

PRINCIPLES, CALIBRATION, AND PROCESS EXPERIENCE
FOR A NEW LASER AEROSOL DETECTOR SYSTEM*

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A particulate detection instrument employing forward scattering of laser light has been applied to in situ measurement of oil aerosol carry-over in oil-injected screw compressor systems, as used in cryogenic refrigeration. For the "micron-sized" aerosol carried through the control equipment, the instrument provides a voltage approximately proportional to particulate mass concentration, with a response time of about one second. The measurement is first-order independent of particle composition and particle size distribution if the particles have mass median diameter d_m in the $0.3 \leq d_m \leq 3 \mu m$ range. Equally important, this direct measurement of particulate matter transported by the process flow is inherently independent of pressure, velocity, temperature, and composition of the process gas.

The instrument employs a calibration/zero concept which permits automatic and continuous compensation for unavoidable changes in electro-optic system transfer gain and baseline zero; both of these variations have previously characteristically limited the application of electro-optic transducer means.

Considerable effort has been made to establish the accuracy of the system's calibration. Several approaches have been implemented but emphasis will be upon cross correlation of the instrument's response with gravimetric analysis of three types of particle: (1) ambient air pollution (2) vertically-elutriated cotton dust and (3) oil aerosols collected from the cryogenic process stream itself. Discussion of the absolute accuracy of the calibration and factors limiting that accuracy will be given. This discussion will also include experience on other process particulates, such as sulphuric acid mist.

This paper also emphasizes operational experience with the BNL 500 watt (7 K) helium refrigerator for the Power Transmission Project and the LASL 700 watt (4.5 K) helium refrigerator presently installed at the Brookhaven Isabelle Project. Representative start-up, operating, and shut-down results are given. Examples wherein the instrument has proven extremely useful in system refinement are given.

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I. INTRODUCTION

An in situ particulate detection and measurement instrument utilizing a laser is described. It is capable of continuous measurement of the level of particulate matter and oil aerosols directly in a gas stream, including high pressure streams. Although this specific instrument was designed to monitor helium at 15 atmospheres (1.5MPa) that is used as a cryogen in a cryogenic refrigerator, it has applications in many other areas.

Laser particulate monitors are not new as, for example, they have been used for mass concentration measurements in applications related with air pollution caused by emissions from smoke stacks. Desirable characteristics of the present laser measurement technique include a response that is independent of particle size over a broad, useful range, in situ measurement capability with no disturbance of the particles being sampled in the stream, good repeatability, and long term reliability.¹ Additional specifications required for the helium monitoring application covered by this paper included a 1000-fold increase in sensitivity, an increase in long term stability to many months of unattended operation, and packaging for use in a high pressure, exceptionally clean process.

The need for particulate monitoring of helium cryogenics came as a consequence of switching to oil injected screw compressors. These are now being designed into most large cryogenic systems starting with a 500 watt (7 K) supercritical refrigerator for the Brookhaven Power Transmission Project in 1974. This system uses a 350 HP oil injected screw compressor capable of handling 130gm sec^{-1} of helium at 15 atmospheres (1.5MPa). Use of screw compressors has increased mechanical reliability over reciprocating compressors to tens of thousands of hours. However, the many-fold increase of oil contact with

the helium has increased the possibility of oil contamination at the input to the refrigerator above the permissible level of a few parts per billion.²

Practical applications of superconducting transmission cables by utility companies requires helium refrigeration equipment to operate reliably for time periods of from many months to one or more years.³ These time periods are much longer than is usually the case at present. Impurities in helium have always been a major problem in cryogenic refrigerators as they freeze out and plug up flow passages. Helium contains impurities when purchased and other contaminants are picked up from the piping, storage vessels, dewars, leaks, etc., including oil from compressors. Discussion of measurement and removal techniques for impurities other than oil are beyond the scope of this paper.

To determine if oil contamination would become the predominate source of helium impurity, a series of experiments were run over a period of months with a 150HP oil-injected screw compressor pilot operation. First, the oil was processed in a vacuum vessel to remove moisture and volatile components. The oil was heated to 120°C and sprayed and vacuum-pumped for about 150 hours. The oil was then transferred by compressed helium, thereby precluding any further contact with air, from the processing vacuum vessel to the compressor oil sump. The pilot operation was operated as a complete closed loop except no refrigerator was involved.

Measurements for vaporous oil were conducted at several points in the loop with a columnless, total hydrocarbon analyzer using a flame ionization detector with a detectability limit of 9×10^{-13} gm sec⁻¹, which in this system would represent approximately 6ppb. No oil, vaporous or otherwise, was detected. Since oil was observed coating the sample tubing walls and collecting in driplegs in sampling lines from the helium process stream

to the input of the hydrocarbon analyzer, it was concluded that any oil remaining after filtration was in the form of an aerosol. The reduction of the helium sample pressure, from 15 to 1 atm (1.5 to 0.1 MPa), and velocity required for injection of the sample into the flame analyzer, probably was responsible for the removal of the aerosol and its resultant nonappearance at the flame detector.

From these observations it was evident that an in situ measurement was required, and a technique using forward scattering of laser light would permit the most sensitive detection of particulate matter.

Accordingly, a particulate detection instrument was designed, which accomplished both high sensitivity and stability by using unique automatic compensation of the entire electro-optical train. It is installed in situ and gives continuous analysis of the helium in the process stream. High sensitivity and stability permit the measurement of particles of between 0.3 and 3 μ m diameter in quantities down to 10 micrograms per cubic meter, corresponding to 5 parts per billion by weight. Since, for this cryogenic system, most particulates having a diameter greater than a few micrometers are presumably removed by 1 μ m "absolute" filters and since, presumably, little mass will be found for particles having diameters smaller than a few tenths of a micrometer, this response range was regarded as very satisfactory.

This instrument is used on a system in which the screw compressor operates at a flow rate of 11.2×10^3 kg day⁻¹ or 4.1×10^6 kg year⁻¹ of helium. An oil contamination level of one part per million by weight would give 4.1 kg year⁻¹; ten parts per billion would give 41 g year⁻¹ to be frozen out in the refrigerator. The latter level was set as the sensitivity requirement.

The instrument provides an analog voltage proportional to particulate mass

concentration with a response time of about one second. The signal can be displayed on a chart recorder or a digital voltmeter or used for alarm purposes if preset concentration levels are exceeded. The direct measurement of particulate matter transported by the process flow is independent of gas pressure, velocity, temperature and composition.

II. PRINCIPLES OF OPERATION

Figure 1 shows the general configuration and location of the sensor unit. The valves and pumping ports permit installation and removal of the instrument without process interruption. The source and receiver outer pressure vessel shells are filled with helium at a pressure of one atmosphere (0.1MPa) and are designed to contain the 15 atmosphere (1.5MPa) process helium if either of the two optical interfaces fails. The control/read-out unit contains a chart recorder and various threshold annunciators for both process monitoring and instrument self-monitoring. One threshold level can be set as a warning; another threshold, set at a higher level, can be used to shutdown the whole process, if desired. Any failure of the instrument itself will be annunciated.

Figure 2 gives the functional block diagram. Coherent radiation from a pulsed GaAs laser operating at wavelength $\lambda \cong 0.9\mu\text{m}$ is formed into a collimated beam and transmitted through the first pressure interface into the process. Oil and other particulates in the optically-defined sampling volume scatter the laser light; these photons, in an angular range $\Delta\theta$ and a mean angle θ , are collected by the receiver optics assembly at the second pressure interface and focussed onto the detector. The laser source and detector electro-optics are at ambient temperature and pressurized to one atmosphere (0.1MPa) with helium.

Upon detection and electronic processing, the received photons produce a signal voltage v_1 which is proportional to the total mass of the particles in the optically-defined sampling volume. Thus the basic transducer function of the instrument converts mass per unit volume into voltage. The receiver optics, in addition to defining the sampling volume and the acceptance cone $(\bar{\theta}, \Delta\theta)$, rejects the main laser beam and stray laser light.

This instrument employs a calibration/zero element which permits automatic and continuous compensation for unavoidable changes in transfer gain and baseline zero; both of these variations characteristically limit the application of electro-optic transducer means in making accurate measurements. In order to rely upon the output signal as truly representative of the process, the instrument's transfer function must be stable and independent of laser power, lens cleanliness, minor optical misalignments, detector gain, amplifier gain and drifts, etc. If the transfer function is not within tolerance, the instrument annunciates and alerts the process operator.

Note that signal v_1 is of a time multiplexed nature and has either signal, calibrate, or zero value, depending on the calibrate/zero element position. Thus the same electro-optical train generates v_1 in all three conditions. This permits more accurate compensation. The positions of the calibrate/zero element are sensed and used to set the electronic demultiplexing switch.

In summary, an automatically compensated, analog signal, typically 0-1V in range, is presented to the user for whatever readout or control functions are desired. If either the calibrate or zero compensation ranges are exceeded, or if any component fails, the instrument annunciates.

The interested reader may refer to reference 4 or to manufacturer's literature for more complete explanation of the electro-optics. Also,

interpretation of the forward-scattered light signal has been covered in detail in references 4 and 5 and will not be repeated here as this paper is more concerned with results of using the instrument.

III. CALIBRATION

Four methods have been used to establish the instrument's calibration. As Method 1, the instrument's response to a monodisperse distribution of particles was evaluated. 30 μ m glass calibration spheres on glass slides were placed in the instrument's sampling volume and their yield extrapolated to an equivalent mass concentration of 3 μ m particles. This procedure is only "ball-park" and was used to coarsely establish system gain.

In Method 2, experiments were performed with natural polydisperse distributions wherein the instrument's output, from a chart recorder, was correlated to contemporaneous gravimetric data obtained by collecting air pollution particles. Calibration by this method was effected by drawing ambient air inside research facilities through the instrument and collecting the entrained particles on standard air pollution sampling filters. Linearity of the instrument response was excellent, from about 7 to 600 μ g/m³. Absolute results from Methods 1 and 2 were in fair agreement even though no attempt was made to determine the particle size distribution of the sampled particulates. It was assumed that their mass mean diameters were in the range 1-5 μ m.

In Method 3, a more controlled calibration was conducted with two identical particle detection instruments in tandem and sampling the effluent from a vertical elutriator measuring the mass concentration of cotton dust in a knitting mill. The purpose of the elutriator is to preseparate or "filter out" particles having diameters larger than about 15 μ m. That is, larger

particles cannot ascend to the collection filter because their settling velocities exceed the slow, presumably laminar, vertically-rising air flow in the elutriator.

Data from both polydisperse Methods 2 and 3 are shown in Figure 3. The cotton dust data were used to set the gains of both instruments which, again, were in tandem. The solid line with 45° slope represents this calibration. The cotton dust calibration was chosen over Method 2 because the experiments were more controlled, with more replications, and both instruments were simultaneously calibrated. Further, a cursory check of particle size with an electron microscope revealed the particles to be fairly spherical and to be a few micrometers in diameter. However, the linearity of Method 2 is better. A fourth air pollution point (square symbols) at $\approx 7\mu\text{g}/\text{m}^3$ (not shown in Figure 3) is equally close to the 45° line.

The air pollution calibration sensitivity of Method 2 was found to be higher than the vertically elutriated cotton dust calibration. Work is now in progress which may resolve the $\approx 50\%$ difference in calibration sensitivity. There is some recently-generated evidence⁶ that the vertical elutriator may emit particles larger than $15\mu\text{m}$, thereby increasing the mass mean diameter \bar{d}_m above the expected value of about $5\mu\text{m}$ ⁷ at the outlet of the VE. Although this result is somewhat contradictory to the cursory electron microscopy, if \bar{d}_m was significantly larger than $5\mu\text{m}$, and especially, if \bar{d}_m increased with increasing cotton dust loading, the discrepancy between gravimetric calibration might be explained.^{4,5}

The difference in calibration response between Methods 3 and 4 is not particularly serious and is typical of such work, when widely different

particulate types are used for calibration, and when many months separate the experimentation. Indeed, the general agreement between Methods 1, 2, and 3 are encouraging.

Generally, a more precise calibration is obtained when the process particulates being monitored are collected for a process-specific calibration. For the BNL cryogenic system, gravimetric measurements were regarded as exceedingly difficult and impractical because, by design, the oil aerosol levels are normally low. This is why it was necessary to employ Methods 1, 2, and 3. The independence of the forward scattering measurement to composition provides the basis for calibrating on one type of particulate and then measuring another.

As Method 4, an exploratory process-specific calibration was attempted at BNL after the oil mist removal system had been significantly upgraded. Prior to the installation of new filtering equipment, a marginally successful gravimetric experiment might have been run, with several hours exposure. Insufficient oil mass was collected on the 47mm filter papers for analysis since the oil aerosol mass concentration in the helium was reduced substantially. "Zero" calibration point was obtained. That is, both the gravimetric analysis as well as the light scattering instrument gave $0 \pm 5\mu\text{g}/\text{m}^3$.

Thus both laser particulate instruments are now operating with the cotton dust calibration of Method 3. These data, coupled with observations of instrument performance for nearly 2,000 hours, have led to the following results for sensitivity (noise) and baseline stability:

noise:	$\delta\chi$	(minutes)	\lesssim	$5\mu\text{g m}^{-3}$
drift:	$\Delta\chi$	(hours)	\lesssim	$10\mu\text{g m}^{-3}$
drift:	$\Delta\chi$	(2000 hours)	\lesssim	$20\mu\text{g m}^{-3}$

As a matter of interest, the latter instrument drift result was obtained with the instrument looking at a vacuum $\approx 10^{-3}$ torr (0.1 Pa) for about half the time and at 4 atm (0.4 MPa) of clean, stationary helium for the balance.

IV. APPLICATION EXPERIENCE

RESULTS OF TEST NO. 1 ON BNL 500 WATT (7 K) HELIUM REFRIGERATOR

The BNL 500 watt (7 K) cryogenic system has an oil removal filtration system which uses a combination of separators, coalescers, demisters and adsorption beds to achieve a desired final purity level of 10ppb ($24\mu\text{g}/\text{m}^3$) of oil aerosol, or less. The adsorption bed is a tower filled 50% with 6 x 8 mesh activated charcoal and 50% molecular sieve 4A and is located as the next to last filter in the series. The laser particle detector was first installed upstream of the adsorption bed. No data were taken at this location as the aerosol mass concentration during operation was in excess of 1200ppb, the upper limit of the instrument. Higher levels can be measured by the instrument by changing ranges but this was not done as this information was of no interest.

The laser particle detector was then installed downstream of the adsorption bed with the results of mass concentration χ versus mass flow rate \dot{M} as shown in Figure 4.

The pressure (P_{He}) was approximately 10 atm (1 MPa) and the mass flow rate was measured using a precision orifice. After stabilization of the χ reading for $\dot{M} \approx 129 \text{ gm sec}^{-1}$, the flow was reduced in discrete steps, as seen on the figure. The "overshoot" following each reduction was due to manually actuated flow adjustments.

After stabilization for $\dot{M} \cong 50 \text{ gm sec}^{-1}$, the compressor was shut down. Note the rapid decrease in χ and, after about ten minutes, that χ rises to a residual value of about $180 \mu\text{g m}^{-3}$ with $\dot{M} = 0$. It does eventually decay to zero.

Figure 5 shows a plot of these data at $P_{\text{He}} = 10 \text{ atm (1MPa)}$ and for 15 atm (1.5MPa) when plotted against \dot{M} . The decreasing behavior of χ with increasing \dot{M} , with constant P_{He} , is expected if the adsorption bed operated by inertial collection. Similarly, increased χ (for constant \dot{M}) with increasing P_{He} is not unexpected. Interestingly, the oil aerosol mass flow rate past the instrument is fairly constant with helium mass flow rate but increases with increasing P_{He} , being $\approx 1.3 \text{ mg min}^{-1}$ for 10 atm (1MPa) and 3.0 mg min^{-1} for 15 atm (1.5MPa) .

It was obvious from the data that the oil removal system was unsatisfactory when mass concentrations at full flow rates were 30 times those desired. Results of experiments with the pilot system indicated that a new design of demister could alleviate the problem. Two were constructed and installed ahead of the adsorption bed. Preliminary results indicate that χ levels are now satisfactory. (See Method 4, above.)

RESULTS OF TEST NO. 2 ON BNL 500 WATT (7 K) HELIUM REFRIGERATOR

The addition of a second laser particle detector allowed for simultaneous measurements upstream and downstream of the adsorption bed in this test run. The instruments had been previously calibrated against each other as discussed under CALIBRATION, Method 3.

A several hour run produced no measurable level of particulates upstream of the adsorption bed (noise level $\approx 3 \text{ ppb}$). Downstream, a level in the order of 20 ppb was measured. This higher downstream reading indicates that

charcoal/mol sieve dust is being carried out of the adsorption bed past the dust retention filters by the helium gas flow. Experiments where puffs of gas were passed through the bed produced dust levels in excess of 1000 ppb. In the future, the adsorption bed will be by-passed and if the upstream filter components continue to be 100% effective in removing oil, then the adsorption beds will be removed altogether. Probably at least 1000 hours of running time will be accumulated before that decision will be made, since the pilot system tests did demonstrate higher equilibrium aerosol levels after 1000 hours. Unfortunately the laser particle detectors were not available during the pilot tests to quantify the measurements.

RESULTS OF TEST ON LASL 700 WATT (4.5 K) HELIUM REFRIGERATOR

During acceptance testing of the LASL 700 watt (4.5 K) cryogenic system, currently on loan to the Brookhaven Isabelle Project, the laser particle detector was used to measure particulate levels. Tests were run for several hours per day over a three day period at helium pressures P_{He} of 15 atm (1.5 MPa) and flow rate M up to 123 gm sec^{-1} . Particulate levels were measured downstream of the adsorption bed and averaged 20 ppb at full flow. No attempt was made to determine if the particulates were oil or charcoal/mol sieve dust. Figure 6 shows the results of mass concentration χ versus time as flow rate M is increased from 0 to 123 gm sec^{-1} .

ACID MIST DETECTION

Figures 7 and 8 describe the flow loop and calibration results for repackaged electro-optics applied to sulphuric acid mist detection. One purpose of this exploratory test was to evaluate the linearity and repeatability of response to g/m^3 mass concentration levels of H_2SO_4 mist. The acid mist particulates were expected to have \bar{d}_m of about $0.4 \mu\text{m}$. Water was injected into a hot,

turbulent, SO₃-rich process stream to produce the aerosol. A sample stream was extracted, transported, and measured as shown. Volumetric flow rates up to about 15 SCFM could be achieved.

Figure 8 shows a typical calibration response. Linearity is fair and repeatability was good so long as the sampling train wall temperatures was constant.

Another purpose of the test was to evaluate the reliability of the repackaged oil mist system. The instrument proved worthy of the hostile acid plant environment over several months of evaluation.

Unfortunately, problems were experienced with the sampling train in the sense that the mass concentration seen by the instrument depended too sensitively on the temperature of the piping. The origin and "fix" for this unknown wall temperature effect are currently under investigation.

V. CONCLUSIONS

A new type of laser particulate detection instrument has proved extremely valuable in the design and monitoring of oil removal systems for cryogenic refrigeration systems. When located at the inlet of a refrigerator, any aerosol or particulates that could be harmful to the operation of the refrigerator would be detected.

Its main features include: in situ measurement of particulates in the high pressure helium, continuous monitoring with alarm capabilities which permits automatic shutdown of attended or unattended systems, sensitivity down to a few parts per billion, long-term stability on the order of 10ppb, and straightforward data interpretation.

The concept of continuous automatic calibration and baseline zero adjustment relieves the operator of any manual adjustments or loss of confidence in the signal levels recorded. The one second response time allows instant turnoff if the filters experience an oil breakthrough or if an operator inadvertently opens bypass valves around the oil removal system. The strip-chart recording allows for long-term comparison of particulate levels to determine if a buildup is occurring and its rate of buildup. It will also aid in predicting the amount of contaminants frozen out in the refrigerator and the operating time remaining before refrigerator shutdown and warmup is required.

Applications to other processes, such as acid mist detection or workplace dust monitoring, are similarly useful and currently under active development.

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CAPTIONS

- Figure 1. Installation Diagram of Laser Aerosol Sensor.
- Figure 2. Block Diagram of Laser Aerosol Detector and Monitor Electro-Optical System.
- Figure 3. Mass Concentration Data: Laser Particle Counter vs. Gravimetric Measurements.
- Figure 4. Representative Chart Record of Mass Concentration χ at Various Mass Flow Rates \dot{M} , BNL 500 Watt Refrigerator.
- Figure 5. Plot of Mass Concentration χ Versus Mass Flow Rate \dot{M} , BNL 500 Watt Refrigerator.
- Figure 6. Plot of Mass Concentration χ Versus Mass Flow Rate \dot{M} , LASL 700 Watt Refrigerator.
- Figure 7. Flow Loop for Acid Mist Tests.
- Figure 8. Instrument Response Versus Acid Mist Concentration.















