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HAZARDS CONTROL PROGRESS REPORT NO. 45
January Through April 1973

June 1, 1973

Prepared for U.S. Atomic Energy Commission under contract No. W-7405-Eng-48

**LAWRENCE
LIVERMORE
LABORATORY**

University of California/Livermore

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Foreword

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

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

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Progress Reports

INDUSTRIAL HYGIENE

Calculation of Respirator Cartridge Service Life

We determined the service life of organic-vapor cartridges for seven classes of solvent vapors.¹ The experiment consisted of assaulting the cartridge with an upstream concentration of 1000 ppm at 50% relative humidity and monitoring the downstream concentration with a flame ionization detector. In this manner a history of the cartridge efficiency as a function of time is obtained for each solvent vapor of interest. However, it would be useful to be able to predict mathematically when the cartridge efficiency began to deteriorate.

Initial cartridge breakthrough can be calculated from

$$t_b = \frac{10^6 V_m w W_c}{MQC} \quad (1)$$

where:

- t_b = initial cartridge breakthrough time (min)
- V_m = molar volume at the system temperature and pressure (24.1 liters/mole at 20°C 760 Torr)
- w = weight of solvent adsorbed per gram of activated carbon (g/g)
- W_c = weight of activated carbon
- M = molecular weight of the contaminant (g/mole)
- Q = airflow rate (liters/min)
- C = upstream gas concentration (ppm)

All the terms can be measured or calculated except w . The weight of solvent adsorbed is extremely difficult to predict since it is a complex function of the nature of the adsorbent and solvent vapor. Therefore, Eq. (1) is only useful if the weight is determined experimentally. However, estimations of the breakthrough time can be calculated from the adsorption isotherm and the Mecklenburg Equation.

The adsorption isotherm yields the maximum weight of solvent adsorbed at total carbon saturation and can be calculated by:

$$w_s = \rho W_0 \exp \left\{ \frac{-BT^2}{\beta^2} [\log(p_s/P)]^2 \right\} \quad (2)$$

where:

- w_s = equilibrium static adsorption capacity per unit weight of charcoal (g/g)
- ρ = density of solvent (g/cm³)

- W_0 = total volume of the adsorption space (cm³/g)
- B = microporosity constant for the carbon
- T = temperature (°K)
- β = affinity coefficient of solvent vapor carbon
- p_s = saturated vapor pressure for the solvent at temperature T (Torr)
- p = equilibrium partial pressure of the solvent vapor (Torr)

The W_0 and B terms are a function only of the carbon, while β is governed only by the solvent vapor.

The β term can be determined experimentally or approximated by

$$\beta \approx \frac{v}{v_s} = \frac{\rho_s M}{M_s \rho} \quad (3)$$

where:

- v, v_s = molar volume for the unknown and reference solvent (usually benzene) cm³/mole
- ρ, ρ_s = solvent density for the unknown and reference solvent (g/cm³)
- M, M_s = molecular weights for the unknown and reference solvent

Comparison of the actual and experimental values of W_s are shown in Table 1. Note that the more volatile materials show the greatest deviation from the calculated values. This is primarily due to the presence of water vapor being preferentially adsorbed rather than the organic component of the assault gas mixture. Currently there is no satisfactory method of correcting for this interference.^{2,3}

As previously mentioned, w_s is the weight adsorbed at total saturation. However, breakthrough occurs long before the equilibrium adsorptive capacity is established. Nevertheless, such breakthrough times can be estimated from the Mecklenburg Equation once w_s has been determined.

The Mecklenburg Equation states that

$$t_b = \frac{v_s \rho_c A n}{Q C_0} \left[Z + \frac{1}{a_c \rho_c} \left(\frac{dG}{\eta} \right)^{0.41} \left(\frac{\eta}{\rho_s D} \right)^{0.67} \times \ln(C_b/C_i) \right] \quad (4)$$

Table 1. Comparison of calculated and observed adsorption capacity for several solvents^a

	Adsorption capacity, W_s			
	Affinity coefficient (θ)	Observed (g/g)	Calculated from Equation 2	Percent high (+) or low (-) of observed (%)
Type 1 cartridges				
Benzene	1.00 ^b	0.327	0.498	+ 25
Toluene	1.33 ^b	0.473	0.507	+ 7.2
m-Xylene	1.38 ^d	0.536	0.540	+ 0.7
Methanol	0.40 ^b	0.018	0.048	+167
Ethanol	0.61 ^b	0.168	0.251	+ 49
Propanol	0.84 ^c	0.384	0.429	+ 12
n-Butanol	1.02 ^c	0.512	0.495	- 3.3
n-Pentanol	1.22 ^c	0.552	0.525	- 4.9
Methyl Chloride	0.56 ^b	0.068	0.016	+100
Ethyl Chloride	0.76 ^b	0.048	0.129	+169
1-Chloropropane	0.91 ^d	0.196	0.293	+ 49
1-Chlorobutane	1.09 ^d	0.391	0.426	+ 9.0
Chlorobenzene	1.18 ^d	0.562	0.669	+ 19
Dichloromethane	0.66 ^b	0.101	0.216	+114
1,2-Dichloroethane	0.90 ^d	0.456	0.555	+ 22
1,4-Dichlorobutane	1.28 ^d	0.755	0.739	- 4.6
o-Dichlorobenzene	1.36 ^d	0.911	0.851	- 6.6
Chloroform	0.86 ^b	0.415	0.518	+ 25
1,1,2-Trichloroethane	1.08 ^d	0.773	0.822	+ 6.3
Carbon tetrachloride	0.96 ^b	0.677	0.690	+ 1.9
Perchloroethylene	1.21 ^d	1.01	0.965	- 4.5
Methyl acetate	0.89 ^c	0.214	0.333	+ 56
Ethyl acetate	1.10 ^c	0.350	0.442	+ 26
Propyl acetate	1.30 ^c	0.507	0.513	+ 1.2
Butyl acetate	1.48 ^c	0.584	0.550	- 5.8
Pentyl acetate	1.63 ^c	0.616	0.577	- 6.3
Type 2 cartridges				
Acetone	0.88 ^b	0.135	0.236	+ 75
2-Butanone	1.01 ^c	0.295	0.360	+ 22
2-Pentanone	1.19 ^c	0.483	0.427	- 12
2-Heptanone	1.57 ^c	0.560	0.493	- 12
Pentane	1.08 ^b	0.228	0.225	- 1.3
Hexane	1.295 ^b	0.334	0.319	- 4.5
Heptane	1.46 ^a	0.432	0.375	- 13
Nonane	1.85 ^d	0.490	0.434	- 11
Decane	2.05 ^d	0.553	0.442	- 20
Methyl amine	0.72 ^d	0.041	0.061	+ 49
Ethyl amine	0.91 ^d	0.179	0.166	- 7.3
Butyl amine	1.29 ^d	0.429	0.371	- 14
Dibutyl amine	1.25 ^d	0.480	0.466	- 2.9

^aThe equilibrium partial pressure, p , at 1000 ppm is 0.76 Torr. The saturated vapor pressure, p_s , is taken from handbook values.

^bExperimental values taken from Ref. 12.

^cCalculated from Eq. 3.

^dCalculated from parachors using Sugden's equation.

Table 2. Comparison of predicted and actual 1 percent breakthrough times^a

Time required to achieve one percent breakthrough (10 ppm)			
Material	Actual time measured using FID (min)	Calculated from Mecklenburg equation	
		Using experimental weight adsorbed (min)	Using calculated weight adsorbed from adsorption isotherm (min)
Type 1 cartridge's			
Benzene	73.3	74.2	92.6
Toluene	94.3	89.1	95.5
m-Xylene	98.7	82.0	82.6
Methanol	0.2	10.9	29.1
Ethanol	28.0	67.6	101
n-Propanol	70.4	115	128
n-Butanol	115	120	116
n-Pentanol	102	104	99.4
Methyl chloride	0.05	3.0	5.9
Ethyl chloride	5.6	13.2	35.6
1-Chloropropane	24.5	43.0	64.3
1-Chlorobutane	72.3	70.8	77.1
Chlorobenzene	107	83.8	99.7
Dichloromethane	10.1	21.6	46.2
1,2-Dichloroethane	54.0	81.2	98.8
1,4-Dichlorobutane	108	96.4	91.9
o-Dichlorobenzene	109	99.6	93.0
Chloroform	33.2	61.0	76.1
1,1,2-Trichloroethane	71.8	98.7	105
Carbon tetrachloride	77.0	75.8	77.3
Perchloroethylene	107	104	99.3
Methyl acetate	32.8	51.7	80.5
Ethyl acetate	66.8	69.1	87.3
n-Propyl acetate	78.8	83.9	84.9
n-Butyl acetate	77.3	81.8	77.0
n-Pentyl acetate	72.6	74.5	69.8
Type 2 cartridges			
Acetone	37.1	58.3	102
2-Butanone	81.9	101	123
2-Pentanone	104	137	121
2-Heptanone	101	116	102
n-Pentane	60.7	77.6	76.6
n-Hexane	52.3	93.7	89.5
n-Heptane	78.2	103	89.1
n-Nonane	76.2	89.4	79.2
n-Decane	70.8	90.3	72.2
Methyl amine	12.4	33.6	50.6
Ethyl amine	40.5	99.3	92.1
n-Butyl amine	110	145	125
Di-n-Butyl amine	75.5	86.9	84.4

^aTest conditions were 53.3 liters minute, 50% relative humidity, and 20°C.

where:

- ρ_c = carbon density (g/cm³)
- A = cross sectional area of the adsorbent bed (cm²)
- n = number of cartridges tested
- C_0 = assault concentration (g/liter)
- Z = bed depth (cm)
- a_c = specific surface area (cm²/g)
- d = diameter of granule (cm)
- G = mass velocity through cartridge (g/cm²-sec)
- η = viscosity of the air gas stream (g/cm-sec)
- ρ_a = air density (g/cm³)
- D = diffusion coefficient (cm²/sec)
- C_b = breakthrough concentration (ppm)
- C_i = assault concentration (ppm)

The predicted and actual breakthrough times are compared in Table 2. In general, the predicted times are somewhat more optimistic than those determined experimentally. This again can be explained by the contribution of water to the available adsorption sites.

The data in Table 2 show conclusively that approximate breakthrough times can indeed be calculated, but only if the activated carbon is well characterized. Also, a best approximation can only be made if the contribution of the water vapor is taken into account. This is especially true for solvents that are miscible with water or have a relatively high vapor pressure.

FIRE SAFETY

Performance Characteristics of Three Types of Heat Actuated Detectors as Tested in LLL Experimental Duct System

Two series of tests (A and B) were run on fusible rods, fusible plugs, and sprinkler heads with fusible links. Series A tests were run with the temperature varying according to the standard time temperature curve.⁴ These tests were run to determine the time and temperature at which the heat actuated detectors would operate under "standard" fire conditions. In the Series B tests the temperature was raised very slowly

from ambient until the detector actuated. Series B tests were conducted to determine the reliability of the detector ratings.

The detectors were tested in the ductwork of the LLL Experimental Duct System.⁵ The velocity of the air passing the detectors was 160 linear ft/min (2.67 lfs), which was in the turbulent flow region ($Re = 10^4$). The detectors and thermocouple were arranged as shown in Figs. 1 and 2.

The results are shown in Tables 3 through 11 and Figs. 3 through 6.

Table 3. Summary of results

Detector	Series A	Series B
Fusible rods	Slowest detector to actuate	Significant differences between activation temperature and detector rating.
Fusible plugs	Fusible plugs detector activations and temperature detector activations occurred between fusible rods and sprinkler heads activations	Significant differences between activation temperature and detector rating.
Sprinkler heads	Fastest detector to actuate	No significant difference between activation temperature and detector rating.

Table 4. Detector tests, Series A^a

Detector	Detector rating, °F	Temp. at actuation, °F	Actuation time, min.	Lag time, ^b min.
Fusible rod	174	492	1.34	1.23
Fusible rod	197	540	1.56	1.43
Fusible rod	228	627	1.93	1.75
Fusible rod	253	692	2.17	1.89
Fusible plug	168	401	0.93	0.84
Fusible plug	210	444	1.38	1.27
Fusible plug	283	595	1.94	1.61
Sprinkler head	160	290	0.36	0.27
Sprinkler head	212	344	0.53	0.40

^aSeries A tests were run with the temperature varying according to standard time-temperature curve.

^bThe actuation time minus the time to reach rated temperature.

Fig. 1. Fusible rods and plugs experimental schematic.

Fig. 2. Fusible rod and head setup.

Fig. 3. Fusible plug and fusible rod detector tests. Series A - Temperature varied according to ASTM standard time - temperature curve.

Fig. 4. Fusible plug and fusible rod detector tests. Series A - Temperature varied according to ASTM standard time - temperature curve.

Fig. 5. Fusible plug and fusible rod detector tests. Lag time - Actuation time minus time to reach rated temperature. Series A - temperature varied according to ASTM standard time - temperature curve.

Table 5. Detector tests, Series B^a

Detector	Detector rating, °F	Actuation temp., °F
Fusible rods	174	213
Fusible rods	197	240
Fusible rods	228	293
Fusible rods	253	329
Fusible plugs	168	188
Fusible plugs	210	292
Fusible plugs	283	331
Sprinkler heads	160	167
Sprinkler heads	212	210

^aSeries B tests were run with the temperature being increased very slowly from ambient until the detector actuated.

Table 6. Series A^a fusible rod detector tests

Detector rating, °F	Temp at actuation, °F	Actuation time, min.	Lag time, ^b min.
174	510	1.33	1.22
174	554	1.61	1.48
174	505	1.30	1.20
174	438	1.19	1.10
174	455	1.25	1.15
AV	492	1.34	1.23
197	564	1.62	1.50
197	668	1.90	1.72
197	483	1.40	1.28
197	490	1.51	1.39
197	493	1.39	1.25
AV	540	1.56	1.43
228	656	1.82	1.71
228	670	2.03	1.82
228	609	1.85	1.69
228	595	2.02	1.82
228	604	1.92	1.70
AV	627	1.93	1.75
253	707	1.98	1.76
253	714	2.23	1.91
253	693	2.11	1.83
253	655	2.17	1.89
253	692	2.35	2.04
AV	692	2.17	1.89

^aSeries A tests were run with the temperature varying according to standard time-temperature curve.

^bThe actuation time minus time to reach rated temperature.

Fig. 6. Fusible plug and fusible rod detector tests.
Series B - Temperature increased very slowly from ambient until detector actuated.

Table 7. Series A^a fusible mount filled pipe plug test

Detector Rating, °F	Temp. at actuation, °F	Actuation time, min.	Lag time, ^b min.	Position of fused plug ^c top/bottom
168	408	1.02	0.91	Top
168	399	0.88	0.79	Top
168	408	0.82	0.74	Top
168	389	1.01	0.90	Both
AV	401	0.93	0.84	
210	483	1.31	1.22	Top
210	424	1.68	1.55	Top
210	428	1.32	1.20	Top
210	440	1.21	1.09	Top
AV	444	1.38	1.27	
283	535	1.72	1.40	Top
283	598	1.98	1.66	Top
283	642	2.10	1.72	Both
283	604	1.96	1.65	Top
AV	595	1.94	1.61	

^aSeries A tests were run with the temperature varying according to standard time-temperature curve.

^bThe actuation time minus time to reach rated temperature.

^cIn all cases the bottom plug had begun to fuse also top plug 5 in. above center line, bottom plug 5 in. below center line.

Table 8. Series B^a fusible rod detector tests

Detector rating, °F	Actuation temp., °F
174 ^b	201
174	222
174	216
AV	213
197	234
197	243
197	243
AV	240
228	275
228	313
226	290
AV	293
253	304
253	317
253	365
AV	329

^aSeries B tests were run with the temperature being increased very slowly from ambient until the detector actuated.

^bIn one test on 174°F rated detector, the detector did not actuate the trip valve although the detector had operated.

Table 9. Series B^a fusible metal filled pipe plug test

Detector rating, °F	Actuation temp., °F
168	188
168	188
168	189
AV	188
210	296
210	305
210	277
AV	292
283	317
283	342
283	334
AV	311

^aSeries B tests were run with the temperature being increased very slowly from ambient until the detector actuated.

Table 10. Series A^a sprinkler head tests

Detector rating, °F	Temp. at actuation, °F	Actuation time, min.	Lag time, ^b min.
160	292	0.33	0.24
160	288	0.33	0.29
AV	290	0.36	0.27
212	337	0.53	0.40
212	330	0.53	0.40
AV	344	0.53	0.40

^aSeries A tests were run with the temperature varying according to standard time-temperature curve.

^bThe actuation time minus time to reach rated temperature.

Table 11. Series B^a sprinkler head tests

Detector rating, °F	Actuation temp., °F
160	167
212	210

^aSeries B tests were run with the temperature being increased very slowly from ambient until the detector actuated.

HEPA Filter Fire Protection. - Smoke Plugging Problems

Introduction. In our report⁶ we related the results of our work in plugging HEPA filters with smoke; the use of wetting agents and "best" arrangements of sprays, scrubbers, and demisters in efforts to wash out smoke particulates; and the technical points raised at a December 1972 meeting regarding realistic burning rates for plastics of interest as well as agglomeration-aging effects on smoke particulates. In the same report, we also suggested a meeting of concerned AEC- and Contractor-personnel to reevaluate the scope of the project and suggested modes of attack.

Such a meeting was held early in April 1973; it is being reported by AEC Headquarters. Prior to the meeting, our work involved the following activities:

- We made a detailed resurvey of the combustible loading of a pertinent laboratory building with particular attention to those materials that yield dense smokes on burning.
- Using Harmathy's approach⁷ and the data obtained from the survey, we calculated the burning rates of the fuel loading and the fire duration.
- Using the NBS/LLL Smoke Chamber, we attempted to age smokes from three materials of interest and determine the changes in particle size distribution.
- Employing burning neoprene as a source of smoke in the experimental duct system, we evaluated the effects of different prefilters on system flow rates and the plugging of the HEPA filters.

This report will deal with the results of these efforts and outline our proposed future work in the field.

Building Survey. The results of the building survey are shown in Tables 12, 13, and 14.

The maximum fuel loading in any one laboratory is about 3 lb/ft². Table 13 shows the maximum fuel loading in the various rooms. This table also shows the fuel loading adjusted to the wood equivalent.

Using Harmathy's equations for one laboratory (Room 139), the peak burning rate was computed to be 16.56 lb/min. The fire duration was calculated to be about 2 hr.

Subsequently, additional calculations were made using filter plugging rates furnished by S. T. Ewing.⁸ These results indicate that by using a Δp -vs-fuel burned curve developed experimentally, and Harmathy's burning rates, the pressure drop across the filter at a given time may be estimated.

Smoke Loading. Examination of Table 14 shows that on the bases of weight of materials and areas exposed, the glove box window material

Table 12. Fuel loading analysis plutonium research building at Lawrence Livermore Laboratory

Heavy smoking materials	Light smoking fuels
P - Polymethyl methacrylate	Paper
V - PVC film and pipe	Cardboard
R - Chlorinated rubber	Wood
B - Pressed board	Polyethylene
U - Polyurethane foam	

Table 13. Fuel load summary

	Maximum fuel load, lb/ft ²	
	Actual	Adjusted ^a
Labs	2.97	3.45
Offices	29.7	27.0
Equipment area	0.0127	0.0132
Hall	0.146	0.167

^aAdjusted to the equivalent of wood based on heat of combustion.

polymethylmethacrylate is the most prevalent. Next comes chlorinated rubber (gloves), followed closely by plasticized vinyl film (bag-out material). One laboratory room contained a significant weight and area of pressed board (workbench tops). Unknown quantities of the glovebox window material are of the fire retardant type. Fire-retarded methacrylate, plasticized vinyl, and chlorinated rubber yield dense smokes when burned (see next section). We have been using these materials in our HEPA-filter smoke plugging tests.

Smoke Chamber Studies. Using the NBS/LLL Smoke Chamber^{9,10} under slightly ventilated conditions, samples of the three above-mentioned materials were exposed to a radiant heat flux of 2.5 W/cm² with and without the presence of a pilot flame. The smokes evolved were analyzed for

- Specific optical density vs time of exposure
- Particle size 1) as freshly generated in the chamber, and 2) as aged 1 to 3 min by passage through a 2-in. diam tube
- Concentration of carbon monoxide, carbon dioxide, and oxygen

Results of these experiments are shown in Figs. 7 through 12 and Table 15. The following observations are made from these figures.

Table 14 Quantities of heavy smokers exposed in laboratory areas

Material	P	V	R	B	U
Maximum area in one lab (ft ²)	319	450	316	119	10
Per floor area (ft ² /ft ²)	0.33	0.46	0.22	0.12	0.008
Maximum weight in one lab (lb)	1200	40	180	350	2
Per floor area (lb/ft ²)	1.23	0.04	0.19	0.35	0.002
Distribution - Area					
% of labs with					
>200 ft ²	19	6	0	0	3
>100 ft ²	24	19	25	5	0
>50 ft ²	71	35	55	5	0
>25 ft ²	81	60	80	10	0
Distribution - Weight					
% of labs with					
>700 lb	19	0	0	0	0
>300 lb	35	0	0	0	0
>100 lb	80	19	25	10	0
>50 lb	86	20	40	10	0

Totals for heavy smoking materials

Area exposed (ft²/ft² of floor area) - 0.11

Weight exposed (lb/ft² of floor area) - 0.26

P = polymethylmethacrylate

R = chlorinated rubber

V = PVC film and pipe

B = pressed board

U = polyurethane foam

Table 15. Same gaseous components of smokes formed by pyrolyzing or combusting materials of interest

Pyrolysis smokes			Material of interest	Combustion smokes		
Quantity/time				Quantity/time		
Max	Max	Min		Max	Max	Min
CO	CO ₂	O ₂		CO	CO ₂	O ₂
(ppm)	(%)	(%)		(ppm)	(%)	(%)
600/20	0	21	Neoprene	1100/15	2.0/10	18.8/10
200/12	0	21	PVC film	1800/6	2.0/6	21
1000/14	0.2/14	21	FR methacrylate	8500/8	3.3/12	18/12

Fig. 7. Smoke density development for materials of interest.

Fig. 8. Particle size distribution by weight of fresh smokes and smokes aged 1 to 3 min for materials of interest.

Fig. 9. Smoke density and weight-loss vs time for neoprene (non-flaming exposure).

Fig. 10. Smoke density and weight-loss vs time for neoprene (flaming exposure).

Fig 11. Smoke density and weight-loss vs time for polymethylmethacrylate (non-flaming exposure).

Fig 12. Smoke density and weight-loss vs time for polymethylmethacrylate (flaming exposure).

- All three materials rapidly produce very dense vision-obscuring smokes. (Note the comparison with pyrolyzing and flaming red oak.)
- Under burning conditions these three materials, most particularly the methacrylate, produce significant quantities of carbon monoxide with only a slight diminution in the oxygen concentration.
- There is a tendency for the pyrolytically-obtained smoke particles to grow in size during the limited aging available in the experiments. This tendency is not evident in the combustion smokes.
- Burning rates are on the order of 0.1 to 0.2 lb/min-ft² exposed for the given heat flux plus pilot flame. Pyrolysis rates are lower.

Duct System Tests. It is common practice to use some kind of a relatively inexpensive prefilter in the exhaust ventilation system ahead of the HEPA filter to minimize the number of replacements of the latter under normal atmospheric dust loadings. In our early work in protecting HEPA filters against heat, we found that a particular prefilter was readily destroyed by temperature of about 1000°F. Hence, we did not use a prefilter in our subsequent work in this area.

However, in the early stages of a fire where temperatures are low (say less than 1000°F), smoke plugging of the prefilter may be important. Therefore, we employed the following three prefilters in the smoke plugging tests:

- One with an indicated efficiency of 90+%
- One with an indicated efficiency of 80+%
- A 1-inch thick household furnace filter

In each test a new prefilter was inserted in the duct upstream of a new HEPA filter, and the ambient air flow rate was adjusted to 1000 cfm. Cold smoke from burning neoprene (fuel rate = approximately 50 g/min) was introduced upstream of the prefilter and the air flow rate and pressure differentials across the prefilter and HEPA filter were monitored.

Typical results of these tests are shown on Figs. 13, 14, and 15. The 80%- and 90%-prefilters are loaded up with smoke, thus protecting the HEPA filters, but reduce the ventilation rate to a very low value, i.e. 40-50% of the original flow. The same phenomenon is true but in a somewhat different manner for the furnace filter. In this case, the furnace filter loads up while passing some of the smoke particles through to the HEPA filter, which in turn starts to load up. As smoke accumulates on the furnace filter the air flow decreases markedly, until this prefilter suddenly ruptures. At this point the air flow rises sharply and

Fig 13. Pressure drop and air flow of ventilation system with 90+ % pre-filter and HEPA filter subjected to neoprene smoke.

Fig 14. Pressure drop and air flow of ventilation system with 80+ % pre-filter and HEPA filter subjected to neoprene smoke.

then decreases as the HEPA filter becomes clogged and its pressure differential increases.

Of interest is the particle size analysis (on a weight basis) of the neoprene smoke. This was measured at two different points in the duct system without any filters, scrubbers, etc. in place. Note the practical identity of the size distribution, and also that 60% of the smoke weight consists of particles smaller than 1 micron in size (see Fig. 16).

Discussion. The smoke chamber tests reported here suggest that in an actual fire situation smoke particles

*The efficiencies of these prefilters were measured by the NBS stain test. This is not comparable to the DOP test used in evaluating the efficiency of HEPA filters.

Fig 15. Pressure drop and air flow of ventilation system with furnace-type pre-filter and HEPA filter subjected to neoprene smoke.

may agglomerate somewhat as they pass from the point of generation to an exhaust ventilation inlet some distance away. The degree of agglomeration may well depend upon the fuel source and the

pyrolysis-combustion ratio as well as the time of travel. In addition, smoke particulates (depending upon the fuel and mode of generation) will contain a certain condensable fraction that can precipitate out on cooler

Fig. 16. Smoke particle analysis for neoprene smoke in duct work.

surfaces within the room of origin. This suggests further work in this area as follows: 1) Exposing materials of interest to different heat fluxes (with and without a pilot flame) in the smoke chamber and allowing the generated smokes to pass through a simulated room (a steel box), and 2) measuring the quantity of smoke particulates and particle size distribution entering and leaving the box. To obtain further information in the same experiment, small prefilters and HEPA filters can be placed in an exhaust ventilation system drawing air and smoke from the box. Thus, materials of interest can be studied on a small scale in regard to their burning rates, smoke generation and aging properties, and prefilter- and HEPA-filter plugging characteristics. Besides the three materials of concern at LLL, there are a number of materials used at other AEC-Contractor installations that should be examined in this manner.

At the same time, Harmathy's equations will be applied to these other materials in an effort to determine their theoretical burning (hence smoke producing) rates. This information will be compared to laboratory and larger scale data as the latter are developed.

As a check on the calculations and laboratory tests, we propose to build a 15 X 15 X 15-ft steel box, and insert it into the large-scale, experimental duct system. This would be used to pilot-scale certain results obtained in the laboratory.

As the experimental information becomes available, means will be sought and tried to abate the smoke-filter-plugging problem. It is obvious that substitutes for heavy smokers should be used wherever possible, but there are some items, e.g. elastomeric gloves that must remain. Further, there is the cost of replacing existing materials that must be considered.

For the immediate future we propose to do the following:

- Evaluate theoretically the burning (smoking rates) of materials of interest.
- Use the smoke-chamber variable-burning-rate, aging-agglomeration-condensation, and filter-plugging technique described above.
- Confirm smoke-chamber findings with larger scale tests.
- Try various smoke abatement techniques in a systematic manner.

Progress in Multisphere Neutron Spectrometry

It is very important to determine the dose equivalent (D.E.) per unit neutron fluence for sources and source-moderator configurations necessary for dosimeter calibrations, as well as determining the spectra encountered in daily radiation protection work. Survey instruments have been developed which have energy response functions that approximate the D.E. curve.^{11,12,13} However, the use of these instruments to characterize radiation fields have at least two drawbacks. In the first place, they over-respond in the intermediate energy region, by as much as a factor of two or more at some energies. In addition, the response of these units has been chosen to match a curve based on RBE data provided by radiobiological experiments and is not, therefore, a physically measurable quantity. When more complete data of this sort becomes available, particularly in the intermediate energy region, the D.E. curve is very likely to change, making it difficult to compare D.E. estimates made with instruments of this type. As a result, we feel that it is important to determine the neutron spectrum, whenever practical, to make accurate D.E. estimates.

Neutron spectrum measurements at the low levels necessary for radiation protection are difficult to make, particularly in "field" situations. Neutron spectrometry systems generally lack adequate sensitivity, as well as capability for spectral resolution at energies below 10 keV. One system that can be used to overcome these difficulties is the multisphere spectrometer.¹⁴ The multisphere system is not a spectrometer in the true sense, but involves use of a set of polyethylene spheres to moderate the spectrum before it reaches the neutron detector. The effect is to have a set of neutron detectors with rather broad response functions whose peak energy response increases with increasing sphere size. The detector usually used in these systems is a ⁶LiI scintillation crystal with output to a scaler. The potential energy resolution of such systems is poor, but it is adequate for neutron protection purposes.

The use of the multisphere system requires that accurate response function data be available not only for the bare detector but for the detector-moderator combinations. The data analysis also requires a spectrum unfolding capability. (This can be provided by an available computer code.)

Available response functions for ⁶LiI scintillation polyethylene spheres have been calculated by other experimenters.^{15,16,17} The experimental data taken in response of this system is insufficient for spectrometry over the full range of energies necessary.

Recent calculations by Sanna¹⁸ have demonstrated the importance of considering polyethylene density in sphere response calculation. In fact, the calculations show that for some detectors an error of 10% in polyethylene density would lead to an error of more than a factor of two in the response function at low energies. Measurement of our own set of moderators show a spread of 5% in sphere density.

Sanna's work has additional value in that it includes response calculations for water moderators as well as polyethylene. This leads to the possible use of water-loaded metal shells that could be filled in place, thus eliminating the transport of heavy polyethylene spheres. Sanna's calculations also include response functions for gold foil detectors which are less sensitive than the ⁶LiI scintillator. This would eliminate the need for the electronics associated with scintillation detectors, allow for measurements with all spheres at once, and permit overnight or weekend measurements.

One set of calculations that we plan to make to augment the available data will involve the use of a ²³⁵U thin foil as a detector. This foil could be used as a radiator in a track detection system and should give us a sensitivity comparable to that obtained with the ⁶LiI scintillator, but again without the necessity for the associated electronics.

Most of our effort on the multisphere spectrometer system has been spent on obtaining an unfolding code. We have looked at the LOUHI code,¹⁹ which was developed at the Lawrence Berkeley Laboratory. LOUHI was attractive to us because it appears to be quite feasible with potential use for other unfolding problems, such as the use of activation foils.

For neutron protection purposes, we feel the best index of unfolding code performance is how well it can reproduce the D.E. weighted spectrum. In this type of analysis, determining the high energy (>0.1 MeV) portion of the spectrum is most important. To make this analysis with LOUHI, we began by calculating the multisphere responses for a series of 28 neutron spectra, most of them taken from the SAND II²⁰ computer code library. These calculated responses were then given to LOUHI using the same detector response functions. It can be seen that the accuracy of response function values for this test is not important, since it is a paper experiment and the responses are cancelled out in the process. Both the LOUHI calculated spectra and the original spectra were weighted by the D.E. response function. Figures 17, 18, and 19 show the comparison of the D.E. weighted spectra for three cases. The D.E. per unit neutron fluence were compared, and we found the maximum difference for the 28 spectra selected was 8.6% with a standard deviation of 3.4% for the entire group.

Fig. 17. Dose equivalent weighted spectrum - Reciprocal energy. SAND II input spectrum - 0.675×10^{-4} rem/n $\cdot$ cm $^{-2}$. LOUHI calculated spectrum - 0.660×10^{-4} rem/n $\cdot$ cm $^{-2}$.

Fig. 19. Dose equivalent weighted spectrum - Fermi thermal shield. SAND II input spectrum - 0.295×10^{-4} rem/n $\cdot$ cm $^{-2}$. LOUHI calculated spectrum - 0.281×10^{-4} rem/n $\cdot$ cm $^{-2}$.

Fig. 18. Dose equivalent weighted spectrum - Fission through 90 cm H $_2$ O. SAND II input spectrum - 2.09×10^{-4} rem/n $\cdot$ cm $^{-2}$. LOUHI calculated spectrum - 2.13×10^{-4} rem/n $\cdot$ cm $^{-2}$.

As a result of these tests, we feel that the LOUHI computer code is capable of unfolding multisphere spectra with sufficient accuracy for neutron protection purposes, and certainly more simple codes may also produce adequate results. The emphasis now will be to evaluate our multisphere system and available response function data in order to provide us with a working system useful for "field" measurements. At the same time, we will evaluate the possible use of water moderators and passive detectors for easier field measurements.

Technical Notes

INSTRUMENT DEVELOPMENT

GAMANAL Now Available for Gamma Ray Spectral Analysis

The computer code GAMANAL developed by Niday and Gunnink²¹ has been adapted for execution on the CDC-7600 computers. The adapted version has been compiled and stored on tape for access by Hazards Control users.

Complete documentation including user's manual, listing, and general description are contained in Ref. 21. Briefly, the code is written for the analysis of high-resolution, gamma-ray, pulse-height spectra taken with Si(Li) or Ge(Li) detectors. All photopeaks in the spectra are located and the continuum is determined. Precise photopeak areas and centroids are

determined directly. The user may also supply energy, timing and efficiency data in which case photon energies and intensities are determined.

Options exist for automatic identification of escape peaks, for radioactive decay corrections, and for geometry corrections. If requested, a tape library of decay scheme data may be searched in order to provide automatic identification of nuclides whose gamma rays are observed in the spectra.

At present input data for GAMANAL can be prepared on cards, magnetic tape, or via a disc file created by any other media. In the latter case, data can be input via tape cassette as soon as this capability of the RJET terminal is realized.

Publications

P. H. Gudiksen, C. L. Lindeken, J. W. Meadows, and K. O. Hamby, *Environmental Levels of Radioactivity in the Vicinity of the Lawrence Livermore Laboratory 1972 Annual Report*, Lawrence Livermore Laboratory Rept. UCRL-51333 (1973).

The Lawrence Livermore Laboratory continuously monitors the levels of radioactivity within the Livermore Valley and Site 300. Results of analyses performed during 1972 for gross radioactivity and for specific radioisotopes of interest in a variety of environmental samples are presented in this report. In all cases, the levels of radioactivity are found to be below the concentration guide values in AEC Manual Chapter 0524.

Airborne particulate samples showed gross beta activities comparable to those measured during previous years. Gamma spectral measurements of these samples revealed detectable activities of typical gamma emitters observed in global fallout. The samples were also analyzed for ^{239}Pu , ^{238}Pu , ^{235}U , and ^{238}U activities. The monthly averaged ^{239}Pu activities were in the range of $5 \times 10^{-17} \mu\text{Ci/ml}$ while the ^{238}Pu activities were about an order of magnitude lower. These activities are typical of those due to global fallout.

Air samples taken at Site 300 were analyzed for uranium. These analyses showed a lower than normal ratio of ^{235}U to ^{238}U . This is most likely due to the resuspension of "depleted" uranium (uranium specially processed to lower the ^{235}U content) used at the Site. The airborne uranium concentrations are well within the standards set by the AEC.

Soil samples collected within the Livermore Valley and Site 300 were subjected to uranium, plutonium, strontium and gamma spectral analyses. Samples collected within several hundred meters of the firing bunkers at Site 300 were depleted in ^{235}U ; however, samples collected on the site perimeter and off-site showed normal $^{235}\text{U}/^{238}\text{U}$ ratios. The mean ^{239}Pu deposition value on soil is about $1 \times 10^{-3} \mu\text{Ci/m}^2$ which appears to be typical of global fallout in this area.

Traces of plutonium were detected in one of the dry holding ponds at the Livermore Sewage Treatment Plant through which the Laboratory's sanitary sewage is processed. The maximum ^{239}Pu activity in the plant effluent was less than 3/100 of one percent (0.03%) of the AEC drinking water standard.

Gamma spectral analyses of vegetation samples revealed no gamma-emitting radionuclides other than those present naturally or in global fallout. The vegetation samples collected in areas generally

downwind from the Livermore Laboratory revealed tritium activities that were 10 to 100 times higher than those collected in areas where the Laboratory's contribution should be minimal. However, these activities are within recommended standards.

Water samples collected within the Livermore Valley exhibited normal background gross beta and tritium activities. Similar levels were observed at Site 300, with exception of one spring containing a higher than normal tritium concentration. Although elevated in comparison with the others, the tritium activity in this spring is below the concentration guide value of drinking water set by the AEC. The off-site radiation exposure rates measured by thermoluminescent dosimeters are within the range to be expected from natural radiation sources.

Assessment of the radiation doses to an individual from the observed environmental activities indicates the contribution from artificially produced radionuclides is small in comparison with the approximately 100 mrem/yr dose received from natural sources. An adult who breathes air, drinks water, and eats vegetation containing the radionuclides at the concentrations shown in this report will receive less than 1 mrem/yr from the artificially produced radionuclides, with the exception of ^{144}Ce from global fallout, which may deliver several mrem/yr to the organs of interest.

G. O. Nelson, *Preparation of Static Calibration Mixtures*, Lawrence Livermore Laboratory Rept. UCRL-74441 (1972).

A summary of the techniques and methods for producing known concentrations of gas and solvent in air is presented. Major topics include contaminant introduction, chamber configurations, calculations and sources of error.

This paper will be included as a book chapter in "Methods of Air Sampling and Analysis" published by the American Public Health Association.

J. S. Luce, H. L. Sahlin, and T. R. Crites, *Collective Acceleration of Intense Ion Beams In Vacuum*, Lawrence Livermore Laboratory Rept. UCRL-74681 (1973).

We have developed a diode system with an insulated anode and rear target that operates in vacuum and produces 4×10^{10} neutrons per pulse. The system forms a pinch discharge that ejects a cluster of collectively accelerated ions that attain full energy in 7 cm. Target neutron data and activation analysis with multiple foils show that pulses of 10^{14} protons with

energy up to 15 MeV have been achieved. Experiments with electrode materials and diode geometry have lead to large improvements in focusing and ion beams have been propagated 8 ft in vacuum.

The presence of high-energy carbon ions indicates that the pinch has the capability of forming and accelerating multiple ionized high β ions.

This paper has been submitted for publication in IEEE Transactions on Nuclear Science.

Z. E. Macbain, Affirmative Action Training Program for Firemen, Lawrence Livermore Laboratory Rept. UCRL-74605 (1973).

This paper summarizes the "Affirmative Action" training of unskilled minorities for jobs as firemen. The various training approaches undertaken are discussed; some were more successful than others. Hopefully, the information provided in this report can be used by other fire departments as a basis for developing their own minority "Affirmative Action" training programs.

This paper was prepared for submittal to Fire Departmental Instruction Conference at Kansas City, Missouri on March 27-30, 1973.

R. V. Griffith, Trip Report - IAEA Symposium Neutron Monitoring for Radiation Protection Purposes, Lawrence Livermore Laboratory (1973).

The IAEA Symposium was held in Vienna, Austria on December 11-15, 1972. This was an informational exchange for people primarily involved in applied health physics or health physics research and interested in the estimation of neutron dose to laboratory workers. Representatives were present from 30 countries and 9 international organizations. Approximately 64 papers were presented, including two or three unscheduled papers that were included at the last minute. The general topic areas discussed were:

- Neutron spectra - 11 papers
- Dose and dose equivalent - 5 papers
- Instruments and techniques, field monitoring - 9 papers
- Instruments and techniques, personnel monitoring - 25 papers
- Comparison of various methods and monitoring systems - 7 papers
- Standardization and calibration - 5 papers

C. L. Lindeken, P. H. Gudiksen, J. W. Meadows, K. O. Hamby, and L. R. Anspaugh, Environmental Levels of Radioactivity in Livermore Valley Soils, Lawrence Livermore Laboratory, Rept. UCRL-74424 (1973).

Soil samples collected from areas surrounding the Lawrence Livermore Laboratory in the Livermore Valley were analyzed for ^{239}Pu , ^{238}Pu , ^{241}Am , ^{90}Sr ,

^{137}Cs and naturally occurring gamma emitters, as part of a continuing environmental surveillance program. The median ^{239}Pu activity corresponds to a surface deposition value of 1.1 mCi/km^2 . ^{238}Pu and ^{241}Am activities were about a factor of 10 lower than that of the ^{239}Pu . Median ^{90}Sr and ^{137}Cs deposition values were 18 and 31 mCi/km^2 , respectively. All deposition values appear to be typical of those expected from global fallout in this area. At present, ^{137}Cs is the principal fallout gamma emitter observed. The radiation exposure rate at 1 meter calculated from the median ^{137}Cs soil activity was about 0.1 $\mu\text{R}/\text{hr}$. Natural terrestrial exposure rates calculated from the ^{40}K , ^{214}Bi and ^{212}Pb soil sample activities vary from 3 to 7 $\mu\text{R}/\text{hr}$. These values are in good agreement with measurements made using $\text{CaF}_2:\text{Dy}$ TLD dosimeters and those obtained with a field operated $\text{Ge}(\text{Li})$ detector.

F. M. Nuh, D. R. Slaughter, A. A. Shihok-Eldin, and S. G. Prussion, A Rapid Radiochemical Procedure for Fission Product Bromine, Radiochemica Acta 17, 149-153 (1972).

A chemical procedure is described which provides for the separation of fission product bromine from the other fission products in a period of 3 sec. The procedure is based upon the rapid oxidation of bromide by permanganate in sulfuric acid. Bromine yields in the range of 30-50% are typical and the radiochemical purity obtained with the procedure has been tested through separation of 32-min ^{84}Br and the mixture of 55-sec $^{86,87}\text{Br}$. Decontamination factors for iodine and ruthenium are given. Evidence is presented which indicates that ^{86}Br may have a shorter half-life than the accepted value of 54 sec.

R. V. Griffith, Results of the 1972 Survey on Track Registration, Lawrence Livermore Laboratory Rept. UCRL-51362 (1973).

The results of a survey of 118 laboratory groups in 20 countries on the registration of particles in dielectric materials is presented. The survey includes data on track registration applications, as well as materials used, particles studied, analyses performed, and methods of data acquisition and track image enhancement. A section is also included which presents detailed replies by 39 laboratories on techniques of spark counting.

J. R. Gaskill, Smoke Hazards and Their Measurement - A Researcher's Viewpoint, Lawrence Livermore Laboratory Rept. UCRL-74573 (1973).

Since the majority of people who die in fires are killed by smoke, all persons concerned with fire problems must become knowledgeable in the various

aspects of the hazards involved. This requires a

- Proper definition of smoke
- Delineation of smoke hazards
- Measurement of these hazards by appropriate test methods and evaluation of the values obtained.
- Combining of the measured-and-evaluated-values into an index that relates the particular material system to the overall smoke hazard under realistic fire conditions.
- Hazard evaluation and index formulation by a competent interdisciplinary committee.

Various smoke hazards are delineated in this report.

Current knowledge is discussed to determine which smoke hazards can be measured and whether these measurements are significant.

Since evaluation of these hazards will take time, the report considers short-range countermeasures.

Good fire prevention is the most effective way to eliminate smoke hazards, but this approach is not always successful. An alternative is controlled use of materials based on their fire and smoke hazards.

This paper was presented at the Armstrong Symposium held in Lancaster, Pennsylvania, March 26, 1973.

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