TARTNP and MORSE Codes, presented at Ninth DOE Workshop on Personnel Dosimetry; Lawrence Livermore National Laboratory, Livermore, CA, CONF-820668 (1982).

TARTNP and MORSE are Monte Carlo codes currently used at LLNL to solve neutron and gamma radiation transport problems in Health Physics. They are useful in a personnel neutron

dosimetry program for the calculation of dose rates, spectra, and detector responses for various neutron source and detector configurations. Examples include the response function of the PNR-4 remmeter and spectra from various sources and moderators in the LLNL calibration facility.

R. A. da Roza, C. A. Cadena-Fix, G. J. Carlson, K. E. Hardis, and B. J. Held, *Reproducibility of Respirator Fit as Measured by Quantitative Fitting Tests*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-87244 (1982).

The Occupational Safety and Health Administration (OSHA) is promulgating new health standards in which *quantitative* respirator fitting is required. However, there has been controversy and confusion in the past few years regarding respirator fitting methods and their effectiveness. Members of the LLNL's Hazards Control Department employ a quantitative chamber to fit facepieces to users of respiratory protective devices. We are reporting the results of a study to determine the variability in protection factors obtained for multiple facepiece fittings, with particular emphasis on the protection fail/pass limits.

On the basis of this work, the following statistical statements can be made regarding the protection that would be afforded by a facepiece properly fitted to the user:

- The probability density function for the fit obtained by one individual wearer with a given facepiece is well described by a log-normal distribution.

- The geometric standard deviation for the above distribution varies widely among individuals and facepieces, making it necessary to measure it for each wearer/facepiece combination.

- A risk-plot method can be used to calculate the exposure risk to a wearer.

J. W. Stengel, *Environmental Testing of Escape Breathing Apparatus*, presented at Twelfth Intersocietal Conference on Environmental Systems, San Diego, CA, July 19-21, 1982; Lawrence Livermore National Laboratory, Livermore, CA, UCRL-87580 (1982).