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Multiple HEPA Filter Test Methods

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MULTIPLE HEPA FILTER TEST METHODS

January - December 1977

by

Burton Schuster, Thomas Kyle, and Dan Osetek

ABSTRACT

Tandem high-efficiency particulate air (HEPA) filter efficiency measurements have been successfully performed on a large number of 20 000 CFM installations. The testing procedure relies on the use of a laser intracavity particle spectrometer and a very high-volume thermal dioctyl phthalate aerosol generator designed and constructed specifically for this purpose. For systems that cannot be tested in this fashion, work has been initiated on the generation and detection of a fluorescent self-identifying aerosol to eliminate the background problem. General candidate aerosols and methods to disperse them have been uncovered. Two distinct detection concepts have evolved for the measurement of size and concentration of these particles.

A prototype inhomogeneous electric field air cleaner, which uses electrostatic spraying of a water-glycol solution from a high-voltage electrode, was successfully tested. The performance of several different high-voltage electrode structures was measured as a function of voltage and airflow rate. The system operates in a fashion similar to a conventional coaxial geometry electrostatic precipitator, but with enhanced collection efficiency for small particles and a postulated nondependence on particle material. Its principle uses were envisioned to be as a HEPA filter prefilter or as a collector for precious materials for which recovery is desirable.

An He-Ne laser detector for molecular ^{129}I was constructed, which relies on absorption by the molecule of the 632.8-nm laser line in a secondary resonant cavity coupled to the principle laser oscillator cavity. Measured sensitivity of the device is anticipated to be 5×10^{-9} gm/cm³. For the case of organic iodide, in particular CH_3I , a thermal pyrolyzer has been constructed for dissociating the iodine from the methane. As a back-up procedure, an He-Ne laser operating at 3.39 μm has been successfully tested for methane detection in the intracavity mode of operation.

An investigation of the use of a thermoelectric cooler for measurements of humidity in air cleaning systems has been performed. The system operates by cooling a detecting surface below the dew point of the air. The system

to obviating this problem will be discussed in another section). Typically, a challenge aerosol of 10 to 100 times the concentration normally used for single-filter testing is required. The laboratory system of a six-stem Laskin generator and associated compressed air supply, when considered in the multiplicity required for an industrial application, is far too cumbersome. In several tests, commercially manufactured thermal DOP generators* were used, but their individually variable outputs and operating periods made multiple units awkward to work with. The DOP reservoirs in these devices were enlarged, which increased the operation time, but the combined output from the several generators did not behave in a temporally constant fashion and the tending of two to four separate generators proved unacceptable.

So that these difficulties could be overcome, an effort was directed at constructing a thermal DOP generator, which would have an output several times that of the individual commercial generators and use the same basic principles of operation. The successful completion and splendid performance of this system provided confidence in the further scaling to a system providing 10 times the output, that is, 100 g of DOP and 34 L of CO₂ per minute when operated at 30 psi.

Figure 1 is a schematic diagram of the high-volume DOP generator. A 6-L aluminum reservoir of DOP is pressurized by a small regulated cylinder of CO₂. The oil is metered by a needle valve and monitored by a rotometer to provide variable flow rates up to 100 cc per minute. Immediately after exiting the rotometer, the DOP is mixed with a large volume of CO₂ gas. The carbon dioxide is supplied by a large external cylinder that is easily connected to the generator by a neoprene hose and a quick disconnect coupling. The gas is also metered by a needle valve and monitored by a rotometer providing variable flow rates up to 60 L per minute. Downstream of the rotometers, the mixture is carried immediately to the vaporizer or heating block. Figure 2 is a photo of the disassembled unit. The vaporizer is composed of a steel heat reservoir block with a one-inch diameter brass core. Symmetrically surrounding the core are four 400-W heating elements and a variable temperature controller. The DOP-CO₂ mixture is introduced into one end of the brass core and routed to the surface, where it follows a spiral channel through the heat reservoir. This channel is machined into the brass, which is properly clearanced to expand tightly against the steel when heated. The channel has a square cross section of 3.6 mm² and completes 47 wraps along the length of the core. The total volume of the channel inside the vaporizer is then 12.5 cm³. This entire unit is insulated, encased in Transite, and mounted in a steel chassis with the rotometers and other electrical components. The chassis, DOP reservoir, and small CO₂ cylinder are assembled on a two-wheel hand cart, as shown in Fig. 3. The electrical schematic is shown in Fig. 4.

The vapor exits at the nozzle end of the brass core and is allowed to cool and condense at ambient temperature and pressure. Aerosol formation results from homogeneous nucleation as well as condensation of the DOP vapor on condensation nuclei present in the ambient air. A perforated shield is mounted over the aerosol exit, which allows the ambient air to enter the aerosol stream and mix with the DOP vapor. The shield will also support a flexible hose for introducing the aerosol into the ventilation ductwork. Complete vaporization can be accomplished by maintaining a proper CO₂-DOP mixing ratio, sufficient transit time, and a heat reservoir temperature of 316°C. DOP fire hazard is described as "slight" in N. Irving Sax's "Dangerous Properties of Industrial Materials," but the recommended material to fight fire is CO₂. All toxic hazard ratings are zero to one, which corresponds to none or slight.

It is possible that the aerosol size may depend upon DOP or CO₂ flow rate, mixing ratio, or the geometry of the apparatus used to introduce the smoke into the system. Smaller, more constricting ductwork, may tend to enhance coagulation, increase median diameters, and decrease

*"Cloud Maker," manufactured by Testing Machines, Inc., Amityville, New York.

Fig. 1.
High-volume thermal DOP generator.

Fig. 2.
Disassembled high-volume DOP generator.

Fig. 3.
Operational high-volume thermal DOP
generator with CO₂ cylinder.

Fig. 4.
Electrical schematic of high-volume thermal
DOP generator.

total particle count. Evaporation, although slight, is a contributing factor to aerosol size changes, but will tend to decrease median sizes.

Tests were conducted to compare the size distributions of aerosols produced by the Cloud Maker at different operating pressures and the new high-volume generator at different flow rates. Diluted samples of the aerosols were measured directly by the ASAS-300 laser particle spectrometer, and the data were computer analyzed using the HP-1000. Tables I and II summarize the test results. The count median diameter did not vary much, averaging $0.180 \pm 0.02 \mu\text{m}$. The slight broadening of the distribution indicated by the larger geometric standard deviation at the higher DOP flow rates may be due as much to anisokinetic sampling or coagulation as to actual generator output.

The new high-volume DOP generator will produce great quantities of smoke nearly identical in size to the commercial units. Since this median size is nearly the size of maximum penetration of HEPA filters and within the requirements of National Regulatory Commission (NRC) test criteria ($0.7 \mu\text{m}$), the generator is completely acceptable for field testing HEPA filter systems.

TABLE I
HIGH-VOLUME DOP GENERATOR OUTPUT CHARACTERISTICS

DOP mL/min	CO ₂ L/min	Mixing Ratio CO ₂ /DOP	CMD ^a μm	AMD ^b μm	MMD ^c μm	σ _g ^d
2	5.71	2860	.167	.201	.220	1.353
2	5.71	2860	.188	.258	.303	1.490
2	8.74	4370	.162	.184	.195	1.281
2	11.8	5380	.174	.204	.220	1.323
2	15.2	7600	.171	.198	.213	1.312
2	22.1	11050	.166	.190	.203	1.229
3	5.71	1900	.181	.252	.298	1.504
3	11.8	3920	.192	.242	.272	1.406
3	18.7	6230	.171	.196	.210	1.300
3	25.5	8500	.172	.196	.209	1.294
9	11.8	1310	.180	.240	.276	1.460
9	18.7	2080	.182	.227	.253	1.391
9	25.5	2830	.171	.201	.218	1.328
9	33.0	3670	.172	.196	.208	1.286
20	15.2	760	.270	.610	.917	1.894
20	18.7	940	.238	.437	.591	1.734
20	25.5	1280	.241	.403	.520	1.659
20	33.0	1650	.203	.295	.355	1.539
35	18.7	530	.216	.350	.445	1.634
35	33.0	940	.239	.435	.587	1.729
60	29.3	490	.297	.516	.679	1.690
85	36.9	430	.260	.386	.470	1.559
85	36.9	430	.261			

^aCount median diameter.

^bAerodynamic median diameter.

^cMass median diameter.

^dGeometric standard deviation.

The complete variability of the DOP supplied to the heater provides the needed versatility for testing different size systems. The unit has been used successfully for field testing tandem systems.

2. Diluter. Because the large concentration of particles introduced upstream of a tandem filter plenum under test will create excessive coincidence rejection in the laser spectrometer, it is necessary to dilute a small fraction of the challenge aerosol in order to provide an upstream measurement. The diluter shown in Fig. 5, which is actually a small wind tunnel, consists of a 1.5-m length of 7.6-cm-diam metal tubing with a variable speed blower on the exit end and a HEPA filter on the entrance end. Affixed to the front of the filter is a choke to regulate diluter pressure. The tube is instrumented with an orifice meter to measure flow rate, and behind the filter, an inlet port, which admits an isokinetic sample of challenge aerosol to be diluted by the main flow. A manometer between the main installation and the diluter is used to monitor the pressure differential, which provides aerosol flow. The dilution ratio is established from the flows and verified with a conventional photometer, which measures the initial challenge aerosol and that in the diluter. Typical dilution ratio is 0.1%.

The diluter also serves a second purpose in that the dilution ratio accounts for about three orders of magnitude of the PF that is to be measured. The spectrometer measures the diluted upstream challenge as well as that which appears downstream of the plenum. The upstream concentration is the product of the diluter count rate and the reciprocal of the dilution ratio. The

dynamic range of the spectrometer is limited principally by the time available for counting and is not the principal reason for the diluter; although, as explained earlier, it is desirable to keep the challenge concentration high and the counting time low.

3. Detector. An intracavity laser particle spectrometer and counter, the P.M.S.* model ASAS 300, was used as the principal detector. In the laboratory configuration, a minimum detectivity of $0.06 \mu\text{m}$ can be attained. Four selectable ranges of 15 channels each are used to span the range $0.06 \mu\text{m}$ to $2.9 \mu\text{m}$. For field testing, the optical characteristics were changed in order to increase the sample flow rate eightfold. This change was accompanied by a change in minimum detectivity to $0.12 \mu\text{m}$.

C. HEPA Filter Installation Protection Factors

A large number of HEPA filter installations, both single and double stage, were tested with the spectrometer system. For comparison purposes, single-stage testing was also, in many cases, performed with the forward-scattering photometer. Table III displays the results obtained using the Cloud Maker generator and the Los Alamos Scientific Laboratory (LASL) prototype high-volume thermal generator.

The critical series of tests occurred during acceptance testing of the filtration systems at the new TA-55 plutonium facility. These tests were all conducted using the high-volume thermal DOP generator. Measurements were made both with the photometer and spectrometer. Each filter bank was measured individually with each measurement system and, wherever possible, a tandem measurement was made with the spectrometer system. Several of the ventilation systems were designed with multiple exhaust fan capability. These systems had an extraordinarily high downstream background count, probably due to fan shaft or plenum louver leaks. Of the two systems with this design feature, only one could be successfully tandem tested; excessive background in the others effectively masked the measurement. The flow rate for all of the systems was approximately $113.3 \text{ m}^3/\text{min}$ (4000 CFM). Table IV displays the entire series of measurements. The error bars are those associated with an individual measurement. Figure 6 is a display (not a graph) of the single-stage measurements, which compares the results of the two measuring techniques. It should be noted that the second-stage measurements in Table IV for systems H and I are so low as to fail the system. Replacement of these banks yielded the acceptable results shown in Fig. 6. Also note that the ordinate is logarithmic, so that small differences are usually at least a factor of 2 different. In 70% of these tests, the photometer indicated a higher PF than the spectrometer. The possible reasons for this are decreased sensitivity of the

TABLE II

CLOUD MAKER DOP GENERATOR OUTPUT CHARACTERISTICS

Operating Pressure	CMD μm	AMD μm	MMD μm	σ_g
20 psi	.194	.239	.265	1.382
25 psi	.213	.328	.407	1.592
30 psi	.190	.225	.245	1.337
40 psi	.172	.193	.204	1.267

TABLE III

SYSTEM PROTECTION FACTORS

Stages	Generator Type	PF Spectrometer
1	Cloud Maker	$(6.8 \pm .1) \times 10^3$
2	Cloud Maker	$(2.9 \pm 2.0) \times 10^4$
2	Large Thermal	$(7.4 \pm 1.0) \times 10^7$
2	Large Thermal	$(4.1 \pm .5) \times 10^7$

*Particle Measuring Systems, Inc., 1955 South 57th Court, Boulder, Colorado.

Fig. 5.
Upstream aerosol diluter.

Fig. 6.
Single-stage protection factor measured by forward light-scattering photometer and single particle detector.

photometer to submicrometer particles and other reasons that were discussed earlier. Since the spectrometer counts particles directly, the more conservative spectrometer data are considered accurate. The display of the last two columns of Table IV is shown in Fig. 7. In most of these cases, as anticipated, the system test PF is more conservative than the product of the individual stages. This was also borne out during the laboratory testing phase of the program.

Forming the ratio of the product of the individual stage PFs to the overall PF for the nine samples shown results in a mean of $m = 3.28$ and standard deviation of $s = 3.09$. At the 97.5% confidence level, the ratio value is 3.28 ± 2.375 . This is significant in that it essentially rules out a value of unity.

D. Future Work

Specifications have been completed for a new single chassis spectrometer to be used for field testing. The size interval of $0.1 \mu\text{m}$ to $3.0 \mu\text{m}$ will be incorporated into two ranges, $0.1 \mu\text{m}$ to $1.5 \mu\text{m}$ and $1.6 \mu\text{m}$ to $3.0 \mu\text{m}$. Resolution will be $0.1 \mu\text{m}$ with minimum detectivity at $0.1 \mu\text{m}$. The sample flow rate will be substantially greater than the present laboratory spectrometer, resulting in shorter integration time and/or greater accuracy. During the field testing of the complete system, that is, generator, diluter, photometer, and spectrometer, a users manual will be developed so that the technology may be transferred to other Department of Energy (DOE) contractor laboratories as well as industry.

TABLE IV
TA-55 FILTER TESTS

SYSTEM	First Bank PF		Second Bank PF		Overall PF ^a		TANDEM TEST PF
	PHOTOMETER	SPECTROMETER	PHOTOMETER	SPECTROMETER	PHOTOMETER	SPECTROMETER	
C	$1.9 \pm .2$ 10^6	$1.3 \pm .1$ 10^6	$1.5 \pm .1$ 10^6	$3.9 \pm .3$ 10^3	$2.7 \pm .4$ 10^6	$5.0 \pm .5$ 10^7	7.2 ± 1.3 10^7
F	$8.6 \pm .9$ 10^1	$3.3 \pm .3$ 10^3	$1.5 \pm .2$ 10^6	$6.4 \pm .5$ 10^6	$1.3 \pm .2$ 10^6	$2.4 \pm .3$ 10^6	$2.7 \pm .3$ 10^7
A	$1.5 \pm .2$ 10^6	$1.9 \pm .2$ 10^6	$1.3 \pm .1$ 10^3	$2.0 \pm .2$ 10^6	$2.0 \pm .3$ 10^7	$3.8 \pm .4$ 10^6	$1.3 \pm .2$ 10^6
G	$8.9 \pm .9$ 10^3	$2.2 \pm .2$ 10^3	$1.6 \pm .2$ 10^6	$1.3 \pm .1$ 10^3	$1.5 \pm .2$ 10^6	$2.9 \pm .3$ 10^6	$1.8 \pm .4$ 10^7
B	$6.3 \pm .6$ 10^3	$5.6 \pm .5$ 10^6	$1.6 \pm .2$ 10^6	$9.6 \pm .8$ 10^3	$1.0 \pm .1$ 10^6	$5.4 \pm .6$ 10^6	9.8 ± 2.8 10^7
D	$4.2 \pm .4$ 10^6	$3.6 \pm .3$ 10^3	$6.2 \pm .2$ 10^6	$1.8 \pm .2$ 10^6	$2.6 \pm .4$ 10^5	$6.4 \pm .8$ 10^7	5.6 ± 1.6 10^7
E	$1.2 \pm .2$ 10^6	$9.9 \pm .8$ 10^3	$1.1 \pm .1$ 10^6	$1.2 \pm .1$ 10^6	$1.3 \pm .2$ 10^7	$1.2 \pm .1$ 10^6	5.5 ± 1.3 10^7
I	9.5 ± 1.0 10^3	$7.7 \pm .6$ 10^3	$2.5 \pm .3$ 10^1	$3.8 \pm .4$ 10^2	$2.4 \pm .3$ 10^5	$2.9 \pm .3$ 10^6	$4.2 \pm .4$ 10^3
H	$1.9 \pm .2$ 10^6	$1.2 \pm .1$ 10^6	$3.0 \pm .3$ 10^2	$3.4 \pm .4$ 10^2	$5.6 \pm .8$ 10^6	$4.1 \pm .4$ 10^6	$6.2 \pm .7$ 10^6

^aProduct of first bank PF and second bank PF.

II. IDENTIFIABLE PARTICLES FOR HEPA FILTER TESTING

A. Introduction

The testing procedure for HEPA filters involves introducing an aerosol upstream from the filter, then measuring the concentration of the aerosol previous to and after passing through the filter. In the past, the aerosol concentration was usually measured by means of the light scattered from a volume of air large enough to contain many particles. More modern equipment makes it possible to both count and determine the size of individual particles.

The capability of dealing with individual particles relieves some of the previous restrictions, which were required to ensure valid testing. The ability to determine the size of the individual particles allows the test aerosol to have a wide range of sizes. By only counting those within a small size interval and neglecting all other sizes, the omitted sizes will be of no consequence to the test. Although not required, considerations of efficiently generating the aerosol indicate that a reasonably narrow size distribution from the aerosol generator is desirable.

The most important improvement that single particle detection allows is in sensitivity. When light scattering from multiple particles is used, there is a minimum concentration of particles per unit volume that must be present to provide a measurement. In the case of single particle detection, there is no lower limit to the concentration of aerosol particles that can be detected. There is a practical limit in terms of time since only a finite time can be spent in awaiting a particle, and particles can only be counted as they arrive in the measuring volume.

The increased sensitivity means that a single test can be used to determine the PF for multiple stages of HEPA filters. This has been tried and found to work, but an unexpected problem was found that caused the technique to sometimes be unusable. The problem arises because the duct in which the filter is located is at a lower pressure than the surrounding room, and air from the room can leak into the duct. The aerosol particles in the room air are then counted because they cannot be distinguished from the aerosol particles being introduced upstream from the filter. If the aerosol particles from the room air were only a small percentage of the total particles being counted, there would be no problem, but when the PF is high, the opposite case can occur. That is, only a small change in the number of particles is noted when the aerosol generator is turned on or off, because most of the generated particles are removed by the filter, and leaked-in air is the primary source of particles.

The solution to the problem of aerosol particles leaking into the ducting and confusing the filter test is to generate an aerosol that is identifiable and to only count those particles that are identified as coming from the aerosol generator. All particles scatter light to some degree, and so some technique other than light scattering is required.

B. The Use of Fluorescent Particles

There is a method by which the particles can be identified and yet the advantages of light scattering retained. It is the use of fluorescent particles. When these particles are illuminated with light of one wavelength, they emit light at a longer wavelength. Thus, they can be distinguished because the emitted light is of a different wavelength than the illuminating light. The fluorescent nature of the particles causes them to emit light when illuminated by light of the proper wavelength, and this provides a signal akin to scattered light, but distinguishable from it by the shifted wavelength. Therefore, the particles can be measured by much the same techniques as light-scattering measurements.

In operation, one wavelength of light will illuminate the particle, and a longer wavelength will be emitted, and this longer wavelength is to be detected. This requires that something other than white light be used for illumination. Otherwise, scattered and emitted light could not be distinguished, which would prohibit the identification of the aerosol particle as coming from the generator. Very small particles must be detected, and this demands an intense source of illumination. These requirements are best satisfied by a laser, especially the need for high-intensity light.

There are many naturally fluorescent materials, but few of these are highly fluorescent. Rather than search for naturally fluorescent materials from which an aerosol can be generated, it is usually more practical to determine the substance for the aerosol, then locate a fluorescent dye that can be used to make it highly fluorescent.

A dye for DOP is a desirable approach as it would allow all the DOP particle technology to be used, and thus reduce the complexity of the undertaking. The DOP technology includes answers to questions of the toxicity of the materials used for the testing of filters and the safety problems when introducing the test aerosol into the air-cleaning system. These problems are more severe than it might appear, as can be illustrated by an example. The physical properties of glycerol made it attractive as an aerosol material, and an additive for making it fluorescent was known. However, it had to be dropped when it was pointed out that the nitric acid vapor in some air-cleaning systems could react with the glycerol to form nitroglycerin.

A survey of 18 manufacturers and distributors of fluorescent materials resulted in no suggestions as to additives to cause DOP to appear fluorescent. Therefore, an experimental approach was used, during which 113 different dyes were added to DOP as a test of their solubility. Those having a reasonable solubility were also analyzed in a fluorometer to determine the wavelength of maximum absorption of radiation and the emission spectra. The result was six materials that appear to satisfy the present requirements.

C. Particle Generation

Most fluorescent dyes would decompose at the temperatures encountered in thermal DOP generators. The use of a thermal generator is desirable because of the high output of particles this type of generator provides. A method was sought to provide for dispersal of DOP in an aerosol with particle diameters of a few tenths of a micron, but with total throughput of up to a few liters per hour. It is not usually possible to generate droplets this small with a nebulizer or spray system, but at least one system has been found that satisfies the need for a high-volume generator of submicron particles of DOP other than a thermal generator.

The unit is called a sonic atomizer and is commercially distributed by the Sonic Development Corporation, primarily as a spray for breaking fuel oil into very small droplets to promote better burning of the oil. The unit operates by using compressed air to create a standing sonic shock wave when the air flows through a convergent-divergent nozzle into a resonator chamber at supersonic velocity. This can be better visualized by reference to Fig. 8. As the air is reflected from the resonator, it creates a standing shock wave between the cavity and the body of the nozzle. Liquids pumped into the shock wave are vigorously sheared into fine droplets.

Tests were carried out on one of these units using DOP as the liquid and a compressed air pressure of 40 psi. Droplets having a mass median diameter of 0.35 microns were produced. The range of droplet sizes was too great to be considered monodisperse, but not unduly wide. It should be recalled that the particles are to be sized by the instrument so a monodisperse source of droplets is not required. The volume output of DOP in the form of the aerosol droplets was not determined, but on the basis of the manufacturer's specifications for water instead of DOP, a volume flow of 1 or 2 L/h seems reasonable.

Fig. 7.

Tandem HEPA filter protection factor as determined by direct measurement and single-stage measurement.

Fig. 8.

Sonic nozzle aerosol generator.

The detection and sizing of the fluorescently dyed DOP can be carried out in much the same way as the presently used laser single particle measuring system. Two modifications are needed, but these are such that a new instrument would have to be purchased. The primary modification is the use of a helium-cadmium laser instead of a helium neon. The He-Cd laser provides a blue light output rather than red, which is required to induce the fluorescence in the dyes that are soluble in DOP. The second modification is the insertion of filters in front of the detector, which pass light of the wavelength where fluorescence occurs, yet blocks light at the wavelength of the laser. The blocking of the laser wavelength is necessary to avoid signals due to light scattering by aerosol particles that may or may not be fluorescent.

Arrangements were being made for the purchase of a system that could detect and measure the fluorescent particles when it was made known that the work would be ending this year. The purchase of the system was dropped because even though it would have been received before the end of the year, there would not have been enough time to calibrate and test it.

D. Solid and Liquid Particles

It was originally felt that the most desirable system of identifiable particles would involve the use of DOP with fluorescent dyes. The advantage of using DOP was that it is accepted as producing no hazard by being introduced into an air-cleaning system. Some of this advantage is lost when dyes are introduced along with the DOP, because many, if not most, of them are suspected of having undesirable health effects. A more significant advantage of DOP is the small but

finite vapor pressure. This permits large quantities of DOP to be introduced upstream from the HEPA filter, so that a large burden is captured by the filter, with no long-term degradation of the filter, because the DOP will evaporate away rather than remain on the filter. Contrast this to the use of a solid nonevaporating particle. The large number of particles that must be introduced to allow a test of a filter system with a large protection factor could provide a permanent loading of the filter that would be a significant fraction of the total burden, which the filter is capable of accepting. The implicit assumption in this reasoning is that the testing is the same for the solid as for the DOP aerosol.

Let a filter system have PF of P , which means that if N_1 particles are introduced before the filter, a number N_1/P will pass through the filter system. Let the flow of the air through the filter system be F_1 unit volumes per unit time, and let the sampling system extract air at a flow rate F_2 . The number of particles collected by the sampling system will be N_2 and is given by

$$N_2 = \frac{N_1 F_2}{P F_1} \quad (1)$$

If there is a background of particles that confuses the measurement, there is an advantage in supplying the N_1 particles in a very short time, but if the particles are to be identifiable, as in the present case, the rate of introduction is unimportant except for the time that the test requires. The number of particles that must be counted for the test is determined by statistical considerations and can be taken to be of the order of a thousand. The values of P and F_1 are determined by the system under test. The determining factor for the number of particles that must be introduced is F_2 alone. The larger F_2 is, the smaller the quantity of aerosol that must be generated in order to test the system.

The limitation on F_2 is the time that must be allowed to detect and size each particle. Most of the time there is no particle within the sensitive volume of the instrument, but when one does enter, it must remain in the volume for a minimum length of time to allow the signal-to-noise ratio to be large enough to provide a valid measurement. With the single particle laser spectrometer, the flow rates cannot exceed a few milliliters per second.

It is the limited flow rate that the measuring system requires that makes solid particles rather than DOP particles of interest. The DOP particles are liquid and must be measured in the air itself, as they would be absorbed or altered if collected on a filter, but solid particles do not have this shortcoming. By using solid particles, a large flow rate can be used for the sampling system, which will capture the particles on a membrane filter. The filter can then be dissolved in a proper solvent, and the liquid used to dissolve it will then contain the undissolved aerosol particles that were on the filter. Perhaps hundreds of liters of air were put through the filter to collect the particles, but they are now contained in a few milliliters of the liquid. The liquid can be put through the measuring system at a low flow rate and the individual particles sized and counted.

An example will illustrate the power of the collection of solid particles on a membrane filter in testing. Assume a standard (24-in. square) HEPA filter system is to be tested. The flow rate, F_1 , is 2.8×10^4 L/min and the PF is 10^5 . The sample will be collected on a 47-mm filter using a flow rate, F_2 , of 25 L/min and a total of 10^3 particles must be collected for a statistically valid test. Putting these values into Eq. 1 gives the result that a total of 4×10^{13} particles must be released upstream from the filter in order for the test to be statistically valid. If these particles are taken to have a specific gravity of 2, then only 1.14 g of material are required for the test. With this overall quantity of material, there is no significant problem with loading the filter.

It is instructive to contrast the limitations of different approaches to filter testing. When filter systems are tested with light-scattering photometers, the particles must exceed a certain concentration, say 100 particles/cm³, before they can be measured with an acceptable accuracy. In the case of multiple HEPA filters, the concentration of aerosol particles getting through the filter is too low for a measurement, and so the unit cannot be used for multiple HEPA filter tests. The single particle measuring systems have the sensitivity for measuring systems with high PFs, but leaks of room air into the system often contribute more particles than the aerosol generator, which prevents a valid test. This points out the need for an identifiable particle to use in the testing.

It might seem that an identifiable bulk material could serve the same purpose. For example, particles of sodium fluorescein could be generated, fed through the air-cleaning system, and then captured on a membrane filter. The filter could then be dissolved along with the aerosol particles and the bulk fluorescence of the solution measured. In such a case, a concentration of the fluorescent material of one part in 10⁸ would be needed. A simple calculation shows that in the case of multiple HEPA filters, the first filter would become overloaded before the required quantity of material was collected.

A single particle detection system using fluorescent particles can provide the desired measurement using either liquid or solid particles. There are advantages to either of these types of particles, but the solid particle approach is the one being followed at this time.

E. Fluorescent Plastic Particles

The solid aerosol particles being used have a number of desirable properties. The material is manufactured by Day-Glo Color Corporation for use as a fluorescent ink in the printing industry, and, as a result of the large-scale production, is reasonably priced.

The particles are composed of a thermoplastic material with a fluorescent dye included within the plastic. This type of material is quite inert, and thus should retain the dye within it so that no hazard would be expected from breathing air containing the particles.

The particles are approximately spherical in shape and seem to have a smooth surface. Tests on the size spectra have been carried out. These tests give the mass median diameter of the particles in the range of 0.25 to 0.35 microns. The difference in the values determined is due to different experimental arrangements and different methods of measurements. The size spectrum is not wide, being a lognormal distribution with a geometric standard deviation of 1.35.

The operational use of the identifiable particle measuring system with solid fluorescent particles to test filter systems will proceed in somewhat the following manner. The particle dispersing unit will be an atomizer that disperses liquid droplets containing the solid particles. The liquid evaporates leaving only the fluorescent particle.

Air samples will be pulled immediately before and somewhat downstream from the filter system. Air from the system is simultaneously pulled through each of two membrane filters during the time the particles are being dispersed. These samples may be either returned to a central location, or they can be dissolved in a few milliliters of liquid and the particles counted immediately. That is, the system that sizes and counts the particles will be portable and capable of providing the test result in a few minutes; but if it is thought to be a better procedure, the filters can be brought back to a quality controlled situation for the quantification of the test results.

A system such as the one described here should be capable of providing more accurate tests of filter systems since it can test multiple stages of filters in a single test. The testing of multiple stages of filters in a single test should also provide economic savings in the cost of testing the filters.

III. INHOMOGENEOUS ELECTRIC FIELD AIR CLEANER

A. Introduction

Research on the inhomogeneous electric field air cleaner was initiated as the result of an Office of Nuclear Waste Management inquiry into alternate or enhanced (compared to HEPA filtration) air-cleaning devices. The initial concept of uncharged particle migration in an inhomogeneous field was modified when it was discovered that certain fluids (e.g., water, glycol, or a mixture of the two) applied to the high-voltage electrode, considerably enhanced the collection efficiency of particles to the outer electrode of the cylindrical configuration. The mechanism appears to be that of charge accumulation by the fluid, formation of droplets, and the repulsion of these charged droplets from the electrode. In the course of their traversal across the interelectrode region, these charged droplets collide with aerosols that are present, hence charging the aerosols and causing them to be collected in a fashion now similar to that of a conventional electrostatic precipitator. The resultant coating of conducting fluid on the collection electrode aids in charge transfer to the electrode, hence obviating the decrease in effective potential that would occur in the case of a charged dielectric coating.

It was anticipated that such a device would prove useful as a HEPA filter prefilter, hence reducing the volume of HEPA filter waste, or for recovery of valuable materials.

B. Theoretical Description

The configuration to be discussed is that of a system of two concentric cylinders through which aerosol laden air flows along the symmetry axis in a laminar fashion. A conductive fluid on the inner (high-voltage) electrode continually forms small charged droplets, which traverse the interelectrode region. It is assumed that the resulting spatial charge distribution contributes negligibly to the electric field that would exist without this distribution. At some radial coordinate, r , the droplet coalesces with an aerosol particle.

The force, $\vec{F}$, exerted on an electrical charge, q , in an electrical field, $\vec{E}$, is given by

$$\vec{F} = q\vec{E} . \quad (2)$$

The field between coaxial cylinders of radii b and a ($b > a$) is given by

$$\vec{E} = \frac{-V \hat{r}}{r \ln b/a} , \quad (3)$$

where V is the potential difference between the cylinders, r is the radial coordinate, and $\hat{r}$ is the corresponding unit vector. The viscous drag on the particle is given by Stokes law as

$$\vec{F} = 6 \pi \eta R c \frac{d\vec{r}}{dt} , \quad (4)$$

where η is the viscosity of the medium, R the particle radius, c the slip correction factor, and t the time. Combining Eq. 1 and 2 setting the result equal to Eq. 3 results in

$$\frac{-q V \hat{r}}{r \ln b/a} = 6 \pi \eta R c \frac{d\vec{r}}{dt} . \quad (5)$$

Integrating this expression results in

$$b^2 - r^2 = \frac{2 q V t}{6 \pi \eta R c \ln b/a} \approx y t. \quad (6)$$

The flow velocity profile for such a configuration is given by

$$v(r) = \frac{dz(r)}{dt} = \gamma \left[a^2 - r^2 + (b^2 - a^2) \frac{\ln r/a}{\ln b/a} \right], \quad (7)$$

where $v(r)$ is the flow velocity along the z axis of the configuration and γ is a constant involving viscosity and pressure drop per unit length. Substituting Eq. 5 into 6 and integrating between 0 and t , corresponding to some z_0 to z , results in

$$z - z_0 = \gamma \left[(a^2 - b^2) t + \frac{1}{2} y t^2 + \frac{(b^2 - a^2)}{\ln b/a} \left(\frac{[b^2 - y t]}{2y} [1 - \ln (b^2 - y t)] - \frac{b^2}{2y} [1 - \ln b^2] - t \ln y \right) \right]. \quad (8)$$

Using Eq. 5 to eliminate the t dependence yields

$$z - z_0 = \frac{\gamma}{y} \left\{ a^2 (b^2 - r^2) + \frac{1}{2} (r^4 - b^4) + \frac{(b^2 - a^2)}{\ln b/a} \left(b^2 \ln b/a - r^2 \ln r/a + \frac{1}{2} [r^2 - b^2] \right) \right\}, \quad (9)$$

which is the general trajectory equation. Consider the most extreme case, a particle initially at the central electrode ($r = a$). Since $b \gg a$, Eq. 8 reduces to

$$z - z_0 \approx \frac{\gamma}{y} \left[\frac{b^4}{2} \left(1 - \frac{1}{\ln b/a} \right) \right]. \quad (10)$$

The total flow rate, Q , in this approximation can be obtained by the integration of Eq. 6 over the cross section and is given by

$$Q = \gamma \pi \left[\frac{b^4}{2} \left(1 - \frac{1}{\ln b/a} \right) \right]. \quad (11)$$

Combining this and the definition of y (from Eq. 5) results in

$$z - z_0 = 3Q \frac{\eta R c}{q V} \ln b/a, \quad (12)$$

so that the length of collector required for the most unfavorable trajectory is proportional to the volume flow rate and particle radius and inversely proportional to particle charge and applied potential. Depending on space considerations, a given airflow could be sent through one long collector or divided to flow through several shorter collectors operating in parallel.

C. Experimental Arrangement

Shortly after the beginning of 1977, the inhomogeneous electric field air cleaner was redesigned and rebuilt to more closely simulate a small-scale pilot model such as could be used with a glove box. Figure 9 is a schematic of this system. Room air is drawn in through a dry gas meter, which provides flow rate measurement. The air is cleaned by passage through a HEPA filter before being mixed with 1.1- and 0.3- μm polystyrene latex aerosol. This mixture then passes through the collector section, which is located between the two insulators, and then is exhausted by the blower through a filter. The aerosol generator is operated by compressed air. The electrode wetting fluid is metered at a rate of approximately $2\text{ cm}^3/\text{h}$ from a pressurized reservoir. The dimensions of the collector system are $7.62\text{ cm} \times 160.02\text{ cm}$. Initially, difficulties were encountered with the cloth based phenolic insulators, which turned out to be too conductive for their intended use. The problem was corrected with new insulators machined from Teflon. Initial operation of the system at 10 kV, (6 CFM) flow rate and a thread wound 0.32-cm control electrode resulted in an efficiency of 99.9%. After several hours of use, this figure degraded considerably.

Fig. 9.
Inhomogeneous electric field air cleaner.

The cause was thought to be due to surface leakage currents across the wet insulators in combination with a marginal power supply. In order to overcome this problem, a 0- to 20-kV dc power supply was designed and constructed, which, when operated at 15 kV, provides a 100- μ A collection current. The large current capacity of the supply overcomes the problem of current division due to leakage.

D. Collection Characteristics

Previous work had indicated that while both water and glycol were effective in enhancing collection efficiency, water worked better for small particles ($<0.4\ \mu\text{m}$), and glycol appeared to be more effective for larger particles. On this basis, an arbitrarily determined ratio of 25% glycol and 75% water was used as the central electrode coating medium. Data were taken at flow rates of 0.00 m^3/min (0.0 CFM), 0.142 m^3/min (5.0 CFM), 0.283 m^3/min (10.0 CFM), 0.425 m^3/min (15 CFM) and 0.566 m^3/min (20.0 CFM). These correspond to face velocities of 0.0, 31.1, 62.2, and 124.4 m/min, respectively.

The major problem that required solution was that of the physical integrity of the central electrode. The thread wound rod was acceptable for feasibility measurements, but obviously could not withstand the rigors of a harsh environment. What was required from the central electrode was that it be wettable, a property that the thread provided. Figures 11a through 14a display the properties of the system with a thread wound electrode. These were used as baseline characteristics. The characteristic voltage-current curves for this configuration are shown in Fig. 10a. The per cent transmission (100% - collection efficiency) as a function of current for the various flow rates is shown on the remaining figures. As would be anticipated from Eq. 11, the collection penetration increase with increasing flow rate. Physically, this occurs because there is less time for the electrostatic force to provide radial movement as the longitudinal velocity component increases, at least for the laminar flow case. Figures 11a through 14a are reminiscent of similar curves for conventional electrostatic precipitators.

The next electrode configuration to be attempted was that of a 0.32-cm-diam steel rod with threads cut into it. It was thought that capillary forces would hold the liquid in the thread root and that the enhanced field at the thread crest would promote the electrostatic spraying of droplets. Unfortunately, this electrode worked so poorly that no data were acquired.

Another electrode was constructed with wettable ceramic sleeves on a steel rod. This system did not work at all, presumably because the electric field at the increased diameter of the electrode was too small to create the spraying effect.

The next attempt proved more successful. The electrode was a 0.34-cm-diam steel cable composed of seven strands of wire twisted together. Complete data runs at the five different airflows could not be performed because of some internal arcing that developed. Figure 10b displays the voltage-current curves obtained with this electrode and Figs. 11b through 14b display the transmission characteristics. Disassembly of the system revealed a conducting path from the high-voltage electrode through the liquid feed to ground, which was the cause of the arcing and, after reviewing the next data set, probably responsible for some measurement inconsistencies in cases already discussed.

The final data set illustrates the results obtained with a 0.24-cm-diam seven-wire stranded steel cable. The family of voltage-current curves in Fig. 10c are very consistent when compared to Figs. 10a and 10b. The curves are well described by the equation $I = AV^N$, where I is the current in μA , V is the voltage in kV, and A and N are fitting constants. For the data displayed in Fig. 10c, the constants are given by $A = 2.88$ and $N = 5.41$. The classical low current Townsend expression for an electrical discharge from a coaxial configuration goes no higher than the second power of the voltage. The difference presumably lies in the different discharge mechanism associated with the liquid-coated electrode. Comparison of the transmission curves for the three

Fig. 10.
Current voltage characteristics of inhomogeneous electric field air cleaner.

Fig. 10a.
Thread wound electrode.

Fig. 10b.
0.32-cm steel cable electrode.

Fig. 10c.
0.24-cm steel cable electrode.

Fig. 11.
Transmission current characteristics at 0.142 m³/min flow rate.

Fig. 11a.
Thread wound electrode.

Fig. 11b.
0.32-cm steel cable electrode.

Fig. 11c.
0.24-cm steel cable electrode.

Fig. 12.

Transmission current characteristics at $0.283 \text{ m}^3/\text{min}$ flow rate.

Fig. 12a.
Thread wound electrode.

Fig. 12b.
0.32-cm steel cable electrode.

Fig. 12c.
0.24-cm steel cable electrode.

Fig. 13.

Transmission current characteristics at $0.425 \text{ m}^3/\text{min}$ flow rate.

Fig. 13a.
Thread wound electrode.

Fig. 13b.
0.32-cm steel cable electrode.

Fig. 13c.
0.24-cm steel cable electrode.

Fig. 14.

Transmission-current characteristics at $0.566 \text{ m}^3/\text{min}$ flow rate.

Fig. 14a.
Thread wound electrode.

Fig. 14b.
0.24-cm steel cable electrode.

electrodes shows that data a and c (that is, the thread wound electrode and the 0.24-cm stranded cable) are very similar except at the highest flow rate, where the stranded cable is clearly superior and, in fact, is better than for intermediate flow rates. One possible explanation is that at this high flow rate, the entire flow has made the transition from a laminar to a turbulent regime. For the 7.62-cm dimension, the onset of turbulence occurs at 31 m/min and for the maximum velocity used, the appropriate dimension is only 1.8 cm. Why the enhancement did not occur in the case of the thread wound electrode is not at all clear, with the possible exception of adverse effects due to current leakage. The inferior characteristics of the larger stranded cable were almost certainly the results of leakage current, which became so pronounced that data at 1.7 m³/min were unobtainable. Comparisons with the trajectory equation in the nonturbulent regime disclose that charges of about 200 electrons would be required to obtain the measured collection efficiencies. This is not an unreasonable charge to be carried off by a small liquid droplet, which then coalesces with an aerosol particle.

Although the above information is far from complete with regard to all the engineering data that should be acquired for optimization, it was felt sufficient for construction of an operational prototype to be used as a glove box prefilter and for incinerator off gas processing. To this end, discussions for application have been held with personnel responsible for plutonium handling

and those responsible for low-level waste incineration. There is a particular attraction for the latter application because of the variability of incinerated wastes (e.g., wood, paper, plastic) that reduce the efficacy of conventional precipitators.

IV. IODINE MONITORING

A. Introduction

The development of a gaseous iodine detection and measuring system at LASL has been directed to exploiting the coincidence of iodine absorption lines and the output of the helium neon laser. There are stronger absorption lines near the output of other lasers, but these have been neglected because these lasers are expensive and not as rugged or compact as the He-Ne. With this approach, it also appeared possible to obtain a portable instrument having sufficient sensitivity and capable of being manufactured at a reasonable cost. The instrument appears to satisfy the original design requirements and after the final adjustments will most likely exceed these requirements by a considerable margin.

The original concept for the instrument involved introducing the iodine into a cell within the laser cavity. The cavity has mirrors at each end with reflectivities of over 99%. These mirrors cause the radiation to make many traversals of the cavity, the beam intensity being amplified a small amount each time it passes through the plasma of the laser tube. Any iodine in the cell within the cavity absorbs a part of the radiation on each pass. This results in an effective decrease in the amplification of the radiation on each pass and causes a decrease in the output from the laser.

A workbench system was quite sensitive to iodine, but when the unit was operated with no iodine in the cell, the output of the laser had output power variations of as much as 5%. These were due to power supply variations, small changes in the optical alignment, and other factors not fully understood. The variations were larger than expected on the basis of more standard lasers because the length of the cavity had been increased to accommodate the sample cell. In any case, the 5% variation in output power meant that quantities of iodine causing the output power to decrease by less than 5% were not measurable. A modified concept was adopted that avoided the problem completely.

The problem can be stated in the following way. There was no way to monitor what the output power of the laser would be if no iodine were in the cell. On the other hand, if the cell were outside the cavity, the output power of the laser could be measured before and after passing through the cell, but the power change would be very small.

The modified technique adopted and illustrated in Fig. 15. is to measure the laser output power from the beam coming through one end mirror of the laser. The absorption cell is placed at the other end of the laser, so the beam through that end mirror goes into the cell. The central idea is to place a third laser type mirror at the end of the absorption cell opposite the laser. The output laser beam, therefore, resonates between that mirror and the end mirror of the laser, rather than within the laser cavity.

B. Optical System

The system has two cavities. One of these has the laser plasma tube in it, while the other has the absorption cell. The problem to be anticipated with such a system is that if the secondary cavity has nearly zero power loss, then it is almost impossible to put energy into the cavity in the first place. The described technique avoids the problem by sharing a common end mirror.

Thus, as the beam exits the laser mirror, it is already in the secondary cavity, and there are no coupling losses. At the same time, the beam coming out the opposite end of the laser has the intensity that is proportional to the intensity being coupled into the absorption cavity. This beam can be used to monitor the laser power being fed into the absorption cavity.

Let the output intensity of the laser into the secondary cavity be I_0 . The fraction of the intensity striking the mirror surface, M_3 , which is coupled out of the cavity, is T , and the fraction of the intensity lost for a single pass through the system is L . This loss includes all types of losses, including the transmission, absorption, and scattering at the mirrors, which are assumed to have identical losses. The beam intensity after the first reflection is $I_0 (1 - L)$ and after n reflections is $I_0 (1 - L)^n$. If the beam intensity is I_n' before the transmission loss at an end mirror and $I_0 (1 - L)^n$ after the reflection, the intensity coupled out of the cavity is found from the relation

$$I_n' = I_0(1 - L)^n + I_n'T, \quad (12a)$$

or

$$I_n'T = \frac{I_0 (1 - L)^n}{1 - T} . \quad (12b)$$

The total intensity, I , emerging from the cavity is the sum of these contributions over all odd indexed traversals, since only the reflections at mirror M_3 are collected. The value of I is

$$I = \sum_{n=1}^{\infty} I_{2n} + I^T , \quad (13)$$

or

$$I = \frac{I_0 (1 - L)T}{L(2-L)(1-T)} . \quad (14)$$

If the only loss should be due to mirror transmittance, the signal at the detector will be greater than one-half the laser output. In practice, there are several other losses due to imperfect optics. A major loss in the present experimental arrangement occurred at the nonmirrored surface of the mirror shared by the two cavities. Even though a dielectric antireflection coating was applied to the surface, there was still a 0.4% attenuation for each beam transit. For the largest signals expected, L will still be small enough to approximate Eq. 14 by

$$I = C/L , \quad (15)$$

with an accuracy of 2%, with C being a system constant.

One further property of resonant cavities must be considered. A cavity is resonant for one or a small group of frequencies. The resonant frequencies are determined by the cavity length. Two cavities are involved in the present case, and in general, the two will have different resonances, but the two will drift with temperature and other conditions. Any difficulties due to this drift are avoided by oscillating one of the laser end mirrors at a few hundred cycles per second and over a distance of about one micrometer. This scans the laser over the full range of frequencies available to it, and thus gives a signal that is the average over the entire laser line width, not just some particular frequency selected by the resonant properties of the cavity.

The physical layout of the optical arrangement is shown in Fig. 15. There is a narrow band filter in front of each laser beam detector. These let only light near laser frequency through and allow the unit to be operated in full room light without interference from the ambient light.

C. Electronics and Sampling

The sample system is an important part of the total system and is more important than just a method for providing a flow path for the sample. It is important for making the system specific to iodine. Otherwise, any output signal must be regarded as due to iodine or any one of several other substances. Many materials are optically active at the He-Ne laser frequency. The sample system is designed to cause the system to only respond to I_2 molecules and to eliminate electronic or optical drift problems.

This is accomplished by a chopped operation. That is, first the gas that might contain the I_2 is pumped into the cell. Then the same gas, but with the iodine removed by an iodine absorbing filter, is pumped into the cell. Filters are available that are quite specific in absorbing only iodide. At present a sintered silver disk is being used, but a silver exchanged zeolite could also be used. Let some gas other than iodine, which is optically active at the He-Ne laser frequency, be present in the incoming air sample, say some freon gas. The iodine absorbing filter will not affect it, and the same signal will be present when both the sample and the reference gas are introduced into the cell. The output signal is the difference in the two signals, the signal from the sample introduced by the sample pump and the sample introduced by the reference pump. The signal from the freon is the same in both cases and gives a zero difference signal. If iodine were present in the incoming air, it would only be present in the sample pump signal, since the I_2 in the reference pump sample would be absorbed by the iodine absorbing filter.

The detectors are silicon photodiodes. The use of solid state detectors eliminates the need for the cooling of photomultipliers and high-voltage supplies. The electronics is shown in the functional diagram of Fig. 16. This electronics is constructed on printed circuit cards and housed with the readout unit in a small case separate from the optical assembly. It is connected by an electrical cable and could be located a long distance from the optical unit. This can be quite helpful in actual measuring situations.

The signal from the detector measuring the laser output and the detector measuring the power beyond the absorption cavity are first amplified, then the ratio of the two signals is obtained from the ratio amplifier. This provides the output given in Eq. 15. At this point, the dependence on the laser output power has been removed by ratioing the two signals.

The timing generator must now be described. There are four time intervals in each operational cycle. The timing generator provides a positive pulse to the output, controlling that portion of the cycle for the duration of the particular interval. This is indicated in the lower left part of the figure.

When output A is positive, the sample pump is activated and pumps air into the absorption cavity. During this time, and during the time that the reference pump is operating, the output signal from the ratio amplifier is disconnected from the following electronics. This is necessary because the airflow introduces noise due to the turbulent flow in the sample cell but, more importantly, during this time period the signal is due to some portion of the gas that has had the iodine removed and some that has not had the iodine absorbed on the filter. The signal during this period is, therefore, of no interest.

During the second time interval, B, the sample and hold circuit averages the signal from the ratio amplifier. After the end of the interval, it effectively holds this signal during the next three intervals, thus maintaining a signal as if the iodine containing sample were still in the absorption cavity.

Fig. 15.
Dual cavity laser iodine detector.

Fig. 16.
Laser iodine detector functional electronics schematic.

The C and D intervals are a repeat of the process above, except for the gas having the iodine removed by the filter. First, the reference pump introduces the gas into the cell, then the second sample and hold circuit averages it during time interval D.

The difference amplifier provides the difference in the signals from the two sample and hold circuits. This amplifier has a long time constant, so it averages over three to four complete cycles. Each cycle is presently 20 s, so the measurement time is about 60 s.

The final output is given on a digital output unit. It could also be used to sound an alarm at a given level of iodine or to drive a permanent recording device with either analog or digital values of the iodine concentration.

The optics are mounted on a 3.18-cm thick and 11.1-cm wide aluminum beam. This beam is an extruded unit made for electronic heat sinks. The fins on the bottom of the beam provide excellent rigidity with minimum weight. The remainder of the enclosure is 0.64-cm aluminum. Aluminum has a large thermal expansion, but the use of aluminum has not resulted in problems, partially because it is used throughout the construction and partially because the high thermal conductivity minimizes thermal gradients. Experience with using and moving the unit indicates that it is rugged enough to be used as a portable instrument without constant realignment or other problems.

The details of the sample system are shown in Fig. 17. The tube is made with Brewster angle windows, so there are no reflection losses at the window surfaces. The windows are fused to the glass tube so the iodine will not interact with the epoxy that would normally have been used for bonding the windows to the tube. At the present time, some Teflon tubing is used in the sample system, such as for attaching to the inlet and outlet tubations of the sample cell, but this will be replaced with glass after the testing is complete.

Fig. 17.
Laser iodine detector sample system.

D. Results

The critical aspect in the determination of the instrument sensitivity is obtaining air with a known concentration of I_2 . In principal, one only has to maintain air in contact with iodine crystals at a known temperature and a concentration of iodine equal to the partial pressure of iodine at that particular temperature will evolve.

This is not a good approach for a flowing gas system such as the one being described here. The problem is that the time required for the vapor pressure of iodine to saturate the air in contact with the solid iodine is unknown. There is a finite flow rate of the air away from the iodine, so there is a chance that the air being used in the testing contains a smaller I_2 concentration than anticipated.

Testing was conducted by flowing air over and through iodine crystals at room temperature, with excess I_2 allowed to condense from the air at the lower measurement temperature providing assurance that the air is saturated at measurement temperature.

The testing indicates that the noise level of the instrument will be a few nanograms per cm^3 . This is for $^{127}I_2$. The sensitivity for $^{129}I_2$ is expected to be greater, but how much greater can not be estimated at this time.

V. LASER INTRACAVITY METHANE DETECTION

A. Introduction

During the studies associated with the detection of iodine, it became apparent that a large fraction of the iodine was in the form of organic iodide, in particular, methyl iodide. The by-products of the pyrolysis of this compound were expected to be elemental iodine, which hopefully would form molecular iodine (I_2), and methane. Very fortuitously, methane has a strong absorption line almost exactly coincident with a He-Ne laser line in the infra-red at $3.39 \mu m$. A preliminary study on the laser intracavity detection of methane had been performed several years earlier, and it was thought that the detection of methane through the pyrolysis of the iodide might be a necessary, although indirect, method for methyl iodide detection.

B. Experimental Arrangement

Seven samples of methane mixed with nitrogen were prepared. On a partial pressure basis, these ranged from one part in 10 to one part in 10^7 . A 26.3-cm intracavity path length, and for linear absorption measurements, a 23.4-cm extra cavity absorption path were chosen. An appropriate gas handling system was constructed for providing slow flow of the sample mixtures through these regions. Cleaning of the system between samples and for null readings was accomplished with a flushing of dry nitrogen. Data were obtained by measuring the laser power as a function of the mixing ratio within the cavity and also in the exterior path region. The latter measurement was used only to qualitatively determine the additional effectiveness of the intracavity absorption.

The plasma tube for this experiment had been specifically designed to have a very low gain at the 3.39- μ m wavelength so that it would always be operating close to threshold, where intracavity perturbations of the radiation field would be most manifest. The cavity resonators were, of course, chosen to be compatible with 3.39- μ radiation. Unfortunately, the plasma tube developed a fracture just prior to the measurements and had to be discarded. In order to expedite the acquisition of at least some preliminary data, a standard commercial off-the-shelf tube was purchased. This tube, of course, was not optimized for the experiment and so was not expected to yield the sensitivity of the original tube. Nevertheless, as shown in Fig. 18, mixing ratio of 2×10^{-6} could be detected. Complete extinguishment of the laser line occurred at about the 2% mixing ratio level. Further work with an optimized system was not pursued when it became apparent that molecular iodine from the pyrolysis process could be detected.

VI. HUMIDITY STUDIES

A. Introduction

There is evidence that the protection action of HEPA filters may be degraded by high humidity. The effect may be due to the condensation of water, which can creep through the filter and carry materials with it, or some other unknown mechanism. The condensation will only occur when the dew point is very near the ambient temperature, which is a difficult condition for standard humidity instrumentation to detect. The difficulty is that both the dew point of the air and the air temperature must be measured with great accuracy and the two related. It is when the difference in the dew point temperature and the air temperature approach zero that condensation will occur on the filters. If a reliable and practical alarm system could be installed in the air ducts, preventative actions could be taken to avoid condensation on the filters when the humidity becomes too high.

With this problem in mind, a different approach to the warning system offers considerable advantage. The system would cool a surface by a known amount and then detect if condensation was occurring on the surface. The advantage of this approach is that only the amount of cooling is measured, rather than two temperatures. The problem was then expanded somewhat to provide not only an alarm to warn of high humidity, but to also provide a measurement of the humidity in the air-cleaning system. The capability to measure humidity was included as a result of discussions with personnel from the Rocky Flats plant. At times they have a severe high-humidity problem, but have no method for measuring how frequently the problem occurs.

B. Approach and Results

The basic approach, to provide a known decrease in the temperature, was to use a thermoelectric cooler. This is a solid-state device that has one surface cooled when current is passed through the unit. If the second surface of the cooler is caused to be at the ambient temperature by flowing air across it, the temperature of the cooled surface is proportional to the current flow through the thermoelectric cooler, provided that variations in the ventilation of the cooled surface are eliminated. The latter can be accomplished by shielding the cooled surface from direct ventilation and only allowing natural convection to exchange air with the airstream.

The only other active element the humidity sensor requires is a means for detecting when condensation has occurred on the cooled surface. In the first unit constructed, a quartz crystal was used as the detector. The term quartz crystal refers to an electronic element rather than just a piece of quartz material. The crystals are used as the frequency controlling elements in electronic oscillators, where they provide an electrical signal at the frequency of their mechanical vibrations. They are quite stable and sensitive to changes in the mass of material absorbed on the surface. There are gold electrodes on the surfaces so only quartz and gold are exposed to the airstream, a combination that is inert to actions by most chemicals.

If water condenses on the crystal, the frequency of the mechanical oscillations are affected, the change is electronically detected, and the current altered through the thermoelectric cooler. The electronics requires only three integrated circuit sized elements in the case of an alarm system, but more when an actual measurement of humidity is to be obtained.

A system operating on the principles described above was constructed and preliminary testing was carried out. It functioned properly and, although never fully calibrated, seemed to be a desirable solution to the original problem.

This approach was dropped after testing indicated it could not perform for extended periods in the environment of the Rocky Flats air-cleaning system where it was needed. The problem was caused by fumes of hydrofluoric acid in the air circulating through the system and attacking the quartz. It is not known how long the unit could function there, but tests in a combination of nitric and hydrofluoric acid fumes indicated that the quartz would be dissolved by the gases.

A second unit was built with the thermoelectric cooler still being used, but with a different method for detecting condensation, which was intended to overcome the reaction of the detector with the acid fumes.

A schematic representation of the unit is shown in Fig. 19. The detector consists of a 0.005-cm sheet of Teflon with gold evaporated onto the Teflon. Because of the chemical inertness of the Teflon, the gold does not adhere well to the surface and could be wiped away. The small amount of adhesion present is acceptable and offers no problems so long as the surface is not touched.

A sharp pointed tool is used to scribe a thin line through the gold coating and divide it into two electrically isolated regions. It is important that a furrow not be cut into the Teflon in the process of scribing the gold. The weak adhesion of the gold is an advantage in this respect. Gold wires are allowed to touch the two gold covered regions created by scribing the surface, and these serve as electrodes that are not altered by the acid fumes.

The condensation of water on the cooled surface, which is covered by the Teflon, decreases the electrical resistance between the two electrodes to about 5 M Ω . When this change in resistance occurs, the surface is below the dew point, and the current through the thermoelectric cooler is decreased until the resistance increases again, indicating that the water has evaporated since the surface is once more at a warmer temperature than the dew point.

Fig. 18.

Normalized laser power as a function of methane-nitrogen mixing ratio.

Fig. 19.

Humidity detector schematic.

The unit was found to be resistant to attack by nitric and hydrofluoric acid, but it did not function as well as the quartz crystal unit. The quantity of water that had to be condensed onto the surface before it was detected was much greater for the Teflon and gold unit than it was for the quartz crystal. This extended the time required for a measurement, but also had a more detrimental effect. When a film of water formed on the surface, chemicals from the air would go into the solution. When the surface warmed because of the decrease in current through the thermoelectric cooler, the water would evaporate, but a residue of chemicals would remain on the surface. After a moderate number of cycles, the chemical residues could provide an electrical contact between the electrodes and make it appear that a film of water was present even when the surface was dry. Because of the low adhesion of the gold, it is doubtful if the surface could be properly cleaned without damaging the layer of gold. Thus, the Teflon film would require periodic replacement.

Work has now stopped on this system. As stated, the quartz crystal system seems to have held the most promise, but it could not survive the particular environment in which the unit was needed.