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INVESTIGATION OF PARTICULATE MATTER MONITORING USING CONTACT ELECTRIFICATION

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INVESTIGATION OF PARTICULATE MATTER MONITORING USING CONTACT ELECTRIFICATION

by

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ABSTRACT

The theory of the charging of aerosol particles by contact electricity is reviewed, as well as the development of monitors for particulate matter based on this principle. Data on the performance of these monitors is scarce and sometimes contradictory. In the present work, laboratory tests were carried out with a variety of test dusts. The results show that the dynamic response of the contact electricity monitor tracks well with that of an optical detector. The total charge correlates well with the gravimetric mass. Humidity has little effect on the response, but droplets cause failure of the instrument. Pre-charge on the particles did not influence the detector. Some evidence was obtained showing that particle size does not have an important effect on the response, although there is a cutoff for very small particles. Results for the sensitivity of the instrument can be grouped according to the electrical resistivity of the material. It is found that the condition of the surface of the Inconel probe has a major effect on the sensitivity. Additional work is necessary on this aspect of the detector. The theory of Cheng and Soo for the charging of metal particles is discussed. There is at present no theory applicable to insulators.

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SECTION I

CONCLUSIONS

The present experimental results confirm the promising findings reported by others concerning the dynamic response of the contact electricity monitor, as well as the good correlation of total charge to mass determined gravimetrically. No evidence was found for disturbing effects of humidity (below the dew point) or precharge. Particle size was found not to have an important influence on the response, although a cutoff at very small particle size exists.

The sensitivities for various substances can be grouped according to electrical resistivity. This is predicted by the theory of Cheng and Soo for metals and good semiconductors. There is no applicable theory for insulators. Sensitivities ranged over a factor of 60, so that some constituents of a mixture will be difficult to detect.

It was found that the condition of the surface of the stainless steel probe had a major influence on the sensitivity. Additional work is necessary before this surface effect can be understood, or at least controlled.

SECTION II

INTRODUCTION

When aerosol particles come into momentary contact with a surface, they may acquire an electrostatic charge as a result.[1,2] This phenomenon has been used as the basis of a monitor for particulate matter.[3,8] An aerosol stream can be drawn past an insulated probe. Particles colliding with the probe then transfer or carry away charge from the probe. The electrical current from such a probe has been found to correlate surprisingly well with the mass concentration of the aerosol.[6,8-10]

The contact electricity type of monitor offers some important advantages by making possible in-situ measurements in real time. The monitor can respond fairly rapidly to changes in mass concentration and is sensitive over a wide range, particularly to high concentrations. The electrical signal is convenient for data collection and the device is comparatively simple in construction. The history of these instruments goes back more than a decade, and at least two models have been available commercially.

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[1] W. R. Harper, "Contact and Frictional Electrification", Oxford U. Press, Oxford, (1967).

[2] L.B. Loeb, "Static Electrification", Springer-Verlag. Berlin, (1958).

[3] A. Schutz, Staub 24, 359 (1964).

[4] A. Schutz, Staub 26, 18 (1966).

[5] A. Schutz, Staub-Reinhalt der Luft, Band 26 (1966) No. 5, Seite 198/201, 1-4.

[6] R. Prochazka, Staub 24, 353 (1964).

[7] R. Prochazka, Staub 26, 22 (1966).

[8] L. Cheng and S.L. Soo, J, Appl. Phys. 41, 585 (1970).

[9] H. Schnitzler, SchrReihe Ver. Wass.-Boden Lufthyg, Berlin-Dahlem, V. 33, Stuttgart (1970).

[10] IKOR, Inc., unpublished reports, and private comm. from A.H. Gruber.

In spite of this, the contact electricity monitor has not come into general use. This may be partially due to a feeling on the part of potential users of the need for better understanding of the physical principles involved in this type of monitor.[11] The present investigation was undertaken to assess the current state of knowledge of these principles. In addition, laboratory experiments were devised to determine the important parameters involved in the operation of these instruments.

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[11] G.J. Sem, et al, "Instrumentation for Measurement of Particulate Emissions from Combustion Sources", Vol. I: Particulate Mass, Thermo-Systems, Inc. APTD-0733, (1971).

SECTION III

THEORY OF CHARGING OF AEROSOL PARTICLES BY CONTACT ELECTRICITY

General Theory

It is necessary to discuss first the terminology used. "Contact" electricity refers to charge transferred as a result of pure contact with no sliding or rubbing. On the other hand, "triboelectricity" involves rubbing (tribo is Greek meaning rubbing).[1,2] The latter may include the transfer of material and local heating. In the case of the charging of aerosol particles, it is not clear which type of interaction takes place. Probably all types are involved to some degree. In the present work, we shall use the term "contact" in discussing the charging of particles, recognizing that other kinds of interactions may well be present.

Pure contact is difficult to achieve in practice. Harper[1] has succeeded in some careful experiments with large metal spheres. His results agree with the theory for contact between metals. When two metals touch, the difference in the contact potentials of the two materials will cause a flow of electrons to take place. The buildup of charge will continue until the resulting electric potential equals the difference in the two contact potentials.

This charging of metals can also be described in terms of the quantum mechanical energy band theory.[1,12,13] In a metal, the energy levels are filled up to a certain maximum energy, the Fermi level. The additional energy from the Fermi energy up to zero energy is called the work function. When two metals are brought into contact, the Fermi levels will initially be at different heights (energies). Electrons will then flow from the higher to the lower until the two Fermi levels are equalized. At that time, the difference in work functions gives the difference in potential of the two metals.

In the case of metals, some charge can flow even when there is a small gap between the surfaces by the quantum mechanical tunnel effect.[1] A serious complication arises during the separation of the metals.[1] The air breaks down and an appreciable back-flow of charge takes place. The net charge transferred as a result of the contact then depends on the magnitude of the backflow.

The theory for metal-metal contact can be generalized to include semiconductors.[1,12] Whereas for metals the charges reside on the surface, for semiconductors there are fewer electrons available at the surface and there will be a flow of charge from the interior. Since there is now an appreciable resistivity, there will be an associated time delay

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[12]"Static Electrification, 1971", Proc. of the Third Conf. on Static electrification organized by the Static Electrification Group of the Institute of Physics held in London, May 1971. Conf. Series, No. 11, The Institute of Phys., London and Bristol.

[13]"Electrostatics and Its Applications", A.D. Moore, Ed., John Wiley & Sons, N.Y., (1973).

with a time constant in seconds given by $8.85 \cdot 10^{-14} K\rho$, where K is the dielectric constant and ρ the resistivity in ohm-cm. This implies that the amount of charge transferred will depend on the duration of contact. Even for large spheres, the time constant becomes too large for appreciable flow of charge in the case of semiconductors having high resistivity.

Insulators have an even higher resistivity so that there is no flow of charge from the bulk material. Surface states become the important source of charge for transfer. Such surface states are complicated, depending on the detailed physical and chemical condition of the surface. Adsorbed ions giving rise to electric double layers play an important role. Harper[1] states that the charge transferred consists of ions rather than electrons. It may be mentioned also that there is a class of insulators designated electrophobic which apparently have no surface levels available for contact charging, if the surfaces are clean. These materials include some common plastics such as nylon, lucite and teflon.[1]

In the case of insulators particularly, there are other charging mechanisms besides contact charging which may have to be considered, such as electrolytic effects for moist surfaces and frictional effects resulting in transfer of material.[1,2]

The foregoing has been a brief review of the status of knowledge of contact charging. The reader is referred to the bibliography for further information. We can sumarize by saying that our knowledge

is good or quantitative for metals but becomes progressively worse as we make the transition to insulators. This applies to ideal surfaces; in real cases, the charging mechanism may be drastically altered by contamination of the surfaces or by the physical condition.

Charging of Aerosol Particles

When the theory of contact charging is applied to aerosol particles, there are some additional considerations. We are concerned with the impact of the particle with a solid surface, a metal in the practical case of a probe. The charging is a dynamic process;[8] the duration of contact is short, of the order of 10^{-9} to 10^{-10} seconds for our present purposes. Therefore, there will be no time for flow of charge from the bulk material for substances having resistivities greater than about 10^3 ohm-cm.

The surfaces of metal particles will invariably be oxidized. According to Harper,[(1)] such oxide layers behave as a metal with a work function of about 5.5eV. Most oxides are semiconductors; when the layer is thick, it behaves as a semiconductor. If the layer is not too thick then, charge can be transported either by conduction or by tunnelling.

A serious attempt to calculate the contact charging of metal particles has been made by Soo and his collaborators.[8,14] Cheng and Soo[8]

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[14]S.L. Soo, "Dynamics of Charged Suspensions", Topics in Current Aerosol Research, Vol. 2, International Reviews in Aerosol Physics and Chemistry, Pergamon Press Ltd., Oxford, (1971).

consider the impact of spherical particles in some detail. This theory greatly overestimates the magnitude of the charging. Cheng and Soo point out that the theory applies to clean surfaces; surface contamination could have an important influence on the charge transfer. In Appendix I, this theory is discussed in terms of the present application.

In conclusion, some progress has been made in understanding the contact charging of particles, but theoretical predictions are not yet quantitative, and there is no theory available for insulators.

SECTION IV

CONTACT ELECTRICITY MONITORS FOR PARTICULATE MATTER

Review of Instrument Development

An early version of an instrument employing a spherical metal probe was described by Schütz.[3] A later version of his instrument utilized a probe-in-nozzle technique.[4] Prochazka[6,7] has described the development of the Konitest, an instrument which was produced commercially for a time. In the Konitest, the electrode is a tube of a semiconductor, steatite (magnesium hydrosilicate). The gas is introduced radially and the resulting helical path causes the particles to impinge on the walls of the tube. In a second version, the tube is shaped as a Venturi nozzle. Soo and his collaborators[8] constructed instruments using a spherical metal electrode and also a tubular electrode. In the USSR, Kisler[15] has described some contact electricity instruments including a monitor using a wire probe. The IKOR Air Quality Monitor was apparently independently developed and is now commercially available.[10] It utilizes a bullet-shaped Inconel probe in a pipe.

In all of the instruments mentioned above, the transfer of charge to the probe by particles colliding with it results in a current which is continuously monitored with an electrometer. Some other types of instruments have been developed to observe the electrical pulses produced by individual

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[15] S. YA. Kisler, Mekh. Avtomat. Proizvod., 26 (9): 27-28, (1972).

particles. Min, et al. [16] described such a device whereby particles charged by wall collisions induce a voltage on an electrode. Detectors for individual particles, however, appear to be applicable only to large particles carrying substantial charges. Other types of monitors have been developed which utilize electrostatic charging of particles but not contact charging. Grindell [17] described a dust monitor which charged the particles in a high voltage corona. The particles were then precipitated on a collector electrode and the current determined. In the instrument developed by Coenen, [18] the particles acquired charge by repeated collisions with the plates of a high-voltage capacitor.

Characteristics of the Contact Electricity Monitor

Some experimental investigations have been made of the characteristics of the contact electricity monitor. These investigations are few in number; most are under field rather than laboratory conditions. On some of the most important characteristics, there are contradictory reports. The status is summarized below by topic.

(1) Electrical current vs. mass concentration. In most cases the instruments are to be used as an indirect measurement of the particulate mass concentration in the gas monitored. It is most important to establish the correlation between the electrical current from the probe and the mass concentration measured gravimetrically. Schütz [4] reported the

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[16] K. Min, B. T. Chao, and M.E. Wyman, Rev. Sci. Instr. 34, 529 (1963).
[17] D.H. Grindell, Proc. Inst. Elec. Engrs. (London) 107A: 353-365, (Jan. 1960).
[18] W. Coenen, Staub 27, 32 (1967).

following relationship for quartz dust:

$$I = ak^b$$

where I is the electrical current in amperes, k is the mass concentration in mg/m^3, the exponent b ranges between 1.26 and 1.30, and a is inversely proportional to the particle diameter.

On the other hand, Prochazka [6] found the Konitest to give an accurately linear indication for concentrations from 0 to $3g/m^3$. The dusts were sampled at various industrial sites. Ito, et al. [19] also reported a linear relationship for a Konitest monitoring cement kiln exhaust at the exit of an electrostatic precipitator for concentrations up to $10g/m^3$. Schnitzler, et al. [9] found a correlation coefficient of 0.92 between the gravimetric mass and the Konitest reading for effluent from a coal-fired plant. The performance of the Konitest compared very favorably to the best response obtained from transmissometers and beta radiation attenuation monitors at the same location.

Cheng and Soo [8] obtained plots of mass flow vs. probe current using coal dust. The plots show small deviations from linearity, being concave upward.

Field tests of the IKOR Air Quality Monitor show that the integrated current (total charge) has a reproducible ratio to the gravimetric mass for aluminum oxide. [10] Good agreement was also obtained between the

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[19] T. Ito, H. Saito, and N. Furuya, Proc. Japan Soc. of Air Poll., 13th, (1972), p. 247.

total charge and the mass concentration obtained with an EPA sampling train. These tests did not span a wide range of concentrations.

(2) Dependence of the current on particle size. As pointed out above, Schütz[4] found the constant a to be inversely proportional to particle diameter. Thus, higher sensitivity is predicted for small particles. On the other hand, Cheng and Soo[8] predict a current independent of particle size for one experimental configuration. They report that this was verified by Min. Ito, et al.[19] report the Konitest to be little affected by particle size when sampling cement dust.

(3) The effect of humidity. Water layers on surfaces usually contain dissolved impurities. Some authors believe electrolytic ions are important for contact charging.[2] However, Harper[1] does not, and tried unsuccessfully to obtain an experimental correlation between humidity and charging.

Schütz[4] reported no effect on his instruments' performance for relative humidity up to 99%. He did warn that droplets of water cause erroneous indications. Cheng and Soo[8] point out that the conductivity of mineral particles increases with the relative humidity. Ito, et al.[19] state that humidity caused no problems in their tests of the Konitest. Kisler[15] states that the humidity of the gas should be below the dew point.

(4) Temperature effects. Schütz[4] stated that the contact electricity monitor should not be operated above 70°C due to the presence of thermal emf's. However, a modified IKOR AQM has been operated at 593°C.[10]

(5) Probe material. With the exception of the Konitest, most instruments have used metal probes. According to Schütz,[4] the use of metal for the probe ensures that the charging will always be positive. It seems clear that this is a dubious assumption; in fact in field tests of the IKOR the sign of the current has been found to vary with material and the standard IKOR AQM is equipped with an automatic polarity-reversing circuit to maintain positive indication on the recorder.[10]

(6) Composition of dust sampled. Although field tests have been conducted with a variety of dusts, there has been no reported correlation of the response according to some characteristic of the dust sampled. Schütz[4] concluded that the influence of the properties of the material was far less decisive than the particle size. Ito, et al.[19] stated that the current obtained for a given mass concentration depended on the material, but that for cement dust the electrical resistance had little effect on the output. Most users of the contact electricity monitors have simply empirically calibrated the monitor for each type of dust sampled.

According to the theory of Cheng and Soo,[8] the amount of charging depends on the electrical conductivity, the density, and the elastic properties of the materials of both particles and probe. (See Appendix I)

(7) Discussion. The above review indicates that the mass correlation observed by several experimenters is surprisingly good. However, there is need for additional work to clarify some of the conflicting reports

in order to identify the real problems associated with this type of instrument. There is also a need for better understanding of the charging mechanism.

SECTION V

EXPERIMENTAL MEASUREMENTS

Experimental Arrangement

Because of the incomplete state of the theory of the contact electricity type of monitor and because experimental information is sparse and sometimes contradictory, a series of controlled laboratory tests were devised to investigate the response of such a detector under various conditions. The goal was to determine the important parameters affecting the operation and to try to understand these in terms of the theory.

The general arrangement is shown schematically in Fig. 1. Room air is drawn into a metal manifold 70 cm long x 3.2 cm inside diameter. Inlets were provided for adding humidified air and aerosol generated from test dusts. The manifold ended in the inlet of the IKOR Air Quality Monitor.* Another inlet nearby led to an optical analyzer. The main aerosol stream passed the IKOR probe, through the filter (or blank) and then was impelled by the IKOR blower to the exhaust.

The airflow rate through the IKOR AQM was between 8 and 14 ℓ/s (17-30 CFM). The room air temperature was maintained at 23°C by the laboratory air conditioning and the relative humidity was stable over long periods of time, being typically at about 50%. Even on the most sensitive scale, no room dust could be detected by the IKOR AQM.

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*IKOR, Inc. Burlington, Mass.

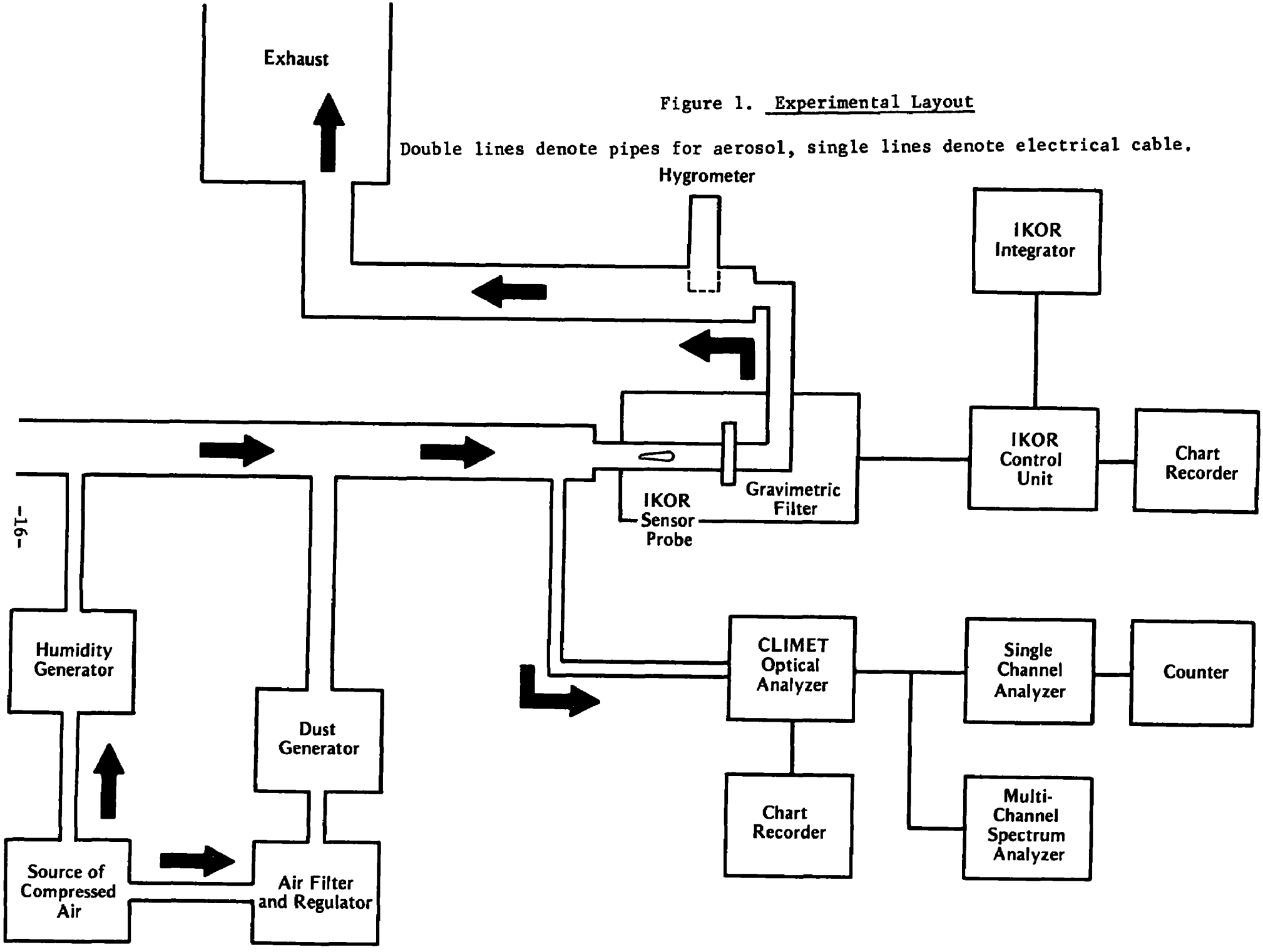

Figure 1. Experimental Layout

Double lines denote pipes for aerosol, single lines denote electrical cable.

The IKOR Air Quality Monitor

A Model 206 IKOR AQM was used for this work. This unit consists of a stainless steel sampling pipe, 2.4 cm inside diameter x 135 cm long, connected to a flexible Teflon-lined hose 318 cm long. The hose conducts the aerosol stream to the sensor unit where it passes through a stainless steel pipe 2.4 cm i.d. containing an Inconel 625 probe shaped like a bullet and centered in the pipe. At the widest part the probe is 2.0 cm in diameter. The probe is insulated and connected to an electrometer circuit which is of a modern operational amplifier (integrated circuit) design. The electrical current is read out on a panel meter and on a Rustrak recorder. The readout is linear with a switch for 5 decades of sensitivity. During the present work, the absolute current was determined with a Keithley 610 C Electrometer; full scale on the IKOR meter ranges from 10^{-12} to 10^{-8} amp. The IKOR unit was also equipped with a current integrator so that the total charge accumulated over a period of time could be determined. The IKOR electronics was very stable, maintaining its zero setting and calibration over two months of operation.

The airflow rate of the IKOR AQM is determined by difference in pressure sensed by pitot tubes. Thermocouples are provided for measurement of gas temperature.

The Optical Analyzer

A Climet CI-201 Optical Analyzer* was used to obtain an independent measure of aerosol concentration. In this instrument, forward-scattered light is sensed by a photomultiplier. The output of a count-rate meter is displayed on a chart recorder with a logarithmic scale. The analog pulses from individual particles were passed through a single channel analyzer and then to a counter. The analog pulses were also processed by a 1024 channel pulse-height analyzer to obtain particle size spectra. The system was calibrated with monodisperse latex spheres. It was determined that the spectrum analyzer operated between 1.0 and 5.5 microns. The counter had a threshold set at 3.7 microns in order to reduce the counting rate to manageable levels.

Dust Generator

The dust generator devised for this work is shown in Fig. 2. Compressed air is filtered, regulated, and passed through copper tubing to a hypodermic needle (hole dia. $4 \cdot 10^{-4}$cm) positioned near the dust sample in the bottom of the test tube. The jet stirs up the dust; the air stream containing entrained particles then rises past the washers (intended to intercept large particles) and leaves via the side arm. The aerosol stream then emerges as a jet which impinges on the bottom of the second test tube. This was designed to break up aggregates and to further disperse the dust.

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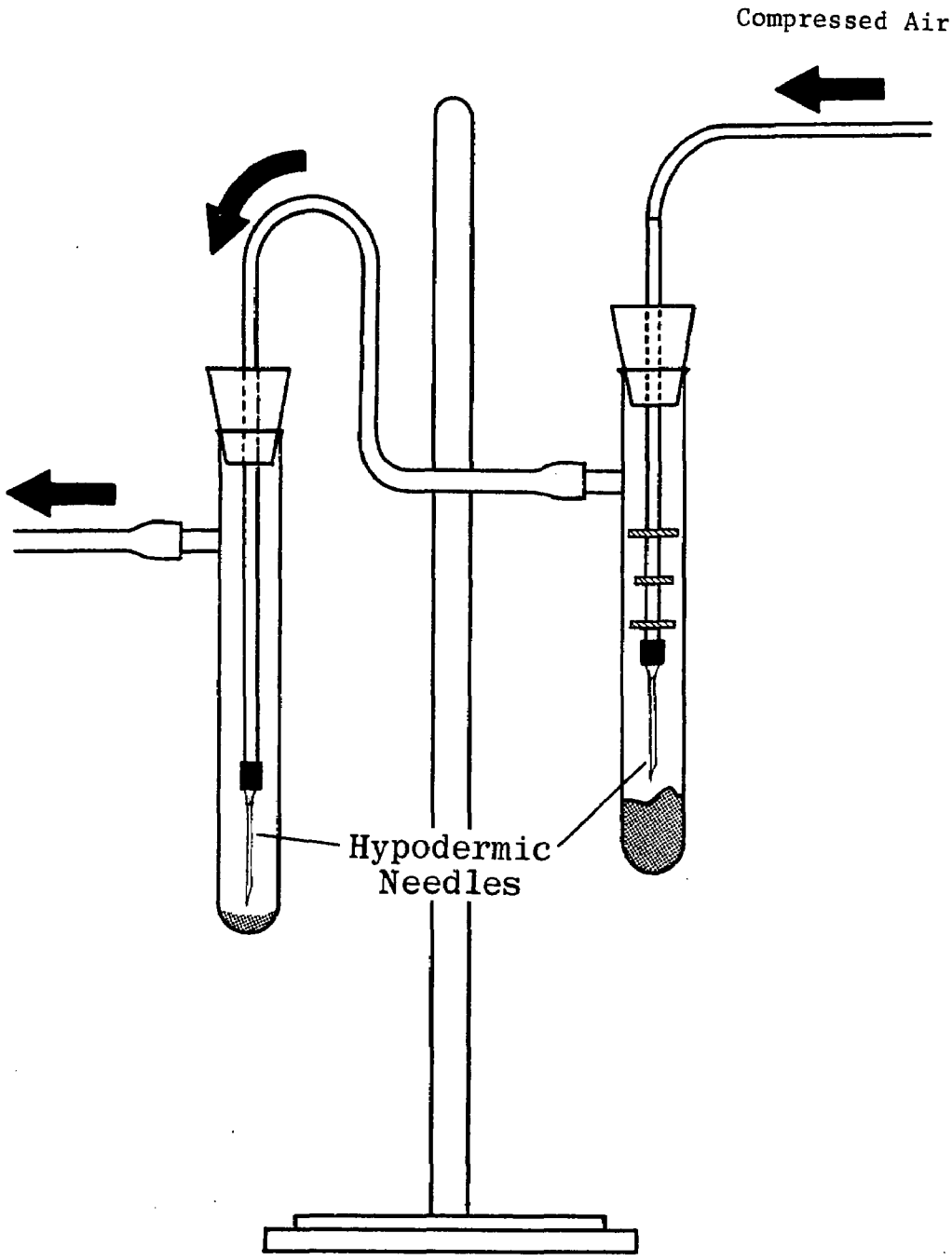
*Climet Instruments Company, Redlands, California

Figure 2

Dust Generator



The generator effectively discriminated against large particles or aggregates. For example, very little output could be obtained with stainless steel particles with diameter 24-48μm. Also, no appreciable output could be obtained from a sample of carbon black particles 0.47μm diameter which agglomerated strongly. For all samples used, the peak of the size distribution as obtained from the optical analyzer was below one micron. This was irrespective of the size information provided by the supplier of the sized samples. In the case of stainless steel samples, a bimodal distribution was obtained; however, the preponderance of the particles were in the less-than-one micron size group.

Samples used included polydisperse aluminum oxide and polydisperse aluminum. These materials were replaced often in order to avoid depletion of the smaller particles. The remaining samples were sized in varying degrees. Table 1 lists the characteristics of the samples.

A maximum flow rate of 11 ℓ/min was achieved by the generator, with dust concentrations ranging up to 0.1 g/m^3. The maximum dust concentration which could be generated depended on the type of sample. Also, the stability of the concentration level depended on the sample characteristics. For some samples, the concentration was stable for extended periods, for others frequent adjustments were necessary. With this generator, tests could be run for about one hour with 1 g of sample.

TABLE 1

SAMPLE CHARACTERISTICS

Sample	Origin	Remarks
Titanium Dioxide	P.I.S.* No. 2-12	0.45μm av. dia.** $10.3m_2/g$ surface area
Red Iron Oxide	P.I.S. No. 25-4	0.3-0.8μm dia., spheroidal oxide
Silicon Dioxide	P.I.S. No. 10-6	1-4.5μm dia.
Fly Ash	P.I.S. No. 14-1	11μm av. dia., 82wt.% less than 44μm
Aluminum Oxide	Matheson, Coleman & Bell	Reagent grade powder, polydisperse
Glass Beads	P.I.S. No. 8-7	3-8μm dia., soda-lime glass
Glass Beads	P.I.S. No. 8-1	0.5-3μm dia., soda-lime glass
Molybdenum	P.I.S. No. 19-6	4.4μm dia., 100% between 0 and 10μm, nonspherical metal
Tungsten	P.I.S. No. 19-1	1.05μm av. dia., 100% between 0 and 3μm, nonspherical metal
Carbon Black	P.I.S. No. 2-6	0.01μm av. dia., 97% between 0.005 and 0.017, surface area $1000m^2/g$
Stainless Steel	P.I.S. No. 27-1B	6-12μm dia., spherical metal
Silver	P.I.S. No. 27-7	6-9μm dia., spherical metal
Copper	P.I.S. No. 27-6	5-150μm dia., spherical metal
Aluminum	P.I.S. No. 27-2A	spherical metal, 0.5-6μm
Aluminum	Unknown origin	polydisperse

*Particle Information Service, Grants Pass, Oregon.
**Size information from P.I.S., however, see discussion in Sec. IV concerning the observed particle size spectra.

Experimental Tests

Tests were performed to investigate the dynamic response of the monitor, the correlation of the total charge to the sampled mass, the effect of humidity, precharge effects, particle size effects, the importance of the surface condition of the probe, and the sensitivity for various substances. These tests are described below.

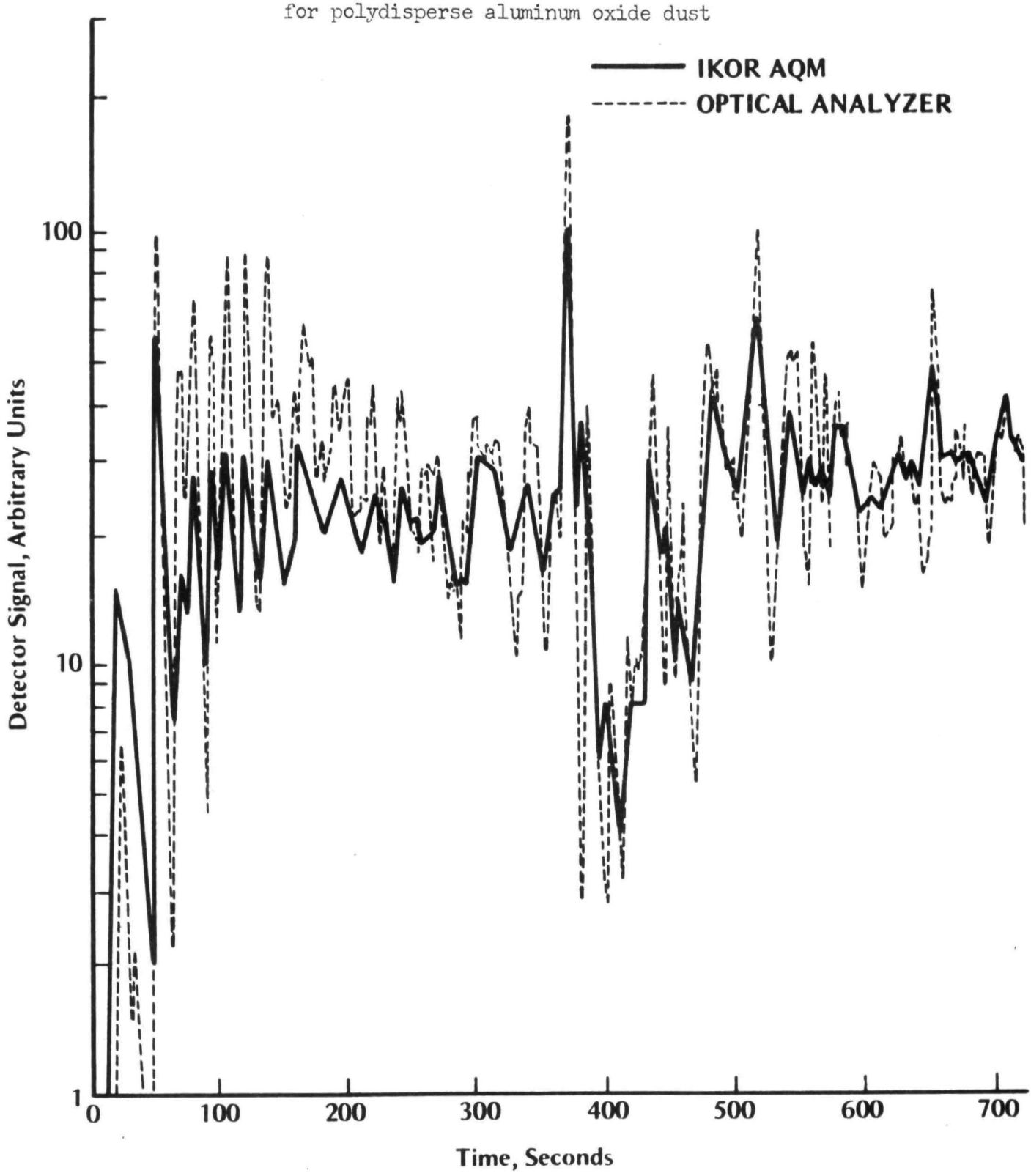
(1) Dynamic response. The time constant (1/e) of the IKOR AQM is approximately 10 sec. Since the Climet optical analyzer is orders of magnitude faster, an R-C network was added to increase the Climet time constant to 4 sec. Fig. 3 shows the superimposed chart recordings of the two instruments for polydisperse aluminum oxide dust. Since the original IKOR record was linear, it was necessary to transform it for the log plot. It has been pointed out before that a logarithmic display is advantageous for a contact electricity monitor.

Some of the differences in the records, particularly the more extreme highs and lows of the Climet trace, can be attributed to the 2.5 times shorter time constant of the Climet. Of course, there can be some real differences in the instantaneous concentrations at the sampling inlets. Also, the Climet sampled only the large end of the size distribution.

On the basis of many such runs, it is concluded that the correlation of the current records is satisfactory. There is one difference which should be mentioned. Sometimes at the very beginning of a run, the IKOR AQM is

Figure 3. Dynamic Response

Comparison of detector signals
for polydisperse aluminum oxide dust



much less sensitive. The sensitivity may increase by an order of magnitude in the first minute or so. This may happen even if the instrument has been previously running on the same dust, sometimes even within the hour. This sensitizing effect will be discussed further below in Sec. (6).

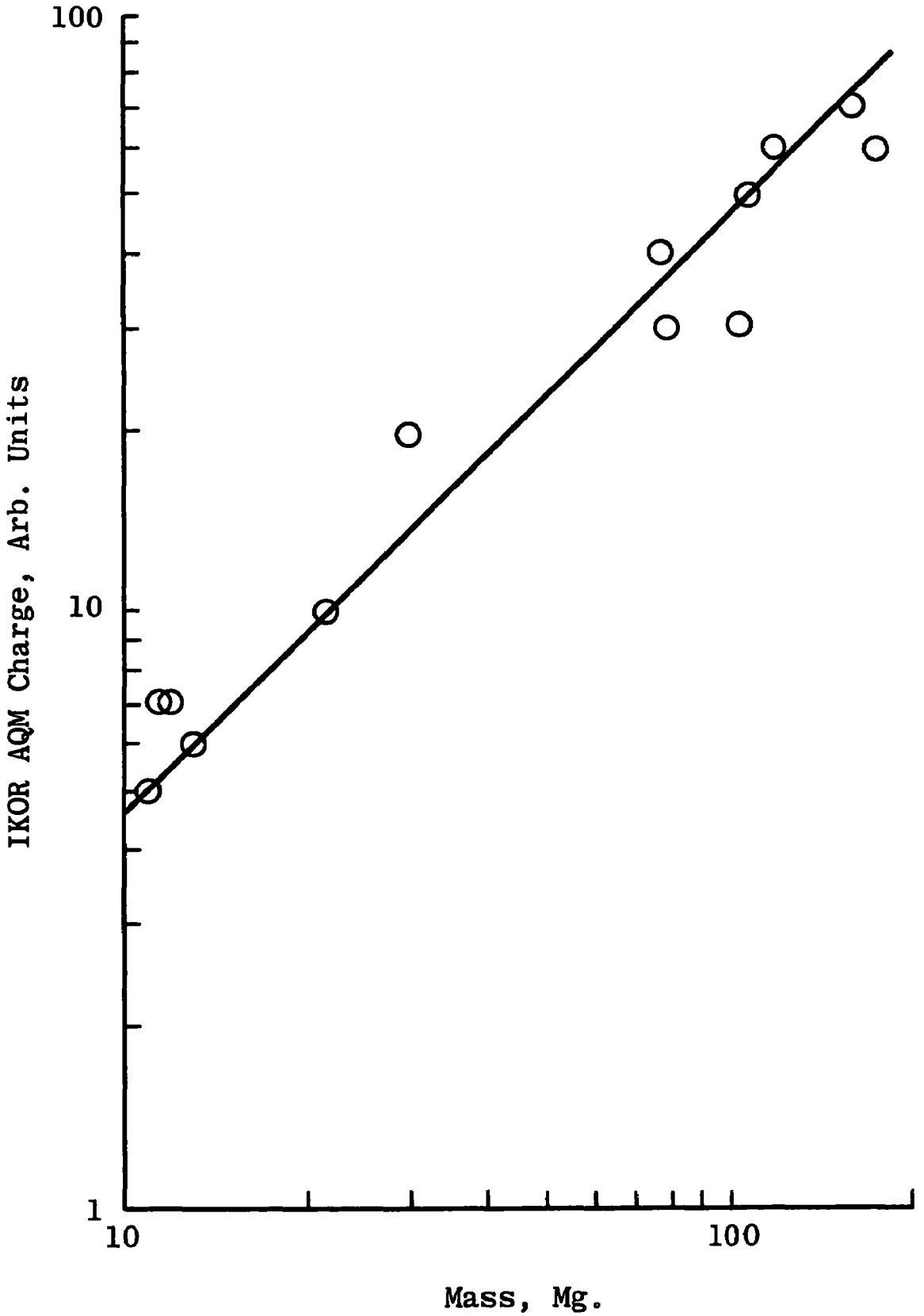
(2) Correlation of total charge to sampled mass. A series of runs were made with polydisperse aluminum oxide to compare the integrated current from the IKOR AQM to the mass determined gravimetrically. Gelman type A, 142mm dia. fiberglass filters were weighed to 0.1 mg. Since the room humidity was very stable, the filters were simply kept open to the room throughout the operation. Reproducibility of the weights was found to be within 0.1 mg in most cases. Runs were about 20 minutes in duration, with the average concentration varied from run to run.

The results are plotted in Fig. 4. The line is drawn with a 45° slope corresponding to true proportionality. The line was located by eye. It is concluded that the proportionality is satisfactory over a factor 20 in concentration.

It is reasonable to expect a strictly proportional response. If each particle independently transfers charge to the probe, that charge will eventually be returned to ground through the electrometer. Since the probe is maintained very near to ground potential by the circuit, there should be no charging effects. One could imagine a buildup of particles on the probe which would then alter the response for subsequent particles.[4] This, however, would shift the entire response curve. This was not observed during the aluminum oxide runs. However, a shift was seen during

Figure 4. Charge vs. Mass for Aluminum Oxide

Plot of total charge from the IKOR AQM vs gravimetric mass of aluminum oxide sample. The line is drawn at 45° (exact proportionality).



the succeeding aluminum measurements. Mass correlations were also run for the aluminum dust. The results are graphed in Fig. 5. The numbers are in sequential order of the runs. There was a very close proportionality for a while, then the sensitivity apparently decreased to the dashed line. Further discussion of this decreasing sensitivity will follow in Sec. (6). It is concluded, however, that the response of the IKOR is proportional to mass for both aluminum oxide and aluminum, i.e., an insulator and a metal.

(3) Effect of humidity. Moisture was added to the air by passing compressed air through warm, moist filter paper. The relative humidity was measured by the wet bulb-dry bulb technique. Using aluminum oxide dust, the relative humidity was varied from 45% to 69%. No effect on the IKOR response was observed. The upper limit to such an effect is estimated at 20%. Incidentally, the IKOR AQM cannot tolerate the introduction of wet steam. This evidently causes electrode leakage and complete instrument failure until the probe structure is dry. Problems from droplets have been mentioned by other authors.[4,15] The lack of humidity-produced effect is an indication that the charge transfer process does not involve surface electrolytic effects or that such effects are not important. The present tests did not investigate possible effects from very low humidity.

(4) Precharge effects. From published work, it is evident that the separation of dust particles as in our dust generator results in electrical charging of the particles.[20] If the particles are charged before

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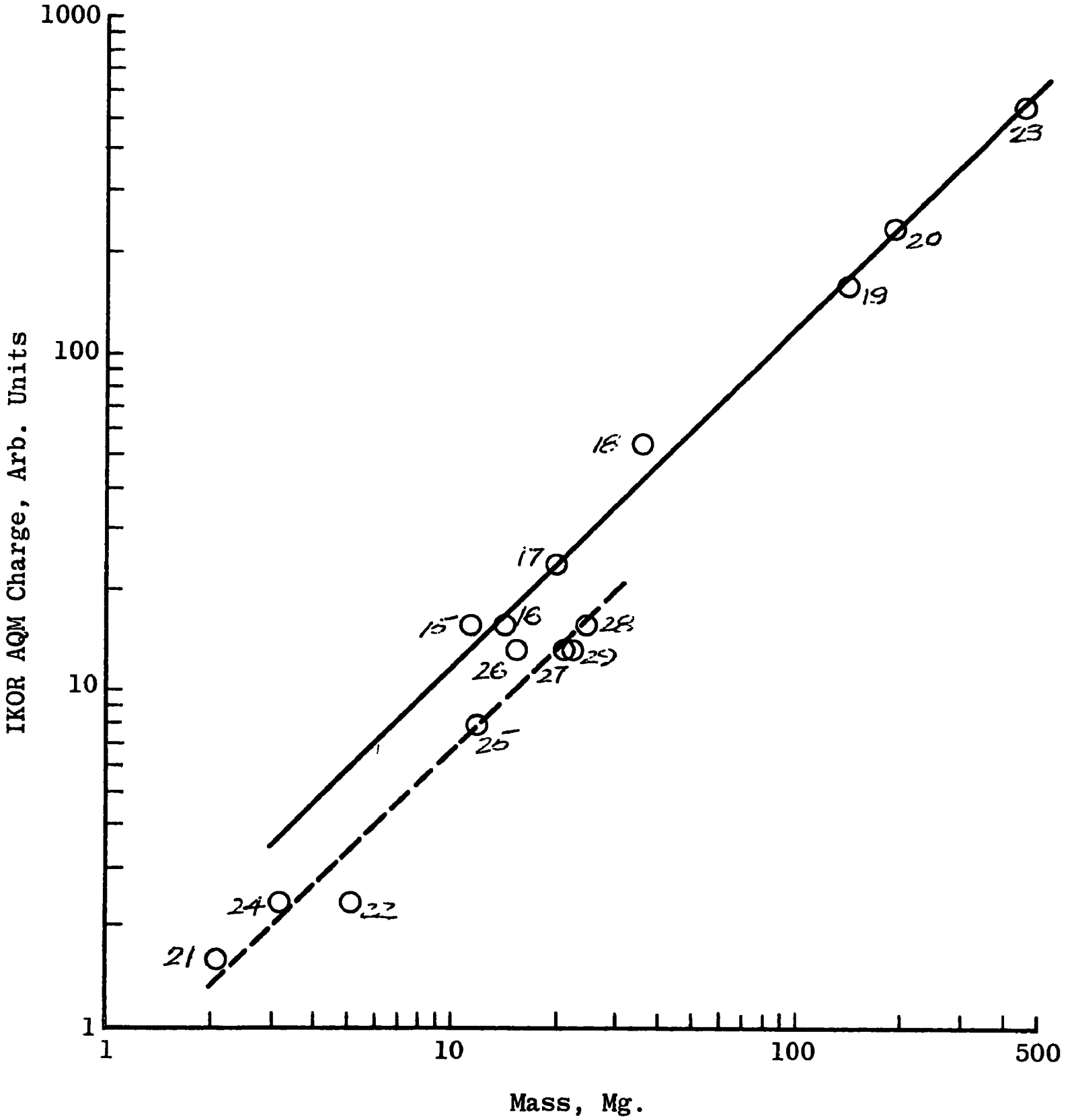
[20] W.B. Kunkel, J. Appl. Phys. 21, 820 (1950).

Figure 5. Charge vs. Mass for Aluminum

Plot of total charge from the IKOR AQM vs gravimetric mass of aluminum sample. The lines are drawn at 45°. Data points are numbered sequentially.



encountering the IKOR probe, this might affect the response. We investigated this possible effect by using a radioactive source to irradiate the aerosol stream emerging from the dust generator. This should reduce the charge through the interaction between charged gaseous ions and the particles.[21] A 1.2mCi Cd-109 source was placed within a centimeter of the 6 mm air tube, which had a thin Al foil window to pass the radiation. It is estimated that 2-5R/hr was incident on the aerosol stream. The ratio of the IKOR current for aluminum oxide dust with and without radiation was 0.97 ± 0.28. It is concluded that no effect was produced.

This finding is reinforced by the observed lack of effect of humidity. If significant precharging effects were present, one would expect humidity to have an appreciable influence on it.

(5) Particle size effects. Direct observation of particle size effects would require the measurement of the complete particle size distributions of the aerosols actually sampled. Since only the > 1μm portion of the distribution was measured in the present experiments, the information on particle size effects is more indirect. However, there are several kinds of evidence which are persuasive as to a surprising lack of dependence of the IKOR AQM response to particle size. Our experimental evidence against dependence on particle size includes:

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[21] D. Keefe, P.J. Nolan, and T. Rich, Proc. R. Irish Acad. 60, 27 (1959).

(a) Many samples of materials were used with various quoted particle sizes. There were obvious variations as judged by eye. The polydisperse Al and a sized (0.5-6 micron) Al sample gave closely similar results for the sensitivity in μC/g. One glass bead sample quoted as 0.5-3 micron diameter and another 3-8 micron diameter yielded sensitivities of 0.46 ± 0.01 and 0.41 ± 0.03 μC/g.

(b) The IKOR AQM flow rate was varied to investigate the effect on sensitivity. A variation of flow rate would affect the probability of impaction on the probe. Also, the charging process could itself be velocity dependent. Both of these effects involve particle size. Over a variation of a factor of 1.7 in flow rate, the sensitivity measured gravimetrically changed by less than 25% (standard deviation) for Al_2O_3, and by less than 18% for Al.

(c) A crude test for response to very small particles was made with cigarette smoke from filter cigarettes. The IKOR AQM barely responded to levels which were off scale for the optical analyzer. This shows there is an effective cutoff in the response to very small particles, no doubt due to the fact that they tend to follow the stream lines and hence fail to impact on the probe. This test shows that the present instrument does not respond inordinately to very small particles; in fact, for sufficiently small particles it does not respond at all.

In summary, these tests are not sufficient to rule out a dependence on particle size, but they do show that such a dependence is not pronounced above the cutoff. In Appendix I, it is shown that the theory of Cheng and Soo predicts the charging to be dependent on particle size. The overall response also depends on the probability of impaction on the probe. This depends on the details of the instrument design. Evidently the IKOR AQM is well designed in this respect.

(6) Surface condition of the probe. Direct evidence was obtained that the condition of the surface of the probe of the IKOR instrument has a very important effect on the sensitivity. It was mentioned above that there is sometimes a sensitizing effect at the beginning of a run. It was also mentioned that the sensitivity changed during the measurements with Al dust. On July 2, 1974, a series of sensitivity runs was commenced with various substances, beginning with Al_2O_3. On July 30, upon returning to Al_2O_3, it was found that the sensitivity had increased by a factor of 5. The sensitivity for Al increased by almost a factor of 10 from July 10 to August 1.

Between runs with different substances, the apparatus was thoroughly cleaned. The probe was wiped clean with a dry cloth and replaced without touching with hands. The probe has a polished surface with less than a mirror finish. Some deep scratches can be seen. After a run, no dust could be seen on the forward surface of the probe even under close scrutiny. A light coating could be seen on the tail surface where the

diameter is reduced, probably owing to the slower stream velocity there. The reassembled apparatus was then run with clean air until no indication could be seen on the most sensitive scale.

Following the last measurement with Al dust which indicated increased sensitivity, the probe was cleaned following the instructions suggested by the manufacturer. This consisted of scrubbing the probe with an abrasive cleaner (Ajax) and a nylon brush. This was followed by a washing with detergent and a thorough rinse. Surprisingly, the result was a lower sensitivity for Al, almost to the first value obtained on July 10.

There would seem to be two possible explanations of the surface sensitizing effect. One would be a cleaning effect or exposure of fresh surface produced by the bombardment by particles, analogous to sand-blasting. The other possibility is that a coating of particles builds up on the surface, altering the charging conditions, since particles would then encounter other particles rather than the surface itself. At present the evidence is insufficient to allow one to decide between these two possible explanations. The author leans slightly towards the sand-blasting hypothesis.

(7) Sensitivity for various substances. A variety of materials were sampled to investigate the sensitivity of the IKOR instrument. The samples were chosen to cover a range of electrical resistivity, particle size, particle shape, and chemical composition. Many of the materials are found in stack effluent.

In Fig. 6, the measured sensitivities have been graphed versus the date of the measurement. Because of the change of sensitivity discussed above in (6), the first measurements of Al_2O_3 and Al have been arbitrarily excluded, i.e., the data from July 13-August 1 is used. When these data are arranged in the order of increasing sensitivity, it is found that they are naturally grouped according to electrical resistivity (see Table 2).

The value for fly ash is consistent with its principal constituents, SiO_2, Al_2O_3, and iron oxide. The resistivity of the glass beads was 10^7 ohm-cm, many orders of magnitude less than the other insulators. Molybdenum and tungsten have about 3.3 times higher resistivity than copper. Both were described as nonspherical particle samples and both are brittle metals. Thus, the area of contact for these particles may be smaller, thus reducing the charging. The other metals were described as spherical particle samples.

Figure 6. Sensitivity vs Date

Plot of sensitivity measurements for various substances vs the date of the measurement.

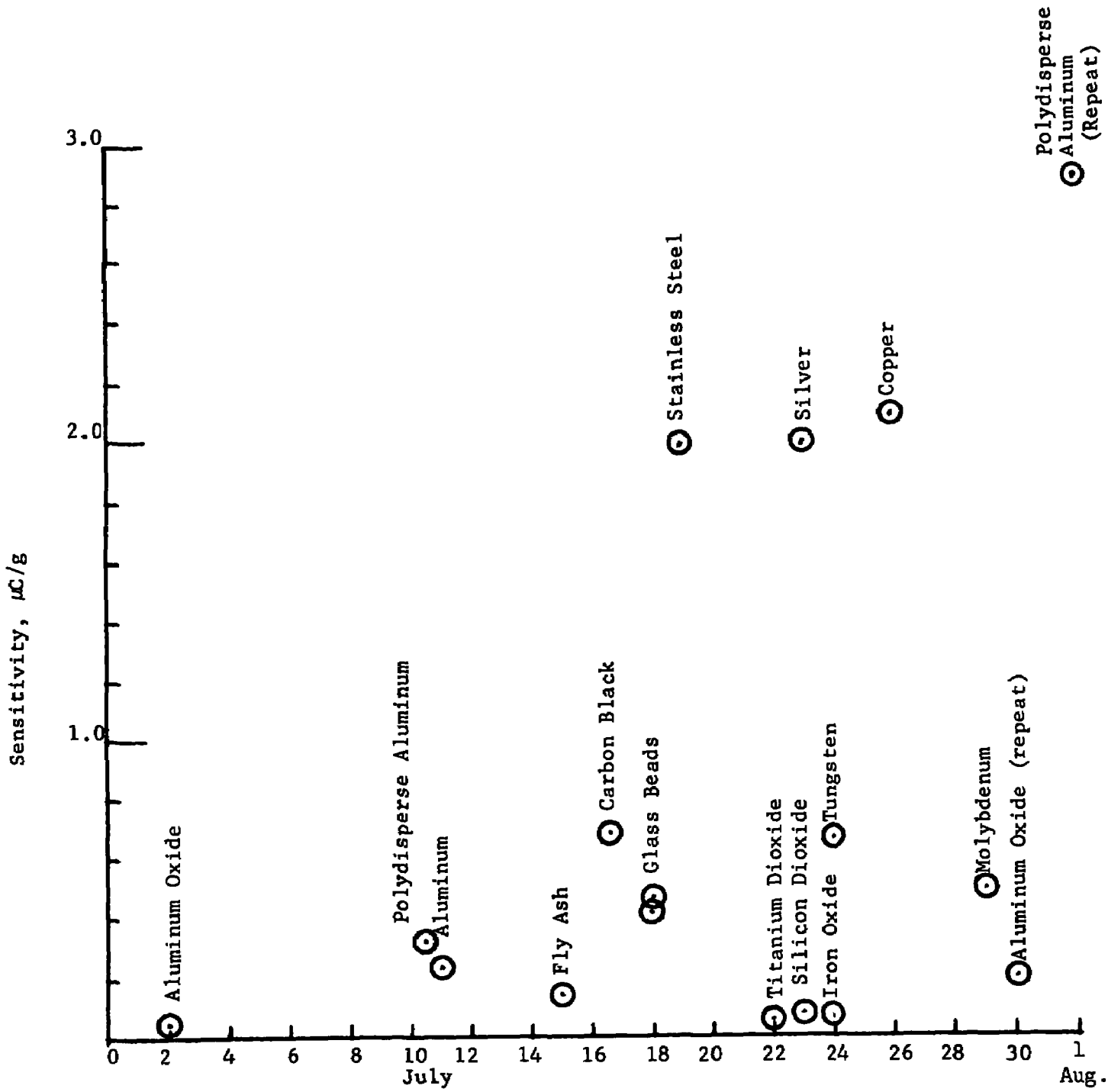

TABLE 2

SENSITIVITY MEASUREMENTS

	μC/g
I. Insulators	
Titanium dioxide	0.051 ± 0.004*
Red iron oxide	0.06 ± 0.01
Silicon dioxide	0.09 ± 0.02
Fly ash	0.148 ± 0.005
Aluminum oxide	0.20 ± 0.01
Glass beads 3-8μ	0.41 ± 0.03
Glass beads 0.5-3μ	0.46 ± 0.01
II. Intermediate conductors, semiconductors	
Molybdenum**	0.51 ± 0.04
Tungsten**	0.67 ± 0.13
Carbon black	0.68 ± 0.09
III. Metallic conductors	
Stainless steel	2.0 ± 0.5
Silver	2.0 ± 0.2
Copper	2.1 ± 0.2
Aluminum	2.9 ± 0.4

*standard deviation

**nonspherical metal particles

Carbon black is a semiconductor with a resistivity intermediate to insulators and metals.

The ranking according to electrical resistivity can be understood in terms of the characteristic time for charge transport. For good insulators, this time is large so that only the area in the vicinity of the contact is involved in charge transfer. The area is larger for semiconductors. For metallic conductors the charge can probably be transported from the entire particle. The oxide layers on the metal particles are presumably thin enough so that they can be penetrated either by electron tunnelling or by conduction with a small time constant. The dependence on electrical resistivity is further discussed from a theoretical viewpoint in Appendix I.

These data provide a basis for predicting the sensitivity for other substances, at least for dry dusts. It is likely that they will fall within the range spanned by the table. The wide range in the sensitivities (factor of 60) has some implications for the detection of components of a mixture. A small amount of an insulator would be difficult to detect in the presence of metals or even semiconductors. The charges per particle implied by the data of Table 2 are not unreasonable. However, we must regard the sensitivities listed as lower limits until the question of the probe surface is cleared up.

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SECTION VIII

APPENDIX

THEORY OF CONTACT CHARGING OF AEROSOL PARTICLES

Review of the Theory

The theory developed by Cheng and Soo[8,14] involves the transfer of charge during a collision between two elastic spheres of radii a_1 and a_2. During the impact there will be a mean area of contact A_{21} lasting for Δt, both of these quantities depending on the kinematics of the collision and the elastic properties of the materials. While the spheres are in contact, the current density will be proportional to the voltage difference between the two spheres. If the current is integrated over the period of impact, the charge transferred is obtained:

$$Q_{21} = \frac{C_1 C_2}{C_1 + C_2} (\varphi_2 - \varphi_1) \left[1 - e^{-\alpha \Delta t} \right]$$

where $C_{1,2}$ are the electrical capacities of the spheres

$\phi_{1,2}$ are the work functions of the materials

and

$$\alpha = A_{21} h_{21} \cdot \frac{C_1 + C_2}{C_1 C_2}$$

h_{21}, the charge transfer coefficient, is given by

$$h_{21} = \frac{\sigma_1 \sigma_2}{d_1' \sigma_2 + d_2' \sigma_1}$$

with $\sigma_{1,2}$ the electrical conductivities and $d_{1,2}'$ the charge transfer lengths.

The transfer lengths are defined by the equations

$$J_{21} = \frac{\sigma_1}{d_1'} (V_c - V_1) = \frac{\sigma_2}{d_2'} (V_2 - V_c)$$

where J_{21} is the current density and V_c is the potential at the point of contact. Thus d_1' and d_2' are the effective distances in the material over which current is transported.

Now Cheng and Soo derive the following expressions for A_{21} and Δt:

$$A_{21} = \frac{\pi\, a_1 a_2}{a_1 + a_2} \cdot \alpha_1$$

$$\Delta t = \frac{2.94}{\Delta u_{21} \cos \theta} \cdot \alpha_1$$

where $\alpha_1 = \left[\frac{15}{16} \pi \left(\frac{1 + r'}{2}\right) \left(\Delta u_{21}\right)^2 \cos \theta \left(\frac{m_1 m_2}{m_1 + m_2}\right) \left(\frac{a_1 + a_2}{a_1 a_2}\right)^{\frac{1}{2}} (k_1 + k_2)\right]^{\frac{2}{5}}$

(note: α_1 is distinct from α)

with r^* the ratio of the rebound speed to the incoming speed

Δu_{21} is the velocity of approach

Θ is the angle of impact

$m_{1,2}$ are the masses of the spheres

and $k_{1,2}$ are elastic parameters given by

$$k = \frac{1 - \nu^2}{\pi E}$$

and ν is Poisson's Ratio, E the modulus of elasticity.

Application to the Present Problem

It is instructive to evaluate numerically some of the above quantities using reasonable values of the parameters. Take sphere 1 to be the aerosol

particle and sphere 2 to be the probe. Then $a_1 << a_2$.

Using $k_1 = k_2 = 3.10^{-13}$ cm^2/dyne

$\Theta = 0$

$\overline{\rho}_1 = 8$g/cm^3 (density)

$a_1 = 10^{-5}$cm

$\Delta u_{21} = 1.5 \cdot 10^3$cm/s (typical for the IKOR AQM)

$r^* = 1$

we find that

$\alpha_1 = 3 \cdot 10^{-7}$cm

$\Delta t = 6 \cdot 10^{-10}$s

$A_{21} \cong \pi a_1 \alpha_1 = 1 \cdot 10^{-11}$cm^2

The short duration of the impact implies that the theory applies only to metals or good semiconductors ($\rho \lesssim 10^3$ ohm•cm) since only then will there be appreciable current flow. The area of contact has a diameter roughly 1/20 of the particle diameter.

Dependence of Q_{21} on the Parameters

The coefficient α depends on h_{21}. If we take $d_1' \cong a_1$ and $d_2' \cong a_2$, the remaining variable is σ_1, since σ_2 is fixed (metal probe).

When $\rho_1 \leqslant 10$ ohm•cm, $1-e^{-\alpha \Delta t} \cong 1$,

and $Q_{21} = 4\pi \varepsilon_0 a_1(\varphi_2 - \varphi_1)$

This equation results because the resistivity is small enough to allow complete charge transfer within the contact time Δt.

If $\rho_1 > 10$ ohm·cm, Q_{21} is reduced by the factor $(1-e^{-\alpha\Delta t})$. For example, when $\rho_1 = 10^3$ ohm·cm, $(1-e^{-\alpha\Delta t}) = 0.05$ and only 5% of the charge is transferred within Δt. The theory, therefore, applies only to metals or very good semiconductors.

Q_{21} is the charge transferred per particle. The total charge transferred to or from the probe is given by

$$Q_T = N_1 \; Q_{21}$$

where N_1, the number of aerosol particles, is given in terms of the total mass of particles, M_1, by

$$N_1 = \frac{M_1}{\frac{4}{3}\pi a_1^3 \bar{\rho}_1}$$

For $\rho_1 < 10$ ohm·cm, $Q_T \propto a_1^{-2}$. Thus the theory predicts that the total charge transferred to the probe is inversely proportional to the particle radius squared. Also, for $\rho_1 < 10$ ohm·cm, the charge transferred is predicted to be independent of the velocity of the particles relative to the probe.

Magnitude of the Charge Transferred

Consider metal particles with a radius of 0.05μm. Assume that $\varphi_2 - \varphi_1 \cong$ 1 volt. Then $Q_{21} = 10^{-17}$ C/particle. Furthermore, $Q_T = 2000$ μC/g.

This is three orders of magnitude greater than observed (See Table 2). The calculated charge could be brought into agreement with experiment by assuming that the particles had an effective resistivity of 5.10^4 ohm·cm which is in the semiconductor range.

The oxide layer on the particle surfaces could easily increase the effective resistance, not to mention contaminant layers on the surfaces of the particles as well as on the probe surface.

Summary of the Theory

While the theory is not quantitative, the model may still be useful in understanding the dependence of the charge on the physical parameters. However, the theory cannot as yet be regarded as verified by experiment.

The above theory applies only to metals or good semiconductors. For materials with $\rho > 10^3$ ohm·cm, which includes most semiconductors and all insulators the charge transferred is that already on the surface in the area of contact. In the absence of detailed knowledge of the physical processes involved, it is not possible to carry out the analysis for the charging of insulating materials.

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15. SUPPLEMENTARY NOTES

16. ABSTRACT

The theory of the charging of aerosol particles by contact electricity is reviewed, as well as the development of monitors for particulate matter based on this principle. Data on the performance of these monitors is scarce and sometimes contradictory. In the present work, laboratory tests were carried out with a variety of test dusts. The results show that the dynamic response of the contact electricity monitor tracks well with that of an optical detector. The total charge correlates well with the gravimetric mass. Humidity or precharge on the particles did not influence the detector. Some evidence was obtained showing that particle size does not have an important effect on the response, although there is a cutoff for very small particles. Results for the sensitivity of the instrument can be grouped according to the electrical resistivity of the material. It is found that the condition of the surface of the Inconel probe has a major effect on the sensitivity. Additional work is necessary on this aspect of the detector. The theory of Cheng and Soo for the charging of metal particles is discussed.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Monitors, Electrostatic charge, Dust, Particles, Fly ash, Konimeters, Sampling probes, Electrostatic measurement methods, Electric charge, Triboelectric charging	Pollution measurement methods, Continuous monitoring methods, Particulate sampling, Contact electricity	1407, 0704, 1302, 2003, 1711, 2102

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