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Foreword

The *Annual Technology Review* highlights some of the research performed in the LLNL Hazards Control Department from October 1986 to September 1987. The sections in this *Annual* report cover scientific concerns in the areas of Health Physics, Industrial Hygiene, Industrial Safety, Aerosol Science, Resource Management, Dosimetry and Radiation Physics, Criticality Safety, and Fire Science. For a broader overview of the types of work performed in the Hazards Control Department, we have also compiled a selection of abstracts of recent publications by Hazards Control employees.

Readers interested in more detail on any of the research described in these articles may contact the authors directly.

Summary of the Bikini Atoll Ionizing Radiation Survey

K. L. Shingleton, J. L. Cate, M. G. Trent, and W. L. Robison

Abstract

The survey was designed to measure the beta dose rate from ^{137}Cs and $^{90}\text{Sr}/^{90}\text{Y}$ relative to previously measured gamma dose rates on Bikini and Eneu Islands. We exposed modified Panasonic-802 thermoluminescent dosimeters (TLDs) in over 100 sites for six months to accomplish this task. The sites were selected to be either in areas of known high gamma dose rates, in areas where the Marshallese would likely spend most of their time upon resettlement, or in areas where experimental environmental changes had been made. Therefore, the beta and gamma dose rates *do not* represent island averages. The mean beta dose rate on Eneu ranged from 23 mrem/yr at 1 cm to 6 mrem/yr at 100 cm, as compared with a mean deep dose rate of 17.5 mrem/yr. The mean beta dose rate around houses and in general areas on Bikini ranged from 425 mrem/yr at 1 cm to 178 mrem/yr at 100 cm, compared with a deep dose rate of about 154 mrem/yr. Because monitoring sites were specifically placed in the most contaminated areas of Bikini and Eneu, the unshielded beta dose rates reported provide an upper limit of radiation dose; actual doses received by the Bikinians would be reduced significantly by clothing, footwear, and ground cover such as crushed coral, which reduces the beta dose rate by 80–90%. The amount of time spent in houses and in the minimally contaminated areas around houses and the lagoon would further reduce the beta dose rates reported here.

Background

Between 1946 and 1958, the United States conducted 23 nuclear tests at the Bikini Atoll in the Marshall Islands. The single largest detonation was from the "Bravo" test, which resulted in extensive radioactive contamination of a number of islands in the atoll and prevented the timely resettlement of the native population. Since 1958, many studies have been conducted to assess cleanup options and the internal and external radiation doses the Bikinians would likely receive should they resettle the islands. These studies have included assessment of the external radiation dose rates from gamma rays; the extent of soil, water, and vegetation contamination; the effect of excavation, fertilization, and irrigation on plant uptake of radionuclides; and the lifestyles and eating habits of the Bikinians.^{1–6}

Although the external dose rates from gamma rays have been previously determined by aerial survey⁴ and a variety of ground measurement techniques,^{5,7} technical constraints limited the assessment of the external beta dose rates that result from the ^{137}Cs and $^{90}\text{Sr}/^{90}\text{Y}$ contamination on the islands. Now, because of the recent development of very thin thermoluminescent dosimeters (TLDs), these external beta dose rates can be measured.

We undertook this survey to determine the beta dose rates and the shallow dose rates (beta + gamma) on Bikini and Eneu islands, as well as to compare the dose rates at heights of 1, 50, and 100 cm. We also determined the effect of various ground covers (e.g., coral gravel and vegetation) on the beta and shallow dose rates.

Survey Summary

This survey was conducted in two 6-month phases, and results were based on data from 800 Panasonic-802 dosimeters. These dosimeters were distributed among 102 monitoring sites (used to determine the beta and gamma components of the radiation field), 11 beta spectrometer arrays (used to assess the maximum and average energy of the beta radiation), and 6 fade-study stations (used to assess environmentally induced fading of the Panasonic dosimeters). Table 1 shows the station distribution by island, phase, and type. Figures 1 and 2 show the station distribution on each island. At each station corroborating data were obtained with LLNL

TLD dosimeters, a Reuter-Stokes Pressurized Ionization Chamber, a Bicon Micro-R meter, and a NaI Field Instrument for the Detection of Low-Energy Radiation (FIDLER) detector associated with a Canberra multi-channel analyzer. The data were all in good agreement. A full report of the study, including raw and calculated data, can be obtained from the authors.⁸

Phase 1 dosimeters were placed in the field in May 1985 and retrieved in November of 1985. Phase 2 dosimeters were deployed in November 1985 and retrieved in May 1986.

Table 1. Distribution of dosimeter stations.

	Bikini			Eneu	
	Monitoring sites	Spectrometer arrays	Fade study	Monitoring sites	Spectrometer arrays
Phase 1	40	5	3	50	4
Phase 2	12	2	3

Figure 1. Distribution of monitoring stations on Bikini Island.

Figure 2. Distribution of monitoring stations on Eneu Island.

General Information

Each Panasonic-802 dosimeter contained four TLD elements, two that were $\text{Li}_2\text{B}_4\text{O}_7\text{:Cu}$ (E1 and E2) and two that were $\text{CaSO}_4\text{:Tm}$ (E3 and E4). Figure 3 illustrates the construction of the TLD. In this report, we will refer to $\text{Li}_2\text{B}_4\text{O}_7\text{:Cu}$ and $\text{CaSO}_4\text{:Tm}$ simply as 'Li' and 'Ca.'

As shown in Fig. 4, Panasonic dosimeters are normally exposed in a special holder that contains plastic and lead absorbers, thereby allowing a rough determination of energy and types of radiation. In this survey we removed half of the dosimeters from their special holders, thereby exposing each TLD element to the full spectrum of radiation.

Li and Ca TLDs have different characteristics. Li TLDs have a relatively flat energy response to beta particles and photons, but are subject to long-term, environmentally induced fading.⁹⁻¹² In addition, they are highly hygroscopic and adversely affected by moisture.

To determine the extent of Li fading, we exposed a number of dosimeters to ^{137}Cs sources. They were placed in several thermal environments and left for the duration of the field exposure. In all cases, the response of the Li and Ca TLDs was statistically equal (i.e., the dose on the Li chips did not fade), presumably because of the packaging configuration we used. However, we were not so successful in protecting the Li TLDs from moisture. Because of the rainy season, many dosimeters initially 1 cm off the ground became buried in damp soil or grasses, and many of the Li TLDs dissolved, erasing much of the measured doses. We eliminated these data.

Also, because Li produces relatively few light units per mrem received, we had some statistical problems when measuring doses in very low background areas.

Ca TLDs have a relatively flat energy response to beta particles and photons with energies greater than about 300 keV. However, these TLDs overrespond to low-energy photons by as much as a factor of 15, with the maximum overresponse occurring at energies less than 30 keV.¹³ However, Ca TLDs are not significantly affected by long-term fading or moderate exposures to moisture, and produce relatively large numbers of light units per mrem received.

To determine if we had to correct for a low-energy Ca overresponse, we compared the data from the Li and Ca in-holder TLDs, which are both covered with a plastic absorber that absorbs only 4% of 30-keV photons. The Ca data exceeded the Li data in only 3.9% of the cases, indicating that the low-energy photons made an insignificant contribution to the total doses measured and that a correction for Ca overresponse was not necessary. We also compared the out-of-holder Li and Ca data and, at the 99% confidence level, found them to be equal 85% of the time. The remaining 15% of the cases were evenly divided: 8% of the Li data were higher and 7% of the Li data were lower than the respective Ca data.

For several reasons, we chose to use only Ca data for the actual dose rate determination: Li TLDs were adversely affected by moisture, heat, and light; Li TLDs had limited accuracy at low doses; and the Ca TLDs showed no low-energy overresponse.

Figure 3. Construction of the Panasonic TLD element.

Figure 4. Packaging for in-holder Panasonic dosimeter.

Fielding the Survey

Transportation

All dosimeters were air freighted to Kwajalein and transported by ship to Bikini. The dosimeters were packed in a shielded box, which reduced the dose rate in Livermore from 8 μ R/h to 1 μ R/h.

When the shielded shipping container arrived in Kwajalein with Phase I dosimeters, it was inadvertently stored next to several tons of high-potassium fertilizer. A small fraction (0.118%) of naturally occurring potassium is radioactive ^{40}K , which emits a 1.460-MeV gamma ray. Though we could not accurately determine the

amount of time the shipping container and the fertilizer were stored together, it did not exceed two weeks.

Because of this possible exposure, we read and annealed the Phase I Panasonic dosimeters before using them in the field, thereby removing any dose from travel and from the high-potassium fertilizer. The LLNL dosimeters were not annealed because the necessary equipment was not available. In retrospect, it does not appear that annealing was necessary, since the fertilizer did not add a significant dose to the LLNL dosimeters, relative to the total doses measured.

Phase 2 dosimeters had an unremarkable trip to Bikini and were deployed in the field without erasure of transportation dose.

Monitoring Stations

Each monitoring station consisted of eight dosimeters: one out-of-holder and one in-holder Panasonic dosimeter each at heights of 1, 50, and 100 cm, and two LLNL dosimeters at 100 cm. The dosimeters were aligned so as not to shield each other from the ground. Figure 5 shows a typical dosimeter station.

Beta Spectrometer Arrays

Each beta spectrometer array consisted of five out-of-holder Panasonic dosimeters at heights of 1, 50, and 100 cm. At each level one dosimeter was left bare, while the other four were covered with aluminum absorbers so that the total absorber thicknesses were 14, 21, 48, 84, and 233 mg/cm², respectively.

Packaging of Dosimeters

All Panasonic dosimeters were heat-sealed in Saran Wrap bags that were lined with aluminized mylar. The Saran bag (2 mg/cm²) provided moisture protection; the reflective aluminized mylar (1 mg/cm²) minimized heat buildup in the bag. As shown in Fig. 6, bagged Panasonic dosimeters were placed between aluminum supports that were stapled to wood blocks. A layer of plastic tape was placed over the top and sides of the dosimeters to protect them from direct rain and sunlight.

Site Selection

Monitoring sites on Bikini and Eneu Islands were selected on the basis of anticipated Marshallese lifestyle with emphasis placed on areas where people would likely spend the most time. For example, as can be seen on Figs. 1 and 2, we placed proportionately more monitoring sites on the lagoon sides of the islands than on the ocean sides, reflecting the Marshallese preference for housing locations.

To establish an upper bound for radiation doses, we used information from previous EG&G overflight radiation surveys to select a number of sites in the most contaminated area of Bikini.⁴ To attempt assessment of the effect of ground cover on radiation levels, we included two adjacent sites in many of the areas monitored, one of which was cleared of plants and debris, and one of which was left uncleared.

The Marshallese often place a 5- to 10-cm-thick layer of coral gravel around their homes, and so we

made measurements over two such areas for comparison with dose rates measured 10–30 m away in backyard areas. We anticipated difficulties comparing the beta data from these sites, since the gamma dose rates varied significantly between them. During Phase 2, we placed a 1-m-radius pad of coral gravel in two highly contaminated areas on Bikini and put a monitoring station in the center of each. Nearby, in an area with the same measured gamma dose rate, we established stations over cleared and uncleared soil.

We also placed considerable emphasis on the Excavation Plot—an experimental garden established in the most-contaminated area of Bikini. All plants and the top 40 cm of soil had been removed from this two-acre plot, where different crops were then fertilized and grown. The Control Plot, equal in size and adjacent to the Excavation Plot, was also stripped of plants and used as an experimental garden, but the topsoil was left essentially undisturbed. A 90-ft-wide Buffer Zone, left in its natural condition, separated the Excavation and Control Plots.

When evaluating data from this survey, especially the mean and median doses, it is important to remember that these values *do not* reflect the mean and median doses for the whole island, since we did not evenly distribute dosimeters across the islands.

Environmental Effects on Monitoring Stations

The typical rainy season on Bikini lasts from June through November. Though we were fairly successful in protecting the dosimeters from rainfall, we were unable to control the plant growth that occurred around the monitoring stations. When we returned to the islands in November, we found a number of sites on Eneu where morning glory vines had completely engulfed the PVC pipe holding the dosimeters. In a few other areas, grasses that had previously been fairly sparse now grew thickly up to the 50-cm monitoring height. This excessive plant growth often provided a continuously damp environment at the 1-cm level, and in many cases the Saran/mylar bags were not capable of sufficiently protecting the Li TLDs.

An interesting phenomenon not associated with the rainy season was the response of the aluminized mylar to prolonged field exposure. In areas that received little reflected light (e.g., in houses or over grassy areas), the Saran/mylar bags appeared unaffected by the six-month exposure. However, in sandy areas exposed to much reflected light, the aluminum often disappeared from portions of the mylar. Locations

Figure 5. A typical dosimeter monitoring station.

with moderate amounts of reflected light produced gradations of effects, ranging from slight mottling of the aluminum to a homogeneous, hazy transparency. All of the dosimeters in which the Li data was statistically less

than the Ca data (i.e., where the Li had apparently faded) were found in bags where the mylar had been affected. We concluded that, when it remained intact, the mylar prevented the Li from fading.

Data Analysis

Data Correction

We made a number of corrections to the raw data before calculating the dose rates. These corrections included:

(1) Application of Element Correction Factors, which were used to normalize the response of each Panasonic element so that all elements gave the same result for a given dose of ^{137}Cs radiation.

(2) A correction for contaminated Panasonic holders, because we found that about 1/3 of the dosimeter holders were made with newly refined lead containing ^{210}Pb , a radioactive daughter product of ^{226}Ra .

(3) A correction for dosimeters exposed out of the holders, because these dosimeters did not receive the scattered radiation from the lead covering E4, which the calibration process includes.

Figure 6. The dosimeter holding device constructed for this study.

We made no correction for transportation dose, because the control dosimeters (which were exposed to 300 mrem before being taken to Bikini and then left in the shielded box during the exposure period) read 309 ± 14 mrem, indicating that the transportation dose was insignificant. We also subtracted a cosmic background of $3.3 \mu\text{R/hr}$ from the data.⁷

Calculation of Deep-Dose Rates

Since beta and low-energy photon radiation do not significantly penetrate the lead and plastic filter covering E4, we used in-holder E4 data to assess the exposure rate in air from penetrating gamma radiation.

For risk estimates, the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR) recommends calculating actual doses to specific organs.¹⁴ Kerr, O'Brien, and Sanna have made extensive measurements converting exposures in air to doses in specific organs.^{15,16} We chose to use Kerr's conversion factor for the testes (0.75 rads in tissue/R in air, at 662 keV),¹⁵ since it provided a conservative estimate for almost all other organ doses, and because the dose to the testes had been reported in previous Bikini publications. Assuming 1 rem/ra $\dot{\text{o}}$, the final conversion of 0.75 rem in tissue/R in air agrees closely with UNSCEAR's value of 0.71 rem to the testes/R in air.¹⁴

Assessment of Beta Spectrometer Arrays

We normalized the data from each beta spectrometer array to the respective 14 mg/cm^2 absorber data, and then plotted the absorber thickness vs dose rate. We compared these curves to ones similarly generated with calibration sources of $^{90}\text{Sr}/^{90}\text{Y}$ ($\beta_{\text{max}} = 2.27 \text{ MeV}$) and ^{204}Tl ($\beta_{\text{max}} = 0.766 \text{ MeV}$). The calculated endpoint energy from the beta spectrometer arrays corresponded to ^{90}Y , but the curves generated with field data decreased faster than the ^{90}Y curve and slower than the ^{204}Tl curve. From this information, we concluded that the average energy of the beta spectrum was somewhere between that of ^{90}Y and ^{204}Tl .

As an additional check, we calculated the beta energy spectrum at the ground's surface using the Monte Carlo transport code SANDYL and a typical Bikini soil analysis.² The curves generated by the Monte Carlo code corroborate our interpretation.

Calculation of Beta Dose Rates

At a depth of 7 mg/cm^2 , the efficiency of the Ca TLDs to ^{90}Y was 85%, and to ^{204}Tl was 72%. Since it is very difficult to fine-tune the calibration beyond these limits, we averaged these values and used a calibration factor of 79%, yielding a calibration error that varied less than 10% from either endpoint.

In this survey, beta dose rates were determined by averaging the E3 and E4 out-of-holder data (which measures beta and gamma radiation), subtracting the corresponding E4 in-holder data (which measures

Calculation of Shallow Dose Rates

At 662 keV, the conversion factor for radiation dose to the skin ranges from 0.685 rad in tissue/R in air to 0.78 rad in tissue/R in air.^{15,16} As a conservative selection, we chose to use 0.75 rad in tissue/R in air, which was the same conversion factor used to convert exposure in air to dose in organs ($D(Or)$). Shallow dose rates were then calculated by adding the beta dose rate to the skin dose rate, which was numerically equal to the deep-dose rate [$D(Or)$] (i.e., $Shallow = \beta + D(Or)$).

Statistical Analysis

The precision of measurements using E3 and E4 was experimentally determined at doses of 25, 50, 100, and 300 mrem. After a total of 800 exposures, we found the standard deviation associated with E3 and E4 at all four dose levels to be $(6.6 \pm 0.5)\%$.

The accuracy of Panasonic measurements was evaluated through use of control dosimeters that, as previously mentioned, were exposed to 300 mrem before being taken to Bikini and were left on Kwajalein during the exposure period. Since these dosimeters and

those used on Bikini and Eneu were exposed to similar temperatures and humidities for a substantial portion of the exposure period, and the average dose reported at the end of the exposure period was 309 ± 14 mrem, we concluded that the measurements made with the Panasonic dosimeters were neither enhanced nor degraded as a result of the experimental exposure conditions.

On the basis of the precision and accuracy of the Panasonic dosimeters, we concluded that the total experimental error on our raw data was approximately $\pm 15\%$ at the 95% confidence level. Using this information, we propagated the errors to report the 95% confidence interval of our calculated dose rates. We assumed that the background value of $3.3 \mu R/h$ and the conversion of 0.75 rad in tissue/R in air were constants, and reported the errors as percents.

Minimum Detectable Beta Activity

Using a one-tailed Student's *t*-test, the minimum detectable beta dose was calculated to be 18% of the deep-dose rate. For example, if the deep-dose rate was $4 \mu R/h$, the minimum detectable beta dose was $0.72 \mu R/h$, corresponding to an annual beta dose rate of 6.3 mrem/yr. If the deep-dose rate was $50 \mu R/hr$ (438 mrem/yr), the minimum detectable beta dose was $9.0 \mu R/h$, or 79 mrem/yr.

Discussion of Results

The dose rates reported in the following discussions do not include natural background, and deep-dose rates refer to the effective dose equivalent¹⁷ (i.e., organ doses consistent with the UNSCEAR methodology of dose rate determination).¹⁴ However, these dose rates should not be used as an absolute indicator of potential personnel doses, since people do not remain in a single spot for extended periods, and doses on Bikini are received from both internal and external sources of radiation. To assess potential doses to people, such factors as the amount of time spent in various areas and the types and amounts of food consumed must be evaluated. Such assessments have been done, and reports on these topics are available.^{1, 35}

Dose Rates on Eneu and Bikini

A summary on the dose rates on Eneu is shown in Fig. 7.

Bikini's radiation profile is more complicated than Eneu's, since Bikini had many unique areas to be

evaluated. To clarify the discussion of dose rates, we divided the data obtained on Bikini into subgroups and calculated the highest, median, mean, and lowest dose rates for each subgroup, as shown in Figs. 8–12.

In Houses. We were surprised to detect significant beta radiation in two of the three houses surveyed, until we found out that the concrete used in some houses had been made from island aggregate (and was therefore contaminated), while concrete used in other houses had been made from uncontaminated coral reef aggregate. Dose rates in these areas are shown in Fig. 8.

Around Houses. This group constituted areas covered with coral gravel, side yards, and areas behind houses where children might play. See Fig. 9.

General Areas. This group comprised all sites that were not in houses, around houses, or associated with the Excavation Plot. This group does not reflect an island average, though, since we purposely selected a disproportionate number of sites in highly contaminated areas. See Fig. 10.

Figure 7. Dose rates on Eneu at 1 cm (left) and 100 cm (right).

Figure 8. Dose rates inside houses on Bikini at 1 cm (left) and 100 cm (right).

Figure 9. Dose rates around houses on Bikini at 1 cm (left) and 100 cm (right).

Figure 10. Dose rates in general areas on Bikini at 1 cm (left) and 100 cm (right).

Figure 11. Dose rates in the Control Plot at 1 cm (left) and 100 cm (right).

Figure 12. Dose rates in the Excavation Plot at 1 cm (left) and 100 cm (right).

Variation of Dose Rate with Height

The data from general areas on Bikini showed that at 1 cm, the mean beta dose rate was about 2.5 times the respective mean deep-dose rate; at 50 cm, it was 1.5 times the mean deep-dose rate; and at 100 cm, it approximately equaled the mean deep-dose rate. These data were valid for heavily contaminated areas, but not for lightly contaminated areas where the beta dose rates more closely paralleled the deep-dose rates at all heights.

Effect of Ground Cover on Beta Dose Rates

Cleared vs Uncleared Areas. We made a significant effort to determine the effect of the natural plant growth on the beta dose rates. Since the gamma dose rates often varied greatly between the cleared and uncleared areas, we normalized the mean beta dose rate to the respective mean deep-dose rate. After normalization the beta dose rates in the cleared and uncleared areas were within two standard deviations of each other. However, when individual sets of sites were compared, some cleared areas had reduced beta dose rates relative to uncleared areas, while others had increased beta dose rates. Unfortunately, the large variations in the beta dose rate that existed within small geographical areas overwhelmed the small botanical differences we were trying to measure.

Coral Gravel Ground Cover. The Marshallese traditionally place a 5- to 10-cm thick pad of coral gravel around their houses. During Phase 1 of this study, we made measurements over such areas, and also over other areas around houses. Since the gamma dose rates varied significantly between these areas, we normalized the 1-cm-height beta and shallow dose rates to the 1-cm-height deep-dose rate. After normalization, the coral gravel reduced the beta dose rate by 29–50%, and the shallow dose rate by 20–32%.

During Phase 2, we eliminated the effects of local dose-rate variations by placing coral pads in two highly contaminated areas of Bikini. We compared the data from these sites to adjacent areas not covered with the gravel and found that the coral provides an effective absorber for beta radiation. In one area the 1-cm beta dose rate was reduced by 89%, from 1015 to 110 mrem/yr; in the other area, the 1-cm beta dose rate was reduced by 77%, from 346 to 79 mrem/yr. The shallow dose rate was reduced from 1488 to 280 mrem/yr (81%) in the most contaminated area, and from 598 to 164 mrem/yr (73%) in the other area. The 1-cm deep-dose rates were also reduced by about 65%, from 37 to 13 mrem/yr in one case, and from 29 to 10 mrem/yr in the other.

Conclusions

The purpose of this study was to assess the external beta dose rates relative to the gamma dose rates on Bikini and Eneu Islands. We have made no attempt in this report to evaluate the consequences of the measured dose rates, or to make any recommendations

relative to cleanup or resettlement options. These matters can only be responsibly addressed by considering many factors, only one of which is the external beta and gamma dose rates.

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INDUSTRIAL HYGIENE

Mutagenic Soot in Ballistic Ranges

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Abstract

We checked soot deposits from burning double-base ballistic propellants to see if we could detect mutagenic activity. Using the Ames *Salmonella* assay, we tested three ballistic ranges and found that these sooty deposits are mutagenic. The evidence suggests that unburned propellants do not contribute to the mutagenicity of the soot under our assay conditions. The majority of the mutagenic potency of the soot seems to be the result of aromatic amine(s) created when the propellants are burned in the low oxygen/pressurized environment of a gun breech. We also found that the soot in the barrel of a pistol, rifle, or shotgun was mutagenic.

Introduction

It has been known for some time that organic materials can form carcinogenic or mutagenic substances when they are burned. In one of the first epidemiological studies of occupational disease, Percival Potts found that scrotal cancer was an occupational disease among chimney sweeps.¹ Automobile exhaust and smoke from burning leaves have been shown to contain carcinogens, and well-cooked hamburger contains potent aromatic amine mutagens.²

With this background we felt it reasonable to believe that the soot in ballistic ranges might contain mutagens. The Ames *Salmonella* test was used to see if mutagens had been deposited on surfaces inside ballistic ranges at LLNL.

Ballistic ranges are essentially enclosed facilities in which guns fire projectiles into targets. Both single-stage and two-stage guns are used at LLNL. Single-stage guns are little different from conventional artillery, except that the bores are not rifled. Projectiles can be accelerated to several thousand feet per second. Two-stage guns use a smooth-bore artillery piece—the first stage—to fire a piston into the first stage's barrel, which is filled with a suitable gas (hydrogen, helium, or nitrogen). The piston compresses the gas until the pressure exceeds the yield strength of a diaphragm on the opposite end of the barrel. The diaphragm bursts and gas enters the second stage, a smaller bore gun. The speed of sound is higher

in hydrogen and helium than it is in the gases evolved by the burning propellant so the gas escaping into the smaller gun moves at high speeds to accelerate a smaller mass down the smaller gun. When driven by hydrogen, the smaller mass can attain velocities of a few tens of thousands of feet per second (velocities similar to those achieved by reentry vehicles and satellites). The projectile enters a target tank that is fitted with instruments to measure projectile speeds and conduct other tests as needed. A catch tank or area is located behind the target tank to slow down and stop the projectiles, using a series of metal baffle plates. These baffle plates are hung in the projectile's path to keep the back of the range from being destroyed by projectiles. We tested the soot deposits in the target tanks of three ranges, two single-stage ranges (Fig. 1) and a two-stage range (Fig. 2).

We also tested the mutagenicity of several materials used to clean the guns, namely "GI bore cleaner" (a gun cleaner from surplus government stocks) and WD-40, a well-known silicon lubricant. The results of these tests were needed to assure that the mutagenic activity found in the soot was not caused by the materials used to clean the guns. No manufacturer or Material Safety Data Sheet (MSDS) could be found for "GI bore cleaner." Fortunately, the mutagenicity of WD-40 (and "Cool Tool" cutting fluid) had already been evaluated at LLNL in 1985 as part of a routine Industrial Hygiene survey in a machine shop.

Figure 1. A one-stage gas gun is basically a smooth-bore artillery piece firing a projectile into a target located inside an instrumented vacuum vessel ("target tank"). The baffles in the "catch tank" keep the remnants of the projectile and target from destroying the catch tank. [Not to scale.]

Methods

All mutagenicity tests were performed using the standard Ames *Salmonella* assay at the Ames Lab in LLNL's Biomedical and Environmental Research Program. They were assayed using the standard Ames test with TA98 and TA100 strains of *Salmonella typhimurium*, which are susceptible to frameshift and base-pair mutations, respectively. (One or more DNA

base pairs are added to or deleted from a DNA sequence in frameshift mutations, while only one pair of bases in a DNA sequence is affected in base-pair mutations.) The Ames tests were performed with and without metabolic activation (in which a metabolizing enzyme extract of rat liver is added to mimic the effect of mammalian metabolism on the material being tested).

Figure 2. A two-stage gas gun fires a heavy piston into a barrel filled with hydrogen, helium, or nitrogen. The piston compresses this gas, causing the pressure to rise until the diaphragm between the stages ruptures. The compressed gas enters the second stage and accelerates the smaller, lighter projectile into the target tank. The piston keeps the mutagenic soot out of the catch and target tanks. [Not to scale.]

Swipes were collected on 100 cm² portions of the surfaces of interest using dry filter paper swipe tabs. The tabs were placed in glass vials with Teflon or aluminum cap liners and treated with 1 ml of dimethyl sulfoxide (DMSO) to leach the sampled material out of the paper.

After each shot the guns at the ballistic ranges are routinely swabbed with cotton cloths that are soaked with WD-40 or isopropyl alcohol and wrapped around a wire brush. The wire brush is attached to a drill via lengths of a shaft. This is used to run the cotton cloth through the length of the barrel to swab it. The barrel must be swabbed out several times before it is clean. Ballistic range personnel cooperated by saving the rags used to swab the guns of two of the ranges. These cloths were saved in separate plastic bags, and we extracted the post-firing residue with solvents for Ames testing. The range personnel also recorded the names and amounts of propellant(s) used in each case.

Bulk samples of GI bore cleaner, Hercules HPC-95 propellant, soot from the breech and catch tank ends of the four-inch gun range, WD-40, and Cool Tool were collected in glass vials with Teflon or aluminum cap liners and taken to the Ames Lab. The bulk samples of HPC-95 and soot from the breech and catch tank ends of the four-inch gun range were subjected to serial dilutions in DMSO to determine the mutagenic potency of the powder and the soots at each end of the range. Results

of the serial dilutions were statistically analyzed using a least-squares-fit test.

After it had become apparent that the soot in the ranges was mutagenic, we burned samples of various propellants in air. We hoped that burning small samples of propellant would be a much cheaper way to generate soot for mutagenicity testing than by actually conducting shots in ballistic ranges. We also hoped that new propellants could be tested in this way before attempting to use them in the guns. We were able to collect sufficient ash to carry out serial dilutions.

Air samples had been collected and tested using the Ames *Salmonella* assay in the 1985 shop study mentioned above. The plume of smoke and mist created by the heating of the cutting fluid was collected on 5- μ m porosity PVC filters, while WD-40 and Cool Tool were used as cutting fluids for machining. The filters were placed in glass vials with Teflon-lined caps, treated with 1 ml of DMSO and taken to the Ames Lab along with bulk samples of the cutting fluids that were also tested using the Ames *Salmonella* assay. MSDSs for the products were also submitted to the Ames Lab.

In addition swabs were collected while patch cleaning a 0.22 caliber pistol and an over/under shotgun/rifle. The patches were soaked with DMSO and submitted to the Ames Lab.

Results

Results of the swipe samples are summarized in Table 1. The numbers are given in multiples of the background derived from a sample of plain DMSO. The swab samples were collected in the four-inch gun after 2140 g of Hercules HPC-95 and 34 g of HP-870 had been burned, while the swab samples in the first stage of the two-stage gun were collected after 4100 g of HPC-95 had been burned. Table 2 gives results of tests from the bulk samples, Table 3 gives results of the ashing of propellants in air, and Table 4 gives the results of the patch cleaning of two small arms.

Compositions of the propellants tested are listed in Table 5. HPC-95 contains nitroglycerin, nitrocellulose, potassium nitrate, graphite, and diphenylamine.¹ Nitroglycerin is mutagenic in the Ames test; *in vivo* cancer assays in rats required doses in the extreme grams or even kilograms per kilogram of body mass range.⁴ The summary of nitrocellulose in the Registry of Toxic Effects of Chemical Substances (RTECS) had no data about its mutagenicity.⁴ Diphenylamine is widely used to stabilize propellants by chemically combining with NO₂ that is slowly generated by the decomposition of the nitrocellulose or nitroglycerine in the propellant before

the corrosive NO₂ can attack the propellant. RTECS does not list the results of any mutagenicity assay for diphenylamine even though it is an aromatic amine. Potassium nitrate is mutagenic in *Escherichia coli* bacteria, but at doses in the five parts per hundred range,⁴ which is so high that results cannot readily be related to human exposure conditions. RTECS contained no mutagenicity data for graphite.

M-6 propellant was tested by burning it in air. M-6 contains nitrocellulose, dinitrotoluene, dibutyl phthalate, diphenylamine, and residual traces of ethyl alcohol and/or water.⁵ Dinitrotoluene (DNT) is a blend of isomers with the "2,4" isomer being the most studied. According to RTECS, 2,4-DNT is mutagenic in the Ames and cultured hamster cell *in vitro* assays at doses in the micro- and milligram per plate range, as well as in mice and rats at doses in the mg/kg of body mass range.² These are doses that can be related to human exposure conditions.

H870 is repacked Olin-Winchester WC870,⁶ which contains nitrocellulose, nitroglycerine, diphenylamine, dibutyl phthalate, tin dioxide, and potassium.⁷ RTECS has no data for tin dioxide, but inorganic tin is not a probable mutagen.

Table 1. Swipe sample results.

Sample location	Date	Enzyme activation	Results (counts $\times$ background)	
			TA99	TA100
Soot in breech end of 4" gun	8/4/87	+	29.7	3.6
	8/4/87	-	2.7	neg ^a
Soot on lower jamb of access door to target tank of 4" gun	8/4/87	+	7.7	neg ^a
		-	2.3	neg ^a
Non-sooty dust in target of 1/4 scale gun	8/4/87	+	neg ^a	neg ^a
			neg ^a	neg ^a
Soot along bore of 4" gun	9/1/87	+	29.1 & 66.7	2.6 & 5
first swab		-	neg & neg ^a	neg & neg ^a
second swab ^b	9/1/87	-	22.8 & 23.5	neg ^a & 4.4
		-	neg & neg ^a	neg & neg ^a
third swab ^b	9/1/87	+		
14.8 & 16	2.4 & 1.6			
		-	neg & neg ^a	neg & neg ^a
fourth swab ^b	9/1/87	+	4.1 & 4.8	neg & neg ^a
		-	neg & neg ^a	neg & neg ^a
fifth swab ^b	9/1/87	+	6.1 & 11	neg & neg ^a
		-	neg & neg ^a	neg & neg ^a
sixth swab ^b	9/1/87	+	2.5 & 3	neg & neg ^a
		-	neg & neg ^a	neg & neg ^a
Soot along bore of 2-stage gun	9/15/87	+	neg ^a	neg ^a
first swab		-	neg ^a	neg ^a
second swab	9/15/87	+	neg ^a	neg ^a
		-	neg ^a	neg ^a
third (final) swab	9/15/87	+	2.2	neg ^a
		-	2.2	neg ^a
Dust from floor of target chamber of 2-stage gun	9/15/87	+	neg ^a	neg ^a
		-	neg ^a	neg ^a
Soot from muzzle of 4" gun (in target tank)	10/16/87	+	10.5	—
Soot at forward end of baffle plate cart of 4" gun	10/16/87	+	3.4	—
Soot at rear end of target tank of 4" gun	10/16/87	+	1.7	—
Soot at bottom of catch tank of 4" gun	10/16/87	+	35.5	—
Three baffle plates removed from catch tank of 4" gun	10/16/87	+	5-22.2	—

^aThe mutagenic activity in the sample was not statistically significant (less than twice the background level).

^bTwo bundles of rags were submitted for each swabbing.

^cSwipes were collected from three sooty baffle plates.

Table 2. Bulk sample results.

Sample location	Date	Results (counts $\times$ background)	
		TA98	TA100
"G" bore cleaner	9/16/87	neg ^a	neg ^a
WD-40	1985	neg ^a	neg ^a
Rapid tap	1985	neg ^a	neg ^a
Pellet of HPC-95 (unburned)	11/3/87	neg ^a	neg ^a
Soot from breech end of 4" gun ^b	"	26,114 rev/mg	6496 rev/mg
Soot from catchtank end of 4" gun ^b	"	3676 rev/mg	1494 rev/mg

^aThe mutagenic activity in the sample was not statistically significant (less than twice the background level).

^bA sufficiently large sample was collected to allow weighing and quantitative evaluation of mutagenic potency. The amount of potency was less than twice that of unadulterated DMSO used for comparison. A sufficiently large sample was collected to allow weighing and quantitative evaluation of mutagenic potency. Units: revertants per milligram of sample.

Table 3. Results of ashing propellants in air.

Propellant	Revertants/mg of burned powder, TA98	
	With activation	Without activation
M-6	1,574	3,846
H-870	895	1,476
HPC-95	1,326	3,461
Shrink tube	8,692	16,597

Table 4. Results of small arms patches.

Source	Revertants/in ² of cleaning patch, TA98	
	With activation	Without activation
0.22 pistol, 1st cleaning	289	neg ^a
0.22 pistol, 2nd cleaning	238	neg ^a
0.22 rifle, 1st cleaning	1494	823
0.22 rifle, 2nd cleaning	1174	neg ^a
12 gauge shotgun, 1st cleaning	2375	2036
12 gauge shotgun, 2nd cleaning	1034	1006

^aThe mutagenic activity of this sample was not statistically significant.

WD-40 is a blend of Stoddard solvent and a more volatile petroleum hydrocarbon blend along with proprietary additives.* Cool Tool contains 1,1,1-trichloroethane (methyl chloroform) and mineral oil, plus proprietary materials.* Methyl chloroform has been found to be mutagenic.*

Ballistic range personnel helped us determine the previously unknown composition of "GI bore cleaner." Mixtures of methylene chloride and phenolic compounds or cresols are used as cold carbon removers and for cleaning aircraft and automobile engine parts. Air sampling disclosed that methylene chloride was present (but the vapor levels were below current and proposed exposure criteria). Gas chromatography/mass

spectrographic analysis of bulk samples disclosed the presence of higher aliphatic hydrocarbons and Stoddard solvent. One of the range operators identified the unpleasant aroma of GI bore cleaner as resulting from coal tars. Another bulk sample was collected and substituted phenolics, indicative of coal tars, were found when it was analyzed. Results of these analyses are summarized in Table 6.

No mutagenic activity was found in bulk samples of GI bore cleaner, WD-40, or Cool Tool, nor in air samples collected while WD-40 and Cool Tool were used as cutting fluids. The mutagenic activity in the air samples of WD-40 and Cool Tool were found to be below 1.5 times the background levels.

Table 5. Propellant compositions.

Propellant	Ingredient	Percentage*
M-6	Nitrocellulose	87
	Dinitrotoluene	10
	Dibutyl phthalate	3
	Diphenylamine	1
	Residual ethyl alcohol	<0.9
	Residual water	<0.5
HPC-95	Nitrocellulose	Balance
	Nitroglycerine	19-21
	Graphite	0.4
	Diphenylamine	0.5-1
	Potassium nitrate	0.5-1.5
	Residual volatiles	<1.0
H-870/ WC-870	Nitrocellulose	Balance
	Nitroglycerine	10
	Residual dinitrotoluene	0.2
	Graphite	0.2
	Dibutyl phthalate	5-5.5
	Diphenylamine	1
	Potassium nitrate	0.8
	Tin dioxide	0.7

* Percentages are approximate.

Table 6. GI bore cleaner results

Sample type	Analyte	Results	Comments
Air	Stoddard solvent	67.5 mg/m ³	
	Methylene chloride	58.1 mg/m ³	
Bulk	Decane	6 % vol	From Stoddard solvent
	Undecane	11 % vol	"
	Dodecane	12 % vol	"
	Tridecane	11 % vol	"
	Tetradecane	11 % vol	"
	Pentadecane	10 % vol	"
	Hexadecane	6 % vol	"
	Heptadecane	4 % vol	"
	Naphthalene	0.15 % vol	Polycyclic hydrocarbon
	2-Methylnaphthalene	0.14 % vol	"
	Fluorene	0.009 % vol	"
	Phenanthrene	0.009 % vol	"
	Phenol	0.26 % vol	From coal tar
	2-Methylphenol	0.26 % vol	"
	4-Methylphenol	0.46 % vol	"
	2,4-Dimethylphenol	0.28 % vol	"

Discussion

Ase et al. studied soots created by firing an M-16 using Olin WC-844 nitrocellulose/nitroglycerine double-base propellant with diphenylamine and dibutyl phthalate additives, and a 105-mm howitzer using a nitrocellulose, nitroglycerine, and nitroguanidine triple-base propellant with ethyl celvolite stabilizer and cryolite additive.¹⁶ In each case they found a number of toxic compounds had been formed. Among these were polycyclic aromatic hydrocarbons, including well known materials such as benzo(a)pyrene and anthracene, as well as nitrobenzene, pyridines, nitriles, and dimethylnitrosamine.¹⁹ The mutagenic activity of the soot assessed in our study is much greater than one would expect from the materials reported by Ase et al.

Subjective observations of soot deposits made during worksite monitoring correlated with the positive mutagenicity test results. Soot was not evident in the target tanks of the two-stage or quarter-scale gun ranges where mutagenic activity was not found. Surfaces in the large, not and catch tanks of the four-inch gun range are, on the other hand, very sooty and considerably more mutagenic.

The quantity of powder used also appears to be related to the amount of mutagenicity. Less powder was burned in the two-stage gun than in the four-inch gun,

and the mutagenicity test results were consequently lower. Much smaller amounts of powder are burned in the quarter-scale gun, which partially accounts for the lack of sooty material and mutagenic activity in its catch tank. The soot is also largely confined to the first stage by the piston.

The data suggest that the mutagens from HPC-95 fired in nitrogen and especially in a vacuum are more active in TA98 than in TA100 bacteria—a characteristic of aromatic amines. This and the presence of nitrocellulose and nitroglycerin in the powder as sources of nitrogen, as well as the probable reducing atmosphere created by firing into a vacuum or inert gas that can convert nitrate to amines, all combine to suggest that the mutagens are aromatic amines. Our first results also showed that metabolic activation dramatically increased the mutagenic potency of the soot in the gun ranges. The mutagens in well-done hamburger, where amino acids and creatinine condense to form heterocyclic amines, may be an analogous case.²

Neat HPC-95 powder does not appear to cause the mutagenic activity in the soot, nitroglycerine notwithstanding, because the bulk sample of unburned HPC-95 had no mutagenic activity under our test conditions. In fact, our preliminary estimates suggest

that the mutagenic potency of the soot in the Ames *Salmonella* assay may be greater than that of any environmentally encountered complex mixture.

Ames results also suggest that the mutagens in the soot from the breeches of the four-inch and two-stage guns are different from those in the ash formed by simply burning the powder in air. Metabolic activation decreased the mutagenicity of the ash formed by burning HPC-95 and other powders in air, while metabolic activation enhances the mutagenicity of soot formed by burning

HPC-95 powder in the nitrogen or vacuum conditions of the gun.

It appears that the mutagens from burning HPC-95 in a vacuum could be readily isolated. (See Fig. 3 for a correlation of ultraviolet absorbance of fractions of the soot dissolved in DMSO, separated by high-performance liquid chromatography vs the mutagenic activity of the fractions.) Only three or four mutagenic fractions were isolated in the soot fractions, though hundreds of ultraviolet absorbing fractions were found.

Figure 3. Correlation of ultraviolet absorbance of fractions of the soot dissolved in DMSO vs the mutagenic activity of the fractions.

Conclusions

Our data shows that the soot formed by burning HPC-95 under pressure in the low-oxygen environment that exists in the bore of a gun fired into a vacuum or into nitrogen at a gun range is mutagenic. The mutagenicity is probably due to aromatic amines. The Ames assay has a 90-95% success rate in predicting the carcinogenic potency of aromatic amines,¹¹ so the possibility that the soot has carcinogenic potential must also be considered.

Upon the advice of Hazards Control the managers of the ranges chose to take conservative measures where mutagenic activity was found (throughout the four-inch range and in the breech and first stage of the two-stage range):

- Using coveralls and, when appropriate, gloves and foot covers to minimize skin contact with the soot.
- Using respirators to prevent inhalation of the soot when doing work that creates airborne soot.

- Limiting access to sooty areas and either packing or cleaning soot-contaminated articles before moving them to other places.

- Disposing of all soot-contaminated waste using hazardous waste procedures.

The soot in the barrels of small arms is mildly mutagenic and appears to be similar to that of large gun barrels. Metabolic activation increases the mutagenicity of the material. Our data suggests that hobbyists and soldiers handling small arms need to wear gloves while stripping and cleaning their weapons, or they should promptly clean their hands afterwards.

This study also demonstrates that the contribution of all combustion byproducts to mutagens in the environment warrants serious consideration.

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Using Smoke Candles and a Tracer Gas for Analyzing Exhaust Stack Performance

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Abstract

We used smoke candles and the tracer gas SF_6 for qualitative and quantitative tests, respectively, to analyze exhaust stack performance in a chemistry laboratory building surrounded by complex topography. We found that smoke candles are easy to use and useful for documenting exhaust reentrainment problems. Smoke candles can also be used for predicting stack performance in the design stage and for verifying stack performance visually. The use of a tracer gas is more difficult as it requires more equipment for sample collection and involves extensive coordination; detailed meteorological data will also assist sample collection efforts. The methodology used and the relative merits of each type of tests are discussed.

Introduction

Many toxic chemicals are used in research laboratories. To minimize and eliminate exposure of toxic chemicals to employees, laboratory hoods (local exhaust ventilation systems) are designed and used to contain and capture the contaminants. In many cases these contaminants are routed through air cleaners such as chemical scrubbers or filter systems for cleaning before being discharged through the stack and into the atmosphere.

Although the use of air cleaners is always advisable, technical and economic reasons often preclude their use. Most scrubbers and air cleaners are effective only for the particular types of contaminant and release rates for which they are designed. In a research facility where many chemicals are used, appropriate air-cleaner and scrubber technologies may not exist or may be too complex or too costly to use. In such instances, tall exhaust stacks are used for releasing the contaminants to be dispersed and diluted by the atmosphere.

Theoretical and empirical methods are available for predicting the dilution characteristics of a tall stack for a given wind condition and direction. However, these

methods generally apply to a simple case, such as a single low building on level terrain with no surrounding obstructions. Some methods can be used for predicting concentrations at a certain minimum distance from the stack; others at locations more immediate to the stack. Factors such as complex topography near a building often increase the uncertainty level of the empirical methods.

We undertook a series of qualitative and quantitative measurements that could be compared with theoretical predictions of potential release conditions to evaluate whether a tall stack of a building is working as designed. The objectives of our present tests were to:

- Use smoke candles as a relatively cheap and easy method for the verification of exhaust stack-height design basic criteria.
- Use a tracer gas to quantitatively verify the preliminary and qualitative results of the smoke candle tests.
- Compare these results to the theoretical results predicted by the Wilson Formula, as described in the next paper.

Background

Building 222 is a chemistry laboratory building oriented north to south with three wings spreading east to west. The laboratory in which experiments with toxic chemicals are conducted is located near the SW corner of the south wing. A seven-story LLNL administrative building (B-111) is located about 500 ft WSW of B-222. A bank of tall trees (40-45 ft high) stands to the west of B-222's south wing (Fig. 1).

The prevailing winds blowing from the southwest are disturbed first by B-111 and then by the bank of trees. Wind can either pass over the top of the trees or through them, depending on the wind velocity and location. The

end result is that a recirculation cavity is created on the leeward side of the trees. The created eddies can hold building exhausts on the roof and cause reentry. In order to avoid or at least minimize reentrainment and the problems related to toxic chemical exposures downwind from the building, the Hazards Control and Plant Engineering Departments were asked to study the problem and make recommendations on the proper height of the exhaust stack. After the new stack was designed and built, tracer gas tests were also performed to ascertain that the dispersion characteristics of the tall stack are within the design parameters.

Qualitative Smoke Candle Testing Procedures

Smoke candles are convenient and widely used for qualitative analysis of ventilation systems and stack-dispersion patterns. The active ingredients of most smoke candles are sucrose, zinc oxide, ammonium perchlorate, and hexachloroethane. When lit, these ingredients react to form a light smoke (composed mostly of zinc chloride, water, and carbon dioxide). Thus, smoke candles can be useful for tracing a plume of smoke as it exits from the stack under study. Other by-products of the reaction may be carbon monoxide and

hydrogen chloride, which are toxic and corrosive and may be health hazards if present in a high enough concentration.

Four different sizes of smoke candles can be purchased to give 0.5, 1, 3, or 5 min of smoke. Candles are normally supplied with a fuse on each end so they can be tied together to provide a continuous supply of smoke. Colored candles are available in purple, red, orange, yellow, green and blue, which will show up well on photographs.

Figure 1. Building 222 and its surrounding area, looking west. Note the abundance of trees.

Smoke candles are relatively easy to use. Because the candles ignite at high temperatures, it is necessary to use a container or a candle holder for containment during combustion. The candles are usually placed inside a hood or a specially designed section of the exhaust system.

Smoke candles were used for the present experiment to qualitatively determine the dispersion characteristics of a laboratory building and its surrounding topography. The dispersion characteristics of the exhaust stack under a particular weather condition were characterized by observing and photographing the smoke released from a laboratory hood. This technique is particularly effective for detecting reentrainment from an exhaust stack that is not tall enough to reach into the free air stream.

Smoke Candle Test Results

Before the new exhaust stack of B-222 could be redesigned, it was necessary to determine the adequacy of the existing stack. A relatively simple and inexpensive test for this was to use smoke candles. We noted that smoke emerging from a low stack tended to be caught in a downdraft and reenter the building (Fig. 2). With assistance from the LLNL Fire Department and using a

fire truck ladder, we conducted additional smoke-candle tests at varying elevations above the roof to find a height from which reentrainment could be minimized.

A scaffolding was subsequently built and a modified exhaust stack was erected at approximately 50 feet above grade. Although the higher stack significantly improves the smoke-dispersion pattern, we observed a downdraft, evidenced by smoke concentrating at roof level.

Although the design method used in the 1985 *Fundamentals* volume of the *ASHRAE Handbook* recommended a stack height of 75 ft above the roof,² we chose a stack of 80 ft in the final design for the following reasons: According to Plant Engineering, the maximum stack height the building can support is 80 ft above the roof. Also, the uncertainties of the model as well as additional complexity introduced by upwind obstructions to airflow added a greater margin of error.

After these design criteria were established, further smoke candle testing was conducted with the aid of a tall construction crane and the smoke candle at 93 ft above grade, or about 80 ft above the roof level. These tests confirmed that at this height reentrainment problems were essentially eliminated.

Figure 2. In the smoke candle tests, we identified a downdraft that reentrained smoke to the building roof.

Following construction of the tall stack, we used qualitative smoke tests to verify that building recirculation problems had been resolved. However, we also needed quantitative test results to verify whether effluents from the tall stack are dispersed and diluted sufficiently to eliminate toxic exposures to people and buildings downwind from B-222.

Summary and Discussion of the Smoke Candle Technique

Smoke candles are useful for documenting existing exhaust stack performance, though the zinc chloride fumes produced are mildly toxic and should be treated with caution. Smoke candles should always be put in proper containers, and the smoke generated should be confined in an enclosed area such as a laboratory hood.

When properly used, smoke candles can give a great deal of information about stack performance. The plume of smoke can be photographed and recorded for later presentation. A visible plume of exhausted smoke can be very convincing to management when pleading a case for a properly designed stack.

Smoke candles can also be used to simulate exhaust performance at various heights. For example, an overhead crane can be used for releasing smoke at various heights. This is a relatively simple procedure for verifying design criteria.

Finally, smoke candles can be used to verify stack performance after the exhaust stack has been built. An entire experiment can be photographed and preserved as evidence that a stack is effective in dispersing toxic contaminants to the free air stream.

Quantitative Analysis Using Tracer Gas

The quantitative tracer gas test involves releasing a known concentration of SF_6 at a known rate through the stack. At various locations upwind and downwind from the stack, gas sample collection bags are placed and samples collected over a given period. Concentrations of SF_6 in these bags are analyzed to determine the dispersion characteristics of the stack for a particular wind direction and velocity.

We released SF_6 through a calibrated rotameter inside a laboratory hood at a rate of 5 cfm. The release duration for each test was 30 min, which was chosen

because we were limited by the 180 ft³ capacity of the gas bottle. The tracer gas was drawn into the exhaust duct inside the laboratory hood, mixed with 3500 ft³ of air and discharged through a 16-in. diameter duct into the stack (Fig. 3).

We chose SF_6 as the tracer gas because of its unique properties:²

- Its very high electron-capture response makes it readily detectable down to one part SF_6 per 10¹² parts air (1 ppt).

Figure 3. The tracer gas was introduced into a ventilation hood through a rotameter and exhausted to the outside.

- The typical natural background concentration of SF_6 gas is extremely low (0.1–0.5 ppt).

- It is nontoxic, non-allergenic, nonradioactive, colorless, and odorless.

- It is chemically inert and thermally stable for atmospheric applications.

- It can be rapidly and precisely released into the atmosphere from a point or area source.

- It is commercially available and can be collected and analyzed by conventional techniques.

Sampling Devices and Analytical Techniques

Samples were collected with 30 liter (30" x 30") Tedlar (polyvinylfluoride) bags and SKC Model HPS 113A pumps at a sampling rate of 1 lpm. The pumps were manually turned on 3–5 min after the tracer gas release and automatically shut off after sampling for a total volume of 20 liters. The samples were analyzed with a LMP-10 portable gas-chromatograph monitor with a 3000-mCi Ti_2 sealed-source electron-capture detector.

At 10 $\times$ sensitivity setting, the LMP-10 has a usable range of 20–2000 ppb with very little drift. It is also built ruggedly for field use and can withstand quite a bit of jarring and some direct sunlight. This instrument was also used as a real-time analyzer in the field. The LMP-10 is not a continuous sampler, however, as it is capable of

sampling only every two minutes with a 1-min sampling and a 1-min purging cycle.

Meteorological Data and Sampling Locations

In order to conduct the experiment and evaluate the results, it was necessary to obtain meteorological data pertaining to the location. Data from 1984–1986 on averages of annual percentage frequency of wind direction for LLNL are given in Tables 1–3 and shown in Fig. 4. These meteorological data were measured 40 m above ground and about 3/4 mile north of B-222. We used the statistics to select preferred test conditions and to predict worst-case dilution problems.

Because SF_6 is invisible and only a finite number of sampling bags are available for sample collection, the correct placement of sampling bags in strategic locations is essential. If the bags were placed in the wrong direction downwind or if the wind changes during the experiment, samples collected will not be representative of actual conditions.

For a given mean wind direction, we located sample collection points 11.25° apart downwind along four 45° arcs originating from the stack. Five sampling points were selected and located at each arc. Arc radii are 200, 300, 400, and 600 ft downwind from the stack. Of all possible sampling locations on the grid, we chose 20 for each test. In addition, samples were taken on the roof of

Table 1. Average annual percent frequency of wind direction vs wind speed (m/s) for 1984.

Direction	0–2	2–3	3–5	5–7	7–9	9–11	11–16	16–20	>20	Total	Avg. speed (m/s)
N	1.6	0.3	0.2	0	0	0	0	0	0	2.1	1.6
NNE	2.1	0.6	0.3	0	0	0	0	0	0	3.0	1.7
NE	3.3	0.7	0.1	0	0	0	0	0	0	4.1	1.5
ENE	2.8	0.8	0.1	0	0	0	0	0	0	3.7	1.5
E	1.5	0	0	0	0	0	0	0	0	1.5	1.0
ESE	1.8	0.1	0	0	0	0	0	0	0	1.9	1.1
SE	2.6	0.1	0	0	0	0	0	0	0	2.7	1.2
SSE	1.8	0	0.1	0	0	0	0	0	0	1.9	1.1
S	1.8	0.2	0.1	0	0	0	0	0	0	2.1	1.4
SSW	3.1	0.5	0.2	0	0	0	0	0	0	3.8	1.6
SW	6.3	3.1	1.8	0.2	0	0	0	0	0	11.4	2.1
WSW	10.2	6.6	3.2	0.2	0	0	0	0	0	20.2	2.1
W	9.2	3.3	1.2	0	0	0	0	0	0	14.1	1.8
WNW	5.4	2.5	0.6	0	0	0	0	0	0	8.5	1.8
NW	2.0	0.4	0.1	0	0	0	0	0	0	2.5	1.4
NNW	1.1	0.1	0.1	0	0	0	0	0	0	1.3	1.4
Calm										15.2	
Total	56.6	19.3	8.5	0.4	0	0	0	0	0	100.0	1.6

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Table 2. Average annual percent frequency of wind direction vs wind speed (m/s) for 1985.

Direction	0-2	2-3	3-5	5-7	7-9	9-11	11-16	16-20	>20	Total	Avg. speed (m/s)
N	1.6	0.5	0.8	0.7	0.4	0.2	0	0	0	4.2	3.9
NNE	1.4	0.7	1.4	0.7	0.3	0	0	0	0	4.5	3.4
NE	1.6	1.0	1.5	0.5	0	0	0	0	0	4.6	2.9
ENE	1.5	0.7	1.0	0.5	0.1	0	0	0	0	3.8	3.0
E	1.8	0.6	0.5	0.1	0	0	0	0	0	3.0	2.1
ESE	1.6	0.4	0.3	0.1	0	0	0	0	0	2.3	1.8
SE	1.4	0.3	0.3	0.1	0	0	0	0	0	2.1	1.9
SSE	1.1	0.2	0.1	0	0	0	0	0	0	1.4	1.7
S	1.7	0.4	0.4	0.1	0	0	0	0	0	2.6	2.5
SSW	2.4	1.0	1.3	0.2	0.1	0	0	0	0	5.1	2.6
SW	3.4	3.2	5.5	3.7	1.1	0.1	0	0	0	17.1	3.9
WSW	3.2	3.6	5.8	2.8	0.7	0.1	0	0	0	16.2	3.6
W	2.4	2.0	5.6	3.8	1.0	0	0	0	0	14.8	4.1
WNW	1.3	0.7	0.9	0.7	0.5	0.1	0	0	0	4.1	3.7
NW	1.3	0.3	0.1	0.1	0	0	0	0	0	1.8	2.0
NNW	1.3	0.3	0.3	0.1	0.1	0.1	0	0	0	2.1	2.5
Calm										10.4	
Total	29.0	15.7	25.8	14.1	4.2	0.7	0.1	0	0	100.0	3.1

Table 3. Average annual percent frequency of wind direction vs wind speed (m/s) for 1986.

Direction	0-2	2-3	3-5	5-7	7-9	9-11	11-16	16-20	>20	Total	Avg. speed (m/s)
N	1.5	0.5	0.8	0.8	0.4	0.2	0.1	0	0	4.4	4.0
NNE	1.5	0.5	1.2	0.4	0.2	0	0	0	0	4.0	3.1
NE	1.7	0.9	2.1	0.6	0	0	0	0	0	5.4	3.1
ENE	1.8	0.8	1.4	0.7	0.2	0	0	0	0	4.9	3.2
E	1.8	0.8	0.8	0.3	0	0	0	0	0	3.7	2.5
ESE	1.7	0.6	0.3	0.1	0	0	0	0	0	2.7	1.9
SE	1.3	0.3	0.2	0.1	0	0	0	0	0	1.9	2.2
SSE	1.4	0.2	0.1	0.1	0.1	0	0.1	0	0	2.0	2.7
S	2.1	0.4	0.2	0.1	0	0	0	0	0	2.9	2.0
SSW	2.6	1.1	0.9	0.2	0.2	0.3	0.1	0	0	5.3	3.1
SW	3.4	2.9	6.0	3.3	1.2	0.4	0.1	0	0	17.3	4.0
WSW	3.5	3.7	5.6	2.2	0.5	0.1	0	0	0	15.7	3.4
W	2.2	2.1	4.6	2.7	0.6	0	0	0	0	12.2	3.9
WNW	1.4	0.7	0.9	0.8	0.3	0	0	0	0	4.1	3.5
NW	1.3	0.3	0.2	0	0	0	0	0	0	2.0	1.9
NNW	1.4	0.4	0.4	0.2	0.2	0	0	0	0	2.5	2.8
Calm										9.0	
Total	30.9	16.1	25.8	12.7	3.9	1.2	0.4	0	0	100.0	3.1

B-222 at the base and 20 ft upwind of the stack. Figure 5 shows all possible sample locations for a range of test wind directions between 202.5–258.75°.

To avoid placing bags in an unfavorable downwind location, meteorological data are useful for predicting the probable wind direction. During this series of experiments, we released and collected tracer gas from 10:45–11:00 a.m. In the Livermore Valley early morning weather conditions are usually unstable because the air over the land mass and the San Francisco Bay heat up at different rates. By 11:00 a.m. the land is usually warmed up enough that wind direction is not likely to change drastically. During the months of May–July (when these experiments were conducted) the temperature is usually too hot by noon for sensitive analytical instruments to operate precisely. We decided that a good compromise was to initiate the tests about 11:00 a.m.

Typical Test Sequence

Collecting the 20 sampling bags requires coordination and a team of six persons to perform individual tasks: four bag placers (each responsible for five bags), one LMP-10 operator, and one SF₆ gas releaser. We followed the schedule listed below.

- 0800–1000 Follow tower MEDATA for wind direction and velocity.
- 0900–1000 Load equipment on truck at B-253 (Hazards Control) and unload at B-222.
- 1010 Use 1000 tower MEDATA for “GO” and wind direction decisions, then communicate to the test crew where sampling bags should be placed.
- 1015–1045 Place and tie-down sampling bags; locate LPM-10 and stabilize machine.
- 1045–1115 Release SF₆ at 5 cfm.
- 1050–1110 Turn pumps on and collect gas in sampling bags for 20 min.
- 1115–1135 Return sample bags and LMP-10 to truck.
- 1145–1200 Unload the equipment and sampling bags to B-253 lab.
- 1300–1600 Analyze tracer gas concentration in sample bags.

A total of five test runs were made. Pertinent test parameters are given in Table 4. The wind direction during test runs ranged from 221 to 264°, which is approximately 45% of the average annual percentage frequency. The wind speeds obtained during our runs included all recorded wind speed ranges on the LLNL site, as shown in Table 5. Quantitative results and comparisons to theoretical models are given in the following paper.

Figure 4. Directional wind patterns for 1984–1986.

Figure 5. Map of the area around Building 222, showing all possible sampling locations.

Table 4. Pertinent parameters of our five test runs.

Run #	Date	Time	Wind Direction (°)	Wind Speed (m/s)
1	7/7	10:55	264	2.1
2	7/8	10:30	261	1.5
3	7/10	10:30	248	6.0
4	7/14	10:25	264	1.6
5	7/15	10:32	221	3.2

Summary and Discussion of the Tracer Gas Technique

The use of SF_6 as a tracer gas in ventilation studies is an effective tool for quantitative analysis of the gas diffusion pattern and stack performance. The tracer gas released can be collected in sampling bags for later analysis.

In order to maximize sample collection efficiency, we placed 18 sampling bags downwind along four 45°

Table 5. Wind speed ranges recorded at LLNL compared with the wind speeds recorded during each of the five test runs.

Range (m/s)	Wind speed during test (m/s)
0-2	1.5, 1.6
2-3	2.1
3-5	3.2
5-7	6.90

arcs. The bags are 11.25° apart at arc radii of 200, 300, 400, and 600 ft downwind from the stack. A team of six experimenters placed the bags at precise locations, which had been previously marked. The experimenters remained in communication with each other through the use of portable radios. After the tracer gas had been released for five min to reach a stabilized condition, all the

sampling pumps were turned on simultaneously, collecting air samples for about 20 min. This process is labor intensive, and the use of remote-control pumps will no doubt lessen the work requirement.

The results of these experiments were compared with theoretical model predictions, as described in the following paper.

Conclusion

Smoke candles and tracer gas can be useful qualitative and quantitative techniques for the analysis of exhaust stack performance, especially in a complex topographic area such as around B-222.

Smoke candles are relatively easy to use. Our tests used smoke candles to document existing recirculation problems with the 9-ft exhaust stack in B-222, and also in conjunction with an overhead crane for a relatively quick simulation of stack performance at various heights. We

also used smoke candles to verify stack performance after a new exhaust stack had been built to its proper height.

SF₆ was used as a tracer gas for quantitative analysis of exhaust stack performance. With the aid of specialized analytical equipment such as the LMP-10 gas chromatograph, quantitative analysis of stack performance can detect the tracer gas in quantities that are only parts per billion.

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Measurements and Predictions of Effluent Dilution from a Tall Laboratory Stack

Ming K. Wong

Abstract

Actual field measurements of downwind ground-level effluent concentrations were compared with the concentrations predicted by two calculational methods—Gaussian plume dispersion and Wilson's design procedure. These are theoretical and empirical methods for the prediction of downwind effluent concentration from a tall exhaust stack. Generally, these methods apply to the relatively simple case of a single building without surrounding obstructions and the release point is above the recirculation region. The Gaussian plume dispersion method is applicable for predicting concentrations at distances greater than 100 m from the stack; Wilson's estimate is for predicting concentration on the roof or a short distance from the stack. We found that Gaussian plume calculations and Wilson's design procedure yield good agreement with experimental data. However, the minimum applicable distances should be considered when using the Gaussian model.

Introduction

Building 222 is a one-story chemistry laboratory building near the southwest corner of LLNL. Since this building engages in research using corrosive and reactive gases in conjunction with highly toxic materials, a catastrophic event has the possibility of occurring. If a vessel containing a reaction were to rupture, a significant amount of highly toxic material could be released through the stack. The Hazards Control, Chemistry, and Plant Engineering Departments share the concern that toxic gases may be reentrained into the building, and that maximum ground-level concentrations downwind from the building during such an event could be beyond acceptable levels.

Building 222 is oriented north-south with three wings spreading east to west. The laboratory where the potentially hazardous experiments are conducted is located near the southwest corner of the south wing. About 550 feet to the SWW end of B-222 is B-111, a seven-story administrative building. A bank of tall

trees (40–45 ft high) is in the immediate vicinity to the west of the building complex. The previous paper provides a more thorough description, including a photograph and a map.¹

The prevailing winds, blowing from the southwest, are disturbed first by B-111 and then by the bank of trees. The end result is that a recirculation cavity is created on the leeward side of the trees above the building. The eddies created by this held building exhausts on the roof and cause reentry. In order to avoid or at least minimize reentrainment and toxic concentration problems downwind from the building, Hazards Control and Plant Engineering studied the problem and made a recommendation as to the height of the stack. This paper compares tracer gas test results with the predicted performance of the stack to ascertain that the dispersion characteristics of the designed stack were operating within the design parameters.

Stack Performance Evaluation

A combination of analytical calculation and qualitative smoke testing was conducted to verify the basic assumption on the B-222 conceptual stack height during design. Analytical methods included theoretical predictions such as the Gaussian plume dispersion models and empirical methods such as Wilson's design procedure. Qualitative smoke testing was used to verify some of the basic assumptions. After construction of the stack, quantitative tracer gas tests were conducted to verify the actual stack performance. Both of these tests are described in the previous paper.¹

Gaussian Dispersion Model

The Gaussian dispersion model is a mathematical model of atmospheric dispersion that attempts to simulate the gross behavior of a plume discharged from a stack. Description and derivation of the model are given in Ref. 2. HOT SPOT, a personal computer program written by S. Homann based on the Gaussian dispersion model,³ is used to calculate the downwind ground-level concentration at various distances from the stack. Standard deviations of various atmospheric conditions are included in the program.

Gaussian plume calculation is for estimating long-distance dispersion (100 m or more); the initial system dilution and subsequent mixing of air into the plume are not considered. The model is also based on

open-country terrain without consideration of local turbulence created by the building wake and trees—local turbulence enhances dispersion and reduces ground-level concentration.

Wilson's Predictions

Wilson's prediction is a design procedure for estimating downwind concentration based on the concept of "available dilution."⁴ This consists of three separate effects:

1. Internal system dilution by combining the released contaminant with the total flow of the exhaust stream.
2. Wind dilution from entrainment of ambient air after leaving a flush roof-level vent with plume rise to a downwind receptor.
3. Stack dilution due to stack height and plume rise from vertical exhaust velocity.

The total available dilution between the stack and the receptor is the product of these three dilution factors.

Wind dilution is based on an empirical correlation of wind-tunnel test data. The prediction assumes the wind dilution process takes place at critical wind-speed, at which absolute minimum dilution takes place.

Test Results

The collected test data for each of the five test runs are shown in Fig. 1. In this figure the analyzed tracer gas concentration from sample bags are plotted in their respective distances and angular locations to the stack. The general wind direction and average wind velocity during the test period are indicated.

The figure also shows estimates of ground level concentrations as predicted by the Gaussian diffusion

model and Wilson's correlation. Gaussian estimates provide ground level concentrations at 50, 100, and 200 m (164, 328, and 656 ft) directly downwind from the stack; Wilson's prediction provides ground level concentrations at 10, 200, 300, 400 and 600 ft at radial distances from the stack.

Discussion of Test Results

Table 1 lists the average measured ground-level SF₆ concentrations and Gaussian predictions at 200, 300, 400, and 600 ft. The calculated values are for locations along the plume centerline directly downwind of the stack, $y = 0$. Since placement of the sampling bags did fall closely in line with general wind direction during the test, a comparison of the tabulated

values is made to indicate the general trend of the prediction. For wind velocity 1.5 to 2.1 m/s, the applicable minimum distance from the stack is between 200–300 ft. For wind velocity 3.2 to 6.0 m/s, the distance is between 400–600 ft. At distances less than 200 ft, concentration prediction by the Gaussian dispersion model is generally much lower than the

Figure 1. Test results for five runs of SF₆ tracer gas. The corresponding model predictions are also indicated.

Figure 1, cont.

Figure 1, cont.

Table 1. Comparing average SF₆ ground-level concentration with the Gaussian prediction (in parentheses) in ppb.

Wind speed (m/s)	Downwind distance (ft)				
	10	200	300	400	600
1.5	60 (—)	148 (32)	190 (140)	120 (230)	53 (230)
1.6	0 (—)	145 (<1)	268 (45)	170 (150)	58 (260)
2.1	34 (—)	43 (23)	106 (100)	54 (160)	20 (170)
3.2	<10 (—)	33 (<1)	62 (<1)	46 (11)	33 (77)
6.0	<10 (—)	11 (<1)	19 (<1)	28 (6)	17 (41)

measured value. The Gaussian estimate predicts the minimum dilution region is about 600 ft from the stack; sampling measurements show the region is about 300 ft. The overall trend appears to indicate that when the dispersion model is used, consideration must be given to establishing the minimum downwind distance as a function of wind velocity.

In Table 2, the average and range of SF_6 concentrations in ppb at the same radial distances from the stack are listed. Wilson's estimates at these distances are given on the bottom row for comparison. The ground-level average concentrations at various distances and mean wind velocities are plotted in Fig. 2.

Examination of Table 2 and Fig. 2 indicates that as the wind velocity increases, the overall average downwind ground-level concentrations decrease. This seems reasonable as a result of increased turbulent dilution of the plume. Sampling measurements show that the minimum dilution region is located about 300 ft downwind from the stack; Wilson's estimate predicts this distance to be around 500 ft from the stack. The concentration distribution patterns seem consistent. The change in minimum dilution location is assumed to be caused by local disturbances created by the building's wake and the trees.

In Wilson's prediction downwind dilution is determined at critical wind speed, at which the maximum concentration will occur. Critical wind speed is defined as:

$$U_{crit} = \frac{20 V_e \sqrt{A_e}}{s}$$

With the stack exhaust velocity (V_e) = 2510 ft/min and square root of stack area (A_e) = 1.18 ft, critical wind speeds (U_{crit}) for various radial sampling distances (s), are given in Table 3. The table shows that the minimum dilution due to critical wind velocity occurred at 200 ft for a wind speed of ~1.5 m/s. The predicted concentration is 150 ppb at this distance, and the average measured concentrations (from Table 2) are 148 and 145 ppb. For other distances the wind velocity was either below or above the critical velocity during the test. Consequently, the measured concentration values should be lower than the predicted values, which agrees well with the average concentration values.

However, the maximum measured values exceeded the predicted values at the 200-ft distance at a wind speed of 1.5 m/s (210 vs 150 ppb). The maximum measured concentrations are a factor of 2 higher than

Table 2. Dispersion of SF_6 discharge from stack. The ground-level concentrations are given in ppb.

Wind speed (m/s)	Downwind distance (ft)				
	10	200	300	400	600
1.5	60	148	190	120	58
		(100/210)	(110/290)	(60/175)	(20/95)
1.6	0	145	268	170	58
		(47/360)	(110/600)	(85/280)	(25/85)
2.1	34	43	106	54	20
		(0/100)	(34/175)	(34/85)	(0/42)
3.2	<10	33	62	46	33
		(0/120)	(0/190)	(10/70)	(0/75)
6.0	<10	11	19	28	17
		(0/25)	(10/42)	(10/70)	(0/42)
Wilson's estimate	0	150	318	380	390

Figure 2. Actual ground-level concentrations at various distances, plotted for different mean wind speeds. Also shown is a curve giving Wilson's estimate for the same parameters.

Table 3. Critical wind speeds for various radial sampling distances.

Distance (ft)	Crit. velocity (m/s)
10	3.0
200	1.5
300	1.0
400	0.8
600	0.5

the predicted values at 200 and 300 ft (360 vs 150 and 600 vs 318 ppb, at wind speeds of 1.6 m/s). Overall ground-level concentrations higher than Wilson's prediction occurred under low wind speed conditions (1.5 and 1.6 m/s) and at 200 and 300 ft distances.

We can draw no clear conclusion for the sampling location on the roof 10 ft from the stack. Measured concentrations are generally higher than predicted for the roof. Since we had only limited sampling bag/pump equipment, and since Wilson's prediction indicates the maximum dilution region occurs between

400-500 ft from the stack, we decided not to place any sampling bag between the 10-200 ft distance. Effluent dilution behavior in this region should be investigated in future study. Otherwise, the test results indicate that, with a wind direction from the west to southwest, Wilson's estimate for predicting dilution from the tall stack of this particular location will have an uncertainty factor of 2. Wilson stated that, "The experimental data correlations used to develop models typically have an uncertainty factor of 2-5 for concentration. This is due mainly to the sensitivity of receptor concentration to small shifts in the trajectory from a point source."⁵

We recorded semi-continuous local tracer gas concentrations in one location during each of the tests; a typical run is shown in Fig. 3. The sampling recorder is 300 ft from the stack. The plot shows instantaneous concentration as sampled by the detector in 20 min. (one 0.1- μ l sample every 2 min). The pattern suggests that the plume diffusion follows a looping characteristic. Large loops of the plume are carried down to

Figure 3. Local tracer gas concentration measured at one location, showing instantaneous concentration as sampled every 2 min. The pattern suggests that the plume diffusion follows a looping characteristic.

ground level and cause momentary high concentrations, followed by the effluent free air as corresponding loops go aloft. The SF_6 concentration collected at the same location as recorded by the 20-min sample is 600

ppb. A method of estimating the average-to-peak concentration ratio is being investigated for health and safety reasons.

Conclusion and Recommendations

A better understanding of stack effluent dispersion in terms of dilution on the roof, downstream ground level, and intake concentration is of great interest for operational health and safety reasons. We performed tracer gas tests that showed consistent results that can be used directly for estimating local concentrations surrounding the test area. Our results can also be used for verifying the applicability and deviations from analytical predictions. However, this verification applies only to the specific tested area. If sufficient tracer gas tests are conducted of various areas at LLNL, correction factors may be generated to facilitate future stack height estimates for the entire site.

Also, for future tracer gas tests for this and other locations, we recommend that samples be collected in a 180° placement, with additional data taken between 10–200 ft. The information gathered will provide a better understanding of air flow around buildings at different locations and will enhance the modification of analytical models.

Recent literature suggests that improvements have been made to the predictive capabilities of the

Gaussian analytical model, some of which can be used to estimate the concentrations near the release stack. Therefore, an effort should be made to investigate and include these methods for prediction.

Caution must be used in applying analytical predictions. Both the Gaussian plume model and Wilson's prediction are based on relatively simple models—uniform wind approaching velocity, simple building shapes, and simple site geography. In situations where these variables are complex, considerable uncertainty will exist. To create the capability to handle future buildings that may involve high toxic materials, a scale model of LLNL buildings and the surrounding terrain should be built and tested in a wind tunnel. Wind-tunnel modeling can be used to optimize building configurations, stack heights, and locations.

The combination of a wind tunnel study, analytical methods, and tracer gas tests will form a firm foundation that will enable LLNL to operate safely with the latest stack design technology.

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INDUSTRIAL SAFETY

The Repetitive Motion Injury Epidemic: A Case Study

Lois J. Halunen

Abstract

During 1987 LLNL experienced a steep increase in repetitive motion injuries, particularly among persons who work at video display terminals (VDTs). Most of the injured workers and their supervisors we contacted were unaware of the strain that repetitive motion or "cumulative trauma" can place on the body. We used a systematic approach to inform the workers and managers about the causes of these injuries and the corrections that are necessary to improve the situation. After a briefing was held for all managers and supervisors, classes in ergonomic adjustment of video workstations were scheduled for all personnel who had jobs that required heavy keyboard use. We offered the assistance of a safety engineer to anyone needing further help in correcting workstation problems. Videotaped material on VDT ergonomics and on repetitive motion injuries was made available for viewing. We distributed pamphlets and articles to encourage further study and discussion. Preliminary results indicate a complete turnaround in the observed incidence of repetitive motion injuries. No new cases have surfaced in the six months since our campaign was concluded. We will continue to track the injuries experienced by this group and will offer refresher classes if necessary.

Introduction

Industrial safety engineers, medical doctors, physical therapists, and others involved in the correction and treatment of workplace injuries have been aware of the ergonomic problems and injuries associated with video display terminal (VDT) workstations for a number of years. This phenomenon has been noted all across the United States and in other advanced countries worldwide where large numbers of workers spend most of their workday at VDTs. However, those persons most affected by these problems—VDT workers and their supervisors—are still largely unaware of the consequences of poor workstation design.¹

Managers of office workers are rarely confronted with disabling workplace injuries and have a tendency to discount the seriousness of repetitive motion injuries. They are not aware that early intervention can prevent

such injuries from becoming permanent, crippling illnesses. Because of this, early symptoms are often not recognized, or the manager is unable to respond promptly and positively to correct the situation. Since prevention and early treatment are critical when dealing with repetitive motion injuries, the manager plays a pivotal role.²

The manager must provide the funds for additional equipment, must assist in rearranging work loads, and most importantly must set the tone to encourage employees to report their symptoms. The most common reaction to "soft tissue" injuries, as repetitive motion injuries are sometimes called, is that people are malingering. This attitude is usually evident from the first-line supervisor to top-level management as well as fellow employees, though studies show that less than 5%

of people affected with these types of injuries are malingerers.³ If the attitude of managers is not supportive, an employee may not mention any discomfort until it requires medical attention and involves lost worktime. At that point, repetitive motion injuries are very difficult to cure and often cannot be reversed. According to some doctors, rehabilitation of a complex case can take 6 to 12 months, and some workers experience a recurrence of symptoms when they return to work.⁴

An employee who works at a VDT must know enough about repetitive motion injuries to recognize the symptoms and make whatever adjustments may be necessary to the workstation to prevent injuries. They must understand the proper ergonomic workstation design (Fig. 1) and be encouraged to take responsibility for their own health and safety. During their career lifetime, they may work at a number of different workstations and must be knowledgeable enough to set up properly with each new move.

Any computer workstation task that involves heavy use of the keyboard, particularly an incorrectly positioned keyboard, can result in a repetitive motion, or cumulative

trauma type of injury. One of these disorders is called carpal tunnel syndrome, which is caused by pressure on the median nerve as it passes through the carpal tunnel in the wrist (see Fig. 2). Repetitive motion, over time, can cause tendons to swell and create pressure on this nerve due to the restricted space in the wrist. Symptoms are numbness and pain in the thumb and first two fingers, often occurring at night. This can progress to loss of some hand functions. Tenosynovitis (inflammation of the tendon sheath) is another repetitive motion injury with symptoms of pain and swelling as well as impaired circulation in the hands. In VDT work these injuries and other soft tissue injuries can be caused by such things as poor positioning of the keyboard or incorrect height adjustment on the chair, resulting in the wrists being held in a strained position while the fingers are typing. Lack of a wrist rest in front of the keyboard can also aggravate the strain on the hands. Since these injuries are extremely difficult to treat at advanced stages, early intervention is critical. Surgery and/or permanent disability can result if action is not taken to ergonomically adjust the workstation.

Figure 1. A well-designed and adjusted computer workstation.

Figure 2. Anatomy of the wrist, detailing the affected areas in carpal tunnel syndrome.

Case Review

A situation that clearly showed the seriousness of this problem was a high incidence of repetitive motion injuries among office workers in one LLNL department. During 4 months, 8 VDT workers out of a group of approximately 100 employees were treated for various repetitive motion injuries of the arms, hands, and wrists. Some of these injuries had advanced enough to require weeks of time off for recovery; in one case surgery was necessary. Anxiety developed as employees saw their fellow workers wearing splints on their wrists or losing work time due to repetitive motion injuries. Doctors in our Health Services Department termed it an "epidemic" and also expressed their concern.

The Hazards Control Industrial Safety Group had been following up on such injuries as they occurred.

After evaluating each case with the employee and supervisor, it became clear that the root of the problem was lack of knowledge about the consequences of poorly adjusted VDT workstations. A related issue was a lack of awareness of the seriousness of the repetitive motion injuries that could result from using these workstations. Employees experiencing symptoms either did not associate the injury with the video workstation or did not think that management would be responsive in correcting the ergonomic problems. Management did not realize the seriousness of these injuries or know how to respond. Management needed to recognize that health complaints associated with VDT work are real, whether physiological or psychological.

Methods

The task for the industrial safety engineer was to provide immediate information to the entire department to assist them in recognizing and correcting the problem. First, a briefing was arranged for all department managers and supervisors. The main purpose of this meeting was to share the collective information about the types of injuries that were occurring and to emphasize the need for prompt action. This was accomplished by describing

specific cases that had occurred within the department and by relating this to the increased incidence of repetitive motion injuries that has been documented worldwide in VDT workers. Characteristics and symptoms of types of repetitive motion injuries were then discussed. We emphasized that these are authentic injuries that can be verified through medical testing techniques. We then discussed causes of these injuries, incorrect work

positioning that placed unnecessary strain on hands and arms, as well as overuse syndrome from poor work habits.

In the briefing we described and demonstrated proper ergonomic workstation design. Specific suggestions were offered on how to correct some of the observed deficiencies, such as providing keyboard drawers to lower the height of the hands, placing wrist rests in front of keyboards, and reevaluating workloads to allow more flexibility in taking mini-breaks from keyboarding. The final segment of this briefing was devoted to encouraging good communication between supervisors and their employees on the issue of repetitive motion injuries and the importance of a positive, supportive attitude from management. Without this, employees may not report their symptoms soon enough for effective treatment. Prevention and prompt intervention were stressed as the only real answer, since medical treatment is limited and not always successful.

Immediately following the manager's briefing, a series of classes were scheduled for all personnel who had jobs that required heavy keyboard use. During the next week, approximately 70 employees attended these classes that emphasized individual responsibility. The goal was to make each employee an expert in correct VDT workstation arrangement so that throughout the employee's career he or she could always have a properly adjusted, personalized workstation.

Ergonomic adjustment of VDT workstations was described and demonstrated. Topics included: chair seat

height, back height, foot rests, wrist rests, document placement, eye strain and glasses, lighting and glare, keyboard height and distance, terminal height and distance from eyes, exercise, and rest breaks. Information obtained from evaluating the employees' workstations within the department was used to suggest ways to deal with specific problems. Employees were assured that management was aware of their needs and was ready to assist in obtaining any necessary equipment or in reassessing workloads.

We followed this with a discussion of the causes and symptoms of repetitive motion injuries, including the injuries reported within their department. We emphasized the value of exercise, rest breaks, and a varied work schedule to avoid constant keyboarding, as well as taking personal responsibility for the employees' own health by adjusting their workstations and promptly reporting any symptoms of repetitive motion injuries. We offered the assistance of a Safety Engineer to anyone needing help in determining the correct positioning of VDT equipment or in identifying problems.

This personalized, concentrated effort to educate all levels of the department was followed by offering additional videotaped material on VDT ergonomics and on carpal tunnel syndrome. Pamphlets and other written material was distributed to encourage further study and discussion within the department.

Preliminary Observations

The initial result of this campaign was the identification of a few additional cases, as employees put their new knowledge to use and reported existing symptoms. However, no new cases have surfaced since the campaign was concluded (approximately six months).

Anecdotal evidence indicates that employees are more aware of the causes of repetitive motion injuries and feel more able and confident in rearranging their workstations to fit their needs.

Conclusion

We will continue to track the frequency and severity of repetitive motion injuries within this group of workers and offer refresher classes if necessary. In addition, this

educational approach may be used elsewhere at ULNL as a preventive measure in departments where large numbers of people work at video display workstations.

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A Study of Burn-Through Times for Laser Protective Eyewear

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Abstract

We studied the performance of laser protective eyewear currently in use at LLNL: a goggle with a cellulose propionate filter from Glendale Protective Technologies and a goggle with a glass optical filter from Spectra Optics. We also studied a new polycarbonate optical filter material from Glendale, and compared its performance with that of the cellulose propionate material. We used three lasers in the study: a single-pulse (variable-pulse-length) Nd:YAG laser with a wavelength of 1064 nm, a pulsed copper-vapor laser with a pulse-repetition frequency of 6.08 kHz and a wavelength of 510.6 nm, and a pulsed xenon-fluoride laser with a repetition rate variable up to 100 Hz and a wavelength of 351 nm. Where possible, we systematically increased the output power of each laser to the damage threshold of the filter material and beyond. Our results showed that the new polycarbonate optical filter suffered less damage at equivalent laser beam challenges than the cellulose propionate optical filter. We also observed that the glass goggle, which was designed specifically for use at the 510.5-nm wavelength, sustained no visible damage from the specified laser light at the highest power levels we could achieve.

Introduction

We conducted a study of laser protective eyewear for three reasons. First, a serious laser-related eye injury at LLNL in July 1985 brought to light the lack of recent scientific literature on current eyewear performance.^{1,2} Second, with the widespread use of powerful lasers at LLNL, safety personnel need access to damage information on eyewear so that they can select the best personnel protection. Finally, quantitative data was needed to verify the performance of protective eyewear already in use at LLNL. We tested this eyewear in the infrared, visible, and ultraviolet wavelength regions—specifically, at 1064 nm (Nd:YAG laser), 510.6 nm (copper-vapor laser), and 351 nm (xenon-fluoride laser).

Three types of eyewear were tested in this study.

The first type was the Glendale Protective Technologies "Laser-Gard" goggle now in use at LLNL with an optical filter material of cellulose propionate (CP) polymer.³ Specific absorbing compounds are added during manufacture to provide defined regions of high optical density. The second type of protective eyewear, also in use at LLNL, was a goggle with a glass filter from Spectra Optics designed for use at wavelengths of 488–530 nm.⁴ These goggles are constructed with a dielectric coating layered between two glass lenses. The third type of eyewear studied was a new polycarbonate optical filter supplied by Glendale. Glendale provided us with three polycarbonate optical filter samples specific to the three wavelength regions described above.

Experimental Details and Methodology

We assessed the mode of failure for each type of eyewear, as well as the time and energy required for each type to reach its failure point. We applied known amounts of laser power up to and beyond the damage threshold

for each material, or up to the maximum power available from the laser. This approach required carefully characterized laser systems, which limited the number of lasers that could be used for our test.

Infrared Region Exposure Studies

We tested a Glendale "Laser-Gard" CP optical filter with a specified optical density rating of 14 at 1060 nm. We used the Nd:YAG laser at a wavelength of 1064 nm with single laser pulses with durations of 1–50 ns at energies of a few joules. The geometry of the Nd:YAG test station is shown in Fig. 1. The beam emerging from the laser is focused by a lens with focal length of 6 m. Sample eyewear under test were placed upstream of the beam waist at a point where the beam diameter was 2–3 mm. To measure the laser power in the beam plane, the converging beam was divided to form equivalent beam planes that were diverted to a vidicon camera and a calorimeter, which measures the fluence distribution (or beam profile) and pulse energy, respectively. Absolute fluence values (peak power) are accurate to within $\pm 7\%$. During the tests our sample was mounted on a stage that allowed it to be moved from the beam into the field of view of a light microscope. The primary method for determining whether the sample had been damaged by the laser was by examining pre- and post-shot photographs taken with the microscope.

We first tested the CP optical filter with single 50-ns pulses. This optical filter was nominally 1.7 mm thick. Light microscopy revealed no damage at exposures of either 1.5 or 3.0 J/cm². When incident exposures reached 5–9 J/cm², a depression became visible in the center of the impingement area, probably caused by a melt in the outermost layer of the optical filter. There was also a slight discoloration in the center of the impingement area, which we speculate indicates some thermal decomposition of the polymer. At higher power larger regions of melting could be seen, until at 22 J/cm² we saw evidence of severe melting of the outer layer and threads of ejected molten plastic.

When we tested the CP optical filter again on the single-pulse system at a pulse duration of only 1 ns, the results were quite different. At the time of the pulse, a

laser-induced plasma was visible to the eye on sites irradiated at 4–8 J/cm². Permanent damage to the test sites consisted of only a few very small (<50 mm) pits; we saw no evidence that the sites had been melted. As we increased the power, we found no increase in surface damage. The formation of a plasma during exposure to the shorter-duration pulse apparently prevents the goggle from absorbing much of the pulse energy. Thus, damage to the surface is less than with a longer-duration pulse of the same total energy.

Visible Region Exposure Studies

We operated a pulsed copper-vapor laser at a wavelength of 510.6 nm and a pulse-repetition frequency of 6.08 kHz to test the CP optical filter. This filter's specifications say it has an optical density of 15 at 488 nm and 11 at 514.5 nm. Figure 2 shows the optical path used in this test system.

We tested the CP filter over a wide range of power densities on two separate occasions. The first test used a beam diameter of 1 mm with 400 mW of average output power (power density = 51 W/cm²). Complete burn-through (a steady beam through the hole) was achieved in two minutes. We then narrowed the beam in focus to 0.2 mm (power density = 1273 W/cm²) and repeated the test. A first burn through was observed within 5 s, accompanied by smoke and a brilliant flash of white light from the exposed surface of the optical filter. As the exposure continued, melting polymer flowed down to block the beam, melted away, and was replaced by freshly melted material. During this sequence of events the full beam intermittently penetrated the optical filter. We saw a steady beam through the hole after 33 s.

When we narrowed the focus further to 0.1 mm (power density = 5093 W/cm²), pieces of molten material were ejected from the beam focus area. Smoke was observed throughout the duration of the test, and upon cooling of the filter, we noticed strands of the melt

Figure 1. Beam characterization of the Nd:YAG laser.

Figure 2. Optical path for the copper-vapor laser used in these tests.

projecting from the damage site. However, the time for complete burn through was 163 s in one experiment and 210 s in another.

We repeated the same tests to see if we could achieve a greater precision. Table 1 lists the data from our second round of tests, which shows that over a wide range of applied power, we found no consistent burn-through threshold. The only observed burn through was at a power level of 382 W/cm². At power levels above and below this value, we found no burn through with exposure times of up to five minutes.

We used the same copper-vapor laser to test the glass optical filter from Spectra Optics. In each case we applied the beam for eight minutes. The power density was increased from 8.5 to 538 W/cm², but still no damage to the goggle was observed. Instead, as the power of the incident beam increased, the specular reflection became

more intense, but the goggle remained unaffected. We were unable to test at higher power levels due to the limitations of the laser.

Finally, we also conducted a series of tests on the prototype polycarbonate optical filter. This optical filter was specified by the manufacturer to have an optical density of 15 at 488 nm, 11 at 514.5 nm, and >4 at 530 nm. No burn through was observed at any power level over a range of 191–3180 W/cm². We observed some smoke at the highest power of 3180 W/cm². In addition, a black spot remained at the target site following the test. Both of these observations indicate combustion and decomposition of the polymer. Nevertheless, the shallow depth of damage on the polycarbonate filter and its integrity after challenge show that the polycarbonate material has a heat resistance superior to that of the CP polymer.

Table 1. Results of the copper-vapor laser (510.6 nm, 6.1 kHz) challenge of the Glendale CP optical filter. The focus diameter was 0.03 cm, and the test lasted for five minutes.

Beam power (mW)	Power density (W/cm ²)	Result
9	13	no breakthrough observed
25	35	no breakthrough observed
44	62	no breakthrough observed
73	103	no breakthrough observed
100	142	no breakthrough observed
120	170	no breakthrough observed
200	283	no breakthrough observed
240	340	no breakthrough observed
270	382	@ 2:05 a brief light broke through, then resealed
320	453	no breakthrough observed
360	509	no breakthrough observed
380	538	no breakthrough observed

Ultraviolet Region Exposure Studies

We used a pulsed xenon-fluoride laser to test a CP optical filter at 351 nm. According to its specifications, this optical filter has an optical density of 15 at 332 nm and 16 at 337 nm. The laser operated at repetition rates of up to 100 Hz with a pulse length of 25 ns. The light path used in this arrangement is shown in Fig. 3. The laser emitted a beam that had good spatial uniformity but a large divergence. To irradiate the sample, we used a rectangular aperture to isolate the uniformly intense center of the beam, which formed a down-magnified image of this aperture at the sample surface. The spatially averaged fluence in the down-magnified beam was computed by dividing the pulse energy by the beam area. Because the beam was not completely flat topped, the fluence at the center of the beam was 25–30% greater than the average.

A preliminary study determined that no permanent or reversible bleaching of the filter material occurred at any time during exposure to the laser beam. (For this discussion, bleaching is defined as a loss of light-absorbing capacity—hence, loss of protection.) A blue fluorescence was visible on the exposed side of the optical filter, and the intensity of this reflected light was observed to be proportional to the power of the incident beam. After this preliminary investigation we systematically challenged the optical filter to evaluate damage at various beam-intensity levels. Results of these experiments are given in Table 2 as separate tests at increasing levels of fluence. An analysis of output power vs burn-through time is shown in Fig. 4.

We conducted a second investigation comparing the CP optical filter with the polycarbonate filter for use at the near-ultraviolet wavelength. The CP optical filter

Figure 3. Optical path for the xenon-fluoride laser used in these tests.

Table 2. Results of the xenon-fluoride laser (351 nm, pulsed) challenge of the Glendale CP optical filter.

Run No.	Fluence (J/cm ²)	Fluence (W/cm ²)	Number of pulses
Test A			
1	6.12	151	190 (burned through)
2	6.10	149	190 (burned through)
3	6.12	151	180 (did not burn through)
Average	6.11	150	
Average burn-through time = 185 pulses + 25 pulses/s = 7.4 s			
Test B			
1	11.2	279	125 (did not burn through)
2	10.8	269	135 (burned through)
Average	11.0	274	
Average burn-through time = 130 pulses + 25 pulses/s = 5.2 s			
Test C			
2	18.3	460	103 (burned through)
3	18.0	451	50 (did not burn through)
4	17.6	442	75 (did not burn through)
5	17.3	433	90 (burned through)
Average	17.8	447	
Average burn-through time = 82.5 pulses + 25 pulses/s = 3.3 s			
Test D			
1	35.1	875	50 (did not burn through)
2	39.1	772	60 (did not burn through)
3	30.1	772	70 (burned through)
Average	31.8	806	
Average burn-through time = 65 pulses + 25 pulses/s = 2.6 s			

was 1.7 mm thick with a specified optical density of 16 at 337 nm and 15 at 332 nm. The polycarbonate filter was 3.2 mm thick with the same specified optical densities at the same wavelengths. We used the rectangular aperture described above and imaged the output beam of the laser onto the front surface of each sample.

Four beam-focus areas were used on the optical filter surface to obtain the desired range of radiation power: 0.260, 0.398, 0.509, and 0.609 mm². Because the beam was formed by imaging, it diverged through the sample, slightly decreasing the power intensity as the beam penetrated the material. The single-pulse fluence levels at the front surface varied from 8–19.0 J/cm². Differences in average power were achieved by varying the pulse-repetition rate. Combinations of beam size and repetition rate were used to obtain fluence levels from 75.0–1900 W/cm². We determined the time required for the laser to burn through the sample by placing a UV-sensitive

paper behind the sample, which produced a visible fluorescence upon exposure to the 351-nm laser light.¹⁰ We stopped the laser at the first visible fluorescence. Laser light power readings were not taken at the back of the sample.

With the CP optical filter, transmittance increased very rapidly after the first penetration, and variations in the burn-through times as large as $\pm 10\%$ were observed with the same fluence and beam widths. These variations can be attributed to slight inhomogeneities within the goggle itself. With the polycarbonate goggle burn-through times were consistent from site to site. Impingement of the laser beam on the new polycarbonate optical filter produced a bright plasma plume and a loud sputtering noise. By contrast, the CP optical filter simply melted with a minimum of noise or light. Tables 3 and 4 give the burn-through data from these experiments. The burn-through times were measured with a series of

Figure 4. Beam output power vs burn-through time for the Glendale CP optical filter.

short-duration pulses. The high peak-power density in the pulse, 0.24 GW/cm^2 at 6 J/cm^2 to 1.4 GW/cm^2 at 35 J/cm^2 , may have contributed to the plasma formation associated with the damage. This plasma may have reflected some of the incident light. Burn through might occur more rapidly if the test were done with a true continuous-wave beam, and the decrease in burn-through

time might be greater for the polycarbonate optical filter. We believe these burn-through times would not be exactly reproduced with a continuous-wave laser because of the differences between our laser with variable repetition rate and the continuous light from a continuous-wave laser.

Conclusions

Results obtained using short pulse durations ($<5 \text{ ns}$) at 1064 nm indicate that the CP goggle is safe protection from a single high-power laser pulse. The plasma plume formed from the beam hitting the optical filter keeps most of the energy away from the filter itself. Using the filter with single pulses of longer duration ($>10\text{--}20 \text{ ns}$) is more hazardous because more light is absorbed by the optical filter than is blocked by the plasma. Even for the larger pulses, though, this work shows that the goggles are safe for single-pulse fluences up to at least 20 J/cm^2 . This is well above that usually found in 1064-nm beams, except for in regions where the beam is tightly focused. If laser systems were engineered to prevent any encounter between the eye and the focused beam, CP plastic goggles would be safe for this use.

At 510.6 nm the glass optical filter from Spectra Optics was safe under all power levels the laser could produce. We were unable to test at the highest power

levels that had been achieved for the plastic lenses. The goggle efficiently reflects away impinging radiation at the test wavelength of 510.6 nm .⁹ However, the reflection that occurred during our test conditions would be hazardous to an unprotected bystander.

The prototype polycarbonate optical filter from Glendale prevented any measurable copper-vapor laser light from penetrating it. No burn through was observed when the optical filter was exposed to a beam of 3180 W/cm^2 for five minutes. Eyewear made from either of these materials should provide protection from laser light at this wavelength under high pulse-frequency conditions or even continuous-wave light. The irregular performance of the CP optical filter during this test series indicates a lower margin of safety, making the glass filter and the prototype polycarbonate goggle better choices for eye protection. It is also possible that the use of a larger challenge beam diameter may lower the

Table 3. Burn-through times observed with the xenon-fluoride laser (351 nm, pulsed) with beam size of 0.609 mm².

Rep. rate (shots/s)	Fluence (J/cm ²)		Fluence W/cm ² burn-through		No. shots for time (s)		Burn-through time (s)	
	Poly.	CP	Poly.	CP	Poly.	CP	Poly.	CP
10	8.0	9.2	80	91.6	1622	91	162.2	9.1
25	7.5	9.0	187.5	224.3	1359	90	54.4	3.6
50	7.5	8.8	375	438.2	1066	96	21.3	1.9
100	7.6	7.7	760	770	943	160	9.4	1.6

Table 4. Burn-through times observed with the xenon-fluoride laser (351 nm, pulsed) with beam size of 0.260 mm².

Rep. rate (shots/s)	Fluence (J/cm ²)		Fluence (W/cm ²)		No. shots for burn-through		Burn-through time (s)	
	Poly.	CP	Poly.	CP	Poly.	CP	Poly.	CP
10	18.4	19.9	184	199.4	1184	55	118.4	5.5
25	17.4	19.7	435	491.5	1039	70	41.6	2.8
50	17.6	19.0	880	974.5	779	89	15.6	1.8
100	17.6	18.8	1760	1878.0	692	123	6.9	1.2

burn-through threshold since an increase in diameter may allow for more efficient absorption of energy by the polymer in the filter. Because of the inconsistent burn-through times observed at 510.6 nm, further research should be conducted at this wavelength to develop consistent safety thresholds for plastic eyewear.

The tests conducted at 351 nm showed that Glendale's new polycarbonate optical filter resisted burn-through for a longer time than their CP material. Under the conditions of our experiment, burn-through times were about a factor of 10 greater for the new optical filter. There are several reasons for this superior performance. First, the prototype polycarbonate sample was thicker, so its absorption coefficient of 115 cm⁻¹ is smaller than that of the thinner CP filter, which has an absorption

coefficient of 215 cm⁻¹. As a result, for a given single-shot irradiation into the optical filters the temperature rise in the new polycarbonate filter was about one half that in the CP filter. Second, the new polycarbonate optical filter appeared to withstand higher temperatures. When the polycarbonate optical filter did fail, it failed by decomposition and removal, whereas the CP optical filter simply softened. Finally, we estimated that about twice as much mass was removed as the beam penetrated the new polycarbonate material, and a correspondingly greater amount of energy was required to do so. The effect of all three factors together accounts for the observed tenfold increase in its resistance to laser light damage at this wavelength.

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The Evaluation of Four Models of Personal Air Samplers

James F. Boyer and Bruce J. Held

Abstract

We have evaluated four makes of personal air samplers to determine which would best serve particular needs of field industrial hygienists and health technicians. The four makes of personal air samplers we studied are the duPont Alpha-1, the Gilian HFS 113A (UT), the MSA Flow-Lite, and the Sensidyne BDX 60. The tables in this report list the physical characteristics of each sampler and also give the results of our laboratory evaluations of each sampler's operating characteristics. We measured the noise levels generated by each sampler, and we inspected the required maintenance procedures. We also determined how long the pumps would operate without recharging or undergoing a change in flow rates and noise levels. With this information field personnel should be able to choose the sampler that best fits the needs of a particular operation.

Introduction

The emphasis placed on minimizing occupational exposure to toxic compounds includes actual sampling of breathing-zone air. One of the best ways to accomplish this is to attach a sample-collection system to the employee with a substance-specific collection vessel (such as an activated-charcoal column, filter paper, or reactant-liquid bubbler) near the employee's face.

The universal component in any sampling strategy is the personal sampling pump. Four makes of personal air samplers were purchased for test and evaluation to determine which would best serve the needs of our field industrial hygienists and health technicians. The four

models of samplers used in this evaluation, shown in Figs. 1-4, were the duPont Alpha-1, the Gilian HFS 113A (UT), the MSA Flow-Lite, and the Sensidyne BDX 60.

We compared the physical characteristics and special features of each sampler and then conducted laboratory tests to determine how long the pumps would operate without recharging or undergoing a change in flow rates and noise levels. We make no conclusions in this report, since it is intended only to guide field personnel in selecting an air sampler that will best suit their needs for a particular operation.

Physical Characteristics

Since workers must wear these samplers up to 8 hours per day, the samplers should be as small and light as possible. Cost is a consideration, but the possible benefits of a more expensive sampler must also be weighed. Air-flow ranges in a sampler are of interest to

the industrial hygienist, as is the recharge time. Table 1 lists the physical characteristics we considered for each of the four makes, and Table 2 gives the special features incorporated into or available for each make of sampler.

Figure 1. DuPont Alpha-1 personal air sampler.

Figure 2. Gilian HFS 113A (UT) personal air sampler.

Figure 3. MSA Flow-Lite personal air sampler.

Figure 4. Sensidyne BDX 60 personal air sampler.

Table 1. Physical characteristics of the four personal air samplers in this evaluation.

Make/Model	Size (cm)	Weight (g)	Approx. cost (\$)	Published flow range (lpm)	Recharge time (h)
DuPont Alpha-1	15 × 15 × 7	1073	1070	0.005-5	14*
Gilian HF 113A (UT)	13 × 13 × 6	970	865	0.5-3.5	14 16
MSA Flow-Lite	11 × 11 × 6	620	522	0.5-3.5	16
Sensidyne-BDX 60	12 × 13 × 7	826	455	1-3	16

* Controlled by built-in microprocessor.

Table 2. Special features of the four personal air samplers.**DuPont Alpha-1**

Programmable start time, length of sample time, and delay time for flow-restriction shutdown
Elapsed-time indicator
Readout of average temperature of sampled air
Pump discharge connection
Elapsed-time indicator for charge cycle shows when charging is complete
Optional external programmer (EP-1) for quick setup of multiple units

Gilian HFS 113A (UT)

Programmable run time
Flow compensation
Actual run time indication (elapsed time)
Built-in Rotameter
Pump discharge connection
Automatic shutdown in case of filter paper clog

MSA Flow-Lite

±3% flow control
Optional low-flow kit—to 10 cm³/min
Automatic shutoff in case of filter clog
Built-in flow meter
Pump discharge connection

Sensidyne BDX 60

Flow compensation
Selectable automatic shutoff in case of clogged filter or other flow restriction
Built-in flow meter

Operating Characteristics

We performed the following tests in the laboratory to determine how well the samplers performed while operating.

Performance Without Recharging

The samplers were each tested to determine how long they could run without being recharged and to determine what changes in flow rates would occur over time. Table 3 shows the percent change in the flow rate after 8 hours of operation when the samplers were

operated at their maximum flow rates using 37-mm, 0.3-μm filters. Table 4 gives the percent change in flow rates when the samplers were operated for as long as possible without recharging, using an asbestos sampling configuration (25-mm, 1.2-μm filter at a flow rate of 2.0 lpm), which puts a fairly heavy resistance load on the pump.

We did not make measurements for higher resistances, such as heavy filter loading or a train of filters and sorbents in line. However, a comparison of

Table 3. Flow control of personal air samplers over an 8-hour shift using 37-mm, 0.8- μ m filters at maximum flow rates.

Make/Model	Beginning flow (lpm)	Ending flow after 8 h (lpm)	Change in flow rate (%)
DuPont Alpha-1	5.011	5.053	+0.8
Gilian HFS 113A (UT)	3.499	3.409	-2.06
MSA Flow-Lite	3.491	3.336	-4.4
Sensidyne BDX 60	3.000	3.003	+0.1

Table 4. Test using 25-mm, 1.2- μ m filters (asbestos sampling configuration) to determine how long each sampler will perform without a recharge.

Make/Model	Beginning flow (lpm)	Flow at end of test* (lpm)	% flow change: beginning to end of test	Hours pump operated w/o recharge
DuPont Alpha 1	2.013	2.005	-0.4	11.5
Gilian HFS 113A (UT)	2.000	2.012	+0.6	19.0
MSA Flow-Lite	2.000	2.029	+1.5	10.1
Sensidyne BDX 60	2.000	2.300	+15.0	18.0

* Flows were measured every half hour, therefore the flow indicated was measured within a half hour of the time when the pump ceased operating.

the manufacturers publications and performance curves indicates that at a flow rate of 3 lpm for 8 hours, the pumps would operate without shutting off at the following resistances:

DuPont Alpha-1	35 in. H ₂ O
Gilian HFS 113A (UT)	15 in. H ₂ O
MSA Flow-Lite	10 in. H ₂ O
Sensidyne BDX 60	15 in. H ₂ O

Noise Levels to Users

We measured noise levels from the four makes of pumps. One set of measurements was made with the pumps operating at 2 lpm with a 25-mm, 1.2- μ m filter cassette installed. A second set of measurements was taken using the same filter cassettes, but with the pumps operating at their maximum flow rates. We hung the pumps on a user's belt and measured the sound level at the user's ear. Table 5 lists the results of the sound levels we measured.

Maintenance Procedures

DuPont Alpha-1. User-serviceable components of this sampler are the battery and internal filter, which can both be replaced. The duPont Alpha-1 owner's manual prescribes filter replacement annually, or sooner if the filter is clogged. The only other prescribed maintenance is to cycle the battery periodically (charge fully, discharge fully, recharge). Flow adjustments are simple and easily done without disassembling the unit. The pump itself must be sent to the duPont Company Instrument Systems for repairs. DuPont offers a ten-day turnaround from time of receipt (for five units or less).

Gilian HFS 113A. Gilian includes a complete parts list and detailed drawings. Prescribed maintenance is to cycle the battery periodically and to replace the internal pump inlet filter when it is dirty or after 250 hours of operation. Flow adjustments are easily done by trained personnel.

Table 5. Measurements of noise levels for personal air samplers.

Make/Model	At 2.0 lpm with 25-mm, 1.2- μ m filter cassette	At maximum rated flow with 25-mm, 1.2- μ m filter cassette	
	Sound level ^a (dBA)	Sound level ^a (dBA)	Max. rated flow
DuPont Alpha-1	53	67	5.0l
Gilian HFS 113A (UT)	55	61	3.5
MSA Flow-Lite	44	57	3.5
Sensidyne-BDX 60	52	52	3.0

^a Sound level reading measured at ear level of wearer, with pump mounted on the belt.

MSA Flow-Lite. The owner's manual includes detailed drawings and procedures for repairs and regular maintenance. A complete parts list is also provided. Regular prescribed maintenance is to cycle the battery and to check and clean the reusable internal filter. Flow adjustments can be done quickly from outside the unit.

Sensidyne BDX 60. The Sensidyne pump manual has complete procedures and a parts list for all repairs.

The recommended maintenance is to "inspect the pump interior and the exterior for damage and ... to make sure case screws are tight before use." The manual prescribes a deep discharge and subsequent recharge as care for the NiCad memory. Flow adjustment and calibration is involved and tedious, however.

Summary

Field industrial hygienists and health technicians should study the details we have compiled about the four makes of personal air samplers described here, the duPont Alpha-1, the Gilian HFS 113A (UT), the MSA

Flow-Lite, and the Sensidyne BDX 60. We make no conclusions in this report, but simply list the results of our evaluations so that field personnel may choose the sampler that best fits the needs of a particular operation.

Leakage of New and Used Filter Cassettes

Arthur H. Biermann

Abstract

A new type of plastic filter cassette from the Nuclepore Corporation can be reused to save costs. We leak-tested both new and used cassettes at flow rates of 1.85 lpm and approximately 17 lpm, using a condensation nucleus counter. Some cassettes were also tested without the normal sealing band applied to them. Two of the new filter holders were disassembled and reassembled more than 60 times to assess degradation due to wear. No significant leakage occurred in any of these tests using the new cassettes, but we did find leakage up to 2% in tests involving the used filter cassettes.

Introduction

Filter cassettes are used to monitor the workplace environment in some areas where toxic materials are machined. Recently, a new type of cassette holder has been introduced for this type of monitoring; to save on costs, industrial hygienists have suggested that these holders be reused. However, the process of reloading the cassettes with new filter membranes introduces the possibility of leakage. The objective of our work was to investigate the occurrence and extent of any leakage in these cassettes.

The filter cassette holders, commercially available from the Nuclepore Corporation, are cylindrical in shape, 3 cm in diameter and 8 cm long (including the inlet cap), as shown in Fig. 1. The holder is made of a black conductive plastic to reduce aerosol collection losses to the walls due to static charge. The filter holder is an open-face design but has an inlet cap, or protection cover, that fits on the front, allowing it to perform as an in-line filter for our leak-check tests.

Cellulose-membrane filters with a 25-mm diameter and a 0.8- μ m diameter pore size were used in these experiments. Our study used 74 filter cassette holders: 49 were new, 24 of which were loaded with filters by the manufacturer and the other 25 assembled by the experimenter. The remaining 25 were used cassettes assembled by the experimenter. We investigated two flow rates—1.85 lpm and approximately 16 lpm.

Leaks were determined by counting the particle concentration downstream from the cassette filter holder with a Thermo Systems Model 3020 condensation nucleus counter (CNC). The CNC is a very sensitive instrument for measuring aerosol particles, detecting as few as 0.01 particles/cm³ at a flow rate of 0.09 lpm. Using the CNC as a detector, the ambient room aerosol could be used as the challenge agent. This is advantageous, since using the ambient air allowed us to perform a regimen of tests without introducing likely contamination from an artificially generated test aerosol.

Experimental Configuration

Figure 2 is a block diagram of the experimental configuration. Atmospheric air may be drawn into the system directly or through a high-efficiency particulate air (HEPA) filter, where it then enters the 25-mm plastic filter holder and passes through the membrane filter. The filtered air travels from the holder to the CNC

through a venturi that is used to monitor the flow direction. The CNC has an external vacuum source applied through a control valve. Since the CNC draws only 0.09 lpm, an additional (or bypass) flow path regulated by a needle valve is provided to achieve the desired flow rate through the filter. A magnetic

Figure 1. The 25-mm Nuclepore filter cassette.

Figure 2. Experimental configuration for leak-testing filter cassettes.

differential pressure gauge monitors the pressure loss across the filter. The high-pressure side of the manometer was left open to the atmosphere so that all readings are of negative system pressure relative to the atmosphere.

We placed the filter housing in the test system by pressing 3/8-in. Tygon tubing over the exit nipple on the bottom support piece. Since the tubing is already connected to the inlet cap of the housing, it can be simply pressed onto the front of the filter holder. With the filter holder in place the bypass and CNC vacuum valves are simultaneously and slowly opened until the desired flow rate or pressure drop is obtained. The apparatus is allowed to flush itself out for a few minutes, and the CNC count data is then manually recorded. Because of the large pressure differentials encountered at the high-flow test condition, separate experiments were performed to determine the CNC's counting efficiency at lower pressures. Although counting losses of 18% were found at the high-flow test condition, such losses were negligible at the low-flow condition. If we observed any counts in a test, we placed a HEPA filter on the inlet to the filter housing and again observed the counts. If there is a significant leak, this determines whether the location of the leak is from the housing or from the filter itself.

Results

Of the 25 factory-assembled cassettes, no leaks were observed at the low flow rate. At high flow one cassette experienced a leak of 1.5×10^{-4} %. This corresponds to our minimum detectable limit for a one-minute CNC accumulation time. Thus, we found no discernible difference in leakage between the two flow conditions. The experimenter-assembled cassettes were tested only at the high-flow condition, 20 with and 5 without sealing bands. Two non-zero counts occurred with these 25 cassettes, one in each grouping. Both leaks were minor, approximately 3×10^{-4} %. When we placed a HEPA filter upstream, the leak rates either remained about the same or increased slightly—this indicates that the leak is into the housing from the outside air. We found no appreciable leakage with either the factory-assembled or the experimenter-assembled new cassettes. However, the cassettes with sealing bands may be less susceptible to separation in the event of mishandling.

As a check for degradation in performance due to wear, two of the filter holders were disassembled and reassembled (without sealing bands) over 60 times. In this testing neither the filters nor the backing material was replaced. Furthermore, to accelerate the wear of the plastic parts, the filter holder body and bottom pieces

Of the 49 new filter cassettes tested, 24 were factory-assembled and encased with shrinkable sealing bands. The other 25 cassettes were disassembled, sonicated in isopropyl alcohol and water for cleaning, and reassembled with the filter. All cassette parts, the inlet cap, body, and bottom support of the various filters were mixed.

To check the effect of the sealing bands on leakage, we assembled some of the cassettes and tested them without the sealing bands. Leak tests were conducted on the new factory-assembled filters at the two flow rates. We chose the lower rate after determining the average measured flow rate of five loaded filter holders while operating on a Gilian high-flow personnel sampler pump (model HFA 113A) calibrated to 2.00 lpm. The filters showed a pressure drop of between 8.6–10.9 in. H₂O at this low-flow rate. In the high-flow rate all 50 of the filter holder assemblies were tested at a constant 100 in. H₂O pressure differential relative to the atmosphere. At this pressure we observed differential flows of 14.8–17.8 lpm. The 25 new and experimenter-assembled cassettes and the 25 used cassettes were tested only at the high-flow condition, which was the worst case.

were purposely misaligned by 20–30° when they were fitted together. We could observe some wear by the accumulation of small, black specks on the filter itself, which is presumably plastic that has worn from the mating surfaces. Despite such harsh treatment and wear, these holders continued to test leak tight. Moreover, we found that the filter holders could even be disassembled during the leak-test operation without introducing a large leak. With the inlet cap and body completely removed from the bottom support, the resulting leakage was only 1% at a pressure loss of 60 in. H₂O.

Leak-test results from the 25 used experimenter-assembled cassettes are given in Table 1. In this test grouping detectable leaks are those having CNC measurements greater than 0.015 counts/cm³. Of the cassettes tested with sealing bands, only one out of five indicated a significant leak. Of those without sealing bands, 12 out of 20 indicated leakage. The highest number of counts incurred (EA2-10-NSB) corresponds to a leak rate of 3.6%; however, the bottom-to-housing fit was noticeably loose. Although a larger fraction of the used filters leaked, this difference is not significant for cassettes with or without sealing bands.

Table 1. Leak performance tests for used cassettes.

Filter No.	High flow (counts/cm ²)	Leak (%)
EA2 ^a -01-SB ^b	0.02	3.95×10^{-4}
EA2-02-SB	0.003	5.92×10^{-5}
EA2-03-SB	0.008	1.58×10^{-4}
EA2-04-SB	0.01	1.97×10^{-4}
EA2-05-SB	4.24	8.37×10^{-2}
EA2-06-NSB ^c	37.0	7.31×10^{-1}
EA2-07-NSB	0.18	3.55×10^{-3}
EA2-08-NSB	0.014	2.76×10^{-4}
EA2-09-NSB	11.10	2.19×10^{-1}
EA2-10-NSB ^d	182.30	3.60×10^0
EA2-11-NSB	4.24	8.37×10^{-2}
EA2-12-NSB	2.48	4.90×10^{-2}
EA2-13-NSB	0.33	6.52×10^{-3}
EA2-14-NSB	1.40	2.76×10^{-2}
EA2-15-NSB	1.63	3.22×10^{-2}
EA2-16-NSB	0.17	3.36×10^{-3}
EA2-17-NSB	0.012	2.37×10^{-4}
EA2-18-NSB	0.00	0.00×10^0
EA2-19-NSB	0.012	2.37×10^{-4}
EA2-20-NSB	0.00	0.00×10^0
EA2-21-NSB	0.16	3.16×10^{-3}
EA2-22-NSB	0.00	0.00×10^0
EA2-23-NSB	0.00	0.00×10^0
EA2-24-NSB	6.73	4.54×10^{-2}
EA2-25-NSB	0.00	0.00×10^0

^a EA2—Second set of experimenter-assembled cassettes.^b SB—Sealing bands.^c NSB—No sealing bands.^d Bottom of housings were loose-fitting.

Conclusion

Our experimental tests indicate that the filter holders can be reused, and the cassette parts can be mixed without the risk of significant leakage. The main area of concern is where the body of the cassette is pressed to the bottom support. Too little applied pressure could result in small leaks, but generally no greater than 1.5%. Too

much pressure could possibly shear the filter itself, although this did not occur in our tests. Very firm pressure applied to the body of the cassette and to the bottom support from two hands works well for assembling the cassette.

Study of the Performance of an Air Sampler for Biological Agents

Arthur H. Biermann and Joseph K. Kwan

Abstract

In recent years airborne-microbe samplers have been developed to offer improvements over the size, weight, portability, and sample-rate limitations of the traditional Andersen cascade-impactor air sampler. We have investigated the collection efficiency of the Biotest air sampler for aerosols and for particle losses within the sampler itself. Results indicate that aerosol concentrations as measured with the Biotest sampler can underestimate the actual concentration at smaller particle sizes (1.5–3 μm). This may be attributed to the decrease in collection efficiency of the device. While the sampler's efficiency increases for larger particles, we found that the great majority of these particles collected on the fan blades of the sampler rather than on the collection strip. Although its higher sample rate is advantageous, the Biotest sampler does not allow simple quantitative interpretation of aerosol concentration measurements.

Introduction

The role of microorganisms in causing environmental and indoor-pollution problems has received much attention over the past few years. Collection and enumeration of airborne microbes is often performed to evaluate the environment in question. Traditionally, Andersen air samplers were used for this purpose.¹ Recently, however, new samplers offering better portability and ease of operation, and higher sample rates, have become commercially available. One such

sampler is the Biotest RCS centrifugal air sampler (shown in Fig. 1). This sampler is light, compact, and easy to operate. Airborne microbes are impacted on an agar-coated collection strip, incubated, and their colonies counted. However, the collection efficiency of the Biotest sampler had not been properly demonstrated. We evaluated the collection efficiency of this sampler as a function of particle aerodynamic diameter.

Experimental Procedure

Monodisperse aerosols of dioctylsebacate (DOS) and fluorescein were generated using a vibrating-orifice aerosol generator. Aerosols were dried by adding dilution air and neutralized by passing through a ^3Kr sealed source. We monitored the aerosol size distributions using a Thermo Systems Model 3300 aerodynamic particle analyzer, which had been previously calibrated with polystyrene latex particles. We readjusted the aerosol generator if doublet or triplet particles were present in the distribution.

Aerosols produced in this manner ranged from 1.5–13 μm aerodynamic diameter with geometric

standard deviations of less than 1.1. This range adequately represents the anticipated range of bacteria-type viable particulates. It also overlaps with the lower portion of the size range expected for spore-like aerosols and the smallest of the pollen-type aerosols. Fluorescein served as a tracer to quantitatively determine the aerosol concentration and the particle deposits within the sampler.

We placed the Biotest sampler along with open faced 47-mm diameter/0.8- μm pore Nuclepore filter samplers inside a 1 m³ aerosol chamber, as shown in Fig. 2. Parallel aerosol samples were taken with the

Figure 1. The Biotest sampler investigated in these tests.

Figure 2. The Biotest sampler and two Nuclepore were placed in a chamber for these tests.

Biotest sampler and a filter, typically with sample times of 24 min. The filters were operated at 10 lpm. We removed the aerosol deposition on the surface of the Biotest collection strip as well as the fan blades and

chamber by washing with alcohol. Particles collected by the filter were similarly dissolved. All samples were then analyzed for the fluorescein tracer.

Results

In our effort to evaluate particle losses within the Biotest itself, the quantities of particles collected on the fan blades and on the collection strip were separately expressed as a percentage of the total. These values were calculated for each of the monodisperse aerosols, as given in Fig. 3.

The total quantity of each monodisperse aerosol collected by the Biotest was further compared to an average of the filter samples. We calculated the sampler flow rate, Q_B , using:

$$Q_B = \frac{C_F \times t_B}{B_f + B_c}$$

where:

B_f = particle collection on the fan blade and housing

B_c = particle collection on the collector strip

t_B = Biotest sample time

C_F = aerosol concentration determined by the filter.

Using this formula and assuming that 100% collection is achieved for aerosols ≥ 5 mm in diameter (those having the greatest collection in our studies), we calculated the average Biotest flow rate to be 284 lpm. The manufacturer reports a sample flow rate of 280 lpm. Macher and First⁷ have found collection efficiencies to be greater than 95% for particles greater than 10 μ m. They determined a sample flow of 210 lpm, but this was accomplished with an artificial chamber attached to the sampler. Using the flow value of 284 lpm, we calculated an overall efficiency for the sampler as a function of particle size, shown in Fig. 4.

Discussion

According to Fig. 4, the collection efficiency of the Biotest sampler as compared to that of the filter samplers can be rather low. For example, less than 50% capture occurs for aerosols < 5 mm in diameter, which is the size range of interest for most microorganisms. Total collection improves for aerosols larger than this. These aerosols would constitute the larger fungal spores and bacteria appearing in clumps as well as bacteria attached to small particles. The Biotest manufacturer recommends a sampling rate of only 40 lpm as a basis for the sample volume. This would be accurate only for aerosols having aerodynamic diameters between 3–5 mm, assuming all collection is on the collection strips.

As illustrated in Fig. 3, over half of the particle deposition occurs on the fan blades for aerosols > 5 mm in diameter. For the 9-mm-aerosol test, nearly all the aerosol was deposited on the fan blades. For the largest particles tested in this study (13 mm) the collector strip

indicated improved efficiency. In our case this improvement is probably due to reentrainment of the liquid aerosol collected on the fan blades, but a similar effect—bounce-off—might occur for solid aerosols. Since particles collected on the blades will not be counted in routine analysis, we find that the actual collection efficiency is less.

The very low efficiencies occurring for aerosols < 5 mm in diameter negate the relatively high sample rate of the Biotest. Also, since the particle distribution of an aerosol being sampled is usually not known, it is difficult to make an efficiency correction. With the actual collection efficiency low and the possibility of large particle losses within the instrument, it is apparent that sampling results should be considered *qualitative* in nature. This is especially true since the poorest efficiencies occur in a particle-size domain of primary interest for airborne-microbe studies.

Figure 3. The fan blades in the Biotest sampler actually collected greater numbers of certain sized particles than the actual collection strip did.

Figure 4. Collection efficiency of the Biotest sampler as compared to the efficiency of the filter samplers.

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Aerosol Sampling Losses in Transport Lines

Arthur H. Biermann

Abstract

Particle losses may occur in transport lines used to sample aerosols. If the losses are significant, the aerosol sampler or detector will underestimate the real aerosol concentration of the source volume. Some of the mechanisms causing particle losses include diffusion, sedimentation, and inertia. Theoretical models predicting losses by these mechanisms under turbulent flow conditions are considered. For a typical transport line with a 0.953-cm diameter and a volumetric flow of 40 lpm, expected losses can be larger than 10% for particles greater than 2 μm in diameter and with densities greater than 7.28 g/cm³. Losses in a single 90° bend can be equally as large.

Introduction

In some aerosol sampling systems, it may be impractical to place aerosol collectors or detectors in or near the source environment. Transport lines are often used to carry the particles from the source to the collector/detector. In such situations particle losses in these transport lines may be great enough that the aerosol detector significantly underestimates the aerosol concentration in the source environment.

Particle losses may occur because of outside forces acting on the particles, or because the particle is unable to follow the flow lines of a gas stream. We will consider some of the mechanisms that can cause such particle losses in general. The deposition mechanisms discussed include particle diffusion, sedimentation, inertia in straight transport lines, and inertia only in bends. Deposition mechanisms are dependent on several characteristics of the particle, such as its size, density, and shape. Where applicable, we compare several models describing losses attributed to the mechanisms. These models can be used in the design of transport lines to minimize the aerosol losses.

Other mechanisms of particle loss are not addressed in this paper, such as convective motion and electrostatic, thermophoretic, and diffusiphoretic forces. In certain situations these forces may cause particle losses equal to or greater than the mechanisms discussed here. For example, large losses may be encountered in the sampling of highly charged particles through nonconductive sampling lines. Inaccuracies in aerosol concentration measurements caused by nonisokinetic sampling are also not discussed in this report.

Our consideration of particle losses is applicable to turbulent (as opposed to laminar) flow conditions. In actual practice laminar flow is difficult to obtain,¹ and either convection or turbulent motion is more than likely. In sample transport lines flows are usually large to the point where the turbulent flow dominates.

We will illustrate the extent of losses by choosing some transport line configurations encountered in sampling systems here at LLNL. Losses can also occur within aerosol collectors and detectors; however, these losses tend to be specific to their respective flow geometries and are not considered here.

Particle Losses

Particle Transmission in Turbulent Flow

The eddies formed during turbulent flow conditions can give rise to significant normal velocity components to the mean flow. If the eddy size and velocity and particle characteristics are such that the particles can follow in the eddy, then particle deposition on the walls can take place. It is assumed that all particles stick to the wall upon impact, although this may not be completely true for all high flow conditions. The rate at which aerosols are transported from a volume to a surface can be defined as the following ratio:

$$V_d = \frac{(\text{amount deposited per cm}^2 \text{ per s})}{(\text{concentration in volume})} \quad (1)$$

where V_d is the particle deposition velocity. In a sample transport line we can think of this as a migration velocity across, or perpendicular to, the free-stream velocity of the gas stream. The amount of aerosol transmission or penetration through a transport line can be determined by writing a material-balance equation over a differential element along the line.² The resulting transmission is given by

$$\frac{C}{C_0} = \exp \left[-\frac{4V_d L}{D_1 U} \right] \quad (2)$$

where

- C = the particle concentration penetrating the line at a distance L ,
- C_0 = the initial particle concentration incident to the line,
- L = the length of the transport line,
- D_1 = the diameter or dimension of the line,
- U = the free-stream flow velocity through the line.

The corresponding fractional particle loss occurring in the line is then

$$\text{Loss}_p = 1 - \frac{C}{C_0} \quad (3)$$

These equations can be used in general to describe the particle penetration and loss in sampling lines having turbulent flow. Expressions describing losses due to laminar flow are readily derivable,^{1,2} but unfortunately, because of the complex boundary-layer phenomena in turbulent flow, analytic expressions for the deposition velocity, V_d , are difficult to attain.

Turbulent Flow

Flow is generally considered to be turbulent if the Reynold's number is greater than 1000-2000. This dimensionless number is given by

$$Re = \frac{U D_1}{\nu} \quad (4)$$

where $\nu = \mu/\rho_a$ = the kinematic viscosity of the gas, μ = fluid viscosity, and ρ_a = fluid density.

For a 3/8-in. diameter sample line with a volumetric flow of 40 lpm, the Reynold's number is 5800. In classical modeling of turbulent flow, the flow is divided in three regions outward from the wall: a sublayer laminar flow regime, a buffer or transition layer, and the main turbulent core. The velocity gradient near the wall is much steeper than in the ideal laminar flow case. This velocity gradient is characterized by the friction, or drag, velocity, given by

$$u^* = U \sqrt{\frac{f}{2}} \quad (5)$$

where f = Fanning friction factor = $0.039 Re^{-0.17}$.

The Fanning friction factor is an empirical factor determined for turbulent flow conditions. The friction factor expression given above has been taken from Davies.⁴ The behavior of particle motion across the velocity gradient is difficult to describe theoretically and leads to difficulties in determining the deposition velocities required for the assessment of particle losses in a sample line.

Diffusional Losses

Diffusional losses are those caused by the Brownian motion of particles. This type of loss is important only for smaller particles, those having larger mobilities and diffusion coefficients due to their small masses. For standard temperature and pressure conditions, losses from diffusion become important at sizes less than 0.1 μm . By making use of the Chilton-Colburn analogy between mass and momentum transfer, Chamberlain⁵ and Davis³ have arrived at the following expression for the deposition velocity:

$$V_R = u^* \sqrt{\frac{1/2}{Sc}} \quad (6)$$

where

$S_i = v/D$ = the Schmidt number for the aerosol particle,

D = the diffusion coefficient for the particle.

Often, in dealing with deposition in turbulent flow, the deposition velocity is transformed to a dimensionless velocity, V^+ , making it relative to the friction velocity, so that

$$V^+ = \frac{V_g}{u^*} \quad (7)$$

Correspondingly, a dimensionless particle relaxation time is defined as τ^+ ,

$$\tau^+ = \frac{\tau u^{*2}}{v} \quad (8)$$

where

$\tau = d^2 \rho_p / 18\mu$ = relaxation time of the particle,
 ρ_p = the particle density.

Using dimensionless quantities permits the description of particle losses for general transport line and particle characteristics. The dimensionless time contains the size and density information for the particle. A shape-factor term could also be incorporated here to account for non-spherical particulates.⁶ The dimensionless deposition velocity as a result of the diffusion transport mechanism is illustrated in Fig. 1. Another formulation for diffusion deposition having a slightly narrower boundary layer for transport given by Fuchs is also indicated in the figure.¹ For diffusion, greater deposition velocities occur for lower relaxation times, corresponding to particles having smaller diameters or lower densities. The point at which deposition by diffusion becomes less important occurs for dimensionless relaxation times >0.1 . For a unit density particle, this is analogous to a particle diameter of approximately $1 \mu\text{m}$.

Sedimentation

Aerosol particles settle from a still atmosphere at their terminal settling velocity, $V_g = \tau g$, where g is the gravitational acceleration.

The velocity is derived by equating the force on the particle due to gravity with that of the fluid resistance. To find the losses associated with turbulent flow through a line, this velocity can be used as deposition velocity. However, since the settling preferentially occurs in the downward direction, the factor of 4 in Eq. (2) must be reduced to 1 for a square duct or $4/\pi$ for a circular duct.

Inertia Losses

Inertia losses occur when the momentum of a particle does not enable it to follow the flow streamlines of the gas. In turbulent flow a particle may get swept into an eddy near the wall and be impacted onto the wall. Theoretical descriptions for the particle loss due to inertia mechanisms are complicated by the velocity profile near the wall. Generally, models assume a diffusion transport mechanism from the turbulent core to some distance near the wall where free flight ensues. Different models assume different velocities during the free-flight portion and different eddy diffusivities. One of the first theoretical descriptions of particle deposition velocities occurring in turbulent flow was that of Friedlander and Johnstone.⁷ Their model gives rise to several expressions for deposition velocity depending upon the stopping distance of the particle relative to the turbulent flow region. Although the expressions are too lengthy to present here, the model is compared with others and the pure diffusional losses in Fig. 1. The discontinuity at $\tau^+ = 40$ is a crossover between two of the flow regimes. A modification given by Forney and Spielman lessens this discontinuity.⁸ Two other models for inertia turbulent deposition are also compared in the figure. Postma and Schwendiman developed a semi-empirical model based their own experimental data and that of Friedlander-Johnstone.⁹ The Davies theory results in the lowest deposition velocities 1-10 τ^+ region.⁴ More recent theories and data agree with the modified Friedlander-Johnstone model.¹⁰⁻¹² This combination is used for calculating turbulent losses due to inertia in the following examples.

Turbulent Loss Examples

As an example, we have taken a sampling transport system similar to those found at LLNL. The line diameter is $3/8$ in. with a volumetric flow of 40 lpm, which is a reasonable flow quantity for filtering through membrane particulate filters. Pertinent parameters required to calculate the deposition velocity are given in Table 1. A comparison of the actual deposition velocities for the diffusion, gravitational sedimentation, and inertia mechanisms is shown in Fig. 2 as a function of particle size. A particle density of uranium oxide was assumed for these calculations. An additional relation is included that indicates the reduction in deposition velocity from the inertia expected at the reduced linear velocity of $1/4 U$, or 234 cm/s. In the latter case, particle losses due to sedimentation are relatively more important.

Figure 1. Comparison of turbulent deposition models. In all cases, $\rho_p = 7.25 \text{ g/cm}^3$.

Figure 2. Particle deposition velocities for different deposition mechanisms.

Table 1. Parameters used in the turbulent loss example.

Line diameter, D_l	0.954 cm
Line length, L	76.2 cm
Bend angle, θ	90°
Free-stream velocity, U	936 cm/s
Reynold's number, Re	5855
Fanning friction factor, f	0.00892
Friction velocity, u^*	62.5 cm/s
Air density, ρ_a	1.2×10^{-3} g/cm ³
Air viscosity, μ	1.83×10^{-4} poise
Kinematic viscosity, ν	0.1525 cm ² /s
Temperature	293 K
Mean free path	6.53×10^{-6} cm
Particle density, ρ_p	7.28 g/cm ³

Total particle losses from these mechanisms are presented in Fig. 3 for a transport line 2.5 ft long. Losses are shown for either horizontal or vertical transport lines at two particle densities, 1 and 7.28 g/cm³. Significant losses begin to occur at 2 and 5 μ m diameter particles for $\rho_p = 7.28$ and 1, respectively. For the 7.28 g/cm³ particles, nearly 100% loss occurs for 8- μ m diameter particles. Note that diffusional losses, which begin to dominate below 0.3 μ m, are independent of the particle density. But these losses are not appreciable until well below 0.1 μ m diameter.

A simple way to minimize losses from inertia turbulence is to lower the free-stream velocity through the line by increasing its diameter. Figure 4 illustrates the effect of increasing line diameters in a vertical transport line for our example—a maintained volumetric flow of 40 lpm for $\rho_p = 7.28$. A doubling of the line diameter can decrease particle losses by a factor of 30. Note that up to a certain line size (3/4 in.), this can be accomplished without a significant increase in diffusional losses.

Losses in Bends

Inertia is the major transport mechanism causing particle losses in short bends. The larger and denser particles are not able to follow the rapidly curving flow-stream lines and are impacted onto the walls. We will compare two models predicting particle losses in bends, a simplified model and a more rigorous one. In the simple model derived by Crane and Evans,¹³ the loss is expressed in terms of the Stoke's number as

$$\text{Loss}_b = St \frac{\theta}{2} \quad (9)$$

where

θ = the angle of the bend in radians,

$St = \tau U/D_l$ = particle Stoke's number (or inertia parameter).

Strikingly, the particle loss is not dependent on either the bend's radius of curvature or the Reynold's number. The percentage loss expected for a range of Stoke's numbers is shown in Fig. 5. With the parameters listed in Table 1, a Stoke's number of 1.09 corresponds to a particle diameter of 5 μ m. A more rigorous derivation of the particle losses occurring in a bend is derived by Cheng and Wang.¹⁴ Figure 5 compares three cases using their rigorous model corresponding to three different radii of curvature to the simplified model. Note that the simplified model agrees fairly well for radii of curvature greater than three times the line diameter. A radius of curvature over the range 1.5–5 in (8–27 line diameters) is not predicted to cause much difference in the amount of particle loss.

Figure 6 illustrates particle losses due to impaction in bends as a function of particles for several density and line-size combinations. Losses reported were calculated using the rigorous model. The effect of transport line diameter is shown for particle densities 1 and 7.28 g/cm³. Note that a doubling the line size can reduce particle losses by nearly a factor of 10.

Discussion

We have summarized some theoretical models that are useful in assessing the particle losses in aerosol sampling transport lines. Mechanisms responsible for particle losses can be diffusion, sedimentation, or inertia. No electrostatic losses were considered here, although it is assumed that all particles will stick to the wall upon contact. Overall particle losses are very sensitive to particle size, density, and the linear velocity through the

transport line. For the sample case cited in this paper, a flow of 40 lpm in a 3/8-in. diameter line, significant losses in straight line flow can occur for particles as small as 2–5 μ m. Losses of similar magnitude can be expected in one 90° bend in such a transport line. Inertial losses can be greatly reduced by increasing the diameter of the sample line.

Figure 3. Losses in vertical and horizontal sampling lines. In all cases, $u^* = 62.5$ cm/s and $L = 76.2$ cm.

Figure 4. Turbulent losses in vertical sampling lines. In all cases, $\rho_p = 7.28$ g/cm³, $u^* = 62.5$ cm/s, and $L = 76.2$ cm.

Figure 5. Comparison of theoretical models for particle losses in bends. R_c is the radius of curvature of the bend.

Figure 6. Comparison of bend losses for various line sizes.

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Recent Improvements in the Measurement of Respirator Fit Factors

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Abstract

Respirator fit factors as presently measured may be as much as two times higher or lower than the true value. Ideally, a fit factor would be measured with a perfectly mixed sample, an aerosol that has no losses, and a detector with the same response to all particle sizes. However, a sample taken from inside the facepiece may be unrepresentative due to streamlining, and the polydisperse aerosol currently used in fit testing is partially lost in the lungs and mask. Our experiments show this loss to be about 65% for particles with a diameter of $1\text{ }\mu\text{m}$. Since the forward-light-scattering photometer normally used for fit testing is more sensitive to these larger particles in the polydisperse aerosol, the photometer reading from inside the mask is about 65% low. We have designed an exhalation-valve sampler to give well-mixed samples and have designed and built a slit impactor with a cutoff of $0.42\text{ }\mu\text{m}$. When compared to the present impactor with a 0.1-inch diameter circular nozzle, ours decreases the concentration of particles larger than $0.55\text{ }\mu\text{m}$ by more than a factor of ten. Using the new impactor in fit testing results in a photometer signal that is much less affected by lung loss and thus gives a truer fit factor.

Introduction

As a continuing effort, the Safety Science Group is investigating various ways of increasing our understanding of respirator fit factors measured in the laboratory. In doing so, we can increase the accuracy of these laboratory measurements and more closely simulate the respirator fit—and hence, protection—that is obtained under actual work conditions. One inaccuracy that was first pointed out by Warren Myers of the National Institute of Occupational Safety and Health (NIOSH) is that the sample taken from inside the facepiece is not necessarily representative of what the wearer breathes.¹ Myers believes that contaminated air leaking into the facepiece through an imperfect face seal follows air streamlines that go directly from the leak site to the nose and/or mouth of the wearer, and the diffusion of the streamlines is so small that the contaminants are mostly missed by the sampling probe.

We reasoned that if Myers is correct, we could obtain a well-mixed sample by sampling the air exhaled from the respirator. Thus, we built an attachment to fit over the exhalation valves of the respirators that we were testing, which allowed us to sample air that had obviously been in the wearer's lungs. The questions then raised are: how much of the contaminant (test aerosol in this case)

was removed by the lungs, and how does this affect the response of the photometer used to get a relative measure of the aerosol concentration? The matter of lung loss not only affects exhalation valve sampling but also any sampling from inside a facepiece—when the wearer exhales, his breath blasts into the void volume between his face and the respirator and mixes with the air already there. All sampling probes pick up this mixture and then the pure exhaled air. The amount of mixture that is sampled before the pure exhaled air is a function of the amount of the void volume. Thus, sampling from a half mask would contain less of the mixture than sampling from a full facepiece. We see now that it would be desirable to know the amount of lung loss, as well as to reduce it, because of differences between masks and the likelihood that the lung loss will change the particle-size distribution and thus the response of the photometer.

The present study focuses on four aspects of the problem: the design of a fixture to sample the air exhaled from a respirator under test, the particle-size distribution of the challenge aerosol, the aerosol loss in the lungs as a function of particle size, and the response of the forward-light-scattering photometer as a function of particle size.

Exhalation Valve Sample Design

The design of the sampler must be such that it fastens onto or around the respirator's exhalation valve or valve cover without restricting the flow of exhaled air. A good rule of thumb is to make any new restrictions larger than those already in the valve or cover. For an MSA half mask we simply cut the exterior tabs off the valve cover and glued a thin-walled aluminum tube around it. The valve cover was glued to the valve body to make an airtight seal.

To sample from the MSA pressure-demand full facepiece, we made a thin aluminum cup-shaped fitting that goes between the rubber of the mask and the valve body, then extends out around the exhaust slits of the

valve. The thin-walled aluminum tube (about six inches long) is then attached.

Every different model of mask will probably need a specially designed attachment. A flexible piece of very light-weight tubing is attached to the exit end of the aluminum tube. We calculate the length of flexible tubing to be sufficient to prevent the challenge agent from being drawn so far up the tube during exhalation that it reaches the sampling probe point in the thin aluminum section. Figure 1 shows the sampling fixture that we used with the MSA pressure-demand UltraVue mask. The fixture has two sampling probes, since we were sampling for both aerosol and gaseous challenges.

Measurement of Lung Loss

The lung loss was measured in conjunction with any loss in the respirator valves, which also took into account the effect of the exhaled air mixing with the air already in the void volume of the respirator. This allowed us to study the total effect on measuring the concentration of challenge agent that leaked into the respirator. Our subject wore an MSA UltraTwin full facepiece fitted with High-Efficiency Particulate Air (HEPA) filters and the exhalation valve fitting described above. To get constant and slightly accelerated breathing, the subject walked on

a treadmill at 3.3 mph and a 2.5% slope. After we measured the concentration of aerosol as a function of particle size (the particle-size distribution) in the test chamber, the subject removed the HEPA filters from his respirator, allowing the challenge aerosol to be breathed into the facepiece unfiltered. After the concentration of aerosol in the exhalation fixture became constant (as measured by a photometer), the particle-size distribution of the aerosol in the exhalation fixture was measured.

Figure 1. Exhalation valve sampler. Note the two sampling probes, used for aerosol and gaseous mixtures.

From these two distributions, the "lung loss," or aerosol deposition in the whole system, was calculated. This is plotted as a function of particle diameter in Fig. 2. The plot agrees well with data from two other subjects who were exercising at a high work rate, and is very

similar in shape to published data of lung loss. We note that the deposition ranges from 20 to 40% between particle diameters of 0.1 and 0.55 μm (a correction we must make), and the deposition increases rapidly above 0.55 μm to 65% at 1 μm and higher for larger particles.

Response of the Photometer

The response of a forward-light-scattering photometer was measured by Biermann and Bergman.² It is presented in Fig. 3 as normalized photometer response vs particle diameter. Below about 0.6 μm diameter the photometer response increased with the cube of the diameter; above that it changes toward the square of the diameter. Again we note the normalized photometer response at the same particle diameters as noted for lung loss—at 0.1, 0.55, and 1 μm the responses are 0.0002, 0.035, and 0.1. Thus, we see that the photometer response

for 1 μm particles that have a lung loss of 65% is three times greater than for 0.55 μm particles where the lung loss is only 40%, and it is 500 times greater than the response for 0.1 μm particles where the lung loss is only 20%. Since it would be desirable to decrease the effect of lung loss on the response of the photometer, we decided to decrease the concentration of challenge particles with diameters above 0.55 μm . To do this, we designed an impactor to remove the larger particles from the challenge aerosol.

Design of the Slit Impactor

The aerosol we had previously used, and which is still used by most respirator test facilities, is produced by a generator that contains one or several Laskin nozzles and some type of baffle that stops very large particles. The design and operating characteristics of some models of Laskin nozzles can be found in Echols and Young³ and Hinds.⁴ This air-operated type of generator produces a

high concentration of an oil aerosol with a wide particle-size distribution. The larger particles are removed by an impactor (see Fig. 4), consisting of a single hole, 0.10 in. diameter, that impinges particles on a plate 0.10 in. away. The particle-size distribution thus produced is shown in Fig. 5 and is labeled as the "standard impactor." This plot shows the number of particles per unit volume in a

Figure 2. Aerosol deposition in lungs and mask as a function of particle diameter.

Figure 3. The response of the photometer increases with particle size.

Figure 4. The circular impactor used to remove large particles in this series of experiments.

Figure 5. Particle-size distributions for the standard impactor and the slit impactor.

given particle-diameter bin. To reduce the number of particles larger than $0.55\text{ }\mu\text{m}$, we needed to design an impactor with the impactor plate closer to the hole, which thus required smaller holes. To handle the required airflow rates, we would need either a large number of small circular holes or a long narrow slit. A slit impactor has advantages over an array of circular holes: The slit length can be extended to keep the pressure drop low; there is no adverse interaction between the air streams on adjacent holes; and the aerosol collected on the impactor plate can be blown off without being reentrained in the air stream.

The impactor design was based on the parameters published by Marple and Willeke.⁵ Using their equations and curves, we calculated the 50% cutoff diameter to be $0.49\text{ }\mu\text{m}$ for the impactor design shown in Figs. 6 and 7 with an airflow rate of $3.02 \times 10^{-2}\text{ ft}^3/\text{s}$ at standard conditions. A slit width of $0.610\text{ }\mu\text{m}$, and slit length of $10\text{ }\mu\text{m}$ were used. Polyethylene glycol oil with an average molecular weight of 400 (PEG 400) was used to produce the aerosol. The particle-size distribution thus produced is shown in Fig. 5, labelled as the "slit impactor." For comparison the two curves were normalized at $0.24\text{ }\mu\text{m}$ diameter.

Detailed consideration of the slit impactor data shows that the concentration falls sharply at the cutoff diameter, with the measured 50% cutoff being $0.42\text{ }\mu\text{m}$, which is 86% of the calculated value of $0.49\text{ }\mu\text{m}$. A comparison of the two curves shows that at the three particle diameters, 0.1, 0.55, and $1.0\text{ }\mu\text{m}$, the ratios of particle concentrations between the slit and standard impactors are 0.96, 0.19,

Figure 6. Cross section of the slit impactor.

and 0.00093. Thus, we have produced a challenge aerosol with a greatly reduced proportion of the larger particles that are more readily lost in the lungs.

Results

We can now put together these four aspects for improving the accuracy of laboratory respirator quantitative fit testing. First, we bring together the response curve of the photometer with the aerosol particle-size distribution. When we multiply the photometer response at each particle diameter by the aerosol number concentration at the same diameter, we get a curve that shows the photometer response to a particular aerosol. This curve is shown in Fig. 8 for both the aerosol from the standard impactor and the slit impactor. Comparing these two curves with Fig. 2 illustrates the importance of reducing the concentration of particles that are more readily lost in the lungs.

We now compare the apparent aerosol loss in the lungs and facemask as measured by the photometer for

the aerosols produced by the two impactors. Using the same subject working at the same rate and using our exhalation valve sampler, we repeated the experiment described under the "Measurement of Lung Loss" section above for both aerosols. This time, however, we measured the aerosol with a photometer, as listed in Table 1. The effect of lung loss with the standard impactor is 40%, whereas it is only 17% with slit impactor. We have succeeded in eliminating the inaccuracy due to loss of the large particles. We have only the 20% loss from Fig. 2 to be the minimum loss at any particle size. We can now correct our quantitative fit test results by this 20%.

Figure 7. Design of our slit impactor.

Figure 8. The particle-size distribution as seen by the photometer.

Table 1. Apparent aerosol loss, by photometer.

Impactor	Photometer response		
	Challenge	Exhaled air	Loss %
Standard	21.7	13	40
Slit	6.4	5.3	17

Conclusions

The accuracy of laboratory quantitative fit testing of respirators was improved by sampling the air that comes out of the exhalation valve, and also by replacing the standard challenge aerosol with one that has fewer large particles. The exhalation valve sampler eliminates errors due to possible streamlining from a leak site to the nose or mouth. The aerosol was improved by passing it through a slit-shaped impactor designed to have a cutoff

diameter of $0.49\ \mu\text{m}$. Larger particles produce a greater response in the light-scattering photometer, but they are also removed to a greater extent by the lungs. Minimum deposition of the aerosol in the lungs occurs between 0.04 and $0.4\ \mu\text{m}$. With this impactor the larger particles are removed from the aerosol, and the response of the photometer is not as greatly affected by lung deposition.

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RESOURCE MANAGEMENT

Determination of the Effectiveness and Proper Funding Level of Safety Programs

O. Clinton Kolar

Abstract

We propose a quantitative approach for planning and justifying resource allocations for safety organizations. We first develop data on the total safety cost (cost of incidents plus program cost) and compare this with the resources allocated to safety at a facility. With a sufficient number of data points from other DOE safety organizations, we can determine an optimum value for the amount of resources a facility should allocate to safety. This paper describes our method and reasoning and shows how the quantitative technique can be applicable throughout the DOE community.

Introduction and Background

This paper describes a quantitative approach for planning and justifying resource allocations for safety organizations. Frequently, the funding and staffing of such organizations is not based on statistical data or quantitative studies, but rather on organizational momentum and past experience. The new approach presented here provides a method for comparing the costs of safety incidents against the investment of time, manpower, and money for a safety program. If successfully administered, this approach could be quite convincing to resource managers attempting to determine appropriate staffing and funding levels for safety organizations at a facility.

We first develop data on the total safety cost (cost of incidents plus program cost) and compare this with the resources allocated to safety at a facility. If a sufficient

number of data points could be developed from other DOE safety organizations, we can then determine an optimum value for the amount of resources a facility should allocate to safety.

In 1985 C. Barnett and O. Kolar developed a model to compare the costs of a nuclear criticality safety program against the estimated cost of a criticality accident if safety measures were not implemented. The intent of this model was to determine an optimum expenditure for the safety program, but it was not developed with actual data for the costs of incidents and safety expenditures.¹ Glenn Briscoe of the System Safety Development Center (SSDC) at EG&G Idaho, Inc., has done a similar study.² The method described in this paper broadens the approach of this earlier work.

Description of the Method

DOE has designated three types of incidents—Type A, B, and C—according to degree of severity, with Type A being the most serious. These incidents occur in categories

related to people, property, environment, or other special circumstances, and can refer to injuries or illnesses, radiation exposures, property damage, the release of

pollutants or hazardous materials to the environment, or other occurrences that affect the operation and the public perception of the operation. A complete description of Type A, B, and C incidents is given in Refs. 3 and 4.

For our determination, we must find the total costs of all safety incidents at a facility. SSDC maintains permanent databases of Type A, B, and C incidents, which include records of costs over the years. Our data sources will be SSDC for DOE data, and possibly the commercial database company Silver Platter for NIOSH (a U.S. database), HSELINE (a U.K. database), and CISDOC (a U.N. database) information. We may discover that a facility experiences many incidents that are too small to be classed even as Type C incidents, but because of their relatively large number, these incidents must be included in the determination of total cost. Cost data for these small incidents may not be available, however, and we have not yet resolved this problem.

Assuming that cost data can be developed, we can create a plot such as Fig. 1. In the figure, C_s is the cost of the safety program as a percent of the facility's operating

budget; C_i is the cost of Type C and unclassified incidents as a percent of the operating budget; the total cost, C_t , is the sum of these two costs, $C_t = C_s + C_i$.

We expect that with more resources devoted to a safety program, an operation will become safer. Mathematically speaking, C_i vs C_s must be monotonically decreasing. Therefore, the total cost, C_t vs C_s , must have a minimum at some value of $C_s < 100\%$.

On Fig. 1 we expect the curves for C_i and C_t to be fairly smooth if they include only high-probability/low-consequence incidents. Relatively speaking, Type C and unclassified incidents probably fit this criterion. Type A incidents, though, are low-probability/high-consequence incidents, which are rare and unique, making generalizations difficult. DOE has had 98 Type A incidents since 1980, six of which occurred at LLNL. Though we will not include Type A incidents in the analysis described above, we do show in Fig. 2 a conceptual representation of how such incidents are likely to be distributed. Despite the large scatter, a downward trend is still apparent.

Figure 1. The total cost of safety at a facility will be the amount spent on a safety program and the cost of safety incidents that occur. This sum should have a minimum at some optimum level of funding for a safety program. Note, this generalization holds only for high-probability/low-consequence incidents.

Figure 2. Low-probability/high-consequence incidents are each unique and do not allow for generalizations. However, they should exhibit a downward trend as the cost of a safety program increases.

In our approach we are looking for a value of C_s that will result in a sufficiently low expectation value of the number of Type A incidents, N_A . With a statistical treatment of the data we can determine the value of C_s ($= C_{s2}$) that yields this acceptably low expectation value $\langle N_A \rangle$. Management must decide how often it is acceptable to have a serious accident, e.g., one per year, or one per ten years, or any other rate. This gives us a parameter, P , that places an upper limit on incident frequency by setting $\langle N_A \rangle = P$. The cost of a safety program decreases as management decides to allow the frequency of incidents to increase. Figure 3 shows qualitatively how C_{s2} varies with P . $\langle N_A \rangle$ is therefore a function of C_s , and we can determine an appropriate level of funding for a safety program by specifying an acceptable incident frequency. As the acceptable frequency goes down, the cost of a safety program will increase.

We now have two values of C_s : C_{s1} from the initial treatment shown in Fig. 1, and C_{s2} from the treatment suggested in Fig. 3. The value that management should use to determine operating levels for a safety program should be determined as:

$$C_{s0} \geq \max(C_{s1}, C_{s2}).$$

Figure 3. Another parameter for determining the optimum C_s is for management to choose an acceptable frequency of occurrence, P , for Type A incidents. As the acceptable frequency of such incidents decreases, the cost of the safety program will increase.

Preliminary Results

Because C_s data is not available to us now, we have resorted to a preliminary analysis. This analysis consists of ranking all 229 DOE organizations in the SSDC database according to number of incidents of a given Type experienced per 10^5 man-hours. The organization with the largest number of incidents is given a rank of 1, the one with the second largest number a rank of 2, and so on.

We have plotted the data points for 229 organizations, given in Figs. 4-6. Our information reflects incidents that occurred from 1980 through mid-1987: 31 organizations had one or more Type A incidents (Fig. 4), 34 organizations had one or more Type B incidents (Fig. 5). The many Type C incidents are shown in Fig. 6. The data point for LLNL is indicated on each figure.

Both Figs. 4 and 5 show a difference of about 2 orders of magnitude between the best and worst incident experience. Note, though, that this information reflects only those organizations that experienced A or B incidents at all. Of the 229 organizations, 198 experienced no

Type A incidents during the time interval, and 195 experienced no Type B incidents. Among those organizations experiencing at least one Type A or one Type B incident, LLNL is in the "good experience" portion of each of the corresponding curves. From Fig. 6 we can see that LLNL does not fare as well among Type C incidents. Figure 7 displays the cost of Type C incidents at each organization.

We investigated whether an organization that experienced a large number of Type C incidents also experienced a large number of Type A incidents. To do this, we plotted lines that contain 90% of the whole population of points for organizations that had both Type A and Type C incidents, and also the 50% lines of the same population of points (Fig. 8). An inspection of the figures shows a loose correlation, but more-detailed statistical analysis will be required to confirm this. In Fig. 9, we have made the same type of plots, attempting to find a correlation between Type B and Type C incidents.

Figure 4. Organizations ranked by number of Type A incidents.

Figure 5. Organizations ranked by number of Type B incidents.

Figure 6. Organizations ranked by number of Type C incidents.

Figure 7. Organizations ranked by total cost of Type C incidents.

Figure 8. Only a loose correlation exists between organizations that experience a lot of Type A incidents and those that experience a lot of Type C incidents.

Figure 9. As in Fig. 8, only a loose correlation exists between organizations that experience a large number of Type B incidents and those that experience a large number of Type C incidents.

Conclusions

Preliminary analyses permit us to look at the variations in C_i , N_{i1} , N_{i2} , and N_{i3} . If these variables change dramatically among the different organizations, we should try to understand why. This could lead to any of several conclusions about the variations.

- Differences in C_i among the organizations
- Differences in the effectiveness of each safety program (after normalization of the C_i differences)
- Differences in the degree of hazard levels among the organizations, or
- Differences in intrinsic costs of incidents among the organizations

Note that this article describes work in progress and all conclusions are only tentative. Our approach, if successful, will apply equally well to *all* DOE organizations and can be used throughout the DOE complex. Given the numbers we hope to develop, other safety programs will be able to determine the appropriateness of their own funding and staffing levels. This method can also provide a gauge as to the effectiveness of various safety programs by comparing safety programs at organizations that have similar C_i values but significantly different incident experiences.

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DOSIMETRY AND RADIATION PHYSICS

Recent Developments in CR-39 Personnel Neutron Dosimetry

Dale E. Hankins, Steven G. Homann, and Joane Westermarck

Abstract

We are continuing to study various technical aspects of electrochemically etched CR-39 foils used as personnel neutron dosimeters. These studies include determining the energy response and the changes in the energy dependence caused by etching the back of the foil. We are also performing measurements of the angular dependence in air and on a phantom as a function of neutron energy. A method to identify the individual CR-39 foils by using laser numbering is also being investigated.

Introduction

For about four years we have been using CR-39 as our personnel neutron dosimeter at LLNL. During this time we have improved the dosimetry system by making

a number of changes in the etching and reading procedures. In this report we discuss these changes and describe the results obtained from additional studies.

Etching Procedure

CR-39 foils can be etched using several different techniques, but the particular procedure used will change the energy dependence, background, and sensitivity of the foils. It is therefore important that the etching procedures and etching parameters be specified, especially if dosimetry results are to be compared.¹

Our etching parameters are given in Table 1. We use the Homann-Bell power supply described previously.² The second etching step (called the "blow-up" step) is very important because it increases the size of the tracks that exist at the end of the first etch step. This greatly improves the precision that can be attained with the

Biotran optical bacterial colony reader (used for track counting), because the tracks are large compared to the imperfections, dirt, and scratches often present on CR-39 foils. The background from the foils is reduced by using a higher track-size threshold on the reader, which allows us to discriminate against most of these small imperfections. Six fields of view, 3×3 mm, are counted by starting at the top-center of the etched area on the foil and moving down the center of the foil. The precision of the Biotran results are typically within $\pm 1\%$ for repeated readings of the same foil. The "Operating Procedures for Electrochemical Etching of CR-39" is given elsewhere.³

Table 1. Recommended etching parameters.

	Etching	Blow up	Post Etch
High Voltage	3000 V	3000 V	0
Frequency	60 Hz	2.0 KHz	0
Temperature	60°C ^a	60°C	60°C
Time	5 hours	23 min ^b	about 15 min
KOH normality	6.5 N	6.5 N	6.5 N

^a The etch chambers and KOH must be left in the oven overnight (or weekend).

^b No adjustment for foil thickness is required if the foils are within 3 mil of 25 mils.

Neutron Energy Response

Tommasino etched CR-39 foils using a low-frequency electrochemical process at elevated temperatures. He found that this produces a different energy dependence than that obtained with high-frequency or chemical etching of CR-39 foils.⁴ The energy dependence he obtained was better for application in personnel neutron dosimetry. In the past two years we have investigated this low-frequency etching technique extensively.

Figure 1 shows the energy response as a function of the etching voltage. These results were obtained using monoenergetic neutrons (with energies below 1.0 MeV) from the Tandem Van de Graaff accelerator at the Los Alamos National Laboratory, and (for neutron energies above 1.0 MeV) an accelerator at the Battelle Northwest Laboratory.

The curves shown in Fig. 1 were obtained using voltages from 0-3500 V for the first etching cycle. The etching time for the blow-up step was varied to give the same track sizes on the foils. The effect of increasing the high voltage is most dramatic in the region below 1 MeV, where the response to the lower-energy neutrons is progressively increasing. At 3000 V the energy response is relatively flat from about 150 keV to about 4.0 MeV.

The lowest threshold energy occurs when 3500 V etching voltage is used. The background tracks increase to an unacceptable level at this voltage, however, eliminating it as a useful etching potential.

The response of CR-39 to neutron energies around 14 MeV is about 42% of its response to a ²⁵²Cf source. We had hoped that changes in the etching potential could increase the sensitivity of the CR-39 to these high neutron energies, but we have found that this cannot be done.

We do not understand the peculiar shape of the response obtained using an etching voltage of 1500 V.

Also the results at 2000 V etching voltage indicate no response to neutron energies below about 200 keV, but a response can be seen when higher or lower voltages are used. The response to ²⁵²Cf neutrons at the left of Fig. 1 is very close to the response obtained with 1.0-MeV neutrons.

We obtained the results shown in Fig. 1 with CR-39 foils that were exposed with the side to be etched oriented toward the incident neutrons. When the side away from the source and next to the phantom is etched, a different energy dependence is obtained. Figure 2 shows this difference in the energy response. We now routinely etch the side of the foil that was next to the wearer because it results in an improved directional response.

The difference between the two curves above 1.0 MeV is caused by protons that enter the foil from the polyethylene protective layer on the foils. This polyethylene prevents damage to the foil by abrasion and alpha particles. (The polyethylene layer is present on both sides of the foil, but the layer on the back, or exit side, does not affect the energy dependence for neutrons incident on the front.) Polyethylene has a higher hydrogen content than CR-39 and therefore generates more recoil protons. Some of the protons originating in the polyethylene from neutrons having energies above 1.0 MeV reach the CR-39 and caused damage that becomes a track when etched. Below 1.0 MeV the protons originating in polyethylene either do not reach the CR-39 or have too little energy to damage the foil.

Foils etched on the back have an energy dependence that is very flat from 1.0-4.0 MeV. They also have a low-energy cutoff at about 150 keV, and have an oversensitivity at 250 keV of about 30%. The oversensitivity at 250 keV is of little concern because

Figure 1. Energy dependence of electrochemically etched CR-39 foils as a function of the etching high voltages using a five-hour etching procedure.

Figure 2. Energy dependence of electrochemically etched CR-39 foils as a function of etching high voltages using a five-hour etching procedure. The energy dependence of foils etched on the front side (incident) is different from the energy dependence of those etched on the back side (against the phantom).

most field spectra contain few neutrons in this energy region; the overresponse may also help to compensate for the lack of sensitivity to neutrons having energies below 150 keV.

With our processing techniques, CR-39 does not respond to neutrons with energies below about 0.05 MeV. CR-39 also has an underresponse to neutrons with

energies between 13–16 MeV (Fig. 1). Our studies have shown that the relative response to high-energy neutrons and to other neutron energies is not affected by changes in the etching parameters. A more accurate estimate of the contribution from neutrons between 13–16 MeV can be made, however, by using the track-size distribution.⁹

Directional Dependence

The directional dependence of CR-39 is a strong function of the neutrons' angle of incidence, as shown in Fig. 3. As the angle approaches 90°, the response of the CR-39 foil drops rapidly. This is caused by the recoil protons entering almost parallel to the surface of the foil. When the foil is etched, no track develops.

Figure 3 shows that the track density is less for neutrons entering the foils through the back (180°), than for those entering through the front (0°). The CR-39 sheets have a 5-mil-thick polyethylene protective covering. As mentioned earlier, polyethylene has a greater hydrogen density than CR-39, and therefore the proton production (n,p) with hydrogen is greater. Consequently, the side of the foil facing the source has a higher track density than the back side by about 25%. By etching the side of the foil next to the wearer, we can use this effect to improve the directional dependence.

Figure 3. Directional dependence of CR-39 foils in air.

Figure 4 shows the directional response obtained from CR-39 foils in a personnel badge located on a phantom and exposed to a ^{252}Cf source. The lower solid curve, obtained by etching the side of the foil next to the wearer, is the best directional dependence attainable at present. This directional response decreases to about 20% at 90°, but this is in reasonable agreement with the ~30% decrease in the 1-cm-depth dose delivered to tissue exposed at 90° (see Ref. 6).

Figure 4. Directional response of CR-39 in a personnel badge on a phantom.

Figure 5. Directional dependence in air (top) and on a phantom (bottom) of CR-39 foils as a function of the neutron energy.

We recently completed a study of the directional response of CR-39 as a function of the neutron energy. We included foils exposed in air and on a phantom to determine the effect of the phantom on directional response (see Fig. 5). The foils were exposed with the side to be etched next to the phantom, or on the back for the foils exposed in air. The shapes of the response curves are similar, indicating that the phantom does not affect the directional response.

Figure 5 shows that for neutron energies between 0.3 and 5.0 MeV, the directional response is essentially the same, and we show a single solid response curve for all these energies. The solid response curve is in good agreement with the directional response shown in Fig. 4 obtained with the ^{252}Cf source. However, at 15 MeV the directional response drops less at large incident angles, resulting in a more favorable directional response. At neutron energies below about 0.3 MeV, the response at larger angles decreases and becomes less favorable.

Stability and Fading

A recently completed one-year study of storage conditions indicates that when the foils are protected from light, little if any fading or change in sensitivity occurs. The background increases at a rate consistent with the environmental neutron background. In contrast, foils that were not protected from ambient light faded, lost sensitivity, and suffered an increase in background to the point where the foils were useless.

CR-39 foils are also damaged by UV light or prolonged exposure to high temperatures. The mechanism for damage is not known, but may be associated with the adhesive that is on the foils' protective polyethylene cover. When the foils are exposed to UV or ambient room light for several months, the surfaces of the etched foils have many small dots or globs of material on them. These dots apparently cause tracks to appear on the foil. As these dots become larger from additional exposure to light or heat, the number of background tracks increases. The background tracks are denser in areas on the foils where these small dots are more dense. We believe that these dots are the adhesive on the

To compensate for the directional dependence, we apply a correction factor to our dosimetry results. We selected a calibration value 20% lower than the results obtained from a face-on (0°) exposure. This means that if a person were exposed face-on to the source, his or her actual dose equivalent could be overestimated by 20%. But for exposures at the higher angles of incidence, the error in underestimating a dose equivalent would be reduced. The disadvantage of etching the back of the foil is that the sensitivity on the back side of the foils is about 30% less than on the front. This reduces the sensitivity of the dosimetry system from about 8 to 5 tracks/cm²/mrem. The background track density on the CR-39 foil is not changed, so the effective dose equivalent of the background is increased. We feel the improvement in the directional dependence justifies the changes in the sensitivity and background.

polyethylene, which has deteriorated and remains on the foils when the polyethylene is removed.

If care is taken to protect the foils from light or excessive heat, the foils are useful for at least one year. The CR-39 foils could be issued for a six-month exchange period. It should be remembered, however, that the background track density on the foils will be increased by the environmental neutron background. To keep the background of the CR-39 foils as low as possible, only fresh foils should be issued.

New foils must be stored in the dark. Earlier we stored the material in a refrigerator or freezer, but the results from our one-year study of storage techniques indicate that reduced temperatures are not necessary. Storage in a refrigerator, however, does provide a convenient guarantee of a dark environment.

We conclude from these studies that fading, changes in sensitivity, or increases in the foil background are not a problem if the foils are protected from light or excessive heat. The CR-39 foils are stable for routine use as personnel neutron dosimeters.

Laser Numbering of CR-39 Foils

In a routine dosimetry program, a rapid, convenient system for foil identification is essential. We have tested laser numbering of the foils using the same laser system that cuts the foils. The power of the cutting laser is reduced to etch letters/numbers at the bottom or the top of the foil. A range of sizes can be used for the letters or numbers. Laser numbering has the advantage that it

clearly indicates the side of the foil that must be etched, and assigns a permanent, unique number to it. This eliminates the handling time required to label each foil with a flow pen or a bar code. We are presently developing a method to read the laser numbers using an image-analyzer system.

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Use of Neutron-Sensitive Personnel TLDs As Flags for CR-39 Processing

Dale E. Hankins

Abstract

To avoid the cost and time required to read all of the CR-39 dosimetry foils issued to personnel, we made a study to determine if we could use thermoluminescent dosimeter (TLD) readings from the thermal-neutron-sensitive TLD in our Panasonic personnel badge as a flag to indicate if a particular CR-39 foil should be processed. We found this to be true: only a small percentage of low neutron doses would be missed if the TLD-flag procedure were used.

Introduction

We reviewed the results obtained with neutron-sensitive thermoluminescent dosimeters (TLDs) in LLNL personnel badges (Panasonic 810 TLD) and our new CR-39 foils over the past nine months. We intended to determine if the thermal-neutron-sensitive TLD in the Panasonic badge can be used as a flag to indicate when a particular CR-39 foil should be read. Such a flag would enable dosimetry organizations that issue large numbers of CR-39 badges to determine which badges had actually been exposed to neutrons. This method would save time and cost in etching and reading CR-39 foils that had not been exposed.

We obtained the neutron dose interpretation from our Panasonic badge by subtracting the reading of one of

the gamma-sensing TLDs from that of the thermal-neutron-sensing TLD. An empirical work-area-dependent conversion factor is used to obtain an estimate of the fast-neutron dose. Although imperfect due to the variation of the fast-to-thermal neutron ratio in a given area, the Panasonic dosimeter can give an indication of the fast-neutron dose in the absence of a CR-39 dosimeter.

The Panasonic neutron reading obtained in this way would not be the same as a reading obtained at other facilities, but the basic principle would apply. A reading from the thermal-neutron-sensing TLD that exceeds a certain threshold value could be used to indicate which CR-39 foils should be etched.

Results

Table 1 summarizes the ratios of CR-39 to TLD dose, estimated for about 110 positive CR-39 indications over a nine-month period. We found that these ratios varied with work location. The vault personnel receive much of their exposures in the Pu vault where the thermal-neutron component is higher than at other locations at LLNL; as a result, these ratios average 0.65.

Work around the ^{239}Pu heat sources, the High-Performance Generators, is shielded by four inches of polyethylene, and the ratios average 1.0. In other areas at LLNL, where the neutron sources are only lightly shielded, the ratios average 1.2.

The differences in the ratios reflect the variation in the thermal-neutron component and again confirm that one cannot use thermal neutrons to accurately measure the fast-neutron dose. The ratios vary from 0.34 to 1.9, which is about a factor of 6. The range of these ratios would be greater if LLNL personnel were exposed to unshielded sources or to heavily moderated sources, as might be expected at some DOE facilities.

One conclusion that can be drawn from Table 1 is that we have been applying the correct conversion factor to the Panasonic thermal-sensing TLD to convert its reading to fast-neutron dose. Both high and low readings were obtained from field results.

Table 1. Ratio of the CR-39 reading to that obtained with the Panasonic badge. We found about 110 positive CR-39 readings (>10 mrem) covering nine months.

Group	Range	Average
Vault personnel	0.34-1.7	0.65
High-performance generators (^{238}Pu heat sources)	0.56-1.7	1.0
Others	0.42-1.9	1.2

Table 2. If a reading of 10 mrem on the Panasonic TLD had been selected as the criteria for etching the CR-39 foils, the following CR-39 readings would have been missed.

Panasonic reading (mrem)	CR-39 reading (mrem)
0 (Gamma = 483 mrem)	17
2	14
9	24
9	10
5	10
7	20
	[total = 5]

Table 2 lists the number of positive (>10 mrem) CR-39 readings obtained from foils in badges where the Panasonic TLD readings were less than 10 mrem. The first reading was from a badge where the worker received 483 mrem of gamma exposure. With this large a gamma reading, the neutron dose of 17 mrem only represents 3.5% of the total dose, and is not considered to be a miss. However, this result shows that if the gamma reading on a badge is high, one should process the CR-39 badge to assure that a small neutron exposure is not being overlooked.

We found five other instances where the Panasonic TLD dose estimates were below 10 mrem and the CR-39 reading was above 10 mrem. The results in Table 2 show that if 10 mrem were selected as the flag for etching the CR-39, we would miss about 4% of the neutron doses ranging from 10 (our threshold) to about 25 mrem. It is unlikely that higher doses would be missed by the Panasonic TLD because these doses give an easily detectable TLD response. Missing only 4% of the smaller exposures would probably be considered acceptable at many facilities. In fact, some facilities are presently using detection thresholds of 20 and 30 mrem.

If missing 4% of the doses is unacceptable, the TLD action level (the flag) could be set at 5 mrem. Then only one of the positive CR-39 readings given in Table 2 would have been missed. However, with the 5-mrem action level we would then need to process an additional 50 unexposed CR-39 dosimeters (Table 3).

Some of the "missed" CR-39 readings in Table 2 may have actually been CR-39 foils with high backgrounds, rather than true doses. In the near future we hope to eliminate this problem by using track-size distributions to distinguish foils with high backgrounds from foils exposed to neutrons.

Table 3 shows the instances in which TLD dose estimates were above 10 mrem, but the corresponding CR-39 foils indicated less than 10 mrem. If a Panasonic reading of 10 mrem is used as the flag for etching the CR-39, 43 badges out of about 500, or less than 9%, would have been etched unnecessarily. Of these, 22 were from vault personnel who work in an environment with a higher thermal-neutron fraction, which creates a larger TLD response. Only eight badges from personnel in other areas of LLNL would have been etched unnecessarily, except for the badges from two health and

Table 3. If a 10 mrem reading on the Panasonic badge had been selected as the criteria for etching the CR-39 foils, the following badges would have been etched unnecessarily. The numbers given are the Panasonic TLD readings in mrem.

Materials Management			H P Techs		Others
			(A)	(B)	
10	20	17	12	16	14
16	16	11	34	2	10
11	14	26	24	14	19
15	22	20	12	22	12
24	10	26	19	10	13
26	16	10	12	6	12
13			7	16	17
			17	17	12
			8	30-(14)	
[total = 22]			[total = 13]		[total = 8]

(If a 5 mrem reading were selected an additional 50 badges would have been etched unnecessarily.)

safety technicians who work in the plutonium building. We have listed the monthly readings from their Panasonic badges. Only one CR-39 foil had a positive reading (14 mrem). The technicians are receiving some neutron exposure in fields containing a large component of thermal neutrons, but the fast-neutron dose is below the 10 mrem threshold for the CR-39 foils.

As mentioned in Table 3, if the action level were decreased to 5 mrem, this would increase by 50 the

number of badges etched unnecessarily. An increase of 50 is less than expected and indicates that setting the flag at 5 mrem would not unreasonably increase the number of badges to be etched. It would reduce to one the number of "missed" positive CR-39 readings given in Table 1. This confirms that we rarely get a positive reading from a TLD that has not been exposed to neutrons.

Summary

Using the results from personnel badge TLDs, we conclude that a thermal-neutron-sensing TLD can be used as a flag to determine if a CR-39 foil should be etched. Only a small percent of the smaller neutron doses would be missed, but the result would be a

significant savings in time and effort. A similar field study should be made at other DOE laboratories (using their personnel dosimeters) to determine what TLD dose level should be used as an action-level flag at these laboratories.

Study of the Spectrum-Dependent Track-Size Distribution of Electrochemically Etched CR-39 Foils

Dale E. Hankins, Steven G. Homann, and Joane Westermarck

Abstract

We have investigated the parameters that affect the track-size distribution on electrochemically etched CR-39 foils. We have found that the largest differences in track-size distribution occur when the foils are etched at 3000 V for 3 hours at 60 Hz, followed by 40 minutes at 2000 Hz. Track-size analysis can be used to determine (within limits) information on the incident neutron spectrum and to determine if the reading on a foil is a true neutron exposure or a foil with a high background. A new image analyzer is being developed at LLNL that will allow track-size analysis without increasing the time presently required to evaluate the foils.

We have shown that useful information on the incident neutron spectrum can be derived from the track sizes on CR-39 foils used for our new dosimetry system.¹ Although the electrochemical etching process alters the track size on the foils, we found that the area of the track is still related to the neutron energy. This is true even after the blow-up stage is completed if the CR-39 has been properly etched. The relationship between track size and neutron energy can provide some information on the incident neutron spectrum, although detailed data cannot be obtained. Track-size distributions can also be used to determine whether the dose indicated by the CR-39 is from a true neutron exposure or simply caused by a foil with a high background. However, these size distributions are affected by the etching parameters including oven temperature, etching high voltage, etching time, and blow-up time.

We measure the foil's track-size distribution by using the Biotran Bacterial Colony counter, which records the area of an object in the field of view. To improve resolution, we increased the magnification of the microscope from 40 $\times$ (used for normal track counting) to 200 $\times$. We usually measure at least 300 tracks on each foil. The measurements are recorded by an HP-85 computer programmed to digitize the track sizes. The digitized track sizes are plotted as a bar graph with a curve drawn connecting the points.

We studied the effects of various etching times on the track-size distributions. We performed a series of etches at 2, 3, 4, and 5 hours with blow-up times of 45, 35,

28, and 23 minutes, respectively. The track-size distributions are shown in Fig. 1. The results from the two-hour etching cycle are not shown since the track-size distributions changed only slightly for the different neutron energies. The greatest difference in track-size distribution occurred for the three-hour etch cycle, and was most noticeable at a neutron energy of 499 keV, where increasing the etching time caused more of the small tracks to appear. When we used a five-hour etch cycle, the track-size distributions from neutron energies of 499 keV and 3.0 MeV could be misidentified, which was not a problem for the three-hour etch cycle.

As the etching time is increased, a shift to smaller track sizes occurs for the higher-energy neutrons, and the track-size distribution flattens for neutrons around 1.2 MeV, as shown in Fig. 1. For all etching cycles the 14–16 MeV neutrons have track sizes that exceed those from ²⁵²Cf sources, making them easy to distinguish. The three-hour etch cycle gives the largest variation in track-size distributions, but we are still investigating changes in the blow-up time and other etching parameters to see if larger variations in the distributions can be obtained. However, based on our results to date, we do not expect to see significant changes in the etching parameters or the track-size distributions.

The differences in the track sizes are caused by a combination of the proton absorption curve and the intrinsic response of the CR-39 to protons. The absorption curve (dE/dx) peaks rather sharply at 60, 70, and 100 keV for hydrogen, carbon, and oxygen, respectively. The

Figure 1. The effect of changes in the etching time on the track-size distributions obtained with mono-energetic neutrons and ^{252}Cf sources.

energy of the recoil protons produced in the CR-39 plastic (or those entering the CR-39 from the polyethylene protective cover) have an essentially uniform energy distribution. However, CR-39 is not sensitive to low proton energies, nor does it respond to protons from high angles of incidence. The combination of these effects is optimized at neutron energies around 500 keV, resulting in the fairly sharp peaking of the track-size distribution from neutrons with these energies.

As the energy of the neutron increases above about 800 keV, the absorption curve drops rapidly; the higher-energy neutrons damage the foil less, producing smaller tracks. The increase in these smaller tracks causes the

peak of the track-size distribution to move toward the smaller track sizes shown in the figure. As the neutron energy is increased further (above 6.0 MeV), the recoil protons are too energetic to damage the foil significantly, and few tracks are formed.

At neutron energies of 14 MeV the tracks formed are from recoil carbon and oxygen atoms that cause considerable damage to the CR-39, producing very large tracks. At higher neutron energies alpha particles and spallation products are produced, which again heavily damage the CR-39 and cause very large tracks.

Figure 2 shows the results obtained from foils exposed to reactor leakage neutrons and high-energy

Figure 2. Track-size distributions obtained from CR-39 foils exposed to leakage neutrons at a power reactor and to high-energy neutrons at the LBL Bevatron accelerator. Also shown are the track-size distributions on CR-39 foils worn as personnel badges at various work locations at LLNL.

neutrons from the Lawrence Berkeley Laboratory (LBL) Bevatron accelerator. The track-size distribution on foils exposed at the power reactor consists mostly of large tracks. The reactor-leakage spectrum contains neutrons with energies primarily in the 100-keV to 1.0-MeV region, which causes this abundance of large tracks. Foils exposed at the LBL Bevatron have track-size distributions that include many tracks larger than can be obtained with fission neutrons. Both the reactor and LBL track-size distributions can be easily distinguished from ^{252}Cf neutrons. The dosimeter readings obtained from the track-density evaluation for both of these spectra are low by about 50% and must be corrected by personnel doing dosimetry evaluations.

The right side of Fig. 2 shows recent track-size distributions obtained from spontaneous-fission neutrons on CR-39 foils actually worn by personnel. The track-size distribution on the badges exposed in the storage vault indicates large numbers of low-energy (scattered)

neutrons. This track-size distribution is very similar to that obtained at a reactor. The track-size distributions labeled "shielded ^{239}Pu " and "radiochemistry bldg" were from exposures through polyethylene and water shielding, respectively, and are very similar. The "Pu bldg" distribution was from essentially unshielded spontaneous-fission neutrons. The track-size distributions in Fig. 2 show enough differences to allow us to determine where an exposure occurred. These distributions are also significantly different from the background foils, which confirms the exposures are real and not from a high-background foil.

The track-size distribution is considerably degraded if the back of the foil is etched—but etching the front of the foil degrades the CR-39's directional dependence. This means that using the track-size distribution method yields a poorer directional dependence of the CR-39.

The etching parameters that can be used to determine the track-size distribution are limited. A high sensitivity

to low-energy neutrons must be retained and (if possible) enhanced, which requires that the etching voltage be kept at around 3000 V. High sensitivity must be retained if the foils are to be used for personnel dosimetry; this requires that the etching time be as long as possible and that the oven temperature be kept high.

Decreasing the etching time from the standard five-hour etch cycle to the three-hour etch and 40-minute blow-up cycle reduces the track density by about 30%. But, to obtain the best track-size distributions, it is necessary to use the front of the foils—which increases the sensitivity by about 30%, although the directional dependence suffers. The net effect is that the sensitivity is about the same as what we presently obtain with our dosimetry system.

We are also investigating track-size distributions on background foils that have unusually high track densities. To date, all but one of the high-background foils have a different track-size distribution than foils exposed to neutrons. However, most of the recent CR-39 foils from American Acrylics have a uniformly low background, making such a study difficult.

Image analysis is preferred if personnel could be exposed to several different neutron spectra, such as fission neutron, reactor-leakage neutron, high-energy neutron (>10 MeV), or heavily moderated fission spectra. For spectra where the CR-39 underresponds (reactor leakage neutrons and 14-MeV neutrons), the track-size distributions are greatly different. This indicates to dosimetry personnel that an exposure to these spectra has occurred, and allows them to correct the low dosimeter readings.

We are developing the software and assembling the hardware for a new image analyzer that will rapidly determine the track density and the track-size distributions on CR-39 foils. This analyzer will determine the track density and display the track-size distribution within the same time now required to determine only the track density using the Biotran reader. This new system is at a cost comparable to that of the biological colony reader.

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The Response of a Liquid-Scintillator Fast-Neutron Spectrometer as a Function of the Angle of Neutron Incidence

John H. Thorngate

Abstract

We have conducted measurements that show the angle of neutron incidence does not affect the fast-neutron spectra measured with a liquid-scintillator if the scintillator has the shape of a right circular cylinder with equal diameter and height. This is true even when efficiency functions used to reduce the data assume a plane wave of neutrons incident along the axis of the scintillator.

Introduction

Our program to develop neutron spectrometers for radiation-protection applications concluded that an organic-liquid scintillator is the optimum detector for measuring the spectrum of neutrons with energies above 1 MeV. Because the scintillator is a liquid, it must be contained in some sort of cell. Normally, these cells are right circular cylinders, most often with the diameter equal to the height. Recently, we published a report describing our work to optimize the detector, the data-acquisition procedures, and the data-reduction programs for this type of spectrometer.¹

The comprehensive report does not include information about the response of a liquid scintillator when the neutrons are incident at an angle to the axis.

Since an isotropic source of neutrons with energies above 1 MeV would be unusual, and since neutrons with such energies usually travel from the source to the detector with a minimum of scattering, we had simplified our calculations for detector efficiency by assuming that the neutrons impinged in a plane wave along the axis of the cell. These efficiency values may not be correct for reducing data where significant scattering occurs, however, since the path lengths traversed in the cell by neutrons impinging at an angle to the axis are different from those traversing the cell parallel to the axis.

This brief report will discuss the results of measurements we made to evaluate the effect of neutrons incident at an angle to the axis of the cell.

Procedures

We used one of our newer 50.8 mm diameter by 50.8 mm high organic-liquid scintillators for this experiment. This scintillator is the white cylinder shown at the right end of the detector assembly in Fig. 1. The photograph also shows the light pipe, photomultiplier tube, and the special tube base used with this detector. For a cylinder this size, the irradiated area (detector face) is 2027 mm². Every neutron in a plane wave incident along the axis has the same 50.8-mm path length within which an interaction can occur. The same beam of neutrons, incident perpendicular to the axis of the cylinder (from the side), would have path lengths that vary from

zero up to 50.8 mm, and the irradiated area (detector cross section) would be 2581 mm². Irradiations at angles between these extremes produce intermediate path lengths and areas.

We designed this experiment to measure the neutron spectrum produced by a small ²³⁹PuBe source located one meter from the center of the liquid scintillator cell. The dynamic range of the pulse-shape discriminator unit limited the neutron energy range for a single measurement between 1.5 and 12 MeV. Measurements were made with the source-detector line at four angles with respect to the axis of the scintillator cell. We defined

Figure 1. Photograph of the detector assembly used in these experiments. The liquid-scintillator cell is the white cylinder at the right end of the assembly.

the source-detector line parallel to the axis of the cell as 0° , and 90° as the line perpendicular to the axis. (See Fig. 2.) Other angles measured were 30° and 60° . In every case, we reduced the data using the efficiency values calculated for 0° incidence.

Our experimental system is shown in Fig. 3. The electronic components we used were a Tennelec TC 951 high-voltage supply run at -1700 volts, a Tennelec TC

202BLR linear amplifier operated with $1\text{-}\mu\text{s}$ pulse shaping and a gain of 52.5, and an Ortec 442 linear-gate/stretcher. We recorded the signals in 512 channel groups using a Nuclear Data ND 620 multichannel analyzer. To separate the neutron-produced signals from those produced by gamma rays, we used a Link Systems PSD 5010 pulse-shape discriminator (PSD) operated at a 30% threshold with a 47% separation and a 29% rotation. For the gamma-ray calibrations of the pulse-height scale, the gamma signals from the PSD unit were used to open the linear gate to allow the output of the linear amplifier to reach the multichannel analyzer. The neutron signals from the PSD operated the linear gate during the neutron measurements.

Before and after each neutron measurement, gain calibrations were made using the 0.570-, 1.064-, and 1.770-MeV gamma rays from ^{209}Bi , the 0.511- and 1.275-MeV gamma rays from ^{22}Na , and the 4.439-MeV carbon deexcitation gamma rays from a PuBe neutron source. For these measurements the sources were positioned about 170 mm from the center of the cell on the same source-detector line used for the neutron irradiation. We recorded neutron data for seven hours at each angle.

After taking a complete set of calibration and neutron data, the pulse-height data was transferred to an LSI-11 computer and stored on a floppy disk. The BASIC program GAMCAL was used to locate the half-heights

Figure 2. The sources were positioned at 0° , 30° , 60° , and 90° relative to the detector.

Figure 3. Experimental setup

from the gamma-ray calibration data, assign a light value to each, and fit a least-squares straight line to the data. Results from the fit were put into a second floppy-disk file. The neutron pulse-height data, the gamma calibration results, and a file of efficiency parameters provided the input for the BASIC program NEWSFC that reduced the data to a neutron spectrum. This program produced a listing of the spectrum and created

a disk file containing the energy uncertainties, the neutron fluences, and the neutron fluence uncertainties. We modified an existing plotting program to create the BASIC program NUNEPL, which plotted the data in the NEWSFC output file. To simplify comparisons, an additional program was written to calculate the ratios of fluence values at each angle to comparable values at 0° .

Results

At 0.5-MeV intervals from 1.5 to 12.0 MeV, we calculated ratios of the spectra at 30, 60, and 90° to the spectrum at 0° . These results are given in Table 1. Within statistical uncertainties, the ratios are unity in every case.

The uniformity of these ratios is somewhat surprising. Intuitively, one would expect decreasing efficiencies at the higher energies for larger angles, due to shorter path lengths in the cell. The essentially uniform response may be due to the symmetry of a right circular cylinder when the diameter is equal to the height, similar to a sphere or a cube. The projected area of the detector increases sufficiently at larger angles to compensate for the decrease in the mean track length.

We performed simple Monte Carlo integrations over the path lengths at 90° , which yielded a higher efficiency for a side irradiation than an end irradiation at

all energies. Unlike the experimental results, the calculated side-to-front ratios depended on energy, decreasing from 1.14 at 1 MeV to 1.06 at 12 MeV. At higher energies the uncertainties in the experimental values could accommodate the difference, but we find no explanation for the discrepancy at the lower energies. Some of the difference between the calculated and measured values may be because the calculations assumed a perfect right cylinder of known dimensions, whereas the actual dimensions might be in error due to imprecisions in manufacturing the glass cell. An additional uncertainty in the experimental efficiency is caused by having an external expansion chamber for the liquid scintillator, which results in a variation in the number of hydrogen atoms in the cell as a function of temperature.

Table 1. Ratios of the PuBe spectra measured at different angles to that measured at 0°, as a function of neutron energy.

Energy (MeV)	Ratio of 30 to 0°	Ratio of 60 to 0°	Ratio of 90 to 0°
1.5	0.95 ± 0.09	0.94 ± 0.08	1.01 ± 0.09
2.0	0.96 ± 0.09	0.95 ± 0.09	0.99 ± 0.09
2.5	0.98 ± 0.08	0.99 ± 0.09	1.02 ± 0.09
3.0	0.97 ± 0.08	0.96 ± 0.08	0.98 ± 0.08
3.5	0.98 ± 0.09	0.97 ± 0.09	1.04 ± 0.09
4.0	0.93 ± 0.09	0.92 ± 0.09	0.98 ± 0.09
4.5	0.93 ± 0.09	0.93 ± 0.09	0.97 ± 0.09
5.0	0.95 ± 0.09	0.94 ± 0.09	1.00 ± 0.09
5.5	0.99 ± 0.11	0.97 ± 0.10	1.03 ± 0.10
6.0	0.97 ± 0.12	0.98 ± 0.12	1.01 ± 0.12
6.5	1.00 ± 0.15	0.99 ± 0.14	0.98 ± 0.14
7.0	0.99 ± 0.16	0.97 ± 0.16	1.02 ± 0.16
7.5	0.99 ± 0.17	0.92 ± 0.16	0.99 ± 0.16
8.0	0.96 ± 0.16	0.95 ± 0.16	1.00 ± 0.16
8.5	1.00 ± 0.18	1.00 ± 0.18	1.03 ± 0.17
9.0	1.03 ± 0.24	0.99 ± 0.24	1.06 ± 0.23
9.5	0.96 ± 0.28	0.95 ± 0.28	0.98 ± 0.27
10.0	0.92 ± 0.25	0.93 ± 0.25	0.98 ± 0.25
10.5	0.93 ± 0.23	0.98 ± 0.23	0.95 ± 0.21
11.0	1.12 ± 0.26	1.03 ± 0.24	1.01 ± 0.23
11.5	1.13 ± 0.31	1.03 ± 0.29	1.04 ± 0.28

Conclusion

The implications of these results are quite favorable. Apparently, we can make measurements using scintillators with this geometry without being overly

concerned about the angular distribution of the neutron field. No correction factors are necessary.

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CRITICALITY SAFETY

Criticality Safety Calculations for a Space Power Reactor Conceptual Design Study

J. S. Pearson

Abstract

We performed criticality safety calculations as part of a conceptual design study for a gas-cooled space power reactor. Most of the calculations are based on a beryllium oxide with uranium dioxide core, although boron carbide with uranium boride (^{11}B isotope), and carbon with uranium dicarbide were also considered. We made a criticality survey as a function of core porosity and size, and studied thermal effects on reactivity. We considered reactor control by means of a burnable poison, a variable-leakage reflector, and internal control rods. Two accident conditions were considered: a flooded reactor and a crushed reactor. We conclude that with the use of retractable neutron poison wires in the core, the space power reactor can be designed to remain safely subcritical even if it is fully immersed in water or crushed.

Introduction

Large amounts of electric power are required for some systems envisioned in the U.S. Strategic Defense Initiative. Some of these systems may benefit from space-based nuclear power reactors.

A criticality study has been performed as part of a larger conceptual design study for a gas-cooled reactor

to be used with space power systems.^{1,2} The reactor design was largely based on previous development experience at LLNL with the 500-MW reactor for the Pluto nuclear ramjet,^{3,4} but a 50-MW reactor design was arbitrarily chosen for this study.

Computer Code

We used the ALICE computer code for our calculations.^{5,6} ALICE is a Monte Carlo (statistical) neutron/photon transport code that uses the probability-table method for handling multiband cross sections; this code properly treats resonance self-shielding effects.⁷ The code has previously been found to be in excellent agreement over a wide range of uranium enrichments when compared with the well-known Monte Carlo code

MCNP, which uses continuous-energy cross sections.⁸ In addition, two cases from our study were independently modeled and run using the MCNP code and yielded good agreement.⁹

The neutron cross-section sets used by ALICE were derived from the Lawrence Livermore National Laboratory Evaluated Nuclear Data Library, ENDL-86. One set of cross sections was used for "cold" calculations at 291 K.

A set of "hot" cross sections was Doppler-broadened for 1400 K (which was selected as an average reactor-operating temperature). "Doppler-broadening" means

that the neutron cross sections were adjusted to account for thermal-motion effects in neutron reactions with nuclei.¹⁰

Reactor Model

Geometry

The standard model used for these calculations is shown in Fig. 1. For the reference reactor core at 291 K, we used an 80-cm-high right circular cylinder. In an actual reactor, this core would consist of a matrix of fuel tubes and flow holes. Gas, such as helium under pressure, would pass axially through the core holes, transferring heat energy for electricity conversion, and cooling the core at the same time. The fraction of the core occupied by flow holes is called "porosity."

A geometry for 13 control rods (1-cm diameter) was also included. Control rods were inserted in combinations of 1, 7, 12, or 13. We also ran variations on this model, such as with different core sizes, the presence or absence of control rods, various control-rod insertion lengths, or with the radial reflector open. For an early set of survey calculations the axial reflector density was (inadvertently) held constant, but for subsequent calculations it was varied with the core porosity.

For some of the reactor flooding calculations, another model was used (shown in Fig. 2). A cell representing two fuel holes was modeled along with a 2-mm-diameter safety wire in each hole. The cell was reflected on four sides, thus providing an infinite planar array of fuel rods. We modeled the reactor axial reflector and 20 cm of external water reflection on the remaining faces.

Material Composition

Three fuels were considered: B_4C+UB , $BeO+UO_2$, and $C+UC$. In some calculations, small amounts of burnable poison—either ^{10}B or natural Gd —were added to the fuel. These are described as a weight percent of the fuel.

Some calculations were made at various reactor operating times. For the reactivity calculations we approximated the results from a fuel burnup code.¹¹ We accounted for burnup of the ^{235}U fuel and burnable poison directly and included production of the most abundant additional actinide elements. The fission-product inventory was approximated by an average cross-section set from the ENDL library developed to simulate such fission products or the average.

The geometric effect of temperature change was modeled in a conservative way by thermally expanding the core and axial reflector, but not the other reactor components. For boron carbide a linear expansion factor ($\Delta L/L$) was determined to be 0.60% from 291–1400 K.¹² For beryllium oxide, 1.07% was the corresponding fraction used.¹³

Several studies were made using fuel loaded with additional uranium compound to initially approximate the fuel needed during operation. This was done by approximating the ^{235}U consumption mass from the expression¹⁴:

$$M_f = (895) (1 + \alpha) PAT/ER$$

where

P = power = 50 MW,

T = time = 365 days,

ER = recoverable energy = 200 MeV,

A = atomic weight = 235,

α = capture-to-fission ratio.

For example, the reference core $BeO+UO_2$ fueled reactor with 80-cm diameter by 80-cm high core using $\alpha = 0.39$ (determined by calculation) yields a consumption mass of 26.7 kg and a total ^{235}U mass of 35.4 kg. The capture-to-fission ratio is the ratio of the relative probability that the compound nucleus formed when a neutron is absorbed will decay by emission of capture gamma rays vs the probability of decay by fission.

The axial reflector is composed of the same moderator as used in the fuel with the same porosity (except for an early survey model, as previously mentioned). The fuel is surrounded radially by concentric cylinders of various materials. The moderator is composed of zirconium oxide plus 8 wt% yttrium oxide. Helium gas at a pressure of 2 MPa at 1000 K is the coolant. The pressure vessel is composed of molybdenum plus 1.9 wt% hafnium. The radial reflector is beryllium. The control rods are assumed to be pure ^{10}B unless otherwise noted.

Figure 1. Standard reactor model for 80-cm diameter by 80-cm high core at 291 K (dimensions in cm). (a) cross section through center, parallel to cylinder center line. (b) cross section through center, perpendicular to cylinder center line.

Figure 2. Cell model of reactor fuel rods used in reactor flooding studies (dimensions in cm). By reflecting on four sides, an infinite planar array of fuel tubes with flow holes and inserted safety wires is modeled.

Survey Study

An initial criticality survey was made of the three fuels ($\text{BeO} + \text{UO}_2$, $\text{B}_4\text{C} + \text{UB}_4$, and $\text{C} + \text{UC}_2$) in a 68-cm diameter by 68-cm high core as a function of core porosity between 0.20 and 0.35. A set of points was also determined for the $\text{C} + \text{UC}_2$ fuel in a 100-cm diameter by 100-cm high core. The results are shown in Fig. 3. For these calculations, except as noted, the axial reflector density was held constant rather than varying its density with porosity. The survey shows several significant effects.

Varying the porosity in the 68-cm core dramatically affects the mass of ^{235}U fuel required to achieve criticality. Increasing the porosity requires more fuel to compensate for the reduced core density. If the density of the axial reflector were also varied with porosity, this effect would be even greater; the critical masses would be somewhat

higher because the axial reflector becomes less efficient at higher porosity. The higher critical masses for the more porous reactors at this core size also lead to a large increase in the median fission energy, E_f , of the neutrons. Nearly all the cases shown are faster than thermal, and most are in the intermediate energy range between a few electron volts to 10 keV. The graphite reactor requires much more uranium fuel to achieve criticality than the other two fuels for a corresponding core size and porosity. The graphite reactor is also the fastest critical reactor of the three fuels (i.e., it has highest median fission energy).

A survey of critical masses is shown in Fig. 4 for various core dimensions (length = diameter) between 68 and 100 cm at porosity 0.30 and temperature 291 K. The critical mass decreases dramatically for the $\text{BeO} + \text{UO}_2$

Figure 3. Critical mass of ^{235}U as a function of core porosity at temperature 291 K (axial reflector density constant). Open symbols are 68-cm $\times$ 68-cm high, solid symbols are 100-cm $\times$ 100-cm high core.

and B_4C+UB_4 fuels as the median fission energy decreases with increasing core dimension. The 80-cm and 90-cm cores for these fuels have median fission energies in the thermal region, whereas the 68-cm core is critical in the intermediate energy range (between a few electron volts to 10 keV). The critical mass for the graphite core, however, varies little between the 68- and 100-cm cores, since the median fission energy remains quite high over this range.

The neutron capture-to-fission ratio, α , is also lower in the larger $BeO+UO_2$ and B_4C+UB_4 critical cores that have lower critical masses. Since fuel consumption is proportional to $1 + \alpha$, the smaller value of α indicates that less fuel burnup is required per unit of power produced

in the larger, more thermal cores.¹⁴ Shown in Fig. 5 are the values of α for both $BeO+UO_2$ and B_4C+UB_4 cores in the 80-cm $\times$ 80-cm core of porosity 0.30 at temperatures 291 and 1400 K over a range of ^{235}U masses. The values of α all lie very close to a straight line.

The thermal effects on reactivity were also studied between the "cold" temperature of 291 K and the average operating temperature of 1400 K over a range of core sizes. This is an important safety consideration, because we want the reactivity to decrease with higher temperature, thus helping to control the reactor and providing a shut-down mechanism in an accident. Such a reactor is said to have a negative temperature coefficient of reactivity.

Figure 4. Critical mass of ^{235}U as a function of core dimension (diameter equal to height) for porosity 0.30.

Figure 5. The neutron capture-to-fission ratio, α_c , plotted as a function of ^{235}U mass for the 80-cm $\times$ 80-cm high core of porosity 0.30.

Table 1. Change in reactivity (Δk_{eff}) resulting from temperature increase from 291 to 1400 K

Fuel	Δk_{eff} (thermal expansion)	Δk_{eff} (cross section)
<u>68-cm core</u>		
BeO + UO ₂	-0.012 ± 0.003	-0.001 ± 0.003
B ₄ C + UB ₄	-0.006 ± 0.004	+0.005 ± 0.004
<u>80-cm core</u>		
BeO + UO ₂	-0.033 ± 0.011	-0.085 ± 0.006
B ₄ C + UB ₄	-0.012 ± 0.006	-0.014 ± 0.007
<u>90-cm core</u>		
BeO + UO ₂		-0.133 ± 0.005
B ₄ C + UB ₄		-0.121 ± 0.004

Results of the temperature study are summarized in Table 1. The changes in k_{eff} due solely to thermal expansion of the core from 291–1400 K are shown separately from the effects due to temperature-dependent neutron cross section changes (Doppler broadening). For the 68-cm core, a small decrease in k_{eff} results from

thermal expansion for either fuel, but the effect due to Doppler broadening is too small to detect within the statistical uncertainties—since the median fission energies are in the tens to hundreds of electron volts, little effect can be expected from Doppler broadening at such high energies.

For the 80-cm core the thermal expansion effect is even greater, and a significant decrease in k_{eff} results from the Doppler-broadened cross sections. The larger effect from Doppler broadening for the $\text{BeO} + \text{UO}_2$ fuel can be attributed to its very low (thermal) median fission energy (0.074 eV) compared to a higher value (2 eV) for the $\text{B}_4\text{C} + \text{UB}_2$ fuel. For the 90-cm core both fuels have quite thermal median fission energies and hence the Doppler-broadened cross sections significantly reduce k_{eff} . Thus, both the $\text{BeO} + \text{UO}_2$ and the $\text{B}_4\text{C} + \text{LB}_2$ fuels have negative temperature coefficients of reactivity for this reactor design, which is an important safety feature.

Reference Reactor Study

For further studies we chose a reference reactor with $\text{BeO} + \text{UO}_2$ fuel, an 80-cm core, and a flow porosity of 0.30. A consumption mass was calculated and added to the 1400 K critical mass to approximate the fuel required to produce 50 MW of power.

The clean critical reactor (3.97 kg of ^{235}U) is quite thermal, with a median fission energy of hundredths of an eV. It shows an 8% decrease in reactivity as the temperature rises from 291–1400 K due to the Doppler-broadened cross sections. With the consumption mass added the 35.4-kg ^{235}U reactor is much faster, with a median fission energy of several eV. The value of α also increased from about 0.26 to 0.40 with the additional fuel. The temperature effects from the Doppler-broadened cross sections are no longer measurable within statistics, although thermal expansion still decreases the reactivity by 2% and thus has a negative temperature coefficient of reactivity.

We studied the effects on k_{eff} of the control mechanisms (control rods and movable radial reflector) for the 35.4-kg ^{235}U reactor (with consumption fuel). The radial reflector decreases k_{eff} by 5% at 291 K from 1.097 to 1.045 (± 0.005) when fully open. We also studied the effects of fully inserting one, seven, twelve, or thirteen 1-cm-diameter ^{10}B control rods. The rods are very effective: 12 rods changed the k_{eff} by 20%, as shown in Fig. 7. A smooth curve results with the highest change in reactivity per change in insertion length occurring around the center of the core. Studying this effect by varying the ^{10}B density of the control rods showed that rods at 50% of normal ^{10}B density are still nearly as effective as full-density rods.

Early in the life of the reactor, burnable neutron poisons could be used to control the excess reactivity of the consumption fuel by removing some neutrons. These poisons would gradually burn up along with the uranium

fuel during operation of the reactor, thus allowing higher reactivity later when it is needed. Two burnable poisons, ^{10}B and Gd , were considered. It was determined that 0.003 wt% of ^{10}B or 0.025 wt% of Gd would reduce the reactivity for a clean new reactor with consumption mass to about 1% above critical, in easily controlled operating level.

Using the results from a burnup code calculation, the reactivity of the core was calculated as a function of time as both the fuel and burnable poison were consumed.¹¹ This is shown in Fig. 8. When the reference core of 35.4 kg ^{235}U (with an initial loading of 0.003 wt% burnable ^{10}B poison) is burned at a power of 50 MW, the large relative production of actinide elements and fission products rapidly poisons the reactor, despite the accompanying rapid burning of the poison, which drops to 5.3% of its initial loading after 180 days, and 0.06% after 365 days. If this reactor is run at only 32 MW, the ^{10}B poison burns more slowly but still is reduced significantly to 18.4% of initial loading after 180 days and 1.3% after 365 days. The production of actinides and fission products is reduced from the 50 MW case, and consequently the reactivity remains much higher but still drops well below critical after about half a year. Reducing the initial burnable poison loading would keep the reactor critical for a longer period, although other modifications (e.g., increasing the fuel loading) would be needed to achieve a full year of critical operation.

The fuel loading of the original reference reactor (50 MW) was then increased to 100.5 kg of ^{235}U to provide a second reference core. Burnable ^{10}B poison (0.030 wt%) was again added to achieve a reactivity slightly above critical at 1400 K. The results of this study are also shown in Fig. 8. The larger loading of ^{10}B poison decreases much more slowly than in the previous two cases, reducing to 42.2% of initial loading after 180 days and 15.9% after

Figure 6. Multiplication factor k_{∞} as a function of reactor reflector thickness; (top) axial reflector; (bottom) radial reflector.

Figure 7. Reactivity worth of twelve ^{10}B control rods as a function of insertion distance.

Figure 8. Multiplication factor k_{eff} as a function of reactor run time for the reference reactors.

365 days. Also, the relative production of actinides and fission products builds up more slowly since a smaller fraction of the original ^{235}U inventory is consumed. As a result, the reactivity remains quite flat and close to critical throughout the year, which is good for operation.

The reactor could be maintained at critical for a full year at or near 50 MW with modifications to the burnable poison or uranium loading, or this reactor could be operated at a somewhat lower thermal power.

Reactor Safety

We considered two accident conditions: a flooded reactor (prior to nuclear operation) and a crushed reactor. First, we modeled the flooded reactor by replacing all voids in the reactor model (including the porosity) with water and closely reflecting the reactor externally by 20 cm of water. The water very effectively moderates the neutrons in the core, which greatly reduces the median fission energy from 7.6 eV in the unflooded reactor to a very thermal value of 0.018 eV for the 35.4-kg ^{235}U reference core. As a result, the reactor becomes supercritical even with the burnable poison and twelve ^{10}B control rods present.

Insertion of 2-mm-diameter wires made of Cd or Hf (good neutron poisons) in various fractions of the core holes was studied as a method to keep the reactor subcritical in the event of such a flooding accident. This was first modeled by homogeneously mixing the wire poison material into the core, although this probably overestimates the poison effectiveness because self-shielding effects of the poison material are not included. Next, we modeled an infinite planar array of fuel elements with wires explicitly placed in only a fraction of them. This probably overestimates the reactivity because of the infinite array and because the control rods are not

included. Thus the actual reactivity should lie between the results from these two models. The results (Fig. 9) indicate that either the 35.4 kg or the 100.5 kg ^{235}U loaded reactor can be kept subcritical when flooded by having such 2-mm Cd or Hf wires in approximately 25–50% of the core flow holes.

We conservatively modeled a radially crushed reactor by removing all voids (including porosity) and close-packing the reactor materials for the reference 35.4-kg ^{235}U loaded reactor. With the burnable ^{10}B poison and twelve ^{10}B control rods inserted, this postulated accident raises the reactivity to a slightly subcritical value ($k_{\text{eff}} = 0.978 \pm 0.007$). The median fission energy ($E_f = 7.7$ eV) was essentially unchanged from the corresponding undamaged reactor. Homogeneously mixing into the core the equivalent mass of 2-mm-diameter Cd wires in half the core holes significantly reduced the reactivity ($k_{\text{eff}} = 0.519 \pm 0.010$). Thus, the crushed reactor is a much less severe accident than the flooded reactor, and both accidents can be prevented from going supercritical by having an adequate number of poison wires inserted in the core during ground and launch operations.

Conclusions

Reactivity calculations have been made for graphite-, boron-carbide-, and beryllium-oxide-moderated gas-cooled reactor designs over a range of core porosities and core sizes. The beryllium-oxide-moderated core appears particularly suitable for space power applications in the power range studied (around 50 MW). We have shown that reactivity control of such a reactor is feasible with a combination of control rods, variable-leakage reflectors, or burnable poisons. Thermal effects were studied, showing that thermal expansion

results in a negative coefficient of reactivity. Doppler broadening of cross sections provides a negative coefficient of reactivity for the more thermal designs, but this effect is too small to measure for the faster reactor designs. We also studied two accident scenarios—if the reactor is crushed or fully immersed in water—and found that the reactor can be designed to remain safely subcritical by the use of retractable neutron-poison wires in the core.

Figure 9. Multiplication factor k_{∞} as a function of the percent of core flow holes containing a 2-mm diameter Cd or Hf safety wires. Reactor model and planar fuel array are described in the text.

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A Procedure for Ranking Fire Performance of Electrical Cables

H. K. Hasegawa, K. Staggs, N. J. Alvares, and A. C. Fernandez-Pello

Abstract

We present experimental results of piloted ignition delay and the upward flame-spread rate over the surfaces of insulated electrical cables under an externally applied radiant flux. The objectives of our experiments are to assess and rank the fire performance of seven types of cable commonly used in electrical installations, and to compare our small-scale results with those previously obtained in a large-scale facility. We carried out our experiments with 46-cm-long single cables that were suspended vertically and exposed to irradiance levels ranging from 0.5–2.5 W/cm². Some of the cables had a conducting core and some did not. Our analysis describes the ignition and flame-spread processes and identifies parameters that characterize these processes. The analysis shows that these parameters are similar to those identified by Quintiere and coworkers in a study of the ignition and lateral flame spread in building materials. We apply a similar method to define the flame-spread properties of the materials in order to assess and rank the fire performance of the seven types of cable. We show that the method can be successfully applied and that it provides a simple and systematic way to rank the cables and to calculate the parameters that control ignition and flame spread.

Introduction

In many facilities housing electronic equipment, a potential source of fire is the various plastic and rubber cable and wire insulations. The study of fire performance of electrical cables is complicated because of the nonuniform composition of insulation materials, the complex geometrical arrangement of cable lines, the interactive effects between cables of similar or different composition, and the interaction between the conducting metal core and the insulation material. Predicting critical conditions for ignition and rates of flame spread in electrical cables is difficult but important for protecting equipment and preventing fire development in a facility. This report gives partial results of a study carried out at LLNL.

We correlated data obtained in a series of small-scale experiments. These experiments burned insulated electrical cables in a modified version of the ASTM E-162 standard test. The data correlation followed the method proposed by Quintiere, et al. to deduce the flame-spread properties of the cables and to rank them according to their flammability characteristics.²⁻⁴ The results show that analyses supporting the experimental procedures and correlations of the data for both upward and lateral flame spread are similar. This indicates that information obtained with the experimental procedure addresses fundamental aspects of the ignition and flame-spread processes and consequently can be applied to different modes of flame spread.

Experiment and Results

Our test apparatus is a gas-fired radiant panel, a specimen-holder frame where the cables are suspended vertically, a pilot burner positioned at the bottom of the specimen, and a water-cooled sliding metal shutter located between the radiant panel and the specimen to precisely control exposure times. The specimen consists of a single 46-cm-long cable suspended vertically in the frame. Two thermocouples are attached to the midpoint of the cable—one on the jacket surface, and one embedded through the jacket to the conductor—to monitor the temperature of the specimen. Each cable type is exposed to irradiance levels ranging from 0.5-2.5 W/cm² to evaluate the effect of increasing flux on flame-spread rate and time to ignition.

The measurements performed include the delay time to preheat the cable jacket to 100°C, the minimum radiant flux for ignition, the jacket temperature at ignition, the ignition delay time, and the flame-spread rate as a function of the radiant flux at the cable jacket surface. Flame-spread and time-to-ignition data for PVC/PE cable is shown in Fig. 1. The indicated ignition delay extends from the time of pilot flame deployment to the time where perceptible flame is sustained on the jacket. The pilot flame is not deployed until the bulk jacket temperature has been radiantly heated to 100°C. Figures 2 and 3 show ignition delay and vertical flamespread data, respectively, for the set of insulated cables analyzed during this test series.

Figure 1. Flame-spread and time-to-ignition data for PVC/PE cable.

Figure 2. Ignition delay time for the set of insulated cables studied in these tests.

Figure 3. Vertical flame spread curves for the set of insulated cables studied in these tests.

Theoretical Model

The theoretical model that supports these measurements consists of an analysis of the transient heating of the cable jacket by the external radiant flux, the ignition of the fuel, and the upward spread of flames over the jacket surface.^{2,5} The model assumes that the cable jacket behaves as a thermally thick fuel, that radiation is totally absorbed at the jacket surface, and that ignition occurs at a specific "ignition" temperature. The flame-spread process is modeled as a heat-transfer

problem. The heat transferred from the flame and external radiant source to the solid causes its temperature to increase until it starts to pyrolyze vigorously. The "pyrolysis" temperature is assumed to coincide with the ignition temperature. The rate of flame spread is considered to be equal to the rate of propagation of the pyrolysis front.

The transient heating of the jacket is described by the equation,

$$T_s - T_i = q'' e \sqrt{\frac{4t}{\pi k \rho c}} \quad (1)$$

which can be used to calculate $k\rho c$ from the data of the time elapsed for the surface temperature to reach 100°C. The ignition temperature, T_{ig} , is calculated with the equation,

$$q''_{ig} = h (T_{ig} - T_i) \quad (2)$$

The value of the critical ignition flux, q''_{ig} , is given by the asymptotic value of the ignition curves in Fig. 2; h is a composite heat-transfer coefficient; and T_i is the initial temperature of the insulation.

The flame-spread rates in Fig. 3 are averaged values of those measured along the cable length (Fig. 1). It is seen that equations of the form

$$V_f = \frac{\phi}{k\rho c (T_{ig} - T_i)^2} \quad (3)$$

or

$$V_f = \frac{1}{C^2 (q''_{ig} - q''_c F(t))^2} \quad (4)$$

describe very well the general characteristics of the experimental data.^{2,5} In these equations, ϕ defines the energy potential for fire spread, $k\rho c$ are thermal constants for the insulators, C is basically ϕ but incorporates convection effects, and $F(t)$ is a form of thermal impedance. The data in Figs. 2 and 3 can also be used to provide information about the fire performance characteristics of the different cables. The comparison of flame-spread rates for a given irradiance level, or of the values of ϕ or C , indicate how hazardous the material is from the point of view of flame spread. The critical irradiance for piloted ignition could also define the ignition propensity for these materials.

Flammability Diagrams

A useful approach to analyzing the above measurements is to plot the ignition time and flame-spread rate measurements for each individual cable as a function of the external radiant flux.^{2,4} A characteristic example of this type of plot is presented in Fig. 4 for PVC/PE coaxial cable. The advantage of plotting the data in this form is that the parameters controlling the ignition and flame-spread processes can be obtained from a single plot, some by inspection, and others in conjunction with the theoretical analysis of the problem.^{2,3}

The results of Fig. 4 can be used to deduce the critical heat flux for ignition, which is given by the asymptotic value of the flame spread and ignition curves.

Once the value of the critical heat flux is determined, the ignition temperature can be calculated from Eq. (2) after estimating the value of the heat-transfer coefficient, h . With the calculated values of $k\rho c$ and the ignition temperature or critical heat flux, Eqs. (3) and (4) can be used, in conjunction with the flame-spread data, to calculate average values of the energy available for spread, ϕ , or the flame-spread rate coefficient, C . For PVC/PE coaxial cable it is deduced that $\phi = 0.92 \text{ W}^2/\text{cm}^3$. The corresponding theoretical flame-spread curve obtained with Eq. (3) is also shown in Fig. 4. It is seen that Eq. (3) predicts quite well the dependence of the flame-spread rate on the external irradiance level.

Figure 4. Plot of the ignition time and flame-spread rate measurements as a function of the external radiant flux for PVC/PE coaxial cable. In a plot such as this, the parameters controlling the ignition and flame-spread processes can be obtained.

Conclusion

Although the predictions from this methodology are not exact, it is encouraging that we can predict a cable's fire performance in terms of time to ignition and rate of flame spread through the use of selected data from small-scale tests. The advantages of using small-scale tests include: economy in test apparatus, economy in small sample size, simple and quick setup time, as well as the capability of performing many tests in a short period of time and rigid control of many test variables.

In its current stage of development this technique can serve as a reasonably accurate wire and cable screening tool for flammability characteristics. From a large number of candidate cables, only one or two would have to be subjected to the great expense of large-scale tests.

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Identification Procedure for Wire and Cable Jacket Insulations

Stephen J. Priante

Abstract

The Fire Science Group evaluated the flammability of various electrical cable insulations and also investigated methods to identify the composition of in-place cable insulations. We used thermogravimetric analysis and differential scanning calorimetry to compile thermograms of various insulation materials. We have compiled this information into a library of thermograms for identifying cable insulations. This paper describes our analysis and gives an index of our library.

Because of the huge inventory of insulated electrical cables used throughout LLNL and other DOE facilities, the Fire Science Group was asked to undertake a study to evaluate the flammability of such cable insulations. We also investigated methods to identify the composition of cable insulations, which would allow the selection of less-flammable insulators.

During an early test series that included both large-scale tests and analysis in the analytical lab, we found that the different "generic" cable insulations based on polymers like polyvinyl chloride (PVC), neoprene, or rubber exhibited different flame-spread characteristics and produced different toxic and corrosive products during the degradation process. Moreover, insulations in the same generic group produced different degradation products. It then became an important aspect of our study to be able to identify the composition of our test cables because we wanted to model cables that had been in use at many DOE facilities for long periods of time.

We realized that attempting to identify cables currently in use, either by visual inspection or backtracking to the LLNL Stores stock catalog, would be a time-consuming exercise that would also be futile in many cases. Many cables in use had no identifying markings or characteristics, and backtracking the cable type through the Stores catalog or to bulk storage was not possible. LLNL Stores is continually updating its stock with the current low-bid cables that meet performance specifications, so the cable in stock will not necessarily be from the same manufacturer, or of the same formulation, as a cable used for the same function but purchased at a later date.

To develop procedures of cable insulation identification, I used two analytical instruments for thermal decomposition research. The first method was thermogravimetric analysis (TGA), which produces a weight-loss history of a representative sample as it is heated at a specified temperature ramp in a controlled atmosphere over a specified period of time. Figure 1 is a typical TGA scan showing specific characteristics of the thermal decomposition of a material such as the onset of degradation, the rate of degradation, and residual weight remaining after thermal degradation is complete.

The second method I used was differential scanning calorimetry (DSC), which is also run at a controlled temperature rate in a controlled atmosphere for a specified period of time. DSC analysis gives the endothermic and exothermic history associated with the transitions taking place as the sample portion heats up. Figure 2 is a DSC plot of the same sample as Fig. 1. The onset temperatures of transition #1, which is in the positive direction (exothermic), and transition #2, which is in the negative direction (endothermic), are important parameters for material identification.

When the results obtained by these two separate analyses are used together, a discrete thermal-degradation fingerprint is produced for each material. This is the basis for the identification procedure of all the cable insulations we measured.

I obtained cables of many generic types that were positively identified by the cable jacket itself or traced from the LLNL Stores catalog. The samples were then analyzed by TGA and DSC to obtain their thermal fingerprint and stored in our cable identification library.

This analysis identified the dominant characteristics of all the insulations we measured and accounted for the differences in generically similar formulations. By the time I completed testing all the insulation materials available for analysis, I had compiled an extensive library of identified insulation thermograms (see Table 1).

With this library, I have been able to identify numerous cables by matching unknown TGA/DSC

fingerprints to known patterns in our library. Table 2 lists the additions to the insulation thermogram library.

Tables 1 and 2 index the collection of thermograms for electrical insulation materials on hand in the analytical chemical laboratory of the Fire Science Group. A file of degradation products for many of these materials is also available in the archives of the analytical lab.

Figure 1. A typical TGA scan showing specific characteristics of the thermal decomposition of a material, including the onset of gross degradation and the residual weight remaining after thermal

Figure 2. A DSC plot of the same sample as Fig. 1. The onset temperatures of the exotherm and the endotherm are important parameters for material identification.

Table 1. Original cable library index.

Cable type	Cable description and use
PVC	Radiofrequency transmittance coaxial
PVC	Radiofrequency transmittance coaxial
PVC	Radiofrequency transmittance coaxial
PVC	Twisted multiconductor telephone
PVC	Twisted multiconductor telephone
PVC	Appliance replacement
PVC	Power cord—stranded copper
PVC	Shielded multiconductor drain wire
PVC	Stranded conductor patch cable
PVC	Three-conductor signal cable
PVC	Shielded multiconductor drain wire
PVC	Stranded conductor control and signal
PVC	Closed-circuit television coaxial
PVC	High flexibility fifty-five conductor
PVC	"Ribbon" cable for multiple connections
PVC	Medium-loss fiberoptic cable
Neoprene	Abrasive-resistant portable power cord
Neoprene	Multiconductor power cable
Neoprene	Moisture- and abrasive-resistant welding
Neoprene	Moisture- and abrasive-resistant welding
Neoprene	Power cable
Neoprene	Welding cable
Neoprene	Abrasive-resistant portable power cord
Rubber	Multiconductor power cable
Rubber	Multiconductor power cable
Rubber	Test product wire
Rubber	Braided cotton-wrap microphone cable
Rubber	Braided cotton-wrap microphone cable
Polyethylene	Multiconductor telephone cable
Polyethylene	Television twin-lead cable
Polyethylene	High-voltage hookup wire
Teflon	Radiofrequency transmittance coaxial
Teflon	Hookup wire
Kynar	Flame, cut, chemical and radiation resistant
Polyurethane	Single fiberoptic fiber cable

Also included in our "known" library were the following virgin polymers (polymers without any additives or fillers):

Polyvinyl chloride—PVC
 Polychloroprene—Neoprene
 Polypropylene
 Polyethylene
 Polyvinylidene fluoride—Kynar
 Polytetrafluoroethylene—Teflon
 Polyethylene terephthalate—Mylar

Table 2. Samples identified by using "fingerprint" library.

Sample designation	Generic identification	Sample origin or purpose
007	Neoprene	Fire Science Group cable fire test
435	Neoprene	Cable overload problem Bldg. 435
#6 AWG	Neoprene	Fire Science Group cable fire test
Welding cable	Hypalon	Fire Science Group cable fire test
#1132	Rubber	Fire Science Group tube fire test
#1138	Rubber	Fire Science Group tube fire test
RG 11 A/U	PVC	Fire Science Group cable fire test
Harry #1	Hypalon	Fire Science Group cable fire test
Harry #2	PVC	Fire Science Group cable fire test
Harry #3	Polyethylene	Fire Science Group cable fire test
Unknown #1	Rubber	Fire Science Group cable fire test
Unknown #2	Polyethylene	Fire Science Group cable fire test
Unk. Inside	PVC	Salvage cable for fire testing
Kirk Unknown (outside)	Hypalon	Salvage cable for fire testing
Kirk Unknown (inside)	Rubber	Salvage cable for fire testing
Hatfield Diesel (outside)	Hypalon	Fire Science Group cable fire test
Hatfield Diesel (inside)	Rubber	Fire Science Group cable fire test
RG-214 coaxial (outside)	PVC	Fire Science Group cable fire test
RG-214 coaxial (inside)	Polyethylene	Fire Science Group cable fire test
Diesel Locomotive (out)	Hypalon	Fire Science Group cable fire test
Diesel Locomotive (in)	Rubber	Fire Science Group cable fire test
Powercord (outside)	Neoprene	Fire Science Group cable fire test
Powercord (inside)	Rubber	Fire Science Group cable fire test
White wire	Silicon rubber	Resistor bank overload Bldg. 431
Plastic screw	Nylon	Resistor bank overload Bldg. 431
Pipe cover	PVC	Welding blanket accident Bldg. 492
VCAL 1	Polyethylene	Fire Science Group cable fire test
VCAL 3 (outside)	Polyethylene	Fire Science Group cable fire test
VCAL 3 (inside)	Polypropylene	Fire Science Group cable fire test
VCAB 1	Hypalon	Fire Science Group cable fire test
VCAB 2	Hypalon	Fire Science Group cable fire test
VCAB 3	Neoprene	Fire Science Group cable fire test

Table 2 continued.

Sample designation	Generic identification	Sample origin or purpose
VCAB 4 (outside)	PVC	Fire Science Group cable fire test
VCAB 4 (inside)	Polyethylene	Fire Science Group cable fire test
VCAB 7 (top layer)	Neoprene	Fire Science Group cable fire test
VCAB 7 (bottom layer)	Rubber	Fire Science Group cable fire test
VCAB 10 (outside)	PVC	Fire Science Group cable fire test
VCAB 10 (inside)	PVC	Fire Science Group cable fire test
VCAB 13 (outside)	Neoprene	Fire Science Group cable fire test
VCAB 13 (inside)	Rubber	Fire Science Group cable fire test
VCAB 16	Rubber	Fire Science Group cable fire test
VCAB 18 (top layer)	Hypalon	Fire Science Group cable fire test
VCAB 18 (bottom layer)	Rubber	Fire Science Group cable fire test
VCAB 21 (top layer)	Nylon	Fire Science Group cable fire test
VCAB 21 (bottom layer)	PVC	Fire Science Group cable fire test
VCAB 24	Hypalon	Fire Science Group cable fire test
VCAB 26	Hypalon	Fire Science Group cable fire test
VCAB 28 (outside)	Polyethylene	Fire Science Group cable fire test
VCAB 28 (inside)	Polyethylene	Fire Science Group cable fire test
VCAB 30 (outside)	PVC	Fire Science Group cable fire test
VCAB 30 (inside)	Polypolyethylene	Fire Science Group cable fire test
VCAB 32	Rubber	Fire Science Group cable fire test
Bock 16 AWC	PVC	Control room survey sample
S/O Oil-resistant	Neoprene	Control room survey sample
Belden 8451	PVC	Control room survey sample
Belden 8473	Rubber	Control room survey sample
Green RG-52	PVC	Control room survey sample
Bec. Co. 6 Pr.	PVC	Control room survey sample
Green #07145	PVC	Control room survey sample
Green RG 580	PVC	Control room survey sample

Table 2 continued.

Sample designation	Generic identification	Sample origin or purpose
Belden 8723	PVC	Control room survey sample
RG 214	PVC	Control room survey sample
Small black	PVC	Bldg. 151 fire sample
Medium black	PVC	Bldg. 151 fire sample
Large black	PVC	Bldg. 151 fire sample
Red wire	Teflon	Bldg. 151 fire sample
ITT Suprenant	Polyethylene	Sandia National Labs sample
ITT Suprenant	Polyethylene	Sandia National Labs sample
ITT Suprenant (inside)	Polyethylene	Sandia National Labs sample
20/10	PVC	Sandia National Labs sample
Diesel Locomotive 2/0	Neoprene	Fire Science Group cable fire test
Green RG-58	PVC	Fermi Labs fire sample
Black coaxial	PVC	Fermi Labs fire sample
Red coaxial	PVC	Fermi Labs fire sample
Grey Multi-conductor	PVC	Fermi Labs fire sample
Ribbon cable	PVC	Fermi Labs fire sample
Blue tubing	Nylon	Fermi Labs fire sample
Green RG-58	PVC	PSG cable fire test
Red cable	PVC	Fire Science Group cable fire test
Green cable	PVC	Fire Science Group cable fire test

Analysis of Fire and Smoke Threat to Off-Gas HEPA Filters in a Transuranium Processing Plant

Norman J. Alvares

Abstract

We performed an analysis of fire risk to the high-efficiency particulate air (HEPA) filters that provide ventilation containment for a transuranium processing plant at the Oak Ridge National Laboratory. A fire safety survey by an independent fire-protection consulting company had identified the HEPA filters in the facility's off-gas containment ventilation system as being at risk from fire effects. We independently studied the ventilation networks and flow dynamics, and analyzed typical fuel loads. We found that virtually no condition for fire initiation exists, and that even if a fire started its consequences would be minimal as a result of standard shut-down procedures. Moreover, the installed fire-protection system would limit any fire and thus would further reduce smoke or heat exposure to the ventilation components.

Introduction

An independent fire-protection consulting company conducted an analysis of the high-efficiency particulate air (HEPA) filters providing ventilation containment for a transuranium processing plant at the Oak Ridge National Laboratory (ORNL). This fire safety survey identified the HEPA filters in the facility's off-gas containment ventilation system as being at risk from fire effects.

The U.S. Department of Energy asked the LLNL Fire Science Group to review and assess these results. Our analysis consisted of two major tasks: to assess fire and smoke risk to the HEPA filters that provide containment for the vessel off-gas and cell off-gas systems; and to define the effectiveness of fire-protection countermeasures for addressing a potential fire threat.

Methodology

Any analysis of fire risk must include:

- Identification of critical areas and/or components.
- Identification of the primary fire source for critical area or component.
- Definition of the fire environment (type, phase, and distribution of fuel; critical dimensions of fire source area; available air volume and ventilation; heat transfer capability of surroundings; and both passive and active fire-protection countermeasures).

- Analysis of fire performance and smoke production using appropriate calculations and models.
- Comparison of analytical results to the perceived fire threat and previous fire experience, if applicable.
- Assessment of resident fire protection.
- Recommendation of changes, if necessary.

In this analysis the HEPA filters in the vessel (and cubicle) off-gas system (VOG) and for the cubicle (and tank pit) off-gas system (COG) are the ultimate containment components of the transuranium processing

plant's ventilation systems. Note that the VOG and COG are separate from the ventilation system for the cold side of the building, which is protected by code-compliant safety systems. The maximum potential fire threat to the fillers would result from fire in a vessel or in one of the tank pits from spilled solvents. Once the environment of fire threat is determined, the fire performance and the smoke production rates can be established.

The approach for this type of analysis (developed by the LLNL Fire Science Group) is to define the time to fire extinguishment by oxygen starvation, using oxygen-depletion calculations for different fire strengths and

ventilation rates. The next calculation uses equilibrium conditions to determine maximum temperature rise for the enclosure, using heat and mass balance considerations. Finally, the rate of initial temperature rise is estimated using the ASET B computer model for early times before heat losses and ventilation effects dominate this algorithm.¹ To assess the production of smoke, I have used fuel consumption for hexane in pool fires and measured smoke partition ratios.^{2,3} The pessimistic smoke partition ratio for straight-chain hydrocarbon fluids is 5% by weight.

Facility Description

Facility Specifications

Figure 1 is a plan view of the transuranium processing plant showing both office and operator's areas, and an isometric drawing of a typical cell in the operations area of the building. The shielded cell bank contains nine 7-ft-wide hot cells each with a 7-ft-long cubicle area, separated from each other by 2-ft-thick concrete walls. Seven cells contain a tank pit area (9 × 22 ft). An inter-cell conveyor housing and the cell-ventilation exhaust duct run through the cubicle pits the full length of the cell bank.

In the first seven cells air enters the south wall of the tank pit through a duct (10 in. diameter), the centerline of which is 21 ft above the floor. The air exits the north wall of the tank pit to the cubicle pit through a slot (2 × 4 ft) ten feet above the cell floor and is drawn into a cell ventilation duct (20 × 40 in.) through an opening (17.5 in. diameter) located 8.5 ft above the cell floor.

In the last two cells air enters the cubicle pit through a similar duct only two feet above the pit floor and exits to a cell ventilation duct. A waste tank pit behind the last two cells and below the first-floor level is connected to each of the last two cubicle pits through a 2-ft-square opening, but there is no air inlet to the waste pit and essentially no air flow between the waste tank pit and the last two cubicle pits.

The air from all of the cells exits the cell ventilation duct through a 40 × 18 in. opening in the bottom of the duct in the Cell 5 cubicle pit and passes under the cell through a 37-ft-long, 30-in.-diam. duct to the COG filter inlet plenum located within the building south of the cell bank. The volume of each pit is 1386 ft³ and the air supply to each pit is 440 ft³/min

Ventilation

The off-gas systems ventilate components of the cell—the tanks, the cubicle, the cubicle pit, and the tank pit. Note that the tank and cubicle pits are served by COG ventilation with metal conduits. The VOG header that collects effluent from the tanks is metal; the header that collects effluent from the cubicles is fiber-reinforced polyester (FRP). Because off-gases from the cubicles and tanks may contain hazardous materials, they are always passed through a caustic scrubber and possibly through carbon bed adsorbers. The scrubber will remove smoke particulates and will further cool diluted combustion gases from any cubicle-kit or tank-kit fire. Thus, the VOG HEPA fillers are not in danger from in-cubicle or in-tank fires.

Carbon adsorber fire phenomenology is not well understood, however. The process is clearly one of spontaneous ignition and should be amenable to analysis, given initial conditions of temperature, air flow, moisture level, and bed packing density.

The COG ventilation system for the pits and waste pits begins with air supply from the limited-access area. The ventilation path comes in high and exits the tank pit through the access opening into the cubicle pit. In the cubicle pit a large metal duct collects this air and conveys it to an exhaust duct under the fifth cubicle where total flow is directed to the HEPA filter system before exiting through the stack. Inlet air is ducted high near the tank pit ceiling, and the air exit is at the half-height level into the cubicle pit. Total air flow through individual pit systems is 440 ft³/min during normal operations.

Figure 1. Plan of the transuranium processing plant (top) and isometric view of a typical cell (bottom).

The pit nearest the filters is the fifth cubicle and is connected to the filter station plenum by 37 ft of stainless-steel duct (not including elbows or other transition pieces). The COG filter complex is redundant, with two filter sets out of four operating in parallel for normal operations. Each set provides three air-flow paths of one roughing and two HEPA filters to accommodate the flow. Because this system serves nine cells, combustion gases from a fire in any single pit will be substantially diluted and cooled before entering the plenum to the filter housings.

Fire Protection

Most areas of the building are protected with standard wet-pipe sprinkler systems. The cells themselves are protected by heat-detector-activated pre-action deluge systems, which operate when the temperature rise is 8-11 C above ambient. When activated, the water supply headers for the pre-action systems are charged, air dampers in the inlet air ducts to the cells close, and fire alarm systems for the building and the ORNL Fire Department are actuated. When any of the detectors reaches 80 C, the deluge system opens and water flows through the sprinklers. Locations of deluge sprinkler heads and thermal detectors are at the top of the cubicle, bottom of the cubicle, and top of the tank pit. Each cell has an individual deluge system that can be actuated manually. Cubicle deluge systems are independent of the cell systems and can only be initiated manually.

Fuel Load

The fuel load per cell is a function of previous in-cell maintenance and the specific processing operations. Residual shoring boards and safety booties from maintenance operations constitute a minimal and passive amount of combustible materials. Processing operations can involve as much as 80 liters of kerosene-type solvents per cell. These liquids are contained in the process tanks at negative pressure from the VOC system. These pressure levels are approximately -10 in. H₂O for all tanks ventilated by the VOC. The inventory of solvent

held in the metal tanks of the waste-tank pit can be as much as 150 liters, generally contained in only one of the tanks. Chance of spill is remote, because of the negative pressure maintained by VOC ventilation. But regardless of the quantity of flammable liquid available, only a limited amount of any spill will support combustion. That amount will depend on fuel surface area and available air.

Structural and Ventilation Component Materials

Pit surfaces (except the floor) are covered by a 30-mil layer of FRP laminate. Since this coating is thin and laminated to the concrete surfaces, it does not constitute a fuel contribution because heat loss to the concrete wall does not allow the FRP to achieve ignition temperature. Thermal impedance of concrete is $kpc = 0.744 \text{ kJ/m}^2 \text{ K}^{\circ}\text{s}$. The VOC header is fiberglass-reinforced resin. Similar ventilation piping at LLNL contains about 40% non-combustible fiberglass, so we assume that 60% of the pipe material is combustible.

Recent tests conducted in the LLNL fire test cell on the flame spread of fiberglass ducts attest to the difficulty of duct ignition. We determined, then, that the VOC duct that passes through the tank pit would not constitute an active fire threat. Because of the sparsity of solid combustible materials in the tank pits, the likelihood of ignition is minimal. The only credible ignition scenario would involve ignition of a solvent or solvent vapor as the first step. Extensive fire growth on the liquid fuel is still needed to ignite the solid fuels. The deluge sprinklers would almost certainly activate before this could happen.

Our background information included a fuel-load log based on heat-of-combustion values for the combustible materials. Although some of the data appears to be conservative, the log provided a basis for a hypothetical fire scenario. The scenario provides a prediction of fire duration and upper-zone temperature rise, which could be used to estimate the threat to adjacent cells and potential smoke exposure to the filter plenum.

Analysis

We applied fire-modeling procedures and oxygen-depletion analysis to the case of fire on a layer of solvent spilled on the floor of a tank pit. Because the burning rate and energy release of liquid fuels is dependent on fuel surface area, we repeated the exercise for a set of fuel

pool diameters that might occur in a catastrophic failure of a tank or tank fitting.

The heat-release rate and smoke-production rate of a pool fire is estimated using the relation $Q_{\text{max}} = 212 d^2 \text{ (kW)}$, where d is measured in feet. We are able to

define the fuel-consumption rate from experimental data and from that result we can estimate heat-release rate and smoke production for these conditions (see Tables 1 and 2). These calculated values of heat and smoke release can now be used to estimate oxygen depletion, air temperature rise, and smoke production.

Oxygen Depletion

A further set of calculations accounted for the amount of oxygen consumed per gram of fuel, based on the heat-release rate of the fuel source. The information required for these calculations includes enclosure volume, vertical position of the fuel source, residual percentage of oxygen, and the ventilation rate. The vertical position of the fuel source determines the available oxygen supply, since without mixing fire plumes generally do not access air from areas below the fuel surface plane.

We used methane as an analog for gas- and liquid-phase fuels, since it has the highest heat of combustion and consumes essentially the same amount of oxygen as most hydrocarbon fuels. The residual oxygen fraction in air at fire extinction is averaged at 12%, a typical level of oxygen depletion for enclosure fires. Table 3 lists the time to oxygen depletion (extinction time) for the tank pits for two ventilation rates, 0 and 440 ft³/min, where these quantities vary with the heat-release rate. Figure 2 is a plot of these data showing a monotonic decrease in extinction time as the heat-release rate gets large. As the heat-release rate increases, the time to extinction converges for both cases, showing that large fires extinguish themselves when resident room air is depleted of oxygen even at moderate ventilation rates. Where the heat-release rate is between 212 and 530 kW, the ventilation air supply is sufficient to sustain burning. However, fire-protection systems will initiate before temperature rise reaches damaging magnitudes. If we take credit for fire extinction only by oxygen depletion, we can estimate total smoke production during the burning phase of the fire. These estimates are listed in Table 4.

The empirical estimates in Table 3 come from experimental results conducted under controlled conditions. To temper these estimates, we include two examples of data collected during conditions that were much less than ideal. The first is from an enclosed kerosene pool fire in the LLNL fire test cell, where the forced ventilation rate was controlled to produce an inadequate air supply. We calculated the heat-release rate from the fuel-consumption rate to be approximately 100 kW, though the pool size would have produced a much larger fire if the ventilation rate had been adequate. The total burn time for this test was 2400 s. Pressure

across the HEPA filter in the downstream duct increased by only 20 Pa. For this test the distance between the cell and the HEPA filter is 19 ft, and previous analysis indicates that over such a distance 57% of smoke aerosol will deposit on duct surfaces. (Note that equivalent burning of certain polymers could produce substantially more smoke aerosol. However, these polymers are not in the materials inventory of the tank pits.)

The next example comes from current work we are doing to pre-load HEPA filters before exposing them to heat and smoke from the designed fire tests. Using a standard American Society of Heating, Refrigeration, and Air-Conditioning Engineers (ASHRAE) dust blower and standard ASHRAE dust (mostly charcoal), we are able to load four pounds of the dust onto the filter, which produces a pressure drop of only 500 Pa. With no decrease in flow rate, the HEPA filters used in these tests are rated for 1000 ft³/min. These examples give practical illustrations of "real world" production of smoke and HEPA filter capability to absorb smoke and dust and still be effective for air filtration.

Equilibrium Temperature

Using a semi-empirical relationship for temperature rise produced from a fire in a fully ventilated enclosure, we are able to calculate the steady-state temperature that the hot layer will attain at various times after an induction period where transient conditions dominate.⁴ This is the temperature that would occur if both oxygen depletion did not occur and fire-protection counter measures were

Figure 2. Results of the oxygen depletion study.

Table 1. Solvent burning parameters.

Pool diameter (ft) (m)		Pool area (cm ²)	Burn rate (cm/min)	Burn volume (cm ³ /min)	Burn weight (g/min)
1	0.3	707	0.31	219	145
1.5	0.48	1810	0.41	724	481
2	0.6	2827	0.48	1357	901
3	0.9	6362	0.60	3817	2535
4	1.21	11500	0.65	7475	4963

Table 2. Heat- and smoke-release rate calculations.

Burn weight (g/s)	Heat release (kW)	Soot rate (g/s)
2.4	212	0.12
8.0	530	0.40
15.0	848	0.75
42.3	1908	2.11
82.7	3392	4.14

Table 3. Results of the oxygen depletion studies.

Q (kW)	vent = 0.21 m/s	vent = 0 m/s
7.2		273
530	261	109
848	106	68
1908	34	30
3392	18	17

Table 4. Fire extinction time and total soot production.

Pool diameter (m)	Ext. time no vent. (s)	Total soot (g)	Ext. time 0.21 m (s)	Total soot (g)
0.3	273	33	NO	0.12
0.48	109	43.6	261	104.4
0.6	68	51.0	106	79.5
0.9	30	63.3	34	71.7
1.21	17	70.4	18	74.5

not initiated. Figure 3 shows these temperature levels for fire durations of 250 and 5000 s. The most extreme air temperature rise, 1050°C, is shown for the 3392 kW fire at 5000 s. This temperature rise, while impossible to achieve, would still be diluted eight times and would experience turbulent heat loss before reaching the HEPA filters.

Initial Temperature Transient

A simple plume-entrainment code, ASET-B, is useful to estimate initial temperature increases during the first stages of fire influence in a compartment fire.¹ This code

relates fire heat-release rate to hot-layer growth and temperature rise, without complication of ventilation interaction or heat losses. Estimates using this code are probably invalid for temperature levels above 300°C. Hot-layer temperature vs time for the previously defined heat-release rate cases is plotted in Fig. 4. The time required for temperature rise to reach 80°C is less than the time for oxygen depletion for all but the largest fire. Extracting these times from Fig. 4 gives the data for Table 5, which gives the time for fire-protection sprinklers to fuse.

Figure 3. Equilibrium hot-layer temperatures at 250 and 5000 s.

Figure 4. Hot-layer temperatures plotted vs time for various heat-release rates.

Table 5. Heat-release rate influence on time to sprinkler fuse.

Heat-release rate (kW)	Sprinkler time (s)
212	89
530	46
848	37
1908	32
3392	26

Conclusions

We used simple analytical techniques to assess the fire threat to the containment ventilation system of the transuranium processing plant at Oak Ridge National Laboratory. Our results of this analysis show that the risk of fire in the tank pits or the waste tank pit is minimal. In the event of a threatening fire, normal shut-down procedures will deny air to the fire, causing oxygen starvation to occur in pit regions. Prior to a normal shut-down, rate-of-rise detectors would alert plant emergency personnel to the unusual condition and pre-charge the deluge systems. Then, if the fire growth rate is large, the

temperature will exceed the deluge system set point, and the deluge sprinklers will activate. Any fire or smoke entering the VOG system would be terminated by the caustic scrubber. Finally, smoke generation by pit fires will never be large enough to compromise operation of the HEPA filters.

Fire risk to the HEPA filters in the COG and VOG systems of the transuranium processing plant is minimal, even in the unlikely event of solvent spill and subsequent pit fire.

References

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R. A. da Roza, C. A. Cadena-Fix, and J. E. Kramer, "The Effect of Beard Growth on Respiratory Protection: SCBAs and Air-Purifying Respirators Under Simulated Work Conditions," presented at American Industrial Hygiene Conference, Montreal, Canada, June 1987.

Beards of various types are becoming more popular with the American worker. The reason may be appearance, facial skin condition, or religion. However, the use of a respirator by a person with facial hair is prohibited by ANSI and OSHA standards. Frequent litigation and noncompliance have ensued because the technical evidence to support the standards is not conclusive. We studied the respiratory protection provided by a modern positive-pressure self-contained breathing apparatus and a negative-pressure full facepiece respirator fitted with HEPA cartridges as they were being used under heavy work conditions. Quantitative fit factors were measured with both aerosol and gaseous challenges for ten subjects walking on a treadmill at 80% of their maximum work capacity. We then tested the subjects as their beards were allowed to grow. The tests were made daily for the first week, weekly for the first month, and finally with two-month's growth. The speed of the treadmill was held constant at 3.3 mph while its slope was gradually increased until the subject's heart rate reached 80% of his cardiac reserve. This technique allowed us to perform each test of the series under identical work conditions. In addition to measuring the fit factor, we measured the facepiece pressure, the regulator air flow, and the air bottle pressure of the SCBA as a function of time.

The penetration of challenge into the facepiece was plotted as a function of duration of beard growth for each subject. There were no observable trends in the SCBA plots. That is, the protection factors for these positive-pressure SCBA did not decrease, even with two-month's growth of facial hair. The fit factors were about 40,000 for several subjects, but one subject had fit factors consistently between 100 and 1,000, while two others averaged about 1,000. The time that an air bottle

lasts was measured for each test. No significant trend was seen for this time as a function of beard growth. The "30 minute" bottle lasted about 20 minutes for most subjects. The pressure in the facepiece did not stay positive all of the time. At the start of each breath, the facepiece pressure became negative for up to 80 ms. However, there is no apparent concentration in the facepiece.

Nine of the air-purifying respirator plots showed no significant increase in aerosol penetration for the first week, then the penetration increased with beard growth until the end of the tests. The average fit factor for a clean-shaven subject was 1000. This decreased to about 100 with a two-month growth of beard.

Under the heavy work conditions simulated in these tests, we found that facial hair growth did not decrease the respiratory protection of an SCBA wearer; however, it did decrease the protection of an air-purifying respirator wearer, which is in agreement with previously published data.

D. E. Hankins, "Results Obtained at LLNL from the European/US/Canadian Neutron Dosimeter Irradiations Nov. 1986," EURADOS-CENDOS Report, KIK 4305, September 1987.

The CR-39 foils used in this study were from a sheet made by American Acrylics in September 1986. The CR-39 foils were placed in personnel neutron badges of the type that is routinely worn by personnel at the Lawrence Livermore National Laboratory (LLNL). The CR-39 foils are about 0.025 in. thick and had a 0.005-in. thick polyethylene protective covering on both sides of the foils. The foils are stored in the dark inside a refrigerator prior to being issued. Three foils are stacked and placed into a recess in the back of the badge. The side of the foil to be etched is oriented toward the person. This orientation is used to improve the directional dependence of the dosimeter. A security badge, beta shield, and Panasonic TLD holder are located between the foils and

the source. No light can reach the foils when they are in the badge. Two badges (six foils) were sent for each exposure but in most cases only five of the foils were etched and the results used in this study. The others are being held for track-size distribution studies.

D. E. Hankins, Track Sizes on CR-39 Foils Vary With Incident Neutron Energy. Lawrence Livermore National Laboratory, Livermore, CA, UCRL-95493, 1987.

We recently discovered that the size of the tracks on electrochemically etched CR-39 foils depends on the incident neutron energy. We have determined the track size distribution on foils exposed to monoenergetic neutrons with energies between 81 keV and 16 MeV. The track size distributions vary greatly with energy, enabling one to determine neutron energy information. The track size distribution also varies with etching procedure, and we are currently conducting a parametric study to determine the etching conditions that give the best track size distribution for neutron spectrum unfolding.

D. E. Hankins, S. Homann, and J. Westermarck, "The LLNL CR-39 Personnel Neutron Dosimeter," to be published in *Radiation Protection*.

CR-39 is used in the LLNL personnel neutron dosimeter to determine fast-neutron dose equivalent ($E_n \geq 0.1$ MeV). Three CR-39 foils are used in each personnel badge. The foils are electrochemically etched in an oven maintained at 60°C. We use a three-stage etching cycle. The first etching step is at 60 Hz, 3000 V for five hours. The tracks are made larger during the second stage (blow-up step) at 2000 Hz, 3000 V for an additional 23 minutes. The third stage is a 15-min post etch which gives the tracks a smoother appearance for more accurate optical counting. A biological colony counter is used to count the tracks. The energy dependence of the dosimeter system is nearly flat from about 150 keV to 5.0 MeV. The sensitivity of the dosimetry system is about 5 tracks/cm²/mrem, with a background equivalent to about 8 mrem. The limit of sensitivity is about 10 mrem. No fading of the latent tracks is observed over a one-year period. Other details of the dosimeter system's performance are discussed including precision, accuracy, linearity, and directional response.

D. E. Hankins, S. Homann, and J. Westermarck, "Neutron Spectrum-Dependent Track-Size Distribution of Electrochemically Etched CR-39 Foils," Lawrence Livermore National Laboratory, Livermore, CA.

We have found that the track-size distributions of electrochemically etched CR-39 foils are related to the neutron source. The etching conditions have a significant effect on the track-size distribution, and we are characterizing the changes in track size with etching times, temperature, and high voltage. This will define the etching technique that gives the best resolution in the track-size distribution for the neutron spectra encountered in operational radiation areas. The goal is to develop a catalog of track-size distributions as a function of neutron spectrum that could then be used to assign more accurate spectrum-dependent calibration factors to the results from our personnel neutron dosimeters.

D. E. Hankins, S. Homann, and J. Westermarck, Personnel Neutron Dosimetry Using CR-39 Foils. Lawrence Livermore National Laboratory, Livermore, CA, UCRL-53833 (1987); submitted to the International Conference of Radiation Dosimetry and Safety, Taipei, Taiwan, Republic of China, March 2, 1987.

We are using CR-39 foils for personnel neutron dosimetry at the Lawrence Livermore National Laboratory. We have developed a three-step electrochemical etching procedure that includes low-frequency electrochemical etching at low frequency and at elevated temperatures. Our first etching step is at 60 Hz, 3000 V for five hours in a 60°C oven using 6.5 N KOH. The second etching step consists of an additional 23-min etch at 60°C, 2.0 kHz, and 3000 V, which increases the track size and makes them relatively uniform. The final step is a 15-min post etch at 60°C, which gives the tracks a smoother appearance.

The foils are etched in chambers that can etch up to 24 foils. Several of these chambers can be used simultaneously with a single power supply. These chambers are rugged and have been used daily for over a year.

The sensitivity of the system is about 7 tracks/mrem with a background of 8 to 10 mrem, giving a lower limit of sensitivity of approximately 10 mrem for the dosimeter (when three foils are used). At 400 mrem, the Biotran reader reproducibility is about 1% and the precision of the foil's results has a standard deviation that averages

about 4%. The dosimetry system is linear up to 400 mrem and can be corrected for doses up to 2 rem. The energy dependence is fairly flat from 80 keV to 4.5 MeV, but drops by a factor of 3 in the range 13–16 MeV. It has no response to thermal or intermediate energy neutrons.

We observe no fading of the latent tracks. The dosimeter has a significant variation in directional response, dropping to about 20% at 90°. This can be improved slightly by etching the back side of the foils. We have studied the effect that changes in the etching parameter have on the results.

These parameters include KOH normality, high voltage, frequency, etch time, oven temperature, and pre- and post-etch procedures. We have been using this dosimeter at LLNL as our personnel neutron dosimeter since January 1985.

D. E. Hankins and J. Westermarck, "Preliminary Study on the Use of the Track Size Distribution on Electrochemically Etched CR-39 Foils to Infer Neutron Spectra," Lawrence Livermore National Laboratory, Livermore, CA, UCRL-95780 (abs); to be published in *Radiation Protection*.

We are studying the relationship between neutron energy and track size distribution on electrochemically etched CR-39 foils using a two-stage etching procedure. Although the electrochemical etching process alters the track size, we now find that the track diameter is related to the neutron energy even after the second stage (blow-up) is completed. This relationship is complex, being affected by the etching parameters, including oven temperature, etching high voltage, and etching time. Current studies are directed at establishing the effect of each of these parameters on the track size distribution and at determining an optimal etching procedure for neutron spectra evaluation.

Byron Odell, *Safety Analysis Report of the Combined AVLIS Full-Scale Demonstration Facility—Bldg. 490, Refurbishment Facility—Bldg. 491, Laser Dye Pump Facility—Bldg. 492*, Lawrence Livermore National Laboratory, Livermore, CA, UCID-20831 (1987).

This report documents the safety analysis performed by programmatic and Hazards Control personnel in accordance with DOE Order 5481.1A for the combined

AVLIS Full-Scale Demonstration Facility, Buildings 490, 491, and 492. We considered various hazards associated with normal operations and the credible accidents that might have adverse on-site or off-site consequences. Appropriate safety features to control these hazards were then incorporated into the facility design, thus providing a safe low-risk operation.

This safety analysis determined that potential hazards associated with the operation of the Separator Demonstration Facility (SDF) section of Building 490 and the operation in Building 491 are categorized as "low hazard" per DOE Order 5481.1A.

This safety analysis determined that the potential fire and/or explosion hazards associated with the large quantities of ethanol flowing in Buildings 490 and 492 could result in a considerable on-site impact to people, equipment, and facilities. Therefore, the 490 Complex is categorized as a moderate-hazard facility per DOE Order 5481.1A, due to the potential ethanol hazard.

This analysis also shows: (1) the worst-case postulated accident for a release of radioactive materials will result in a radiation dose of less than 100 mrem at the nearest site boundary, and (2) a criticality accident cannot occur under any condition. Based on this analysis and operational experience gained at other LLNL AVLIS facilities, the Building 490 Complex does not present a potential for a significant nuclear hazard to employees or the general public. Therefore, the Building 490 Complex is designated as a low-hazard nuclear facility as defined by DOE Order 5480.1A, Chapter V. The designation is consistent with other similar facilities at LLNL.

M. S. Singh, *Radiation Associated with an Inertial Confinement Fusion Laser System*, Lawrence Livermore National Laboratory, Livermore, CA, UCID-94044 (1986); submitted to 20th Mid-Year Topical Meeting of the Health Physics Society, Reno, Nevada, February 8–12, 1987.

The primary objective of the LLNL Laser Fusion Program is to understand and develop the science and technology of inertial confinement fusion. Nova, a 100-TW/100-kJ laser system, has demonstrated efficient compression, ignition, and burn of D-T fusion fuel. The LLNL fusion computer program (LASNEX) and experiments strongly support achieving high gain with the proposed multi-MJ laser system. In this paper, I examine the prompt and residual radiations associated with these laser systems.

M. S. Singh, *Production and Shielding of X Rays From Electron Beam Vapor Sources*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-95043 (1986); submitted to 20th Mid-Year Topical Meeting of the Health Physics Society, Reno, Nevada, February 8-12, 1987.

Electron-beam vapor sources are now widely used in material processing sciences and coating technologies, such as the semiconductor industry, for producing aluminum films on Si wafers; the metallurgical industry for melting, evaporating, and refining metals; and at Lawrence Livermore National Laboratory for vaporizing metals for laser isotope separation applications. Power for these sources ranges from the kW regime in the semiconductor industry to the multi-MW regime in laser separation technology. Operations of these sources can generate copious amounts of x rays by the direct and indirect interactions of the energetic electrons with the target materials. In this paper, we present the results of our calculations regarding the x-ray emission intensity, angular intensity and energy spectrum distribution, and shielding characteristics for vapor sources with acceleration voltages from 10 to 60 kV.

C. E. Walter, J. S. Pearson, W. J. Comfort, III, T. F. Harvey, B. T. Kornblum, B. W. Johnson, and C. F. Smith, *Gas-Cooled Reactor Power Systems for Space*, Lawrence Livermore National Laboratory, Livermore, CA, UCID-21008 (1987).

Application of the reactor technology developed in the nuclear ramjet program of the sixties to the multi-megawatt space power required for Strategic Defense Initiative missions has been studied. That gas-cooled thermal reactor technology is found to be applicable. System performance/mass analyses indicate that the Alert Mode System providing 10 MW, for 1 y would have a specific mass of 6.2 Mg/MW. The reactor would be coupled to an axial-flow turbine in a direct regenerated Brayton cycle using helium as the working fluid. The heat pipe radiator for this system contributes two-thirds of the system mass, again registering the need for a breakthrough in heat rejection technology. Startup from a reactor critical condition to full power in 30 s appears to be readily accomplished because of the thermal, mechanical, and nuclear characteristics of the gas-cooled

reactor. The reference reactor uses BeO-UO₂ as the core material, but other choices are available depending on further analyses of specific mission requirements. Additional information is needed on the effects of radiation on these core materials if they are to be used for times in excess of 1 y. Reactor safety and safeguards issues are ameliorated by the inherently low inventory of ²³⁵U in thermal relative to fast reactors. The control swing required is minimized by using a burnable poison in the core. Analyses of potential water immersion and crushing accidents show that the reactor remains subcritical with safing wires placed in the core. The wires would be removed as part of the startup operation.

The same general reactor design is found to be suitable for the Burst Mode System providing 500 MW, for an integrated operating time of 2000 s comprising up to 25 bursts. The characteristics of the reactor appear to allow increasing power from a low level to full power in 5 s. For this application the reactor would be coupled to an axial-flow turbine operating in an open Brayton cycle discharging hydrogen to space. The large inventory of hydrogen discharged from weapon cooling systems (as presently conceived) at about 300 K is more sufficient to serve as the working fluid throughout of the power system. In the study, inlet and outlet hydrogen conditions to the reactor are assumed to be 400 K at 5 MPa and 1200 K at 4.92 MPa, respectively. With these conditions the reactor design limits are not stressed. The reactor would shut down on loss of hydrogen flow because of the strong reactivity contribution of hydrogen at 5 MPa. This feature would be utilized in the control system architecture. With these hydrogen conditions, it is believed that the Burst Mode System could be built with existing technology. If the weapon hydrogen were unavailable, then there would be an incentive to develop higher-temperature technology for the reactor and the turbine to permit more efficient operation and thus a lower hydrogen inventory. In either case, the contribution of the reactor to the system mass is less than 0.01 Mg/MW. The balance-of-plant and the hydrogen inventory represent essentially the complete Burst Mode System mass. A radiation shield would probably not be required. Because of the very large difference in their power levels and operating times it is not likely that both the Alert and Burst Mode applications can be satisfied with one bimodal system.

C. L. Wang, M. D. Pocha, J. D. Morse, M. S. Singh, and B. A. Davis, *Ultrafast Photoconductor Radiation Detectors*, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-96004 (1987); submitted to *Optoelectronic Materials and Devices Integrated Optical Circuit Engineering V* (part of O-E Fibers '87), San Diego, CA, August 17-21, 1987.

We are developing and investigating homogeneous, photoconductive semiconductors as very fast radiation detectors. We irradiated GaAs, Cr-doped GaAs, and Fe-doped InP crystals with 14-MeV neutrons to produce

lattice defects that act as fast-recombination centers for electrons and holes. Using short-pulse lasers and 17-MeV linear-accelerator electrons and bremsstrahlung x rays and γ rays, we have measured the temporal response and sensitivity of these photoconductors as functions of fluence ranging from 10^{12} to 10^{16} neutrons/cm². The carrier lifetime and mobility decrease monotonically as the neutron fluence increases, resulting in faster detector response at the expense of sensitivity. A resolving time of less than 30 ps (FWHM) was measured for the above photoconductors irradiated with $\sim 10^{15}$ neutrons/cm².

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Cover Photographs

(counterclockwise from front)

Electron micrograph of a HEPA filter fiber,
showing particle loading.

Use of laser eye protection with a copper-vapor
laser.

HOT SPOT Health Physics codes can be taken into
the field for rapid initial assessment of nuclear
accidents.

Explosives safety allows for safe, successful tests
at LLNL's Site 300.

Self-Contained Breathing Apparatus research is
conducted in the respirator test facility.