ELECTRONIC METHODS OF COUNTING AEROSOL PARTICLES[1]

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Former methods of determining the particulate concentration in aerosols have been slow and laborious; but within the last few years two different rapid electronic methods of counting have been developed, the scope and range of which are discussed in this article. The first method depends upon light scattering, and the second upon electrostatic charging of the particles.

In the first method, developed by the authors and their collaborators, a fine stream of the aerosol, protected by a flowing sheath of pure air, passes through a spot under intense dark-field illumination and scatters flashes of light forward upon a photosensitive cell. Each particle about 0.6 micron or more in diameter causes an electrical pulse which is sufficiently large, after suitable amplification, to operate a mechanical counter. This apparatus will count particles weighing 5×10^{-13} g., at rates up to 1000 per minute.

A second type of electronic counter, developed by Arthur C. Guyton, depends upon the electrostatic charging of aerosol particles forced at high velocity through a fine jet to impinge upon a metallic collector. The electrical pulses imparted to the collector by particles of 2.5 microns or larger are amplified to operate a mechanical counter. Guyton found that the pulse amplitude is proportional to the square of the particulate diameter. This empirical fact we have explained in terms of the charging mechanism.

Both counters may be applied to determine the size of the aerosol particles by using suitable electrical discriminators. The electrostatic device may be very useful for larger particles, while improvements in the photoelectronic counter may make it applicable to particles even smaller than those counted at the present time.

INTRODUCTION

The determination of the particulate concentration of aerosols and other colloidal systems frequently is important for the colloidal chemist. Until

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For a fuller discussion of the design and operation of these counters than is presented here, see Chester T. O'Konski, "New Instrumental Methods in Aerosol Studies," Ph.D. Dissertation, Northwestern University, August, 1948.

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recently, the only available techniques were slow and tedious, but two different electronic methods have been developed within the last few years to reduce the time and increase the convenience of these measurements.

One of the earliest methods of counting aerosol particles involves the collection of a sample upon a suitable slide and a tedious visual count under the microscope. Several types of collector are available. One of these is an impactor, in which the aerosol passes through a fine jet at high speed and strikes a collecting plate to which it adheres. Efficient impactors have been described by May (19) and Sonkin (26). Another method of collecting aerosol particles depends upon their motion in the direction of a thermal gradient. This method was discovered by Aitken (1) and the theory of it has been discussed by Cawood (5). Recently the phenomenon has been studied by Rosenblatt and La Mer (24), who determined the velocity of thermal repulsion of individual small spherical particles of tricresyl phosphate. Comparison with the equations developed for radiometer theory indicated that thermal repulsion of aerosol particles is a radiometer phenomenon. This motion causes the dust-free space surrounding heated objects, investigated by Watson (29) and found to be clearly defined, extending to a distance approximately proportional to the square root of the thermal difference. Green and Watson (9) have described a dust sampler in which a wire is strung halfway between two closely spaced parallel cover glasses. The wire is heated so that the dust-free space is slightly greater than the distance between wire and cover glasses. When air is drawn across the wire, the dust precipitates just before it passes the wire. In another form of thermal precipitator, the sample of air is passed slowly between two closely spaced parallel plates differing in temperature by about 75°C., and the particles are deposited on the cooler one. The thermal precipitator is almost 100 per cent efficient.

A second counting method employs an ultramicroscope cell. The Zsigmondy slit ultramicroscope, in which the depth of the field is limited by the light beam, may give counts which are too high, because light scattered by the aerosol illuminates particles outside the desired volume. The effect increases with particle size. To eliminate this difficulty, Whytlaw-Gray and his collaborators (21, 22) developed a special cell of the cardiod type, in which the depth of field is limited by the walls of the cell. The apparatus is shown in figure 1. Light from an arc lamp G passes through a water-cooled slit E_1 and is focussed by means of a lens F upon a second slit E_3, where it passes through a microscope objective D. The light passing through the final slit E_2 provides uniform intense illumination of the space 0.1 mm. in depth between the parallel glass plates comprising the counting cell A. The viewing microscope C has a depth of focus greater than 0.1 mm., so that the depth of the cell limits that of the sample, while the area is limited by suitable stops in the eyepiece, chosen to give an average field of two or three particles. The inlet tube B is grounded and lined with moistened blotting paper to prevent loss of particles by electrification. The outlet tube is connected to suction through the stopcock H rotated by a wheel K connected to a motor. Thus successive samples of the aerosol are drawn in and stopped

momentarily under the microscope. Only about a minute is required to count sixty fields, which contain enough particles to give an adequate statistical average. Since the field of view is 3×10^{-4} cc. or less, this counter is limited to aerosols containing about ten million particles per liter.

The walls of the cell are coated with a high-boiling liquid like *m*-xylene or paraffin oil, which wets glass and also wets any aerosol particles which strike the walls, rendering them invisible. Liquids as volatile as *m*-xylene also have another important effect. They condense upon the aerosol particles and brighten them appreciably. Whytlaw-Gray estimates that particles originally as small as 0.05 μ in radius may be loaded sufficiently to be counted, although otherwise they would be missed if they were below about 0.1 μ.

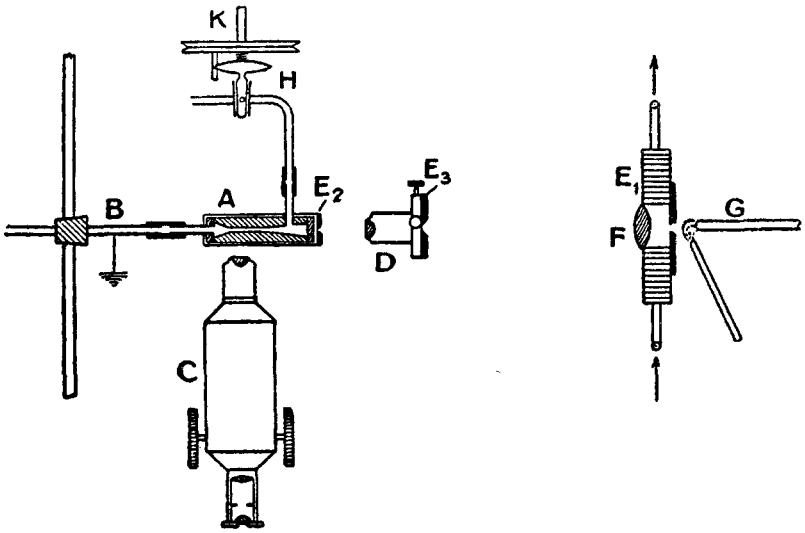

FIG. 1. Ultramicroscope counting apparatus of Whytlaw-Gray and his collaborators (from *Smoke* by R. Whytlaw-Gray and H. S. Patterson, Edward Arnold and Company, London 1932).

An extension of the condensation method of counting has been made by Green (8), who adapted the cloud chamber of Wilson (30) to this purpose. A small expansion chamber is connected to a standard Zsigmondy slit ultramicroscope cell and a cycle of operations is controlled by an electric motor. A sample of the aerosol is drawn into the cell, which is lined with damp filter paper. Then the valve is closed, the piston in the expansion cylinder suddenly drops, and an almost adiabatic expansion deposits moisture on the aerosol particles. The cell is photographed through the microscope, and then flushed out for another sample. A complete cycle takes only 2 sec., so that the counts are rapid. The usual difficulty with the Zsigmondy system is avoided in this case, because condensation builds up all the particles to nearly the same size; hence the volume under direct illumination is easily distinguished.

A third counting method employs bacteriological techniques. If an aerosol

of single viable microörganisms is set up, they may be collected from a known volume of aerosol by means of a suitable cotton plug filter, or an impinger jet operating under water, as developed at Camp Detrick, and described by Rosebury (23). If the organisms are washed from the filter or collected in the impinger and deposited on agar, a colony count will give the number of particles. This process is limited in application and is very slow, ordinarily requiring about a day for incubation. It also involves the uncertainty of viability.

A PHOTOELECTRONIC PARTICLE COUNTER

During World War II gas-mask filters were improved to such an extent that they would remove all but a very small fraction of even the most penetrating

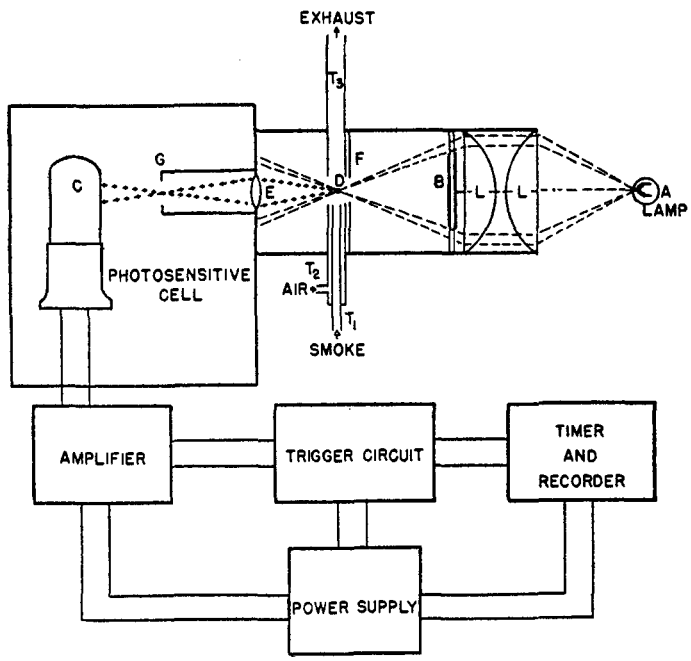

FIG. 2. Schematic diagram of photoelectronic particle counter

aerosol particles (about 0.3 μ in diameter). The penetration of somewhat coarser test aerosols might be only a few particles per minute. In the summer of 1944 we undertook to develop an automatic instrument to count the individual particles in a rapidly moving aerosol stream and thus provide quantitative tests for the best available filters. By June, 1945, we had developed a photoelectronic apparatus which would count particles of dioctyl phthalate (DOP) down to about 0.6 μ diameter.

The general arrangement of the apparatus is shown in figure 2, while further details are given elsewhere (11). The lamp A is a 50-cp. automobile headlight, powered by a heavy-duty 6-v. storage battery. Inside the black-walled cell at D, the aspheric condenser lenses LL form a 4-mm. image of the filament of

A. Two circular black disks B cut out the center of the converging cone of light outlined by the dashed lines, thus providing dark-field illumination at D. The aerosol stream is introduced through the tube T_1, surrounded by a sheath of air flowing from T_2 at nearly the same linear rate. Thus each particle passes once through the intensely illuminated space at D, and is exhausted through T_3 without any chance of recirculating through the cell. Each particle scatters a flash of light, a part of which is collected by the lens E and focussed upon the photosensitive cell C, as indicated by the dotted lines. The electrical pulse from C is magnified up to 200,000 times in the amplifier, and then passed to a thyratron trigger circuit which actuates a mechanical recorder.

The optical system is like that developed in 1941–42 by La Mer, Sinclair, and Hochberg (16) for their study of aerosols carried out at Columbia University under contract with the National Defense Research Committee. It utilizes the light scattered in the near-forward direction, from about 5° to 30°. For particles with radii comparable to the wave length, a greater fraction of the light is scattered in this direction than through the same solid angle at 90°, as predicted by Mie (20) and verified by La Mer and Sinclair (16).

The blackened baffles F and G were designed to reduce background light in the system, and the black disk B inside the cell was found to prevent reflection from the glass plate of stray light which otherwise would find its way to the photosensitive cell C. The unit was built rigidly, and the lamp was held in a heavy clamp to prevent vibrations which vary the background light and may cause "optical microphonics" and spurious counts.

Two photosensitive cells were found useful in the counter. The first was the Thalofide cell, containing a special photoconductive surface of thallous sulfide, developed by Cashman (4) and his associates, working under a contract with the Office of Scientific Research and Development in the Physics Department of Northwestern University. High-resistance cells, with values of 5 megohms or more under background illumination, proved more sensitive than those of a lower resistance, which matched the 2.5-megohm input impedance of the amplifier more closely.

Later we found that a 931-A photomultiplier tube, operated at 30–50 v. per stage, could be used satisfactorily, after the background light had been reduced as much as possible. In this instrument, the stray light current in the 931-A tube was about ten times that due to an 0.8 μ DOP particle, while in the Thalofide cell the ratio was about 800 to 1, probably owing to long-wave stray light, to which this cell is more sensitive.

The electronic circuits of the counter required careful design and construction. The power supply was used to operate everything except the phototube, which was connected to a 22.5-v. dry battery, and the amplifier tube heaters, which were supplied by a 6-v. storage battery. The high-voltage supply was fed through a full-wave rectifier to two independent two-stage choke-input filters, one of which supplied the initial 1603 amplifier tube, chosen for its low microphonics, while the other supplied the two 6SJ7 amplifiers, trigger circuit, and recorder. This arrangement adequately reduced the rectifier ripple and prevented feedback from the second 6SJ7 to the 1603 tube.

The amplifier employed resistance–capacitance coupling between tubes, with wire-wound 5-megohm resistors in the input circuit from the phototube to reduce the noise, and the photocell and 1603-tube circuits enclosed in a desiccated brass box to decrease the possibility of excessive leakage currents and act as an electrostatic shield. The grid leads of the 6SJ7 tubes were made short, and shielded with a woven wire sleeve, soldered to the grounded chassis.

The biasing voltages in all three stages were furnished by cathode resistors, shunted by large condensers to extend the amplifier range down to about 10 cycles per second. This passed the low-frequency components of the non-sinusoidal photocell pulse and prevented pulse multiplication. The input resistors of the 6SJ7 tubes were shunted with suitable condensers to attenuate the high frequencies and improve the stability of the amplifier. This gives a response curve which is essentially flat from about 10 to 5000 cycles and falls to about one-half at 10,000 cycles. The gain of the amplifier was adjusted by means of potentiometers in the cathode circuits of the 6SJ7 tubes, which varied the negative feedback.

The trigger circuit employed a type-885 thyratron tube with a self-quenching choke-condenser plate circuit. The grid bias was controlled by means of a discriminator potentiometer, calibrated in volts with the zero reading at the firing point of the tube. The setting of the discriminator then determined the size of the minimum pulse which would fire the tube.

The time-recorder circuit contained two mechanical counters, which could be used in duplicate experiments, and a standard electric timing clock. Pressing a single button starts the timer and connects the trigger circuit to one of the counters. When the counter hand moves around to 100, it closes a contact in an automatic cut-off circuit which simultaneously stops both timer and counter. The operator then reads the time for 100 counts, which gives the same statistical uncertainty in each experiment. If a larger number of counts is desired, the automatic circuit is turned off, and then each revolution of the primary counter actuates an auxiliary *hundreds-counter*. At any time the automatic circuit can be turned on again to stop the instrument at the end of the next hundred counts.

In operation, the cell first is swept out with carefully filtered air, introduced through the outer tube at 3 l. per minute, until the dust count has fallen to one or less per minute. Two sheets of the best grade service-canister paper, or two complete pleated filters fastened into a canister, were found to give dust-free air *after they have been swept out for several hours or preferably overnight.*

In order to avoid using a flowmeter in the aerosol line, its flow is determined by difference between the effluent flow and the sheath-air flow, each read on calibrated flow meters. The standard rates are 4 and 3 l. per minute, making 1 l. per minute for the aerosol flow.

Careful tests of the counter were made to determine the size of particle and rate of counting for which it was applicable. Since the light scattering of an aerosol particle decreases rapidly with particle size, being proportional to the sixth power of the radius for very small particles, there will be a limit below which no particles will be counted. However, for dilute aerosols in which

the particles are appreciably above this limit, each should register if the whole cross section of the stream at the focus is uniformly illuminated. As the concentration of the aerosol is increased some particles may be missed, owing to coincidences in the field of view and limitations of the photoelectronic circuits and mechanical counters.

Two methods were used to determine the upper limit of the original form of the counter. Both are described in the original publication (11), but only one will be treated here. The test aerosol was made up either of homogeneous spherical DOP particles of about 0.8 μ diameter, from a generator of the type developed by La Mer and Sinclair (15) in their OSRD work, or of spores of *Bacillus globigii* (BG), which are ellipsoidal, with a major axis averaging 1.2 μ and minor axes of about 0.8 μ, as shown by electron micrographs. The flow line using the BG aerosol is shown in figure 3. An aqueous suspension of 0.1 to 20 billion (10^9) BG spores per milliliter was sprayed through a nebulizer[3] into a

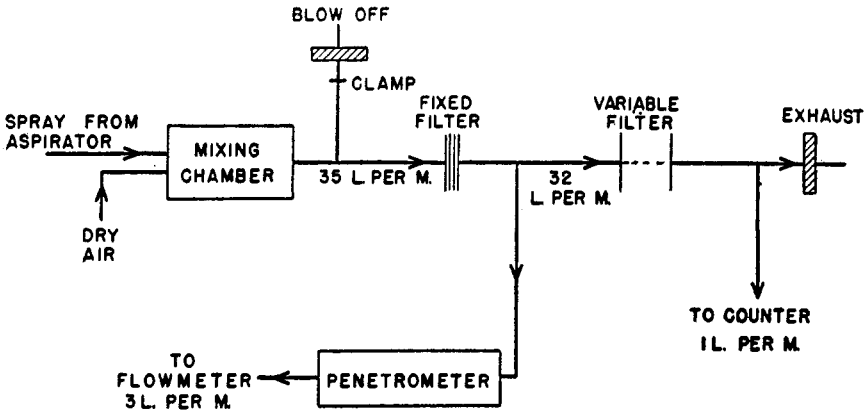

FIG. 3. Flow line for testing counter (from Journal of the American Chemical Society **69**, 2422 (1947)).

chamber where it was mixed with enough dry air to form an aerosol of dry spores. This apparatus was kindly furnished by the group at Camp Detrick, Frederick, Maryland, with which we coöperated. They had found that this arrangement gave particles almost all of which were single spores. The number of viable spores in the suspension was determined at Camp Detrick. Despite the purification of the original material, there were always appreciable numbers of foreign particles from the nutrient medium.

The aerosols were sent to the counter at a pressure somewhat above atmospheric, to avoid contamination with laboratory air, which often contains many thousands of particles per liter. The flow was adjusted by the variable blow-off to 35 l. per minute. The aerosol concentration was reduced by means of a fixed filter to a constant value, which was checked within 5 or 10 per cent by the reading of scattered light in the sensitive photoelectric photometer (penetrom-

[3] Made by the Vapo-Nefrin Company, Upper Darby, Pennsylvania.

eter) developed by our group under an OSRD contract (10, 12). The dilute aerosol then was passed through a variable filter, made of sheets of paper, the transmission of each of which had been measured with the penetrometer, using a higher concentration of the same aerosol. A sample of the effluent aerosol was passed through the counter. From the observed counts per liter, c_2, and the known fractional transmission of the variable filter, P, the influent concentration was calculated as:

$$c_1 = c_2/P$$

The value of c_1 calculated thus was found to increase to a constant value c_0 at low counting rates. This value, divided by that at any higher rate, gives the corresponding count-rate factor, by which the observed count per minute should be multiplied to be consistent with the low-rate value. Figure 4 shows typical results with one of our early counters, obtained with a BG aerosol at Camp Detrick by Cohen and Voelker, on three successive days. Counts up to 1000

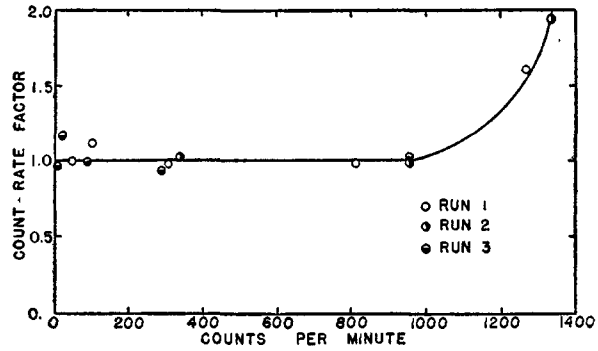

FIG. 4. Count-rate curve for BG aerosol, showing results obtained by Sgt. A. Cohen and Cpl. W. Voelker (from Journal of the American Chemical Society **69**, 2422 (1947)).

per minute were made with a count-rate factor of 1, but the correction increases greatly at higher rates. The practical range of the instrument is 3–1200 counts per minute.

The count-rate experiments did not determine whether all, or only a constant fraction, of the particles above a critical size are counted at low rates, and this is a difficult question to answer with certainty. Early experiments were carried out at Camp Detrick, in which the counts in our apparatus were compared with a colony count of viable spores collected on a cotton impinger, washed onto an agar plate, and incubated for 24 hr. These experiments showed more counts than viable spores, suggesting the presence of non-viable spores and extraneous matter from the cultural medium.

Later a series of experiments was carried out in which the concentration of viable spores reaching the counter was estimated from the concentration of the original aqueous suspension and quantitative dilution of the aerosol. The nebulizer was run with careful control of air pressure and flow rate. The BG

suspension was diluted with water in five steps, to a ratio of 1/16, in order to vary the concentration of the aerosol considerably without changing the relative humidity, which might affect the scattering from the particles. The dilution steps were found to be self-consistent within ±8 per cent, as determined by the light scattered by the raw aerosols, measured in the penetrometer. The flow line was that shown in figure 3, except that the fixed filter was removed, and a three-stage diluting system was substituted for the variable filter, to avoid the selective removal of larger particles in a filter. The dilution factor of each stage was measured individually with the penetrometer, and the over-all transmission factor for the three stages used in these tests was calculated to be 0.030 per cent of the raw aerosol. Thus, the viable spores per liter in the diluted aerosol could be calculated, assuming no loss in the atomizer, and could be compared with the observed counts. Two counters were used. One, containing a Thalofide cell, was constructed in our Laboratory by Leo E. Farr for the use of Dr. Ronald M. Ferry of the Harvard Medical School. The other contained a 931-A photomultiplier. In these experiments, the count-rate factor increased from unity at about 500 counts per minute to 1.3 at 1000. At low rates, the average value of counts per viable spore in the atomized suspension was 0.3 in the Thalofide counter, and 0.5 in the photomultiplier counter. Later the lenses were adjusted to focus more red light on the aerosol stream, and values of 0.6 count per viable spore were obtained with the Thalofide cell. At first (11) we interpreted these results as due to lack of homogeneity in the 4-mm. image of the lamp filament, providing the dark-field illumination of the smoke stream, so that only a fraction of the particles were illuminated and counted. However, Dr. Ferry has pointed out to us recently that this difference may be due to the fact that a considerable portion of the original aqueous suspension fails to appear as aerosol particles in the output from the nebulizer.

Dr. Ferry has just completed a series of careful comparisons of the counts registered by our instrument and numbers calculated from the counts obtained in a two-stage impinger. He used both BG and *Serratia marcescens* (SM). The latter is slightly smaller, occurring in ellipsoidal or often nearly spherical form, 0.5 by 0.5 to 1.0 μ (3). In his counter, he found it necessary to use an 8-v. battery, giving 7 v. across the lamp bulb, in order to increase its intensity. He then found excellent agreement between the two methods, the ratio of counts to impinged specimens being 1.1 ± 0.1 for BG and slightly less for the smaller SM. These results have been given in more detail elsewhere in this Symposium (7).

SCOPE AND LIMITATIONS OF THE PHOTOELECTRONIC PARTICLE COUNTER

The photoelectronic counter was designed as a supersensitive penetration meter for testing gas-mask filters against aerosol particles about 1 micron in diameter. It is sensitive to single oil-smoke particles of 0.6 μ diameter, weighing less than 10^{-12} g. Spurious counts due to electrical noise in the system are kept below one per minute; hence when the counter is used at the present sampling rate of 1 l. per minute with a test aerosol containing 10^8 particles per liter, it

will respond to penetrations of 1 micropercent, providing spurious counts due to dust are eliminated by blowing out the filter before the test. The upper range of the original counter is 1200 counts per minute; hence with the test aerosol of 10^8 particles per liter, the practical range of this instrument is about 3–1200 micropercent.

The lower limit of sensitivity is determined by the concentration of the test aerosol, the rate of sampling, i.e., the flow rate through the counter, the statistical errors which become significant when small numbers of particles are counted, and the dust particles blown from the filter under test. Since increasing the concentration of the test smoke increases clogging or aging effects in filters, this is not a satisfactory way to attain greater sensitivity for the very efficient filters. Increasing the flow rate through the counter also has its attendant problems. This cannot be done by increasing the cross section of the sampling tube greatly, unless a larger light source is used, since all particles must pass through the image of the light source in order to register. Another alternative, increasing the linear rate of flow, shortens the duration of the pulse of light upon the photosensitive cell. This is a disadvantage with both types of cell. Tests showed that the output signal voltage of the photoconductive Thalofide cell decreases approximately in proportion to the decreased length of the pulses of several milliseconds duration. Since the pulses at the present rate are only two to three times the largest fluctuations due to random electrical noise, the linear flow rate cannot be increased appreciably with this cell. The output signal voltage of the photoemissive electron-multiplier tube does not decrease with decreasing pulse length, but the amplifier soon becomes the limiting factor. For shorter pulses, the band width of the amplifier circuits must be increased in proportion to the increase in flow rate in order to maintain the same signal sensitivity. Since the noise increases as the square root of the band width, the signal-to-noise ratio will decrease as the square root of the flow rate. This again precludes use of much higher flow rates with the original counter.

Assuming that larger particles are being counted or that the signal-to-noise ratio can be increased in some other way, the above considerations show that a large increase in linear flow rate through the $\frac{1}{8}$ in. sampling tube of the counter might be tolerated in a counter using the electron-multiplier phototube. Another factor then enters,—namely, the laminar flow of the aerosol stream through the tubes in the smoke cell. At a certain limit turbulence might set in to make a further increase in flow rate impractical.

The upper limit on the counting rate of the present instrument is largely set by the resolving time of the thyratron counter circuit, which is about 10 millisec. This limitation can be overcome by using a scaling circuit with a shorter resolving time. However, after about a threefold increase in counting rate is achieved in this way, further increase is limited by the larger number of coincidences, caused by the presence of two or more particles in the field of view at the same time. According to statistical theory, the per cent of coincidences of randomly distributed aerosol particles in a given illuminated volume of sample goes up linearly with the concentration. This volume coincidence effect is independent

of the flow rate through the cell when considered as an upper limit on the *concentration* of smokes which can be counted. Decreasing the cross-sectional area of the sampling tube will raise this limit proportionally without introducing further difficulties. However, decreasing the length of the flow path over which the particle is illuminated and viewed will have the same effect as increasing the linear flow rate, in causing a decrease in signal-to-noise ratio, as explained in the preceding paragraph.

The lower limit of particle size is likewise determined by the signal-to-noise ratio in the instrument. If this can be improved, smaller particles can be detected. The signal amplitude can be increased by using a more intense light source or by improving the optical system to collect and focus sharply a greater amount of light on the particle and to collect more of the light scattered by the particle. The amount of stray light in the optical system is important for two reasons: (*1*) it determines the output current and noise level in the electron-multiplier phototube, and (*2*) if it is as large as in the original instrument, relatively small variations in the lamp and vibrations of the system may produce spurious pulses. For both reasons, the stray light in the optical system should be decreased as much as possible, preferably without decreasing the signal amplitude for smaller particles.

A new optical system, under consideration for some time, has been constructed and is now being tested. Preliminary results show that the stray light in the system has been reduced so much that the background-light current in the photomultiplier tube is about the value of the dark current. In addition, a differential pulse amplitude selector circuit has been designed and constructed. This will pass only the pulses in a given range of amplitude, and will allow automatic discrimination against the larger pulses from dust particles. Tests are being continued and the improvements and extended applicability of the apparatus are being evaluated according to the considerations discussed above.

APPLICATIONS OF PHOTOELECTRONIC PARTICLE COUNTING

In addition to its original use as a penetration meter to test the best gas-mask filters which can be produced, the photoelectronic counter has many possible applications in other fields.

The counter built for Dr. Ferry is now being studied by his group at Harvard University to evaluate its usefulness in counting bacterial aerosols (7). Other bacteriological applications may be made. It is very convenient in testing for leaks which might bring contamination to a system where sterile conditions are desired. A trace of unfiltered room air will instantly register on the counter, while the accompanying bacterial contamination would require many hours to show up in the usual plate counts. The instrument can be used to count various particles suspended in liquids either by adapting the optical system to liquids, or by atomizing the liquid suspension and counting the particles as aerosols. Particles which differ appreciably in size could be distinguished and counted separately by several settings of the *discriminator*.

The improved counter now under study contains a pulse-amplitude selector

circuit which will permit the counting of pulses within a chosen range of voltage, and may be applied to a rapid determination of particle-size distribution, at least in spherical liquid aerosol particles, where the light scattering can be calculated by means of the Mie theory (16, 20).

AN ELECTROSTATIC PARTICLE COUNTER

Shortly after the development of our photoelectronic particle counter, Guyton (13) developed at Camp Detrick an electrostatic apparatus which can count aerosol particles with a diameter of 3 μ or more. His detector unit consisted of a tube, presumably of glass, tapering at an angle of 45° to a hole 0.8 mm. in diameter, with a collector directly in front of the orifice. This collector was either a metal plate or a copper wire, 0.4 mm. in diameter. The latter was found to be more sensitive and was used in most of Guyton's work. By applying a vacuum beyond the tapered orifice, aerosol particles are drawn through the tube to impinge upon the copper wire, which is connected to a high-frequency amplifier with resistance and choke coupling to give a response above 10,000 cycles per second and an amplification of about 100,000. Individual particles striking the wire produce impulses which are amplified in the first three stages and rectified by the grid of the fourth stage of amplification. This serves to lengthen the pulses from about 50 to 500 microsec., determined by the *RC* value of the resistance-capacitance coupled grid circuit. The pulse then fires a thyratron tube, whose discharge in turn is amplified through a class C triode which energizes a mechanical counter. This instrument was used at counting rates up to about 1800 per minute, or 300 per liter, since the flow rate through the orifice was about 6 l. per minute.

Guyton stated that the various solid aerosol particles he tested all produced on the wire positive pulses which increased in voltage with the square of the particle diameter, or the surface area of the particle. The polarity and size of the pulses he found to be independent of the potential at which the detector wire was maintained. Water droplets behaved very differently. They gave positive pulses with the wire at ground potential, no pulses with the wire at plus 22 v., and negative pulses increasing linearly with the wire potential beyond this value. He concluded that the "surface voltage on water particles is plus 22 volts" and that the "natural electrostatic charge" of water particles can be reversed by an applied voltage.

Guyton was doubtful about the mechanism by which the electrostatic charge is produced, but concluded from the linear increase of the pulse size with the surface area of the particle that the charge may be produced by friction between the particle and turbulent air in the nozzle or between the particle and the wire. He indicated that the pulse duration of 50 microsec. could be decreased to 10 microsec. or less by decreasing the *RC* value in the electrical connections to the hookup wire, and that the theoretical upper limit of the counting rate would therefore be about 10^5 per second. He also concluded that the lower limit of particle size depended upon the particular amplifier he used, which could record only pulses of 60 microvolts or more, and that the design of the amplifier could

be improved "to detect impulses of a few microvolts which correspond to particles of 0.3 to 1 micron in size."

The electrostatic charging of dielectrics, or so-called frictional electricity, has been observed and studied for a long time. Most of the studies have been empirical. Perhaps the first utilization of the electrostatic charging of small particles was that by Armstrong (2), who constructed a steam generator and jet which he used as an electrostatic generator as early as 1843. Subsequently Michael Faraday (6) undertook a study of the mechanism by which the charge was produced. His ingenious experimentation with limited equipment led to many interesting observations and speculations, including the notion that the charging was due to a rubbing or frictional process. Since his time, a great deal of work has been done on electrostatic properties and a review of the phenomena in general by Loeb (17) shows "a most unsatisfactory array of discordant, often non-reproducible and confusing results, whose theoretical interpretation in some cases is quite hopeless."

Whatever the charging process, there is evidently a theoretical upper limit to the amount of charge which can exist on a small particle. If the charge on an aerosol particle is increased, a point is reached where the potential gradient at the surface becomes sufficient to produce breakdown in the surrounding air, which would tend to discharge the particle and prevent further accumulation of electricity. Hence for a spherical particle of radius r the maximum possible charge q can be calculated by making use of the relation $q = CV$, where C is the capacity of the particle, which in electrostatic units equals the radius in centimeters, and V is the potential in volts. Taking the derivative we get for the change of potential with drop size the expression: $dV/dr = -q/r^2$. This may be shown to be identical with the expression for the potential gradient at the surface, which can be set equal to the breakdown potential gradient in air, about 30,000 v. per centimeter or 100 E.S.U. per centimeter. This gives, for the maximum charge, a value of $100\ r^2$ E.S.U., where r is in centimeters, or $3.3 \times 10^{-8} r^2$ coulomb. Hence we observe that the maximum permissible charge on the particle is proportional to the square of the radius or the surface area of the particle. This relation has been used by Vollrath (28) in a description of his high-voltage electrostatic generator, employing finely divided diatomaceous earth blown through copper tubes which become highly charged in the process. Airplanes may build up a high charge as they fly through dust, clouds, rain, or snow, and this causes corona discharges—St. Elmo's fire—which interfere with radio reception. Hall (14) studied the possibility of discharging airplanes by blowing diatomaceous earth through brass tubes filled with metal turnings. He evidently used the same equation for the maximum charge per particle, to show that the maximum amount of charge which could be transferred by 1 g. of finely divided material varies inversely with the radius of the particles. For non-spherical surfaces the same considerations apply and Silsbee (25) computes a limiting charge density of 2.65×10^{-9} coulomb per square centimeter, which is in agreement with the result of our calculations.

SUGGESTED MECHANISM OF THE ELECTROSTATIC COUNTER

Although several mechanisms can produce electrostatic charging effects, as has been indicated by Loeb (17), the authors prefer to explain the action of Guyton's electrostatic counter by means of the mechanism suggested by Vollrath (28) in accord with the views of Helmholtz on so-called frictional electricity. This postulates that a double layer of charge is set up at the surface of contact between the particle and the wire. Because of the small distance between the surfaces (10^{-8} to 10^{-7} cm.) at contact, the effective capacitance is high and a low contact potential can result in the transfer of a substantial quantity of charge according to the relation $q = CV$. After a charge transfer has occurred, the surfaces become separated. As the particle moves away from the wire, its capacity with respect to its surroundings rapidly decreases with distance from the wire. The charge q, trapped on the particle, will cause the potential V to rise accordingly. If the potential gradient at the surface exceeds the breakdown potential of air, ionization of the surrounding air will occur and the excess charge will be lost. This most probably would occur when the particle is still very close to the wire, so it appears reasonable that the excess might be largely returned to the wire, resulting in a net charge transfer equal to the maximum permissible charge on the particle. In this event, the voltage of the pulses on the wire can be calculated directly from the size of the particle and the capacity of the input circuit to ground:

$$V_p = q_{max}/C_i = 3.3 \times 10^{-8} r^2/C_i$$

where r is the radius of the particle in centimeters, C_i is the input capacity in farads, and V_p is the pulse amplitude in volts. Converting to microns, micromicrofarads, and microvolts, we obtain the relation $v_p = 330r^2/c_i$.

INTERPRETATION OF GUYTON'S RESULTS

The preceding equation may explain why Guyton found the wire collector more sensitive than the metal plate, which would be expected to have a higher capacity to ground, increasing the value of c_i.

The empirical relation which Guyton found for particle diameter d in microns as a function of pulse size v_p in microvolts was:

$$d = v_p^{1/2}/3.04$$

Comparing this with our equation we obtain a value of 9 micromicrofarads for the capacity of the input circuit. The rated input capacity of the first tube in Guyton's amplifier, a 1603, is 4.6 micromicrofarads. The difference between these two values could readily be due to the distributed capacity of the detector wire, the blocking condenser, and the grid leads.

The capacity of the input circuit can also be estimated from the duration of the pulses, which Guyton placed at about 50 microsec., and the resistance to ground of the equivalent input circuit, 1.5 megohms. Setting RC equal to the pulse duration, this yields the approximate result of 33 micromicrofarads which also might be possible, depending on the physical arrangement of the grid blocking condenser and the leads in the amplifier. However, this gives in our equation

$v_p = 10r^2$. This is not consistent with Guyton's empirical relation, which can be written $v_p = 37r^2$. The visual estimation of a short pulse on an oscilloscope could be considerably in error, and could yield a result far from the true RC value of the input circuit. Hence further investigations are required to establish whether or not the assumptions introduced above are valid.

To account for a greater charge transfer than expected on the basis of the above postulates, two points merit further investigation: (*1*) the chance that a particle may be charged before striking the wire and (*2*) the possibility that small particles may carry away a greater charge than they could be expected to retain under normal conditions. The first effect might double the charge transfer. If, for example, before a particle came in contact with the wire, it were charged positively to the maximum value given by $q = 3.3 \times 10^{-8}r^2$, perhaps by contact with the orifice wall, it could effect a charge transfer of $6.6 \times 10^{-8}r^2$, providing it left the wire with the maximum negative charge. In connection with the second effect, the processes involving ionization in air, corona discharges, etc., are very complex and subject to statistical variations which are often difficult to interpret, as pointed out by Loeb and Kip (18). The electrostatic detector may prove a useful tool for investigations of these phenomena.

SCOPE AND APPLICABILITY OF THE ELECTROSTATIC COUNTER

In its original form the electrostatic detector can count only particles with a diameter of about 3 microns or more. The above discussion clearly indicates that the lower limit of sensitivity might be extended to somewhat smaller particles, perhaps 1 micron or more in diameter, by improved design of the amplifier. A value of about 5 micromicrofarads for the input capacity is feasible with existing electronic equipment.

Another important consideration must be thoroughly investigated, namely, the input noise of the amplifier circuit. The root-mean-square value of the thermal noise in an amplifier is given (27) by the equation:

$$\sqrt{\overline{E_t^2}} = \sqrt{1.64 \times 10^{-20} R\Delta f}$$

where R is the input resistance in ohms, and Δf is the band width in cycles per second. Thus the RMS thermal noise alone in an amplifier with a band width of 10,000 cycles connected across a 1.5-megohm load is about 16 microvolts. Random peak fluctuations can easily exceed two or three times this value. It therefore appears that little can be gained by increasing the amplification of the amplifier used in the electrostatic counter because, above a certain level, random noise pulses will give spurious counts. Any attempts to increase the counting rate to the proposed value of 10^5 per second will require an amplifier of greater band width and will therefore decrease the sensitivity of the unit.

To investigate the mechanism and the possible improvements in the electrostatic counter, and to compare it with the photoelectronic counter, several detector units and the necessary amplifier and counter circuits have been set up in our laboratory. For larger particles this type of counter might prove useful in discriminating between particles of different chemical compositions, and a

correlation between results obtained with it and our photoelectronic counter might yield additional information on the aerosol system under test.

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