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SRI Technical Report No. 19

PNEUMATIC DISSEMINATION OF DRY POWDERS

Special Report

by

PAUL L. MAGILL
DON PON

July 1967

DEPARTMENT OF THE ARMY
EDGEWOOD ARSENAL
Research Laboratories
Physical Research Laboratory
Edgewood Arsenal, Maryland 21010

Contract DA-18-035-AMC-122(A)

METRONICS ASSOCIATES, INC.
3201 Porter Drive
Stanford Industrial Park
Palo Alto, California

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Contract DA-18-035-AMC-122(A)
Task 1B522301A08101
SRI Project PAU-4900

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FOREWORD

The work described in this report was authorized under Task 1B522301A08101, Dissemination Investigations of Liquid and Solid Agents (U). The work was started in June 1964 and completed in June 1967.

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DIGEST

The object of this study was to obtain basic data on the dissemination of bulk powder into airborne primary particles 1μ to 10μ in diameter. Results indicated that discrete particles are obtained when initially suspended agglomerates are shattered by wall or target impact. Significant reagglomeration can occur later because of triboelectrification and insufficient dilution with primary air. Modification by powder additives proves to be selective in weakening contact forces between particles and changing electrification.

Seventeen powders and sixteen additives were selected for study. These had a wide range of properties: different sizes, smooth and rough surfaces, spherical and irregular shapes, crystalline and amorphous structures, organic and inorganic compositions, and different dielectric constants. Flowability, angle of repose, adhesive force, viscosity of a fluidized bed, and evidences of electric charge separation were measured for various powders and powder-additive combinations. Certain combinations, with the additive at about 1% concentration, showed marked changes in bulk properties and improved dissemination.

Aerodynamic disseminating methods, which permitted control of breakup forces over a wide range of values, were used for tests of aerosolizability. Several assessment methods were evaluated, and a laminar flow settling chamber in which aerosol particles deposit on long horizontal plates, was considered to be most applicable for this type of research. Effects of air velocity, powder to air ratio, and flow geometry, were studied. Electrostatic phenomena were observed, and it was found that these can be modified by powder treatment and by injecting gas ions along with disseminating air.

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I INTRODUCTION

The object of this study was to obtain basic data on the dissemination of bulk powder into airborne primary particles 1μ to 10μ diameter. The initial effort was a critical review of the literature, which has been published ("Aerosol Dissemination Processes - A Critical Review," Volume I, Stanford Research Institute Special Technical Report No. 2, Poppoff, 1965). The research reported here was based on information recovered. The review showed that:

Particles, 1μ to 10μ, adhere firmly to their neighbors in the bulk powder. The adhesion may be the result of any one or a combination of forces resulting from (1) electrostatic attraction, (2) absorbed liquid films, or (3) van der Waal's forces. The smaller the particles are, the greater is the difficulty of producing an aerosol of individual particles.

Methods for making an aerosol from dry powder could be placed in one of the following four categories in which: (1) powder is fed into the intake of a rotary air blower; (2) powder is caused to flow through Venturi throats or capillary tubes; (3) loose or compressed powder is exposed to a sudden burst of gas with or without impaction on a solid surface; and (4) loose or compressed powder is eroded from a powder bed surface by high velocity air jets.

In terms of the surface energy of the particles, the power efficiencies of aerosol devices that have been used were very low, perhaps as low as 0.001%; however, this value criterion has little meaning because most of the energy is used for diluting and transporting the aerosol. At some stage between the time that the particles exist as a bulk powder and the time that they are sufficiently dilute to prevent agglomeration, energy must be used for mixing air. The only concern for energy is that it be large enough and effectively coupled with the powder mass to produce forces that break bonds between individual particles.

Complete assessment is a formidable task; several presentations of the practical and theoretical problems are given in the literature. No single method appeared to offer a complete solution to the problem.

Considerable effort has been devoted to bulk powder modification techniques. Changes in the properties of bulk powder such as tensile strength, shear strength, angle of repose and flowability have been measured in attempts to correlate these measurements with improvement in aerosolizability; the assessment methods used give data on the comparative efficacy of several modifications. Except for a few instances, however, no dramatic improvement in aerosolization occurred as a result of powder modification.

It was apparent from the literature review that the following aspects of powder dissemination should be included in this research program.

a. Use of a wide range of powder types to obtain data on the effect of chemical composition, particle size, size distribution and various powder treatments.

b. Convincing demonstration of improved ability to get individual particles suspended by treatment of powder with additives. New observations were needed to confirm intuitive beliefs.

c. Development of a simple and workable assessment procedure.

d. Employment of a simplified dissemination system amenable to mathematical analysis and capable of operation over a wide range of energy should be employed.

e. Observations of subsidiary phenomena and difficulties that are encountered during powder dissemination, including electrostatic effects.

f. Identification of scientific principles controlling the production of individual particles should be identified.

Results of these investigations are covered in the body of this report.

Throughout the period of the program there was a continued search of the literature for any new and useful information bearing on the conversion of a powder to an aerosol. The information was collected, digested and organized. Appendix B of this report is an expanded list of references to supplement The Critical Review.

II SUMMARY

Powders differ markedly in their behavior in bulk and the ease with which the individual particles can be converted to an aerosol. A general study of powder dissemination must therefore examine many kinds of powders. In this program, seventeen powders and sixteen additives were used in various combinations. The powders had a wide range of properties and the additives were used to modify the bulk properties and to improve the dissemination of the powder particles.

Flowability, viscosity in aerated state, adhesion and angle of repose were measured to characterize the behavior of the powder and powder-additive combinations. The adhesion measurements revealed only general trends, but the other three methods gave data that, in general, correlated with ease of dissemination. For powders susceptible to improvement by use of an additive, 0.5-1.0% hydrophobic Philadelphia Quartz (PQ 2340) gave the best results in terms of flowability, angle of repose, and reduction of viscosity of fluidized powder. Additives in excess of 1.0% were less effective than the optimum amount. The effects of additives are highly specific to the powder and additive combination. For example, glass beads, zinc-cadmium-sulfide and saccharin were benefited by use of an additive, while quartz and aluminum powders were only slightly improved. The viscosity of a fluidized powder is believed to most nearly reflect the properties of a powder associated with its ease of dissemination.

Aerodynamic disseminating methods were employed which permitted control of breakup forces over a wide range of values. These included a) subjecting a small pile of powder to a gentle "puff" of air for low energy application; b) aspirating an air/powder mixture through straight tubes over a range of velocities for soft impact at the tube wall; c) aspirating an air/powder mixture through a curved tube at up to sonic velocity for hard impact.

Some of the larger sized particles (12-18μ) were almost completely disseminated at the lowest energies used. The smaller the particle size, the greater was the difficulty of obtaining complete dissemination. The most complete dissemination of the smaller particles (1-10μ) was obtained by aspirating powder with air at sonic velocity in a coiled tube to insure "hard" impact on solid wall surface. Other results were:

a) The effect of additives was dramatic for some powders not only in improving flow properties, but also in improving dissemination though the measured adhesive forces between particles were reduced only 1/2 to 1/3 by the use of an additive.

b) Irregular shaped particles disseminated more completely than smooth spherical particles of the same size.

c) Small ratios of powder/air in the aspirating system resulted in less reagglomeration following dissemination.

d) Reagglomerated aerosols can result in agglomerates having a greater mass than a single particle of the same material and yet have the same aerodynamic properties. The implication of this result is that control not elimination of agglomeration may be the most desirable approach to dissemination of powders into an aerosol with suitable aerodynamic properties.

e) Theoretical considerations are presented which show that impact of agglomerates against solid surfaces are a necessary feature for successful pneumatic dissemination of particles in the 1-10μ size range.

f) The reagglomeration observed could not be explained on the basis of collision and adhesion resulting from turbulent mixing, but was most likely caused by electrostatic attraction between particles.

g) Electrostatic effects were observed frequently during the course of dissemination tests. They affected dissemination, caused reagglomeration and complicated assessment procedures. Additives were used to alter the charges generated on powder particles during dissemination. Injection of gas ions with the disseminating gas also altered electric charges on particles, and with further development may provide a technique for more efficient dissemination.

A laminar flow settling system was developed for assessing aerosols. This was used in conjunction with microscopic observation and photographic recording of settled particles. Microscopic observation alone did not permit distinction between agglomerates and single particles of irregular

shapes. Particles settled from air can land sufficiently close to each other to render uncertain whether they existed in the air as an agglomerate or as single particles. A method of correcting for this coincidental settling is presented. The use of multiple techniques permitted the acquisition of data not obtainable by any single assessment method.

III RECOMMENDATIONS FOR FURTHER RESEARCH

There is no aspect of the study of powder dissemination followed in our program that could not be extended with benefits resulting from new knowledge. Some judgment must be employed to limit the number of recommended items for future work. Any list will include some individual preferences and ours are as follows. One recommendation is for a fundamental study of triboelectrification and the other pertains to applying what we now know to the design of a scaled up model of a pneumatic powder disseminator.

1. Triboelectrification of Powder Particles

The scientific principles that control electrical charging of powder particles when they separate from each other or from solid surfaces (contact charging or triboelectrification) have not been identified. The electric charging of particles is probably the least understood phenomenon that affects dissemination and reagglomeration of powder particles.

When the scientific principles of triboelectric charging of powder particles are understood, it should be possible to minimize their detrimental effects and perhaps utilize them to advantage. A fundamental study of triboelectrification is therefore recommended.

2. Development of a Pneumatic Powder Disseminator

We have shown that successful pneumatic dissemination of a powder can be best accomplished by shattering agglomerates by wall or target impact. Reagglomeration can be minimized, even with some electrostatic charging, by dilution with primary air.

The work conducted in our program was designed to define the principles of dissemination without regard to constructing an integrated dissemination device. The scale of dissemination was deliberately small to accommodate laboratory facilities.

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There are design problems associated with the development of larger scale devices. These include powder feeding mechanisms, configuration of flow channels and/or anvils to obtain powder impact, minimizing power requirements, and confining the system to a reasonable size envelope.

The design, construction and evaluation of an integrated device using what we now know for disseminating 10 to 50 grams of powder per minute should provide useful data for still larger devices. The successful construction of a powder disseminator will make available useful devices for larger scale laboratory and small-scale field test devices by which active chemical agents can be evaluated.

IV MATERIALS

The dry powders and their additives used in this study are as follows:

A. Powders

The powders are tabulated in Table IV-1, and were selected to have a range of properties such as smooth and rough surface, spherical and irregular shape, crystalline and amorphous, organic and inorganic, dielectric and non-dielectric, etc. Most of the work was performed on organic powders since they simulate many of the characteristics of chemical agents. Saccharin, quinine and Microthene were the most widely used of all the powders and had been employed by other investigators.

The particle size distributions of saccharin, Microthene and quinine, as received, are given in Fig. IV-1 and Fig. IV-2. Quinine blended with 1% PQ 2340 additive in an Osterizer Blender was also used frequently, and its particle size distribution after processing is given in Fig. IV-3. In all the particle size distributions reported herein, the sizes were measured with an optical microscope, and in most cases the powder was dispersed on a microscope slide with an immersion oil to deagglomerate the particles before sizing.

Particle size distributions for glass beads, 1μ to 30μ and for aluminum metal, are given in Figs. IV-4 and IV-5.

B. Additives

The addition of powder modifying agents, or additives as they are designated in this report, was a dominant powder treatment. A large number of additives were assembled, and although all of them were not used in these tests, they are nevertheless listed for possible future use. The additives are listed in Table IV-2 and those additives marked with an asterisk were used in this program.

The additives listed are dry powders which have been found to be useful in small quantities to modify bulk properties. In general, the additives had a primary particle size 10^{-1} to 10^{-3} times the size of the powders they modified.

Table IV-1

POWDERS USED FOR TESTING

Powder	Source	Physical and Chemical Properties
Spherical Shaped		
Aluminum K-3	Valley Metallurgical Process Essex, Connecticut	3μ nmd; roughly spherical; electrical conductor; poor flowability.
Copper	Particle Information Source Los Altos, Calif.	10-150μ size; electrical conductor.
Glass beads	Cataphote Corp. Jackson, Miss.	2 sizes: 1-30μ; insulator; poor flowability; 44-62μ; insulator; good flowability.
Microthene FN-510	U.S. Industrial Chemical Co Cincinnati, Ohio	20μ nmd; good insulator; poor flowability.
Steel	Particle Information Service Los Altos, Calif.	10-150μ size; electrical conductor
Zinc Cadmium Sulfide	U.S. Radium Corp. Morristown, New Jersey	4μ mdd; roughly spherical; poor flowability; fluorescent in several colors.
Irregularly Shaped		
Aspirin	Local Drugstore	Tablets ground and screened to (-)325 mesh.
Black sand (no. 1)	Unspecified	3 fractions: 50-100μ; 100-150μ; 150-350μ; free flowing.
Black sand (no. 2)	Unspecified	Number mean diameter, ~37μ.
Feldspar	United Clay Products Trenton, New Jersey	0.5-50μ; poor flowing; highly agglomerated.
Garnet sand	Unspecified	Number mean diameter, ~320μ.
Ground cast iron	Unspecified	Number mean diameter, ~32μ.
Laterite soil	Unspecified	Number mean diameter, ~4μ.
Quartz	Del Monte Properties San Francisco, Calif.	(-)325 mesh; inorganic.
Quinine	Matheson Coleman & Bell Catalog #6968	11.0μ nmd; organic; poor flowability.
Quinine, treated	--	5.7μ nmd; organic; poor flowability; treated with 1% PQ 2340 in Osterizer Blender for six minutes.
Rhodamine B	Matheson Coleman & Bell Catalog #B1193	Fluorescent dye, organic.
Saccharin	Monsanto Co. St. Louis, Missouri	Number mean diameter, ~8.0μ; organic, poor flowability.

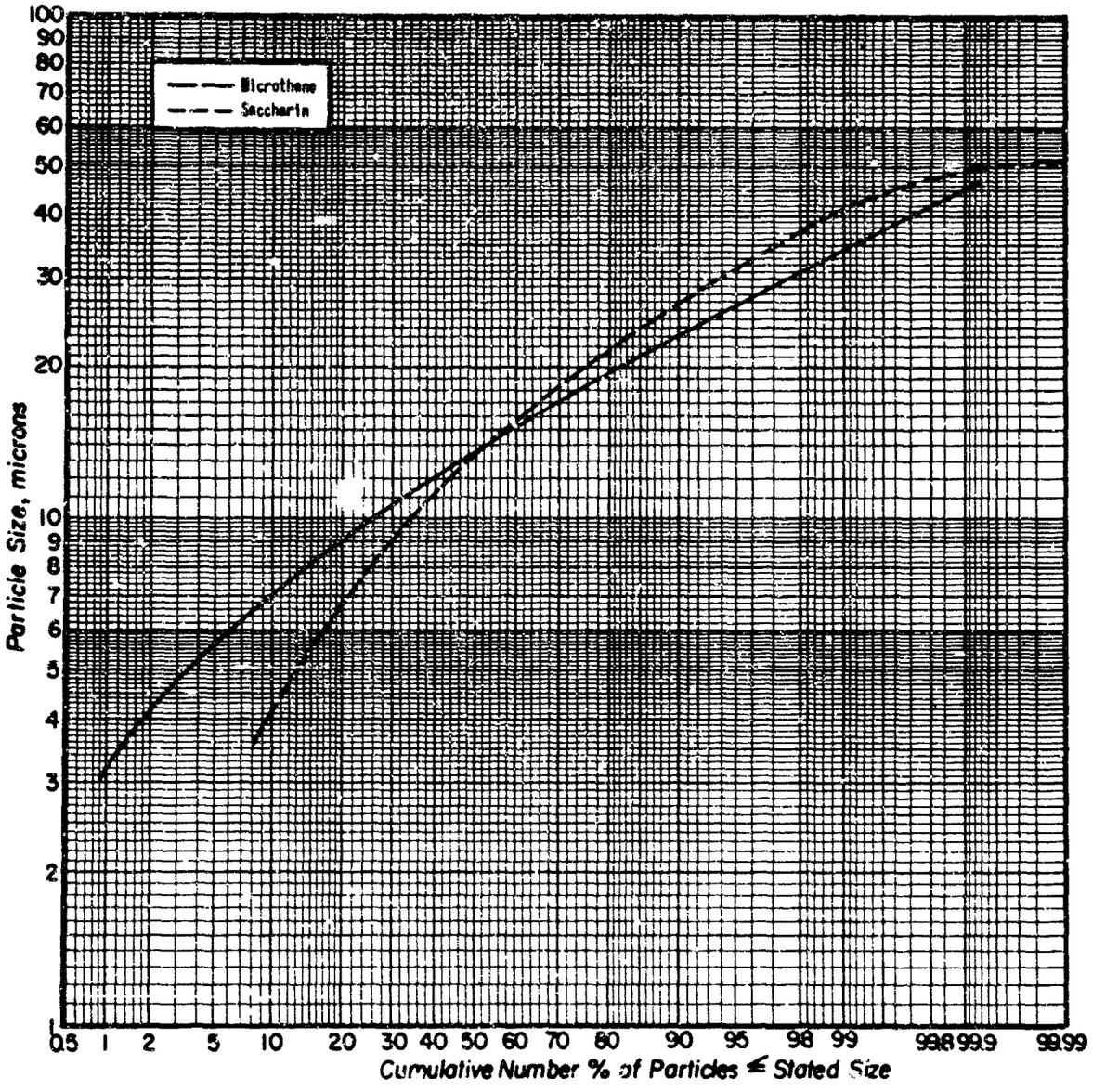


FIG. IV-1 PARTICLE SIZE DISTRIBUTION OF ALUMINUM H-3 DISPERSED IN OIL

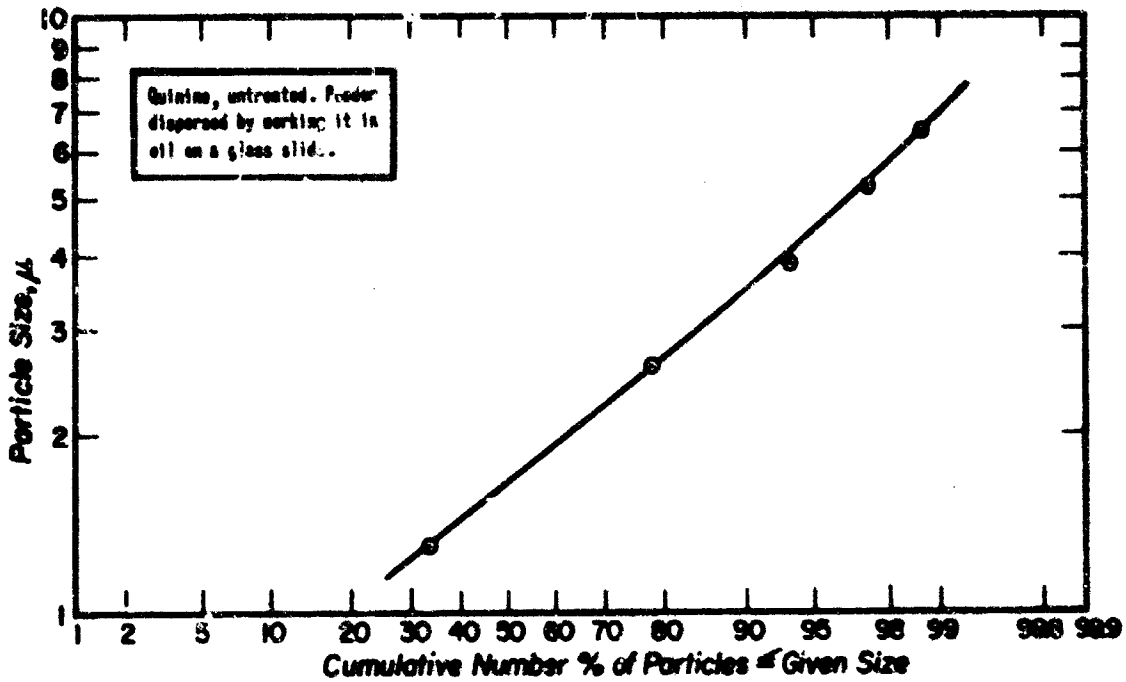


FIG. IV-2 PARTICLE SIZE DISTRIBUTION OF QUININE DISPERSED IN OIL

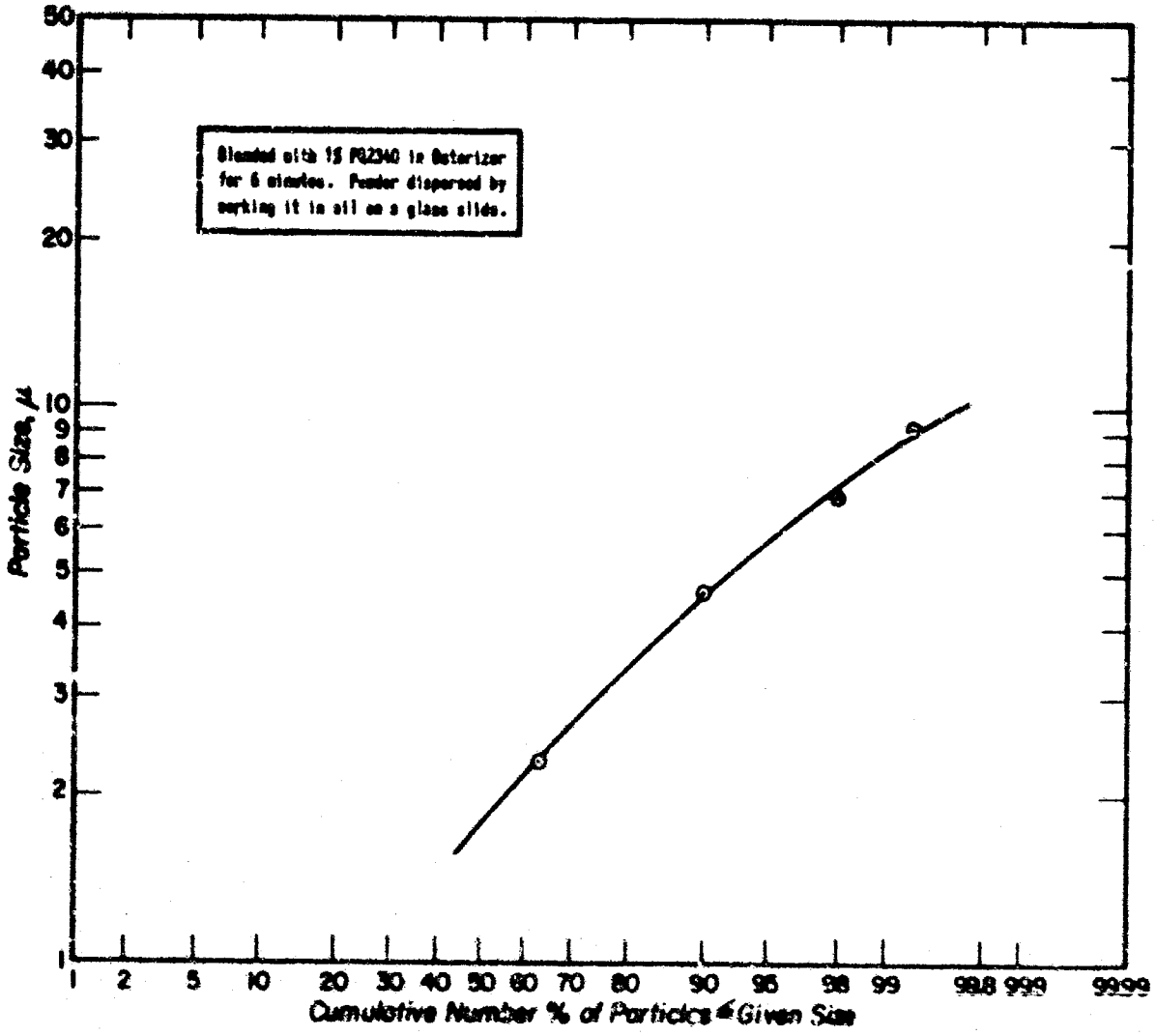


FIG. IV-3 PARTICLE SIZE DISTRIBUTION OF QUININE, TREATED

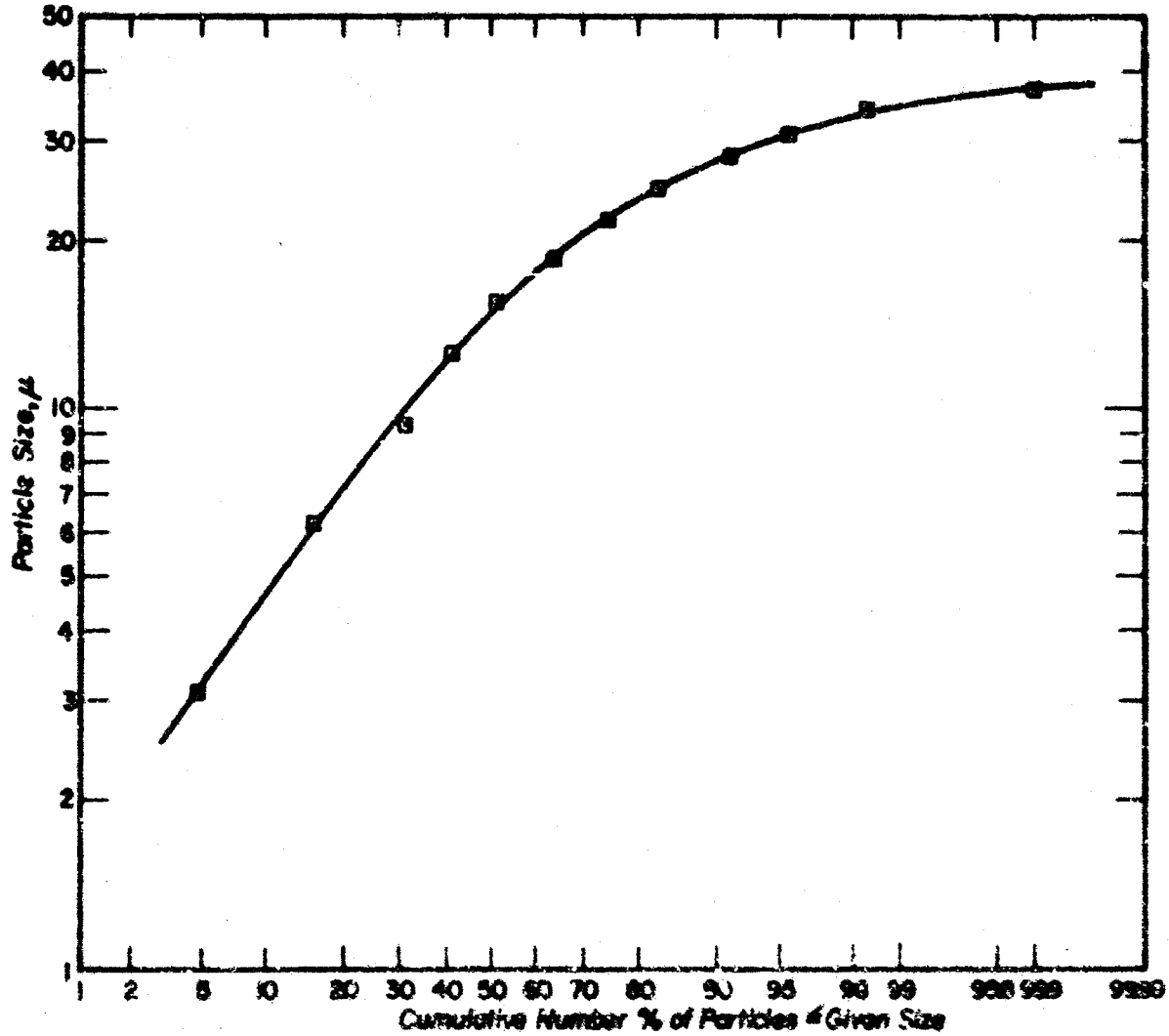


FIG. IV-4 PARTICLE SIZE DISTRIBUTION OF 1-30μ GLASS BEADS

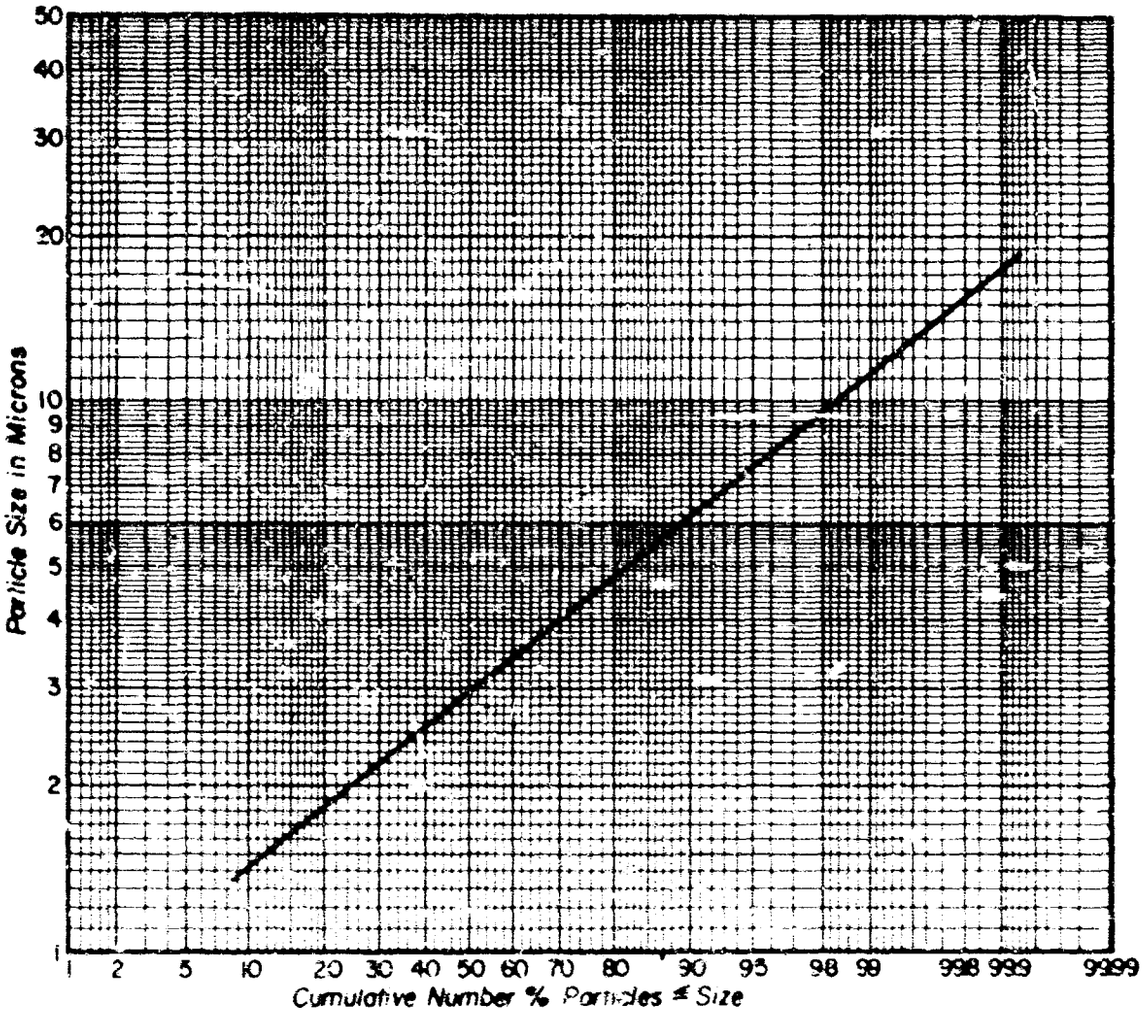


FIG. IV-5 PARTICLE SIZE DISTRIBUTION OF SACCHARIN AND MICROTHENE BY OPTICAL MICROSCOPY

Table IV-2

ADDITIVES USED FOR POWDER MODIFICATION

Additive Trade Name	Chemical Composition	Source	Properties
*Cab-O-Sil M-5	99% pyrogenic silica	Cabot Corporation 125 High St., Boston, Mass.	.015μ dia; 175-225 m^2/gm
*Cab-O-Sil	99% pyrogenic silica	Cabot Corporation 125 High St., Boston, Mass.	.008μ dia; 300-350 m^2/gm
*Organ-O-Sil	99% pyrogenic silica	Cabot Corporation 125 High St., Boston, Mass.	.015μ dia; 175-225 m^2/gm, hydrophobic coating
Alon C	95% pyrogenic alumina	Cabot Corporation 125 High St., Boston, Mass.	.01-.04μ; 80-100 m^2/gm
*Baymal	83% alumina fibrils	E. I. du Pont Wilmington, Del.	Fibrils .008μ dia; 275 m^2/gm; has positive charge on surface
*Ketrosil	99% silica	E. I. du Pont Wilmington, Del.	.01-03μ; has hydrophobic coating
*Hydrophobic silica G8	84-88% silica	E. I. du Pont Wilmington, Del.	.01μ; 200-300 m^2/gm; has hydrophobic coating
Sil-Stone 110	90% silica	Stoner Rubber Company 10792 Knott Ave, Anaheim, Calif.	.0[illegible].1μ; coated with silicone oil
*Quso W-22	99% precipitated silica	Philadelphia Quartz Company Philadelphia, Penn.	.012μ; 325 m^2/gm
*PQ2326	99% precipitated silica	Philadelphia Quartz Company Philadelphia, Penn.	.012μ; 325 m^2/gm; 80% hydrophobic silica
*PQ2340	99% precipitated silica	Philadelphia Quartz Company Philadelphia, Penn.	.012μ; 325 m^2/gm; 100% hydrophobic silica
*Santocel Z	90-92% silica	Monsanto Company St. Louis, Mo.	.01-.02μ; 280 m^2/gm
Santocel 62	94-96% silica	Monsanto Company St. Louis, Mo.	.01-.02μ; 260 m^2/gm

Table IV-2 (Concluded)

Additive Trade Name	Chemical Composition	Source	Properties
ASP 100	84% aluminum silicate	Minerals & Chemicals Philip Menlo Park, Calif.	.55μ; thin flat platelets
*Aerosil	99.8% silica	Degussa Inc. 609 Schuyler Ave, Kearny, N.J.	.01-.04μ; 200 m^2/gm
*Aerosil R972	99.8% silica	Degussa Inc. 609 Schuyler Ave, Kearny, N.J.	.02μ 120 m^2/gm; has hydrophobic coating
*Carbon Black XE-2	99% carbon	Particle Information Service 600 Springer Rd, Los Altos, Calif.	.01-.05μ, 145 m^2/gm
*Carbon Black LA-1	99% carbon	Particle Information Service 600 Springer Rd, Los Altos, Calif.	.005-.017μ; 1000 m^2/gm
*Kaopaque	Aluminum silicate (kaolin)	Particle Information Service 600 Springer Rd, Los Altos Calif.	.20μ, has hydrophobic coating
*Magnesol	Magnesium silicate	West Vaco Chem Div FMC Corporation	.01μ; 315 m^2/gm
Calofort U	98% calcium carbonate	John Sturge Ltd Wheeleys Rd, Birmingham, England	.1μ; [illegible] m^2/gm
Calofort S	99% calcium carbonate	John Sturge Ltd Wheeleys Rd, Birmingham, England	[illegible]μ; 23 m^2/gm; has 3% stearate coating
Super Multiflex	98% precipitated calcium carbonate	Diamond Alkali Company Cleveland, Ohio	.03-.10μ
Multiflex MM	98% precipitated calcium carbonate	Diamond Alkali Company Cleveland, Ohio	.03-.15μ
Purecal U	98% calcium carbonate	Wyandotte Chemical Corporation Wyandotte, Mich.	.03-.04μ

V POWDER MODIFICATION AND BULK PROPERTIES

A. Preparation of Powders with Specific Particle Size

Most of the powders used in the program were obtained in the particle size desired. However, some powders were fractionated into specific sizes for a particular experiment. Fractionation of the powders was almost always accomplished using a double-disc separator of the type shown in Fig. V-1. The high speed (2000-5000 rpm) rotating disc causes the particles from the feed to spiral outwardly. The larger particles because of their inertia accumulate in the clearance space between the casing and the periphery of the rotating double-disc. These larger particles are removed from the spacing by a plough and collected in a flask as shown in the insert to Fig. V-1. The fine particles, because of their smaller inertial forces, are able to pass through the gap between the double-disc and out to a cyclone separator where they are collected. The particle diameter cut-size can be varied by alteration of the air flow rate, or of the rotational speed of the double-disc, or both. This technique was used to fractionate glass beads and quartz powder (irregularly shaped) into four fractions from 30 to 4.5 microns. It was also used for separating quinine into fine and coarse fractions.

Another method for altering the particle size of a powder was to grind it in a ball-mill. This was done with quinine powder. Using petroleum ether as the dispersing medium, quinine was ground with and without a chemical treatment (1% hexamethyldisilizane plus 4% carbon tet.) for approximately 24 hours. After air drying the powder was screened through a 30# mesh screen.

B. Addition of Powder Modifying Agents

Powder modifying agents, or additives, were incorporated into powders by adding the additives and then either hand shaking the mixture or by blending the mixture in an Osterizer blender. With some powders, the speed and duration of osterizing had to be carefully controlled to prevent

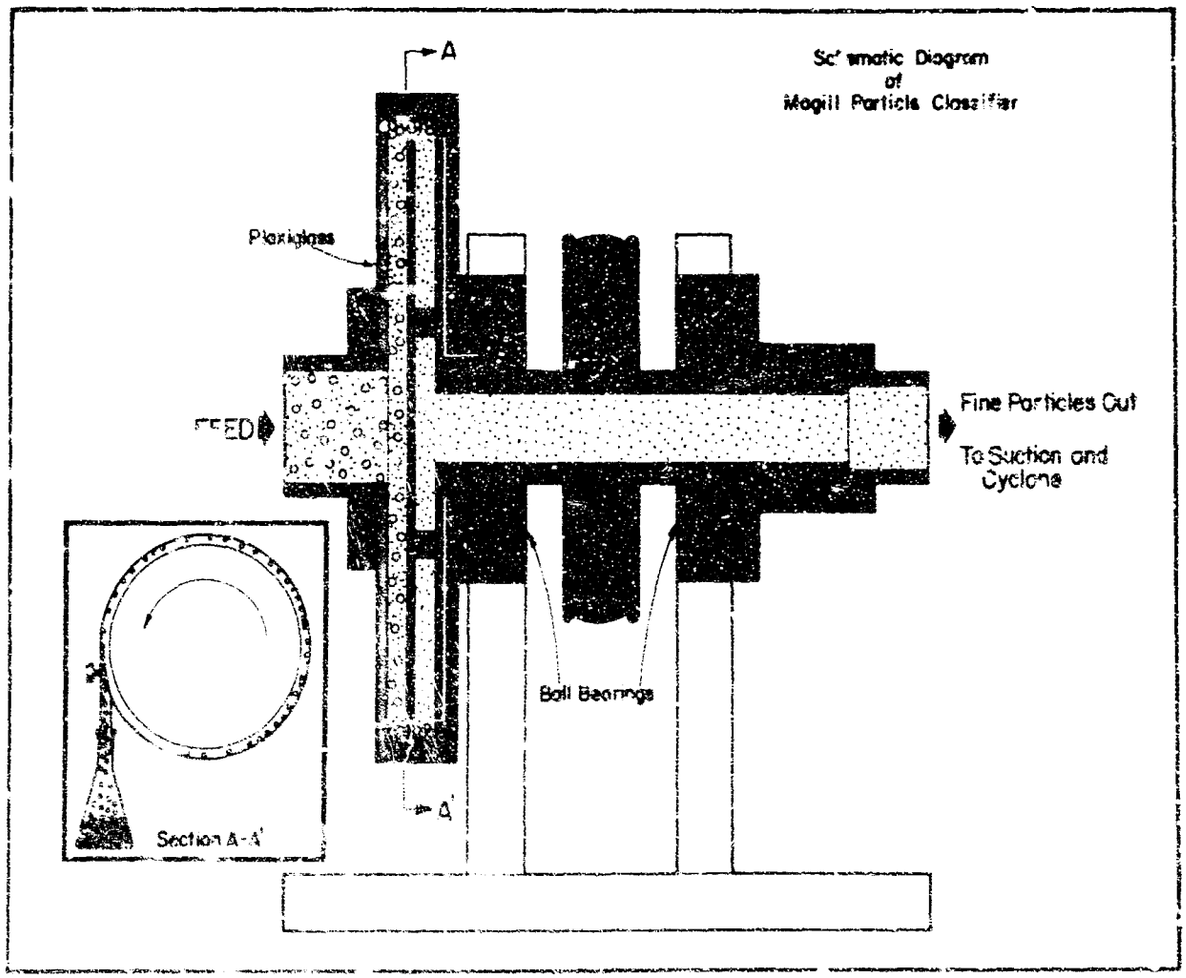


FIG V-1 DOUBLE-DISC PARTICLE SIZE FRACTIONATOR. The particle diameter cut-size can be varied by alteration of the air flow rate or the rotational speed of the double-disc or both

excess local heating and fusion of the powder particles. For example, saccharin osterized for 3 minutes continuously in a small container had a larger particle-size distribution than saccharin osterized for 3 minutes at one-minute intervals in a large container. The additive added was always measured on a weight basis.

C. Procedures for Measuring Bulk Properties

There is a voluminous mass of publications on the modification of bulk powder characteristics with a variety of solid, liquid and gas additives. The effectiveness of the powder treatment was then measured in terms of the particular property of interest, either flowability, adhesive force, tensile strength, or angle of repose, etc. Although the property of interest in this program was the aerosolizability of the powder, it was desirable and often revealing to learn about bulk properties of the powder other than its aerosolizability.

1. Flowability

Finely divided powders less than 30 microns often act sticky. They adhere to each other and to other things. Some powders are so sticky they block apertures when attempts are made to transfer them from one container to another or to meter their flow into disseminating equipment. Yet, in most practical applications of pneumatic dissemination, the powder must somehow be transferred from the container to the disseminating source. Therefore, unless the powder is already free-flowing, the powder must be treated or modified to flow, and means must be available to control and measure this flow.

The first measurement of "flowability" was made with a readily available powder feeder device used in Metronics' Aerosol Generators. The powder feeder, Fig. V-2, is a hopper with a notched wheel at the bottom. As the wheel rotates, the powder fills into the notches and then drops out on the underside of the hopper. The delivery rate is regulated by the speed of rotation, the deepness of the notches, and the number of notches. The weight of powder delivered in a designated time interval is a measure of flowability.

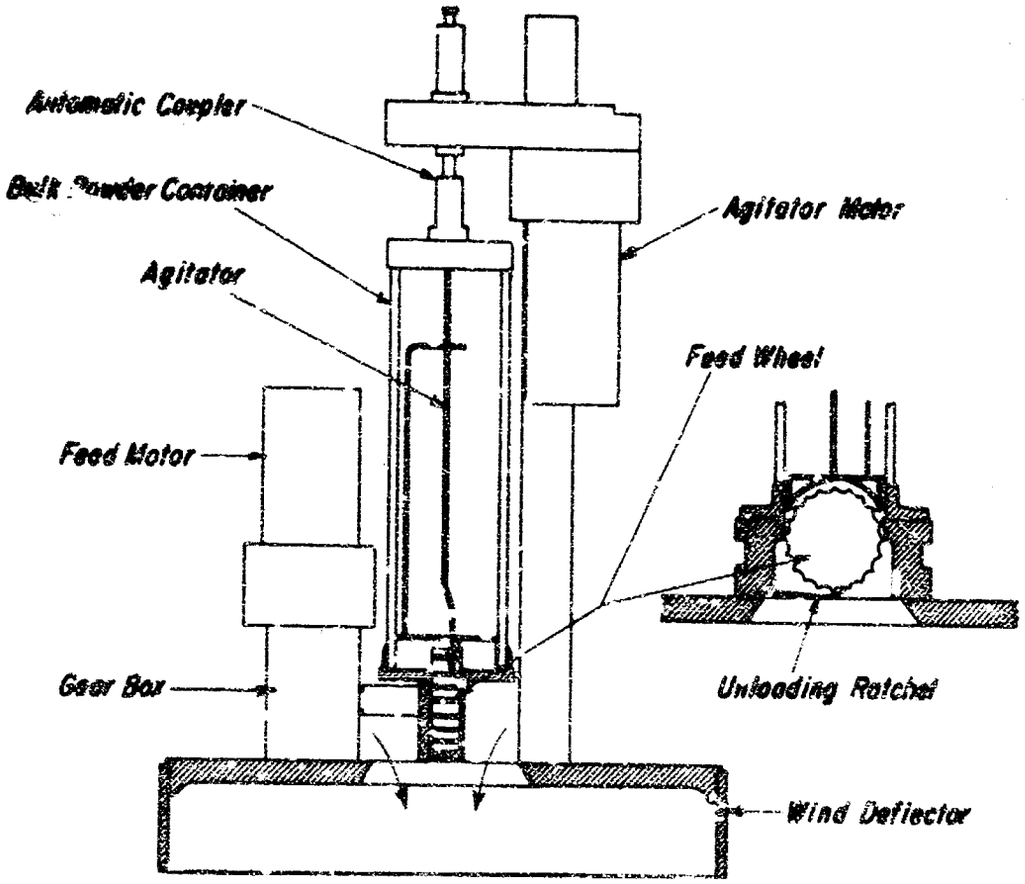


FIG. V-2 ROTATING NOTCHED-WHEEL TYPE POWDER FEEDER USED FOR MEASURING FLOWABILITY

2. Angle of Repose

Another measurement of bulk properties is the angle formed by a powder when it flows through an aperture (the angle of internal friction) or the angle subsequently formed by a pile of the powder beneath the opening (the angle of repose). Figure V-3 illustrates the difference between these two angles. In their discussion on powder rheology, Zenz and Othmer (1960) and Crosby (1980) consider the angle of internal friction as probably the most important and fundamental property of a powder, despite the fact that it is not commonly measured. The simplest method of determining the angle of internal friction is as follows:

A narrow, deep rectangular trough, shown in Fig. V-4, fabricated from clear plastic, with a small, stoppered hole at the bottom of the trough, was used for measurement. With the hole closed, powder is evenly and gently poured into the trough up to a designated level, after which the hole is opened and the powder allowed to flow out. The angle subtended by the stationary solid inside the trough and the horizontal is the angle of internal friction. However, with certain powders, another angle, the angle of repose, is also formed. With free-flow powders, the two are identical, but with powders that do not flow as freely, there is a difference.

Attempts to use this method to measure "sticky" powders were not too successful, because the powder did not flow out of the hole. Therefore, the range of method was limited and often did not apply to powders, such as untreated saccharin and Micro Cene. It was found, however, that if the hole was left open while the trough was being filled, the powder would flow out, even with a sticky powder, to give a characteristic angle.

A review of the literature revealed various definitions for this particular angular property, but since the technique is similar to building up a pile of powder, it was designated as the angle of repose rather than the angle of internal friction.

Measurement of free-flowing powders with this technique had a reproducibility within $\pm 2^{\circ}$. With sticky powders, the reproducibility decreased, and the procedure became sensitive to the means by which the powder was

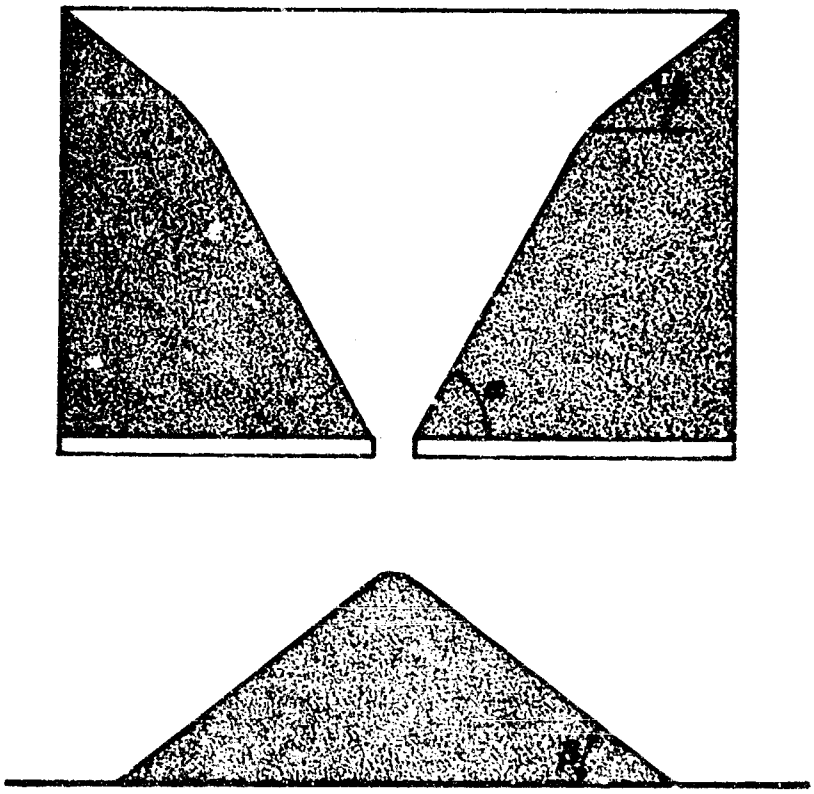


FIG. V-3 DIFFERENCE BETWEEN ANGLE OF INTERNAL FRICTION AND ANGLE OF REPOSE ACCORDING TO ZENZ AND OTHMER[5]: α = ANGLE OF INTERNAL FRACTION AND β = ANGLE OF REPOSE

FIG. V-4 APPARATUS FOR MEASURING THE ANGLE OF REPOSE OF POWDERS

loaded into the trough. A vibratory feeder was found to be the most suitable means for feeding the powder into the trough, because it avoided undue compaction of the powder.

3. Adhesive Force

The technique of Cremer et al. (1952) has been the most commonly used method for measuring the adhesive force of a bulk powder. It has been used by Baerns (1966), Nash et al. (1963), Orr et al. (1957), Patat and Schmid (1960), and Whitnah (1965) for measuring a variety of different powders. Orr et al. (1957) found the results to be too widely scattered to be of any value, but Baerns (1966) obtained consistent results with the technique. Others obtained fair to good results. In the main, the technique can be used to establish a trend, although at times, an indefinite one.

The method involved tilting a plate containing powder and measuring the angle at which the powder slid off as a bulk due to the force of gravity. Figure V-5 illustrates the apparatus with a bed of powder sliding down the plate. To avoid sliding between the particles and the plate, a multilayer of particles was first glued to the surface of the plate. The bed of powder measured 8.8 cm by 6.3 cm, and it could be packed up to a height of 2.5 cm. Following the recommendation given by Corn (1961), a contact time of five minutes was allowed for the powder bed to become equilibrated with the permanently fixed layer of powder. After carefully leveling the powder bed, the plate was gently vibrated to insure uniformity of contact and packing of the bed. Then the plate was tilted at approximately 1.5 degrees/sec until the bed sheared and slid from the plate.

The relationship among the angle of slide, θ, the adhesive force F of the powder bed, and the interparticle friction coefficient, μ, is given by the equation:

$$mg \sin \theta = \mu mg \cos \theta + F,$$

where m is the weight of the powder bed and g is the gravitational constant. By repeating the measurements for different weights of material

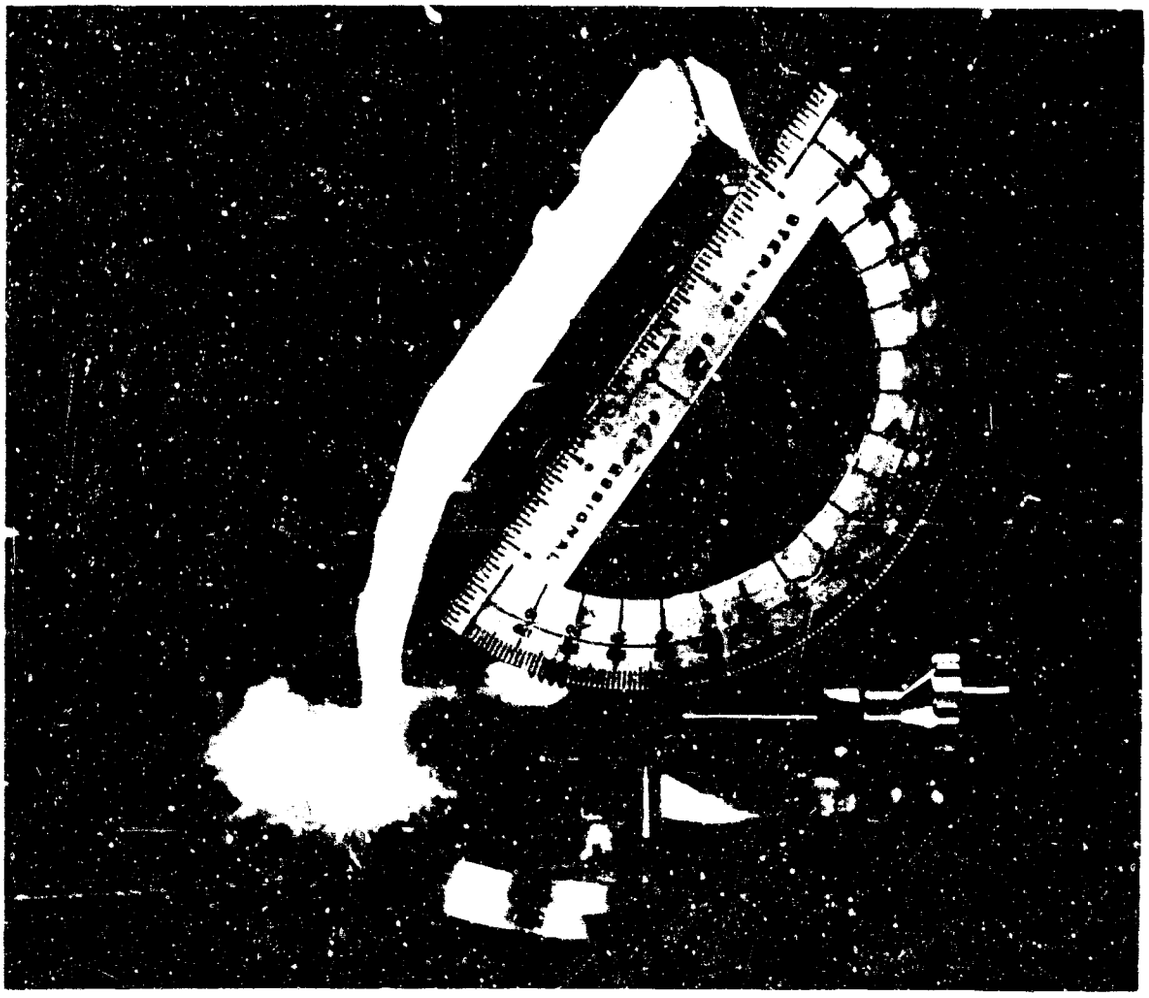


FIG V-5 TILTING PLATE FOR ADHESIVE FORCE MEASUREMENTS OF BULK POWDERS

and plotting mg sin θ versus mg cos θ on Cartesian coordinates, μ is obtained from the slope of the line and F from the intercept of the ordinate. To determine the interparticle adhesive force, the adhesive force of the powder bed, F, must be divided by the number of interparticle contacts between the bed and the fixed layer of particles. This number can be approximated by dividing the area of contact by the square of the mean particle diameter.

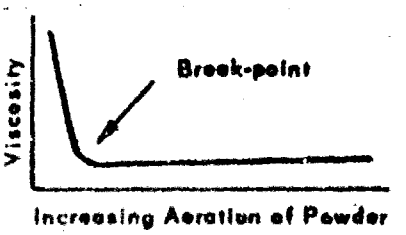
To test the reproducibility of the procedure, measurements of four inorganic powders, black sand, garnet sand, laterite soil, and cast-iron powder were made. These results, plotted in Fig. V-6, gave fair results. Cast-iron powder, probably because of its small particle sizes, gave the highest adhesive force. Both the garnet and black sand gave low adhesive forces and the laterite soil gave intermediate results.

4. Viscosity of Aerated Powders

In Quarterly Report No. 6 (Poppoff, 1965d), the overall aerosolization process was described as one in which the individual particles are separated and the air is placed around each particle to create a two-phase system (solid and air) with the desired ratio of phases. The process can be visualized as occurring in stages, as follows:

Stage	I	Breaking of bonds between particles
Stage	II	Separation of particles to an aerated mass
Stage	III	Conversion of the aerated particle-air mass to an aerosol of some desired dilution.

The transformation from one stage to another occurs without sharp boundaries or definitions. Nevertheless, a break does occur, and it was proposed that this break might be defined as the break-point in a curve in which the viscosity of a powder-air mixture is plotted against the amount of aeration applied to the powder. An ideal curve would be like the one shown below.



Powder Viscosity vs. Aeration

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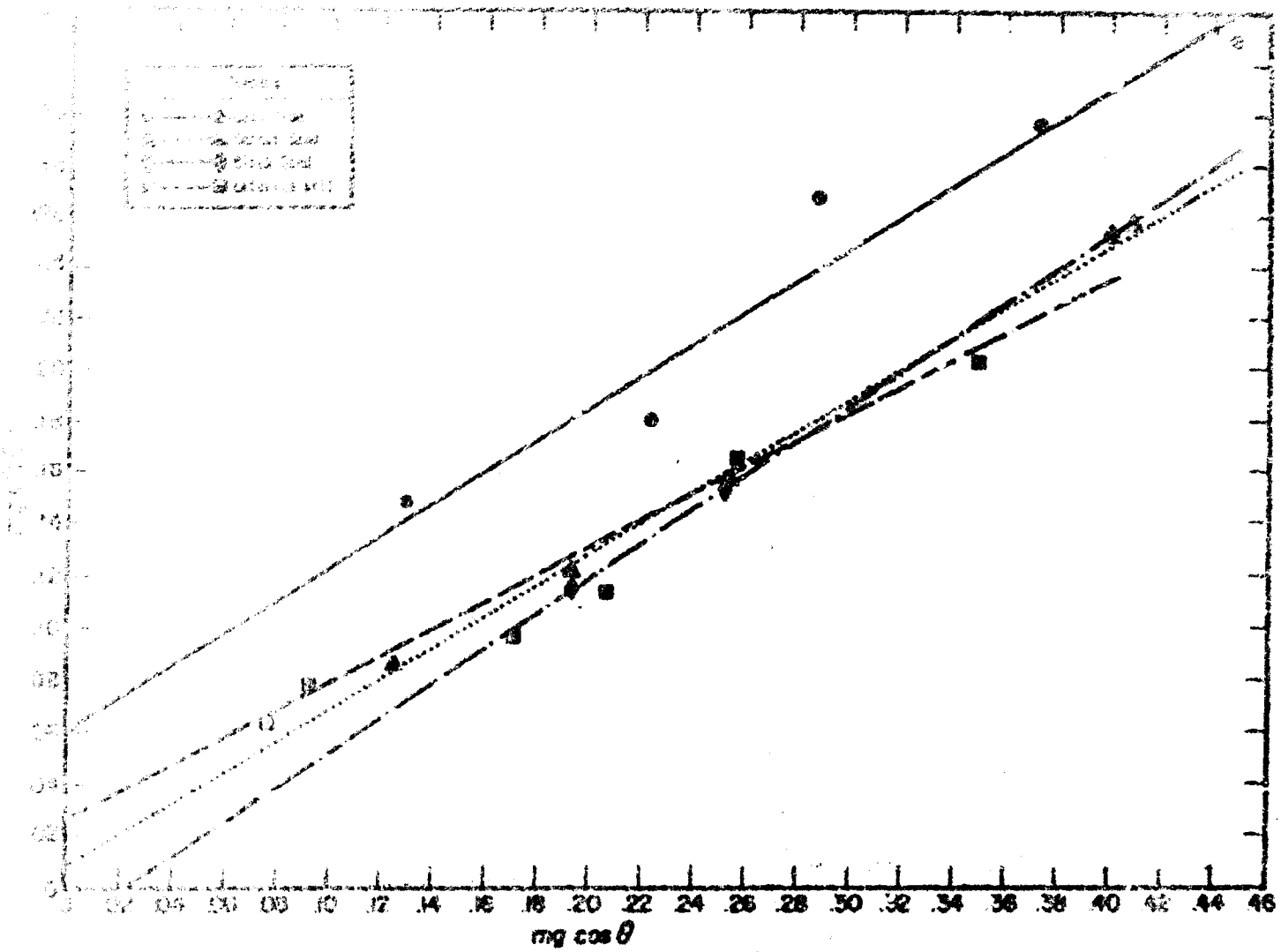

FIG. V-5 ADHESIVE FORCE MEASUREMENT OF SEVERAL INORGANIC POWDERS WITH THE TILTING TABLE APPARATUS

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The viscosity of the bulk powder drops rapidly at the beginning of the aeration until it reaches the break-point between Stage II and Stage III (conversion of the aerated powder mass to an aerosol). After that point the viscosity remains more or less constant.

This formation of an aerosol from a bulk powder by applying air may be likened to stirring into suspension a friable solid that converts to a viscous fluid which eventually acquires the viscosity of the fluid. First the shear stress applied by the fluid must be sufficient to overcome the yield-point of the plastic stress, then the shear stress must disrupt the viscous partially aerated powder and finally mixing must proceed to equilibrium.

To investigate this concept, a general purpose Stormer Viscosimeter of the type shown in Fig. V-7 was employed for measuring the viscosity of aerated powders. The system was patterned after that used by Matheson et al. (1949) in their study of the characteristics of fluid-solid systems. The dimensions of the powder column and the stirring paddle are given in Fig. V-8. The powder to be tested was added to the aerating chamber (fluid bed) until it completely covered the stirring paddle. The flow of air through the porous plate and into the powder bed was adjusted to provide fluidization of the powder. Weight in 5 gram increments was placed on the pulley so that 10-20 seconds were required for the paddle to make 100 revolutions. The air flow was reduced and the time required for 100 revolutions was recorded. This procedure was followed at successively lower air rates until flow was reached at which the paddle would not turn. The measurements were repeated at increasing air flow and the average of the two measurements was taken as the time required for 100 revolutions at the designated air flow.

The above time can be translated into absolute units by making time measurements on solutions of known viscosities as follows: The porous plate was removed and replaced by a flat flange. Glycerol solutions in concentrations varying from 0 - 100% were prepared and the time required for 100 revolutions was recorded for driving weights of 1, 5, 10, 25, 40, and 50 gram loads. A family of curves was obtained in which the time required for 100 revolutions was plotted against viscosity for the

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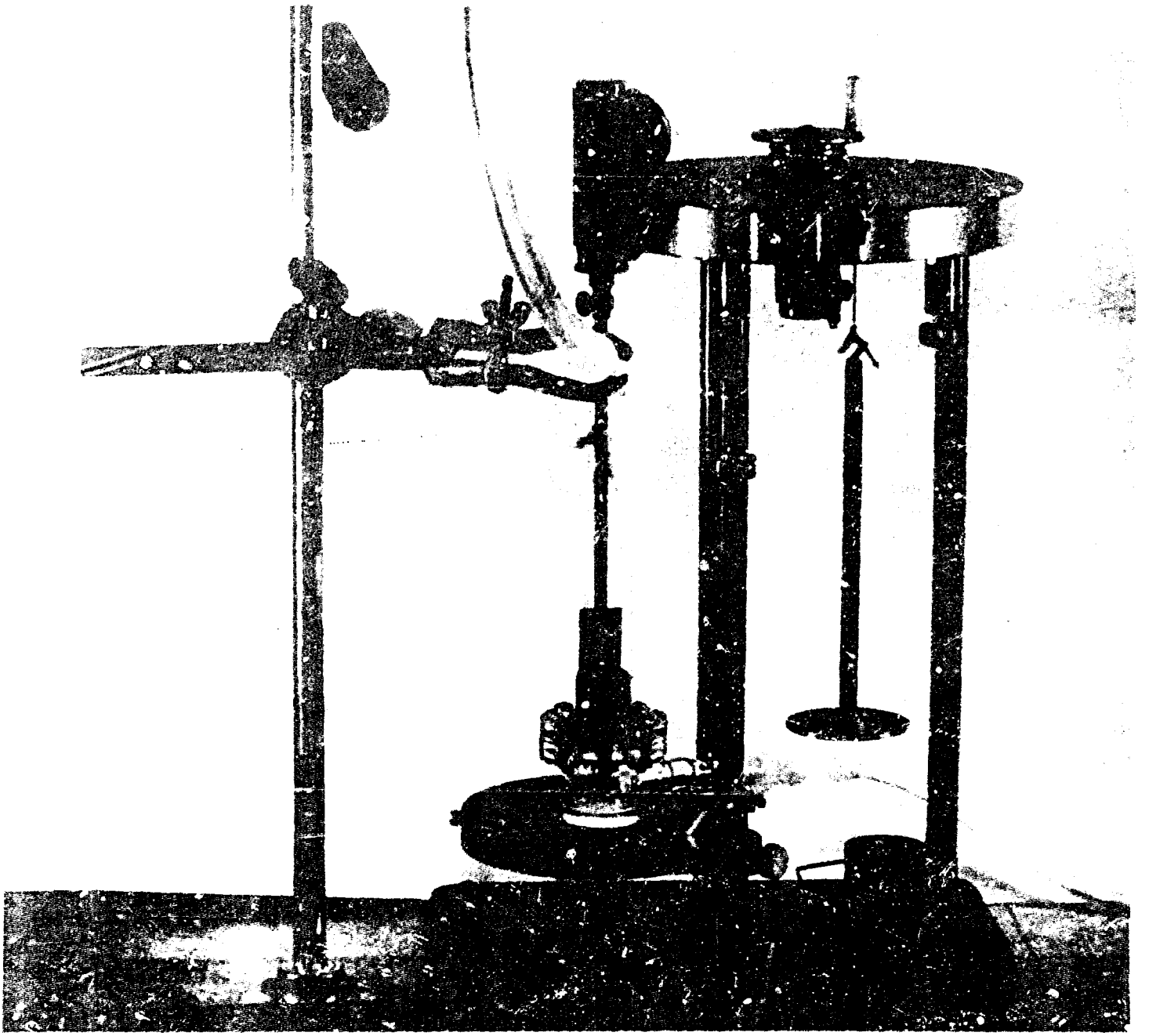

FIG. V-7 STORMER VISCOSIMETER APPARATUS WITH POWDER COLUMN FOR MEASURING VISCOSITY OF GAS-SOLID SYSTEM

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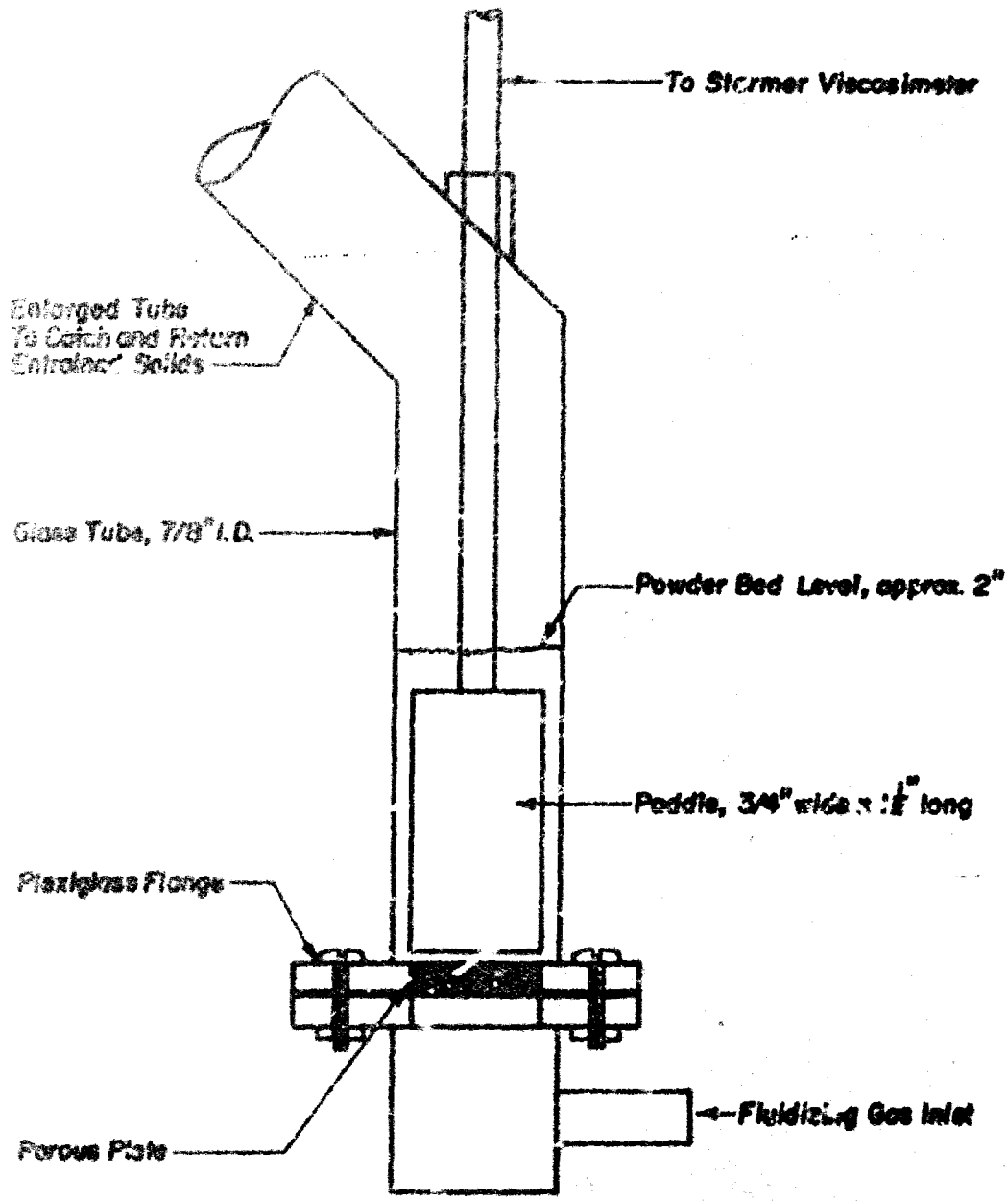

FIG. V-8 FLUID BED USED FOR MEASURING VISCOSITY OF GAS-SOLID SYSTEM

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various driving weights. A typical family of calibration curves shown in Fig. V-9 is obtained. Therefore, a specific time and driving weight will correspond to a specific viscosity.

Preliminary measurements with the viscosimeter were made on three fractions of black sand. The fractions, obtained by screening, were 80-100μ, 100-150μ and 150-350μ. The results are plotted in Fig. V-10 and they show that the break-point was easily defined.

B. Effect of Powder Modification on Bulk Properties

As noted in the critical review, the use of dry additives for modifying bulk properties of powders is practiced extensively, primarily because the additives can be incorporated into the powder very simply. At the present state-of-the-art, additives still remain as the predominant means for modifying the physical properties of a powder without altering its size or chemical properties. Additives were used widely in this program, and it was obvious that they can bring about useful change in the bulk properties of the powder.

1. Flowability

Tests on the effects of different additives on the flowability of zinc cadmium sulfide powders were made. These powders were chosen because in the untreated state they are sticky and did not flow easily. The flowability tests were performed with a notched wheel-hopper combination described earlier.

Approximately twenty additives were evaluated. The various additives used and their effects are shown in Tables V-1 and V-2. The results reflect not only the ease with which the toothed wheel was filled and emptied, but also changes in the bulk density as may be influenced by the additive. The results showed that some additives strongly altered the flow properties while others did not. There is, however, a definite trend. The additives which caused the greatest change in the powders had three characteristics in common: their particle sizes are in the millimicron size range; they have hydrophobic surfaces, and they have high melting points.

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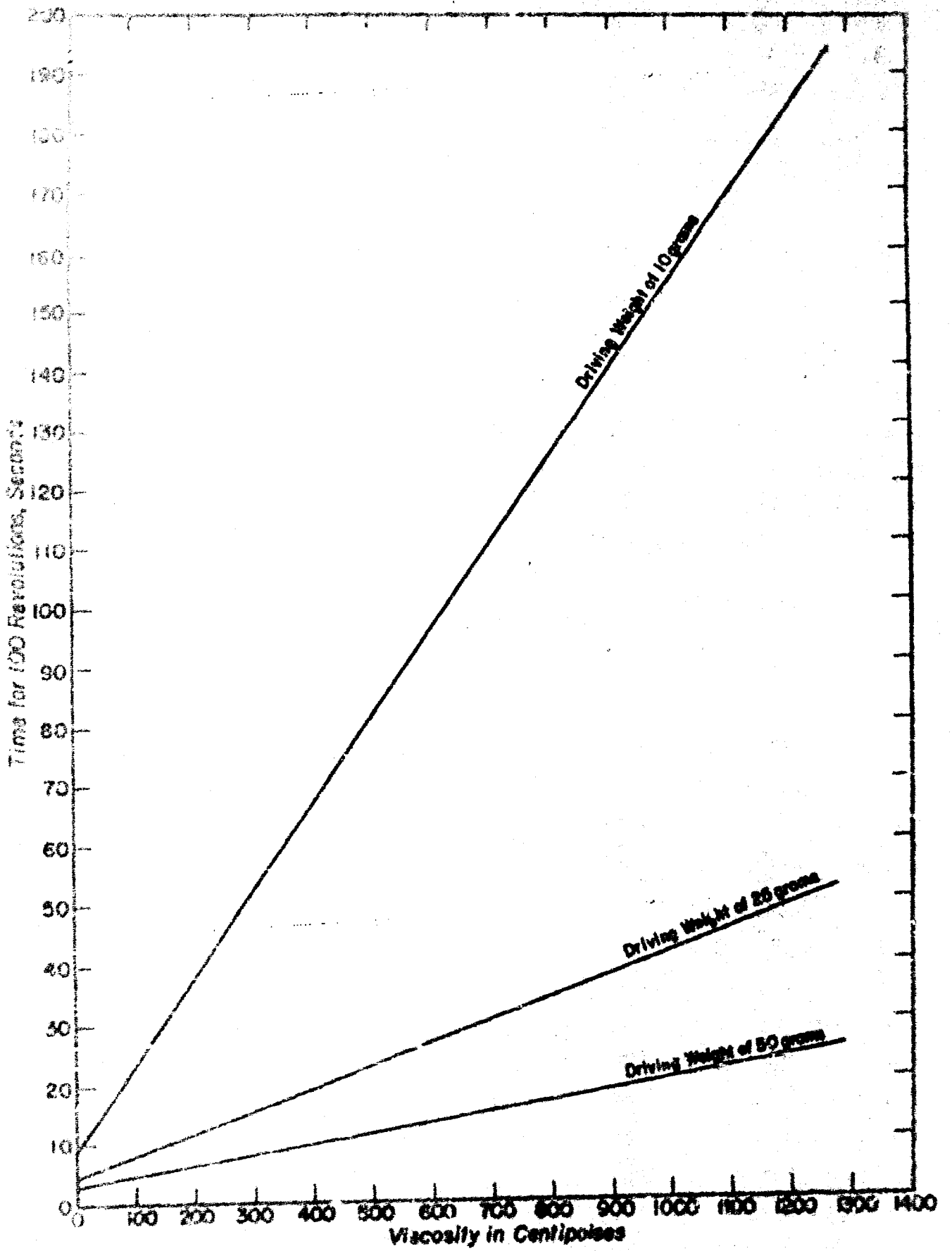

FIG. V-9 TYPICAL FAMILY OF CURVES FOR CONVERTING TIME AND DRIVING WEIGHT TO VISCOSITY

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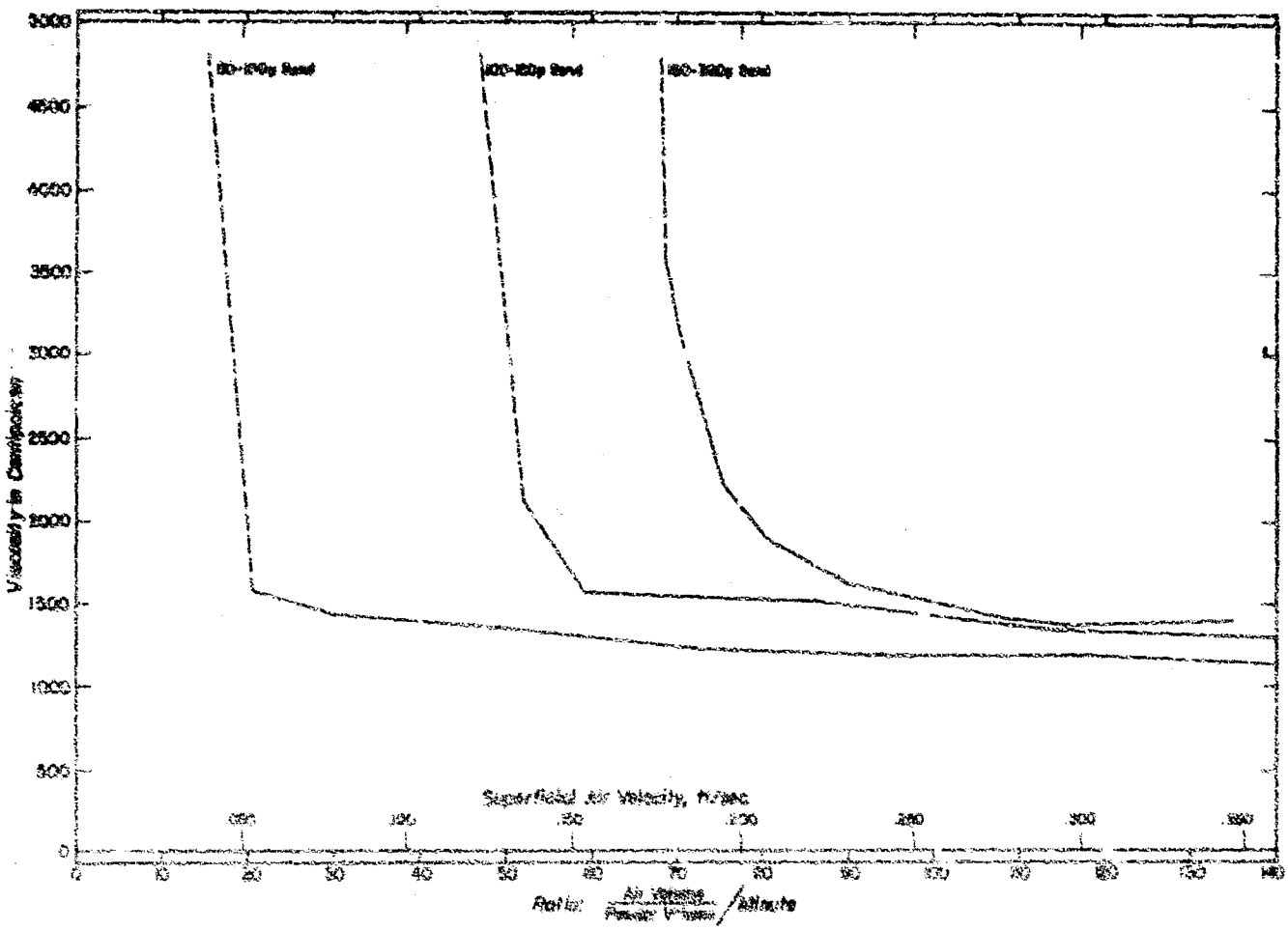

FIG. V-10 VISCOSITY OF THREE FRACTIONS OF BLACK SAND ILLUSTRATING THE BREAK POINT IN A GAS-SOLID MIXTURE

Table V-1

MEASUREMENT OF FLOWABILITY OF ZINC CADMIUM SULFIDE* WITH THE NOTCHED-WHEEL POWDER FEEDER†, SERIES #1.

Additive	Concentration	Feed† Rate (gm/min)	% Increase Over No Additive Powder
None	--	3.3	--
Valron Estersil	1/4%	6.0	82%
Valron Estersil	1/2%	6.4	94%
Valron Estersil	1%	6.8	106%
Hydrophobic Silica GS	1/2%	6.4	94%
Baymal	5%	5.2	57%
Cab-O-Sil M-5	1%	4.4	33%
Cab-O-Sil M-5	1%	4.8	45%
Cab-O-Sil M-5	1-1/2%	4.6	39%
Organ-O-Sil	1/2%	7.2	118%
Sartocel Z	1%	4.2	27%
Quso F-22	1/2%	5.2	57%
Quso F-22	1%	5.6	70%
Silstone 110	1%	3.9	18%
Magnesol	8%	3.8	15%

* Fluorescent powder #MLS-170 zinc cadmium sulfide was employed as the untreated material.

† The feed mechanism was Metronics Associates Model III B-3, Serial #10, operated at 15 volts.

Table V-2

MEASUREMENT OF FLOWABILITY OF ZINC CADMIUM SULFIDE* WITH THE NOTCHED-WHEEL POWDER FEEDER. SERIES #2.

Additive	Concentration	Feed† Rate (gm/min)	% Increase Over No Additive Powder
None	--	3.9	--
Hydrophobic Silica GS	1/2%	9.8	151%
Organ-O-Sil	1/2%	9.2	136%
PQ 2340	1/2%	8.9	128%
PQ 2326	1/2%	8.2	110%
2-2 Carbon Black	1/2%	7.7	98%
2-4 Carbon Black	1/2%	8.0	105%
2-6 Carbon Black	1/2%	8.8	126%
Kaophobe	1/2%	6.5	67%

* Fluorescent powder #MLS-218 zinc cadmium sulfide was used as the untreated material.

† The feed mechanism was Metronics Associates Model III E-3, Serial #10, operated at 15 volts.

It was desirable to determine the concentration of additive which would impart the maximum flowability. Using Hydrophobic Silica GS as the additive, mixtures containing 0.2%, 0.5%, 1.0%, 2.0% and 5.0% by weight additive were tested. The results, plotted in Fig. V-11, showed that there is an optimum concentration, and this optimum is around 0.5%. Later the bulk densities of these zinc-cadmium sulfide mixtures were measured, and it was found that the density curve closely followed the feed rate curve. Consequently, because of the method of assessing flowability, the changes in the feed rate observed in seeking optimum additive concentrations may have been due in part to changes in the bulk density, and changes in the "flowability" of the powder. For this reason, measurement of flowability with the notched-wheel hopper was discontinued.

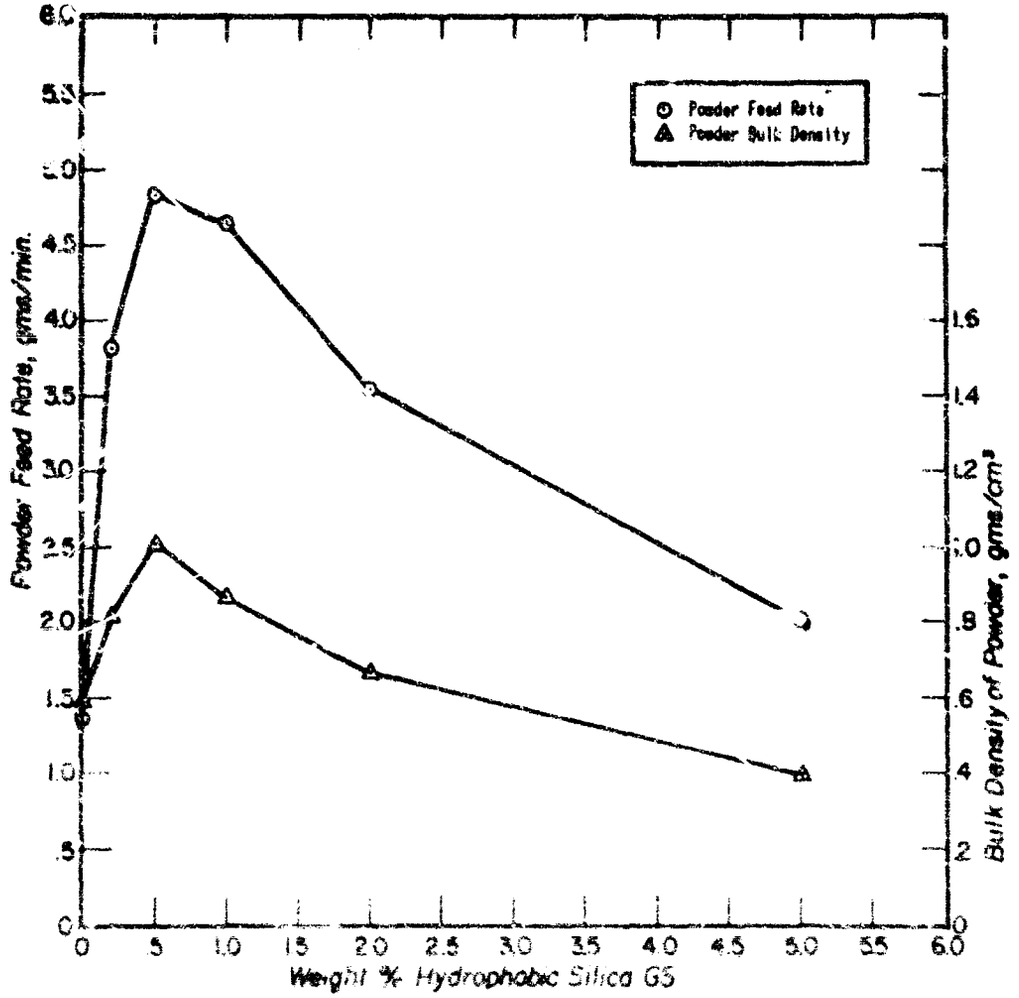

FIG. V-11 COMPARISON OF POWDER FEED RATE AND BULK DENSITY FOR ZINC CADMIUM SULFIDE TREATED WITH VARIOUS PERCENTAGE OF A HYDROPHOBIC SILICA ADDITIVE

3. Angle of Repose

a. Glass Beads

Angle of repose measurements on 44-62μ glass beads and 1-30μ glass beads treated with various percentages of Cab-O-Sil are shown in Fig. V-12. The effect of the silica additive on 44-62μ glass beads was negligible in the range 0% to 3% additive; at excessively large additive concentration, the angle of repose increased slightly. In comparison 1-30μ glass beads were influenced markedly by the silica additive. The angle of repose decreased steadily from 68° without additive, to 35° with 1% additive. Again at larger concentrations of additives the angle of repose increased.

b. Microthene

Curves for the angle of repose for Microthene treated with Cab-O-Sil H-5 and PQ 2340 are shown in Fig. V-13. Both additives showed a marked effect on the angle of repose, and both showed an optimum concentration of approximately 1.0%. Although PQ 2340 gave a smaller angle at the optimum concentration, the difference between the two additives was slight. Again at high additive concentration, the angles of repose increase, indicating that the frictional and/or adhesive forces between particles increased when excessive amounts of additive were present.

c. Saccharin

Angle of repose measurement for saccharin treated with various percentages of PQ 2340 silica is shown in Fig. V-14. The optimum concentration of additive is approximately 0.7%. The angle starts to increase at excessive additive concentration as was the experience with glass beads and Microthene.

The most obvious conclusion from this series of tests is that there is an optimum concentration of additive for a given powder. At concentrations greater than and/or less than the optimum, the angle of repose increases, indicating that the frictional and/or adhesive forces of the bulk powder have increased. Thus, these angles of repose measurements support a conclusion suggested by the flowability tests.

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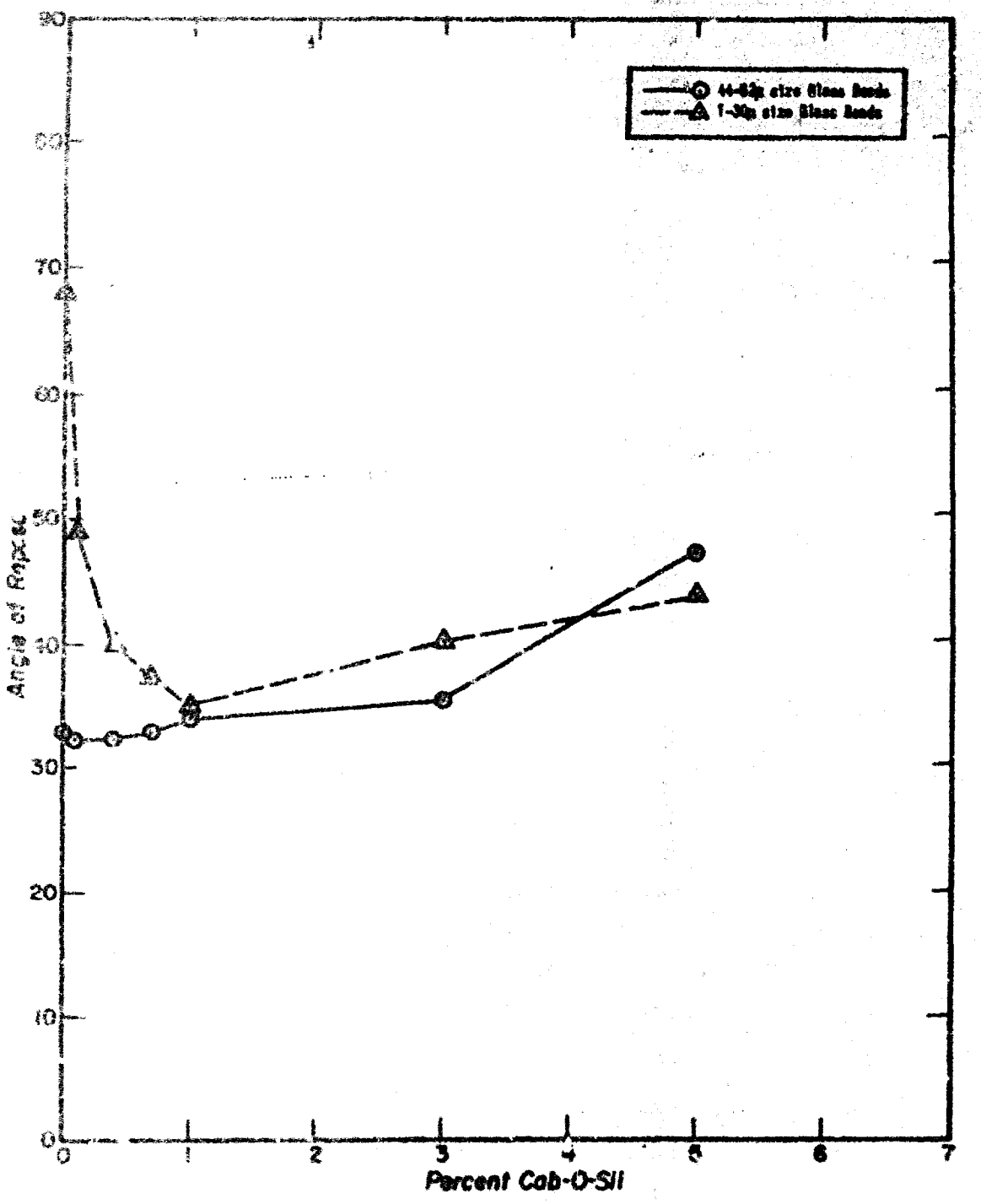

FIG. V-12 CHANGES IN THE ANGLE OF REPOSE FOR GLASS BEADS TREATED WITH CAB-O-SIL

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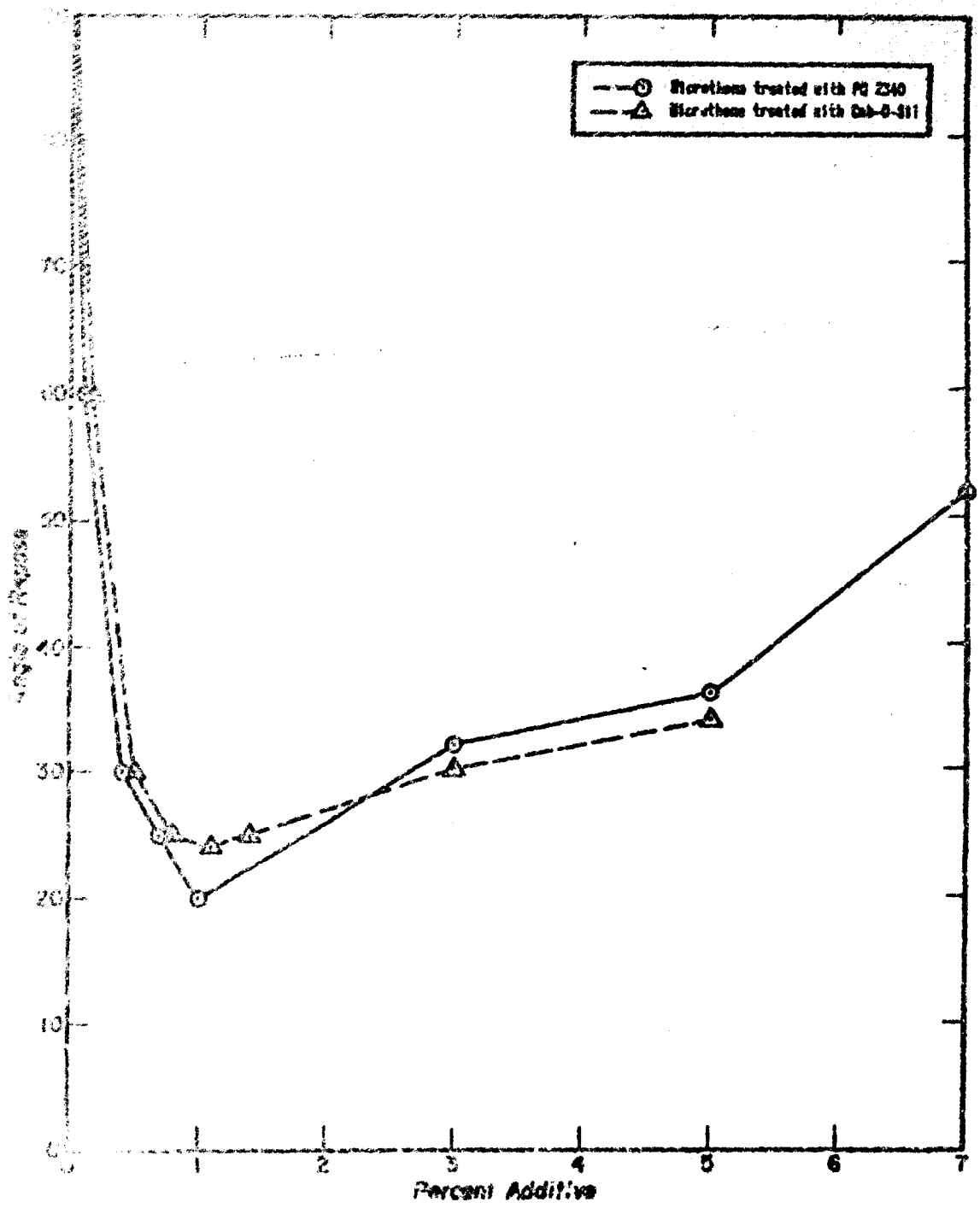

CHANGES IN THE ANGLE OF REPOSE FOR MICROTHENE TREATED WITH CAB-O-SIL AND PQ 2340

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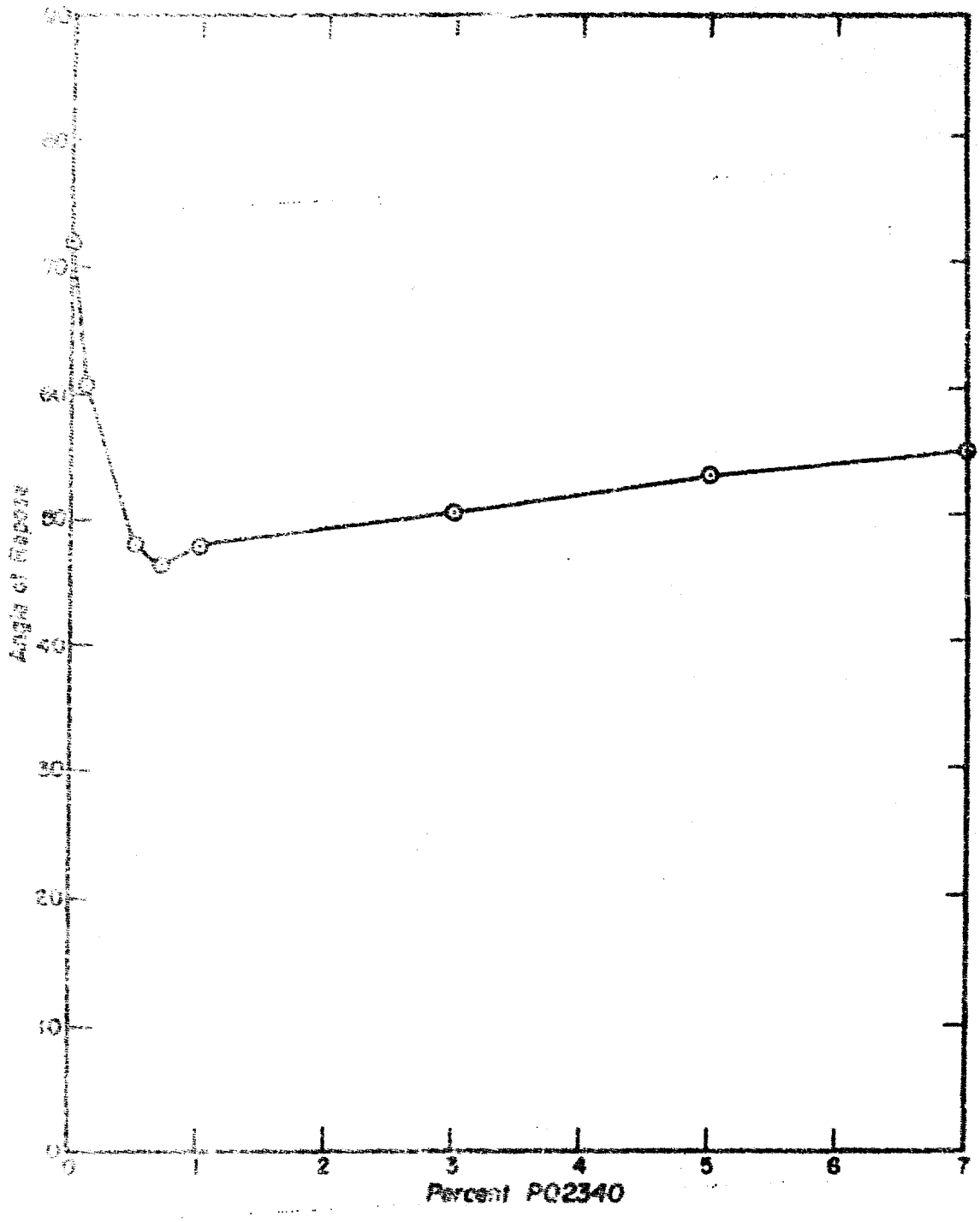

FIG. 7-14 CHANGES IN THE ANGLE OF REPOSE MEASUREMENTS FOR SACCHARIN TREATED WITH PQ 2340

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c. Effectiveness of Different Additives

The results of the bulk properties measurements indicated that the angle of repose was the simplest and the most reproducible measurement to make. Consequently this procedure was employed to explore the effect of different additives on powder behavior.

In the first series of tests, ten different silica additives, with slightly different properties, were added to saccharin in 1% quantities and the mixtures tested. The results are shown in Fig. V-15. It is seen that all the hydrophobic coated additives produced a lower angle of repose than any of the non-hydrophobic ones. The averaged angle of repose for hydrophobic additives was 47.2° with a standard deviation of ±5.4. For the non-hydrophobic additives, the average was 59.8 ±3.4°. The obvious conclusion from these tests was that a good powder flow additive for saccharin must have a hydrophobic coating. It should be noted that in the flowability tests with zinc cadmium sulfide (an inorganic powder), the additives which produced the most flow also had hydrophobic coatings.

In the second series of tests, it was decided to expand the effect of different additives by interchanging four base powder and four additives, each with widely different physical and chemical properties.

The powders were: (1) Saccharin, 3 to 40μ, irregularly shaped, organic, crystalline, soft. (2) Glass beads, 1 to 30μ, spherical, inorganic, noncrystalline, hard. (3) Quartz, 1-40μ, irregularly shaped, inorganic, crystalline, hard. (4) Aluminum, mmd = 3μ, semi-spherical, metallic, electrically conducting.

The additives were: (1) PQ 2340, 0.02μ, hydrophobic silica, amorphous. (2) Cab-O-Sil, 0.02μ, non-hydrophobic. (3) Magnesia, 0.05μ, non-hydrophobic, crystalline. (4) Carbon Black, 0.015μ, hydrophobic, amorphous, conducting.

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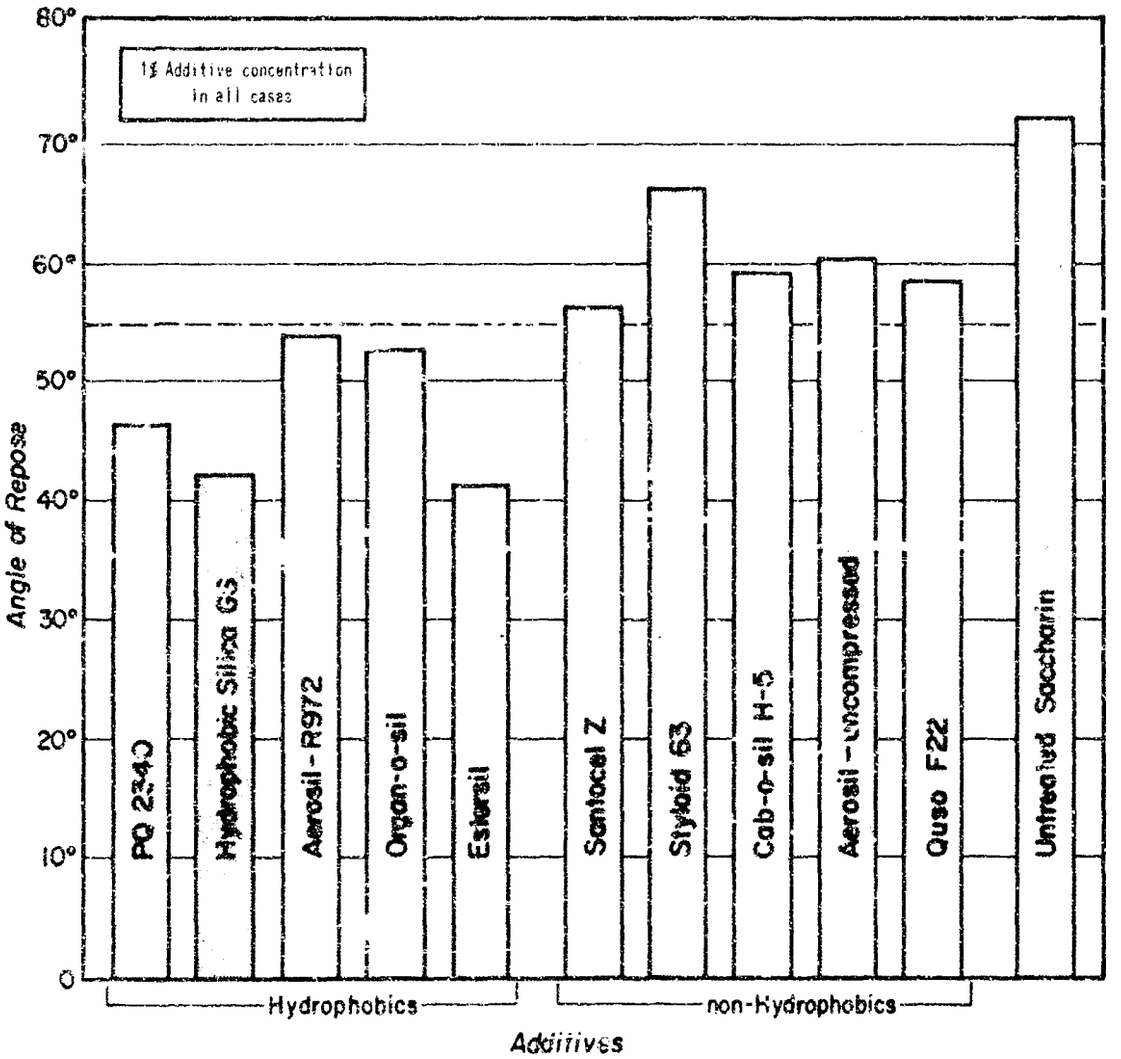


FIG. V-15 COMPARISON OF EFFECTIVENESS OF HYDROPHOBIC AND NON-HYDROPHOBIC SILICA ADDITIVES ON ANGLE OF REPOSE OF SACCHARIN POWDER

The results are given in Table V-3 below

Table V-3

EFFECT OF ADDITIVES ON ANGLE OF REPOSE

Powder	Additive	Angle of Repose
Saccharin	none	72°
	PQ 2340	46°
	Cab-O-Sil	59°
	Magnesia	63°
	Carbon Black	58°
Glass Beads	none	68°
	PQ 2340	29°
	Cab-O-Sil	35°
	Magnesia	35°
	Carbon Black	34°
Quartz	none	85°
	PQ 2340	68°
	Cab-O-Sil	70°
	Magnesia	plugs up
	Carbon Black	plugs up
Aluminum	none	plugs up
	PQ 2340	plugs up
	Cab-O-Sil	plugs up
	Magnesia	plugs up
	Carbon Black	plugs up

The trends are easier to see when presented in the following way.

CHANGE IN ANGLE OF REPOSE
(+ increase, - decrease)

Powder \ Additive	PQ 2340	Cab-O-Sil	Magnesia	Carbon Black
Saccharin	-26	-13	- 9	-14
Glass Beads	-39	-33	-33	-34
Quartz	-17	- 5	+ 5	+ 5
Aluminum	0	0	0	0

Aluminum was unaffected by any of the additives. Quartz showed slight changes; it was adversely affected by the magnesia and carbon black, but the two silicas, PQ 2340 and Cab-O-Sil, produced a positive effect by lowering the angle of repose. All four additives improved the flow of

glass beads substantially. The effect of the additives on saccharin was more selective; the PQ 2340 was the most beneficial, the magnesia was the least. Overall the PQ 2340 produced the largest reduction in the angle of repose of the powders tested.

3. Adhesive Force of Powders

a. Microthene

The results of adhesive force measurements with untreated and treated (with PQ 2340) Microthene are shown in Fig. V-16. It can be seen that the adhesive forces (intercepts on the ordinate) of untreated and 0.1% treated Microthene have higher adhesive forces than the 1.0% and the 5.0% treated powder. The calculated interparticle adhesive forces of the powder mixtures were 5.4×10^{-4} dynes, 7.0×10^{-4} dynes, 2.4×10^{-4} dynes, and 2.2×10^{-4} dynes, respectively. Therefore in a very general sense the effect of the hydrophobic silica additive was to reduce the calculated force between the particles by factors of from 1/2 to 1/3.

b. Saccharin

Measurements of adhesive forces for saccharin powder, treated with 0%, 0.5%, 1.0% and 5.0% PQ 2340 are plotted in Fig. V-17. The results were not reproducible as three determinations of the 1.0% treated saccharin gave widely different curves.

Further experimentation with the powders was made in an attempt to improve the technique, but it was finally concluded that this method for measurements on these materials was too erratic to produce good results. At best, it can only yield general trends.

c. Individual Particles

Although the measurements of adhesive force of bulk powder did not reveal any definite trends, they did suggest that the additives were effective in reducing the adhesiveness between particles. To supplement the above tests, a simple procedure, previously employed by Bailby (1921), was used to measure the adhesiveness of individual particles to a glass surface. His procedure was to drop onto a microscope slide mixed sizes

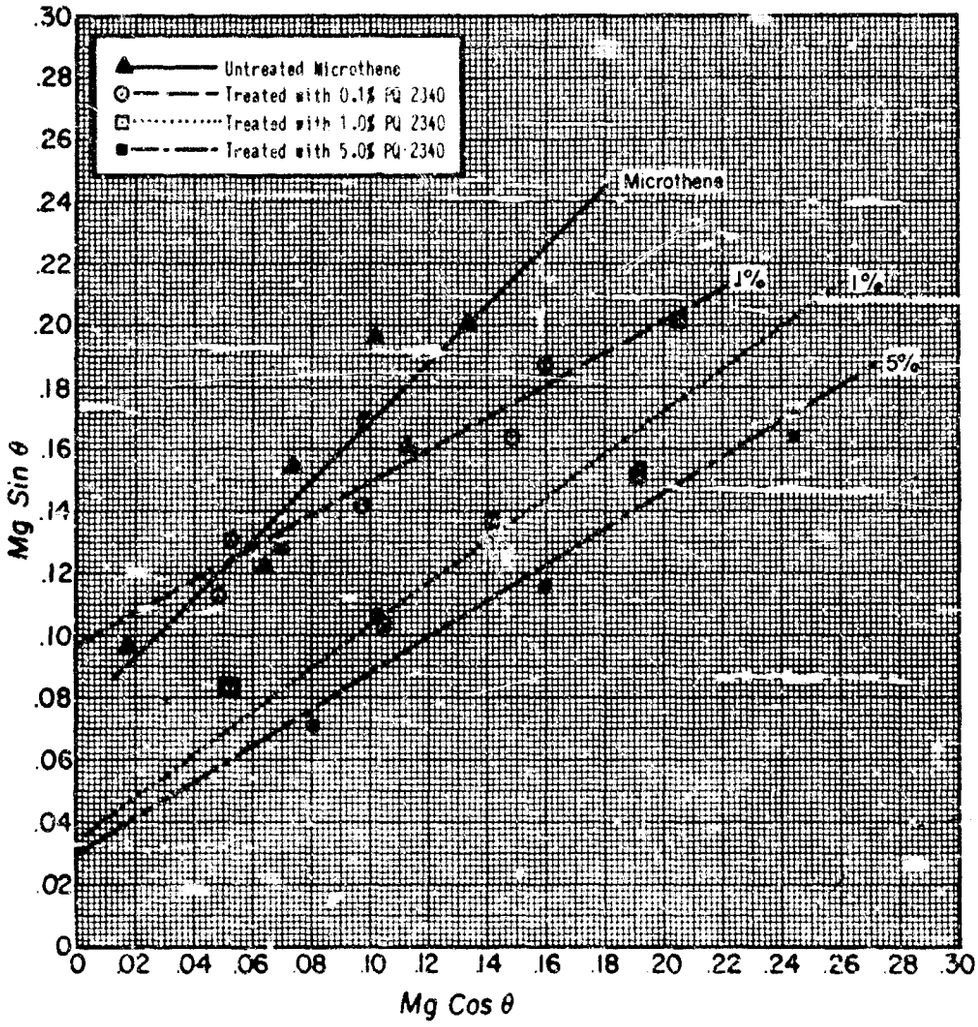
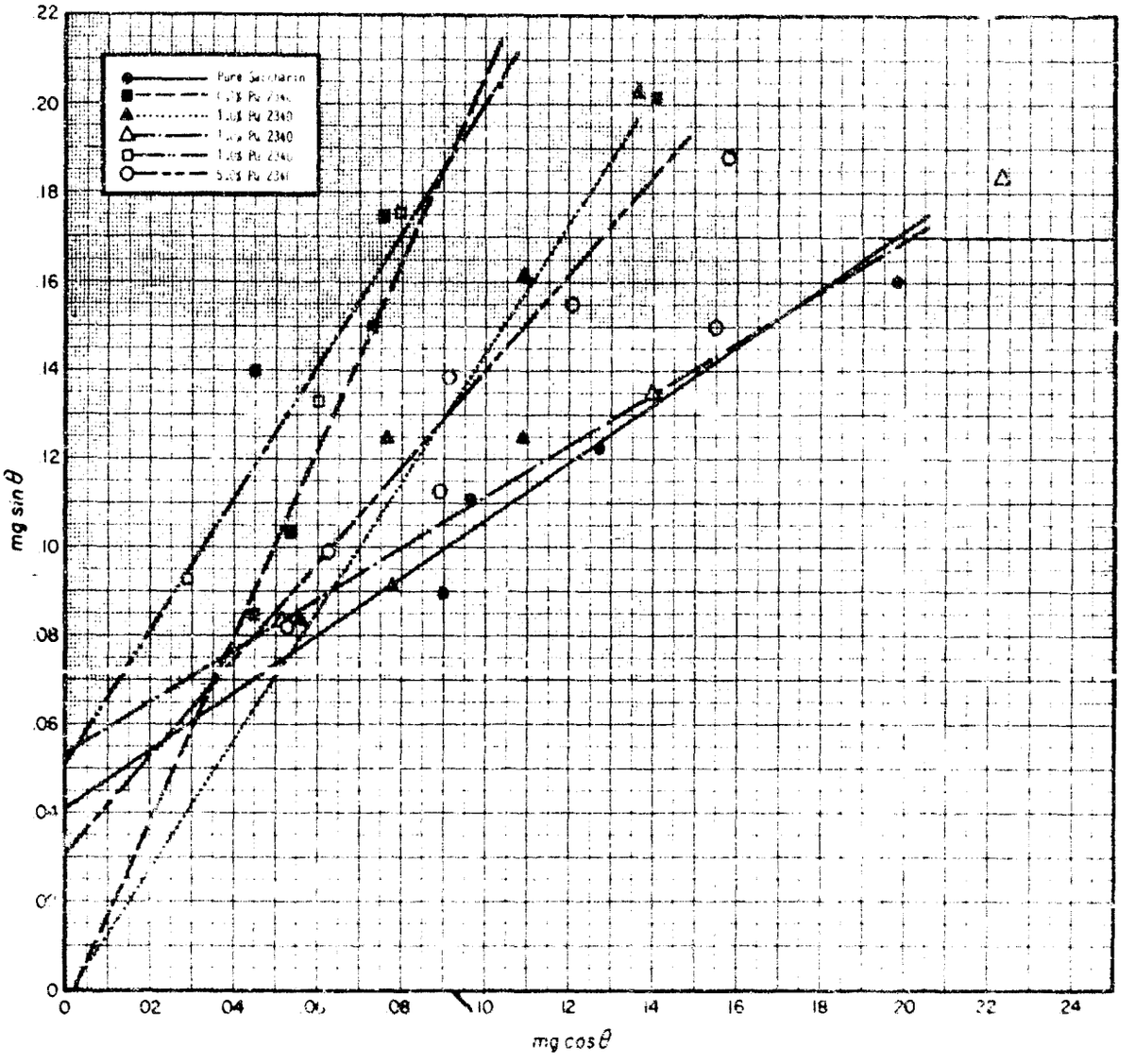


FIG. V-16 ADHESIVE FORCE MEASUREMENTS OF MICROTHENE TREATED AND UNTREATED, USING THE TILTING TABLE METHOD



F.G. V-17 ADHESIVE FORCE MEASUREMENTS OF SACCHARIN, TREATED AND UNTREATED, USING THE TILTING TABLE METHOD

of metal spheres, invert the slide, and measure the largest metal particle that would be retained against the force of gravity by the glass-metal adhesion. By assuming an area of contact, he could then calculate the adhesive force per unit area.

Beilby's measurements were repeated using spherical powders "as received" and powders which had been coated with one of the currently used additive agents to determine if there was a change in the adhesive force due to the additive. Tests were made with metal spheres: copper, 5-150 microns; stainless steel, 5-150 microns; glass beads, 1-50 microns; and Microthene 1-30 microns. The additive used was 5% PQ 2340. The following observations were made:

1) The metal spheres as received showed no agglomeration and the largest single particle adhering to the glass was approximately 15 microns diameter.

2) The silica additive increased the agglomeration tendency for metal spheres so that agglomerates in the range 50-75 microns were seen. Some tendency to form chain-like agglomerates was observed. However, these agglomerates were weak and would disintegrate when the glass slide was tapped.

3) The presence of the additive did not cause a significant change in the size of metal spheres that would support their weight by adhesion.

4) The glass bead and Microthene spheres as received were highly agglomerated, particularly the latter ones. The largest glass bead agglomerate adhering to the glass was approximately 200 microns in diameter, and the largest Microthene agglomerate was about 600 microns.

5) The silica additive decreased the agglomeration tendency for glass and Microthene spheres drastically, reducing the agglomerates to approximately 75 microns.

6) The untreated glass and Microthene spheres adhered firmly to the glass and showed no motion under the microscope when the glass slide was lightly tapped. But the treated spheres would readily slip over the surface of the glass slide on tapping and agglomerates would form and disintegrate with continuous tapping.

The original purpose of the test, detecting changes in the normal contact force between glass and primary particles, was observed by the complex forces between particles in agglomerates. There was no effect of

additive in the case of metal spheres; a single normal contact force could not be measured in the case of glass beads and Microthene Spheres. However, in the agglomerate cases, the shear force for sideways movement was certainly lowered by additives.

4. Viscosity of Aerated Powders

Measurements with the Stormer Viscosimeter were made on glass beads, saccharin, and Microthene.

a. Glass Beads

Glass beads, 44-62μ, were used for the viscosimetric measurements. The shape of the curve that is obtained by plotting the number of revolutions per second of the paddle of a Stormer Viscosimeter against the mass used to drive the rotor indicates the type of viscosity exhibited by the medium being investigated. Figure V-18 reveals the results of such measurements with 44-62μ diameter glass beads. A family of curves is obtained, each one representing a different rate of aeration. Plotted on the same coordinates is the viscosity of glycerine (1167 centipoises) obtained with the same viscosimeter. Glycerine has true Newtonian flow. The rate of shear starts from zero and increases at a constant rate as the stress increases. The rate of shear-stress curve therefore is a straight line. The shape of the curves obtained for aerated glass beads are those characteristic of a dilatant fluid. In this case, the apparent viscosity increased with increasing rate of shear. From the trend of curves, it appears that the aerated powders become less dilatant as the amount of aeration is increased.

Figure V-19 is a comparison between untreated and treated glass beads, 44-62μ diameter. Measurements were made for three different rotor weights: 50 gms, 25 gms, and 10 gms. It is seen that there is little difference between the two samples; therefore the effect of the additives was negligible on beads of this size. This confirms the results obtained with the angle of repose measurements, in which little effects were observed by the addition of the additives on 44-62μ glass beads.

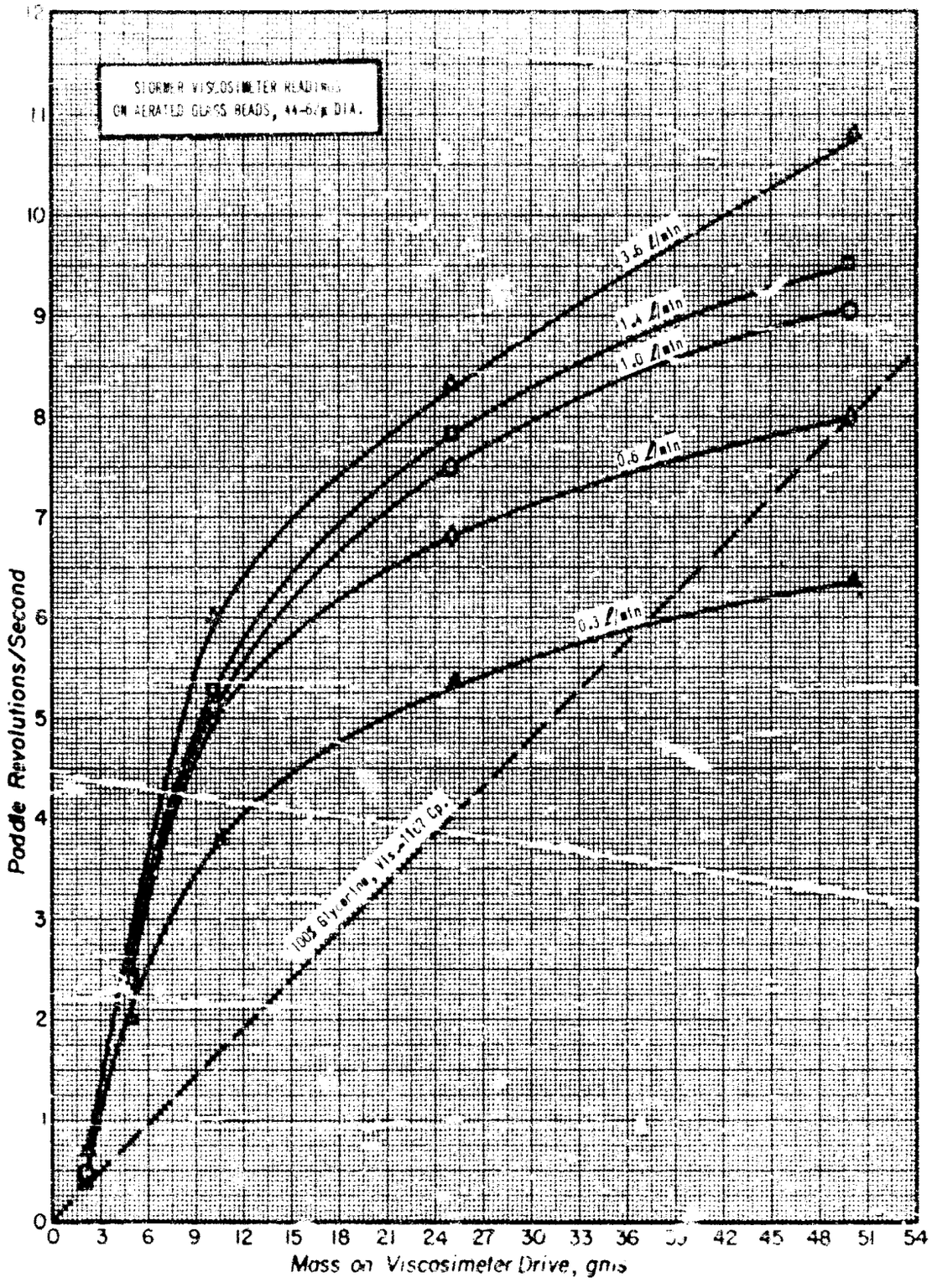


FIG. V-18 STORMER VISCOSIMETER READINGS ON AERATED GLASS BEADS, UNTREATED FOR DIFFERENT DRIVING WEIGHTS

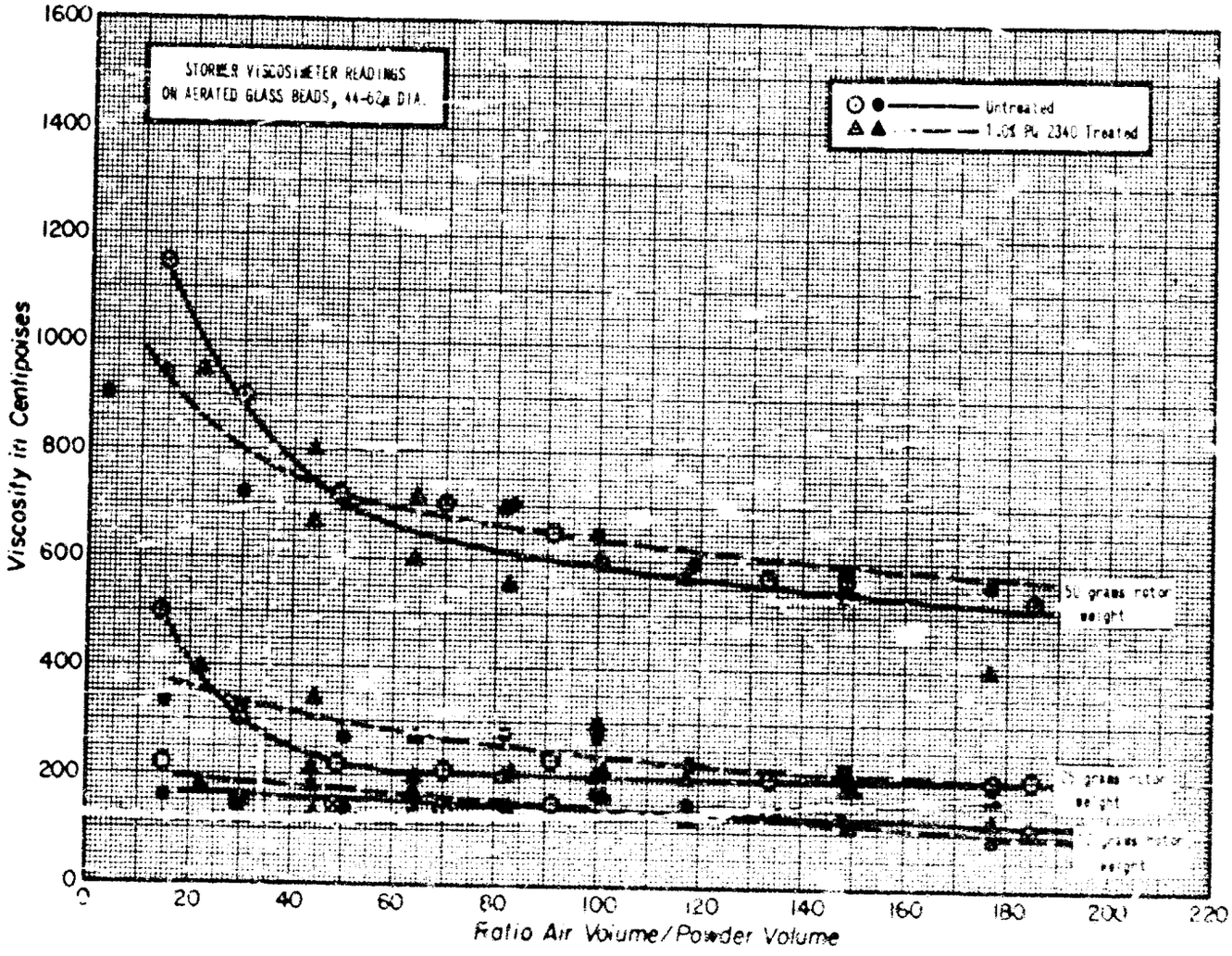


FIG V-19 STORMER VISCOSITY CURVES FOR TREATED AND UNTREATED GLASS BEADS

b. Microthene

Tests with Microthene treated with various percentages of PQ 2340 are shown in Fig. V-20. Even a small addition, such as 0.1%, was sufficient to cause a reduction in the viscosity measurements. Further additions, 1.0% and 5.0%, produced mixtures which behaved nearly the same, and both reduced the viscosity to approximately 20 centipoises at an air/powder volume ratio of 15. Thus the PQ 2340 additive caused the Microthene to become extremely fluid.

c. Saccharin

Untreated saccharin and saccharin treated with 1% PQ 2340 also gave sharply contrasting results, as shown in Fig. V-21. The untreated saccharin exhibited a very high viscosity and had a break-point at a high air/powder ratio. On the other hand, the treated saccharin was very fluid, exhibiting a low viscosity and a low air/powder ratio.

It would seem that the same processes which are responsible for this change would be of importance in the aerosolization processes. For example in Fig. V-21, the untreated saccharin required approximately 250 air volume/min/powder volume before it exhibited the break-point and progressed into the aerated powder-mass stage. On the other hand the treated powder required only 15 air volumes/min/powder volume, before it become an aerate powder mass. Stated in another way, the additive is causing the saccharin powder to become airborne at a much lower rate of air flow and with smaller shear stresses in the "disseminating device."

E. Discussion

The flowability, viscosity, adhesion and angle of repose measurements are useful to characterize the behavior of a powder. The adhesion measurements revealed only general trends, but the other three methods gave valuable information on the bulk powders. The most notable trend obtained from these tests was that a 0.5% - 1.0% addition of PQ 2340 additive to a powder gave the best results in terms of flowability and fluid motion. Additives in amounts in excess of this were found to be detrimental to the flow properties. This value is in agreement with the 1.0%

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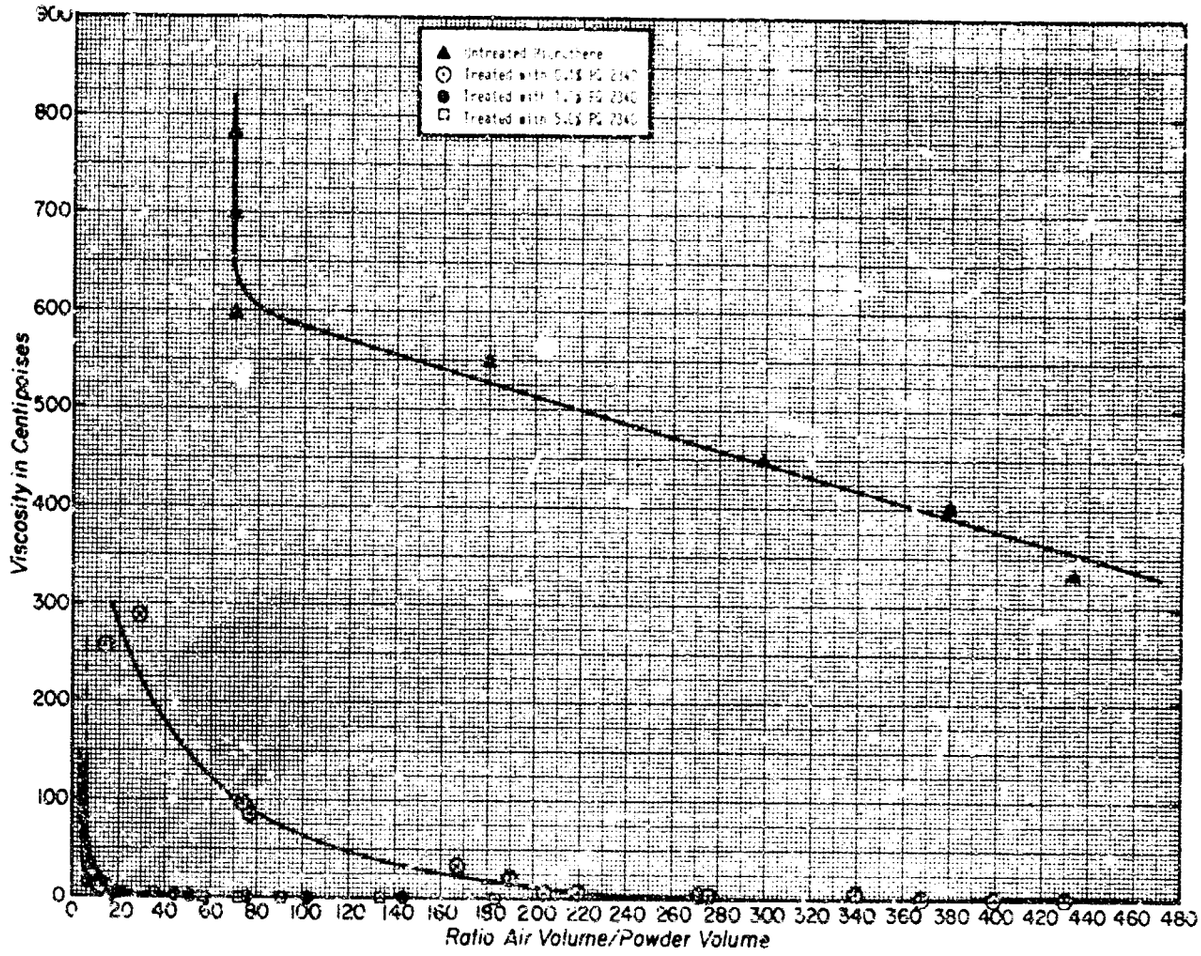


FIG. V--20 VISCOSITY CURVES FOR MICROTHENE POWDER, TREATED AND UNTREATED

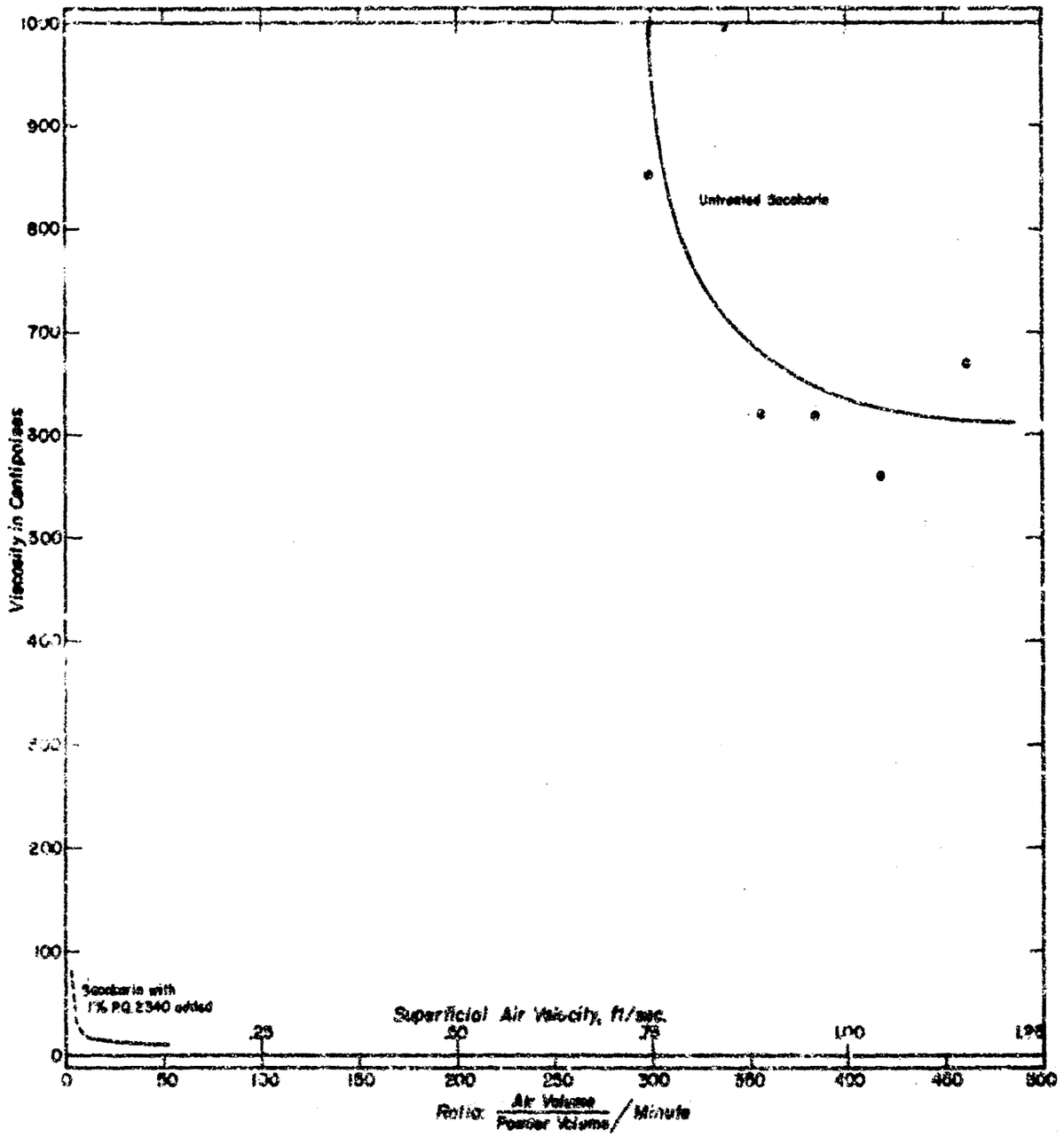


FIG. V-21 VISCOSITY CURVES FOR SACCHARIN, UNTREATED AND TREATED

value found by Craik (1958) when he treated starch powder with a magnesium oxide additive. His angle of repose measurements are reproduced in Fig. V-22 below. Based on their shear strength tests, Nash et al. (1965) found that a 1.0% addition of Cab-O-Sil to Carbowax was the optimum concentration of additive. Nash et al. (1965) calculated that a 1.0% addition was just sufficient to provide a layer of coating of Cab-O-Sil particles around each Carbowax particle.

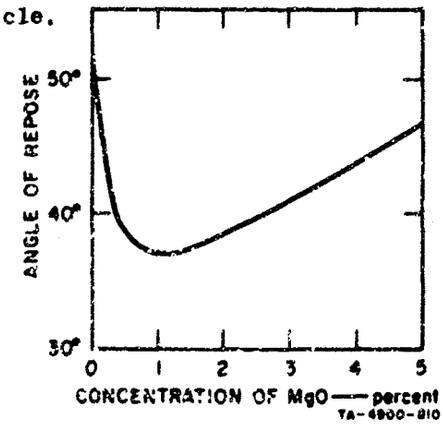


FIG. V-22 GRAPH OF ANGLE OF REPOSE MIXTURES OF STARCH POWDER AND LIGHT MAGNESIUM OXIDE IN VARYING PROPORTIONS (from Craik, 1958)

A discussion on the mechanisms by which additives function is given in the literature review on adhesive forces in Appendix B.

As will be shown in Section VIII, there are correlations between powder properties and aerosolizability. The correlation is not surprising, since the ease of flow, viscosity, angle of repose, adhesiveness and aerosolizability all depend on how much attraction there is between particles, and how much force is needed to separate them. The angle of repose measures this quality in a slightly different way than do the other tests. The angle of repose is controlled by the downward pull on a particle due to gravity and the motion is overcome by adhesion between particles in the powder mass.

A characteristic of particles in the size range 1-10μ is that arching or bridging of particles is common, forming powder beds with high void volumes and resistant to flow. The void volumes can be reduced and flowability increased by inducing the particles to move or "roll" into more stable position. A measure of the "roll" of the particles is reflected in the manner by which the flowability tests were carried out. The relative "flowability" of powders is easily observed subjectively.

but quantitative measurements vary widely depending upon the methods employed. In practice the "flowability" is best measured in the flow of a powder in the devices and under conditions designed for their use.

Finally the viscosimetric measurements reflect the movements of particles in an "expanded" powder bed. In comparison to angle of repose tests, fluid bed tests seldom have been used to characterize a powder. Yet, because of the interaction between powder and air, fluid bed tests probably come closest to simulating those properties of a powder which are associated with its aerosolizability.

[redacted]

VI DISSEMINATION METHOD

The experimental dissemination of various powders was accomplished using two general types of equipment. An effort was made to keep the equipment simple in design, and the experimental products amenable to analysis, and to permit the application of a range of energies in the experiments.

A. The "Puffer" Tube

Much of the work that had been conducted by others in the past on the aerosolization of dry powders was done at relatively high energy inputs. However, some powders may be aerosolized at very low energy inputs; for example, lycopodium powder merely needs to be dropped through a short distance in air for practically complete aerosolization. As a consequence, the difference in the aerosolizabilities of several powder treatments may be obscured by high energy inputs. Often the agglomeration tendency of the powder is more dramatic when it is disseminated at low energy such that the agglomerates dominate the aerosol. With this in mind, a low energy disseminating system was set up. In this system a vertical glass cylinder, 12.5 cm in diameter and 100 cm high was used as the settling chamber. At the top of the chamber was a small shallow cup for containing the powder. Spaced 1/2" above the cup was the outlet for a small reservoir of compressed air. The air, approximately 25cc, was released from the reservoir by means of a quick-opening valve and "puffed" at the powder, with just enough force to make the particles airborne. At the bottom of the 100 cm chamber was a slide arrangement for quick changing of microscope slides to collect the particles and agglomerates after they had settled. Figure VI-1 shows the completed assembly. Figures VI-2 and VI-3 show the top and bottom respectively.

A measure of the energy in the air blast was made by directing the air against an aluminum rod suspended in a manner so that the rod behaved as a ballistic pendulum. By measuring the maximum amplitude attained by the pendulum it was possible to calculate the energy of the air directed

FIG. VI-1 "PUFFER" SYSTEM FOR DISSEMINATING POWDERS AT LOW ENERGY. The glass settling chamber is 12.5 cm diameter and 100 cm high.

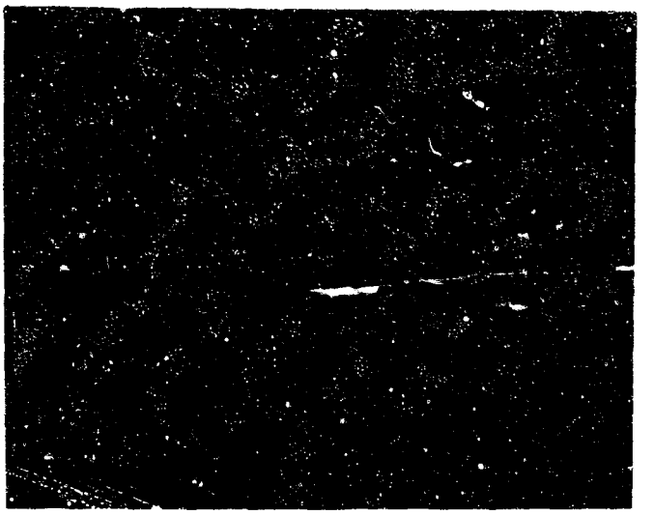


FIG. VI-2 TOP OF THE PUFFER SYSTEM SHOWING THE CUP FOR THE POWDER AND THE TIP OF THE "PUFFER" TUBE

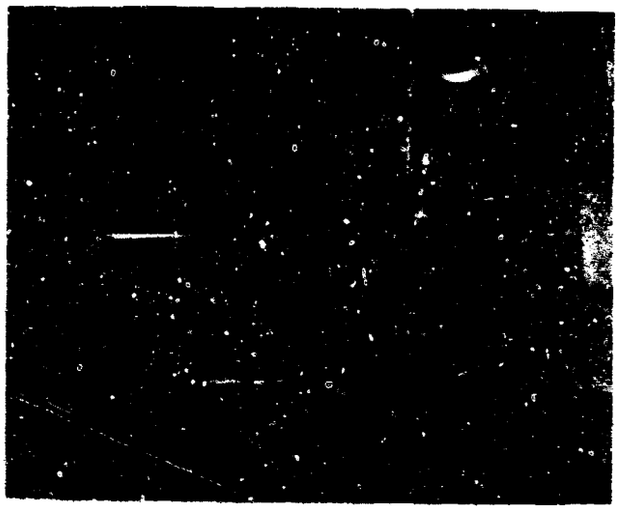


FIG. VI-3 BOTTOM OF THE PUFFER SYSTEM SHOWING THE MICROSCOPE SLIDE HOLDER CHARGER

at the powder. When the "puffer" was pressurized to 15 psi, a release energy of approximately 1.6×10^{-3} joules was obtained.

An important consideration in the choice of the puffer tube was the reproducibility of the energy released. Repeated measurements of the amplitude of the pendulum showed a variation of ±10%. An independent check of the reproducibility was made by measuring the air volume released utilizing water displacement techniques. The reproducibility was found to be ±5%.

B. The Aspiration System

The aspiration system utilizes a greater amount of energy and was used in tests where this was required. The powders were aspirated by two methods. In one, a Venturi tube was used to suck up the powders, and in the other a pressurized box with a tube outlet was used. An essential feature of these aspirators was a rotating horizontal disc for metering the powder flow. Figure VI-4 shows the Venturi tube picking up powder from a rotating disc. At times it was expedient to use the Venturi tube within a pressurized box to facilitate powder pickup, and such an arrangement is shown in Fig. VI-5. Powder was evenly spread in a narrow line near the circumference of the disc. The entrance to the tube or Venturi throat through which the powder was ejected was arranged directly over but not touching the powder.

The most frequently used powder feed and disseminator was the one shown in Fig. VI-6. Inside the 5-1/2" diameter pressurized cylinder is a 3-1/4" diameter rotating disc. The apparatus could be taken apart quickly, cleaned and reassembled. It was used extensively to test out the different types of disseminator tubes.

Whereas the puffer was a batch type disseminator, the aspirators disseminated powder continuously and over controlled air/powder ratios. The ratio of powder to air could be varied by changing (1) the air flow rate, (2) the weight of powder per unit length placed on the rotating disc, and (3) the speed of rotation of the disc.

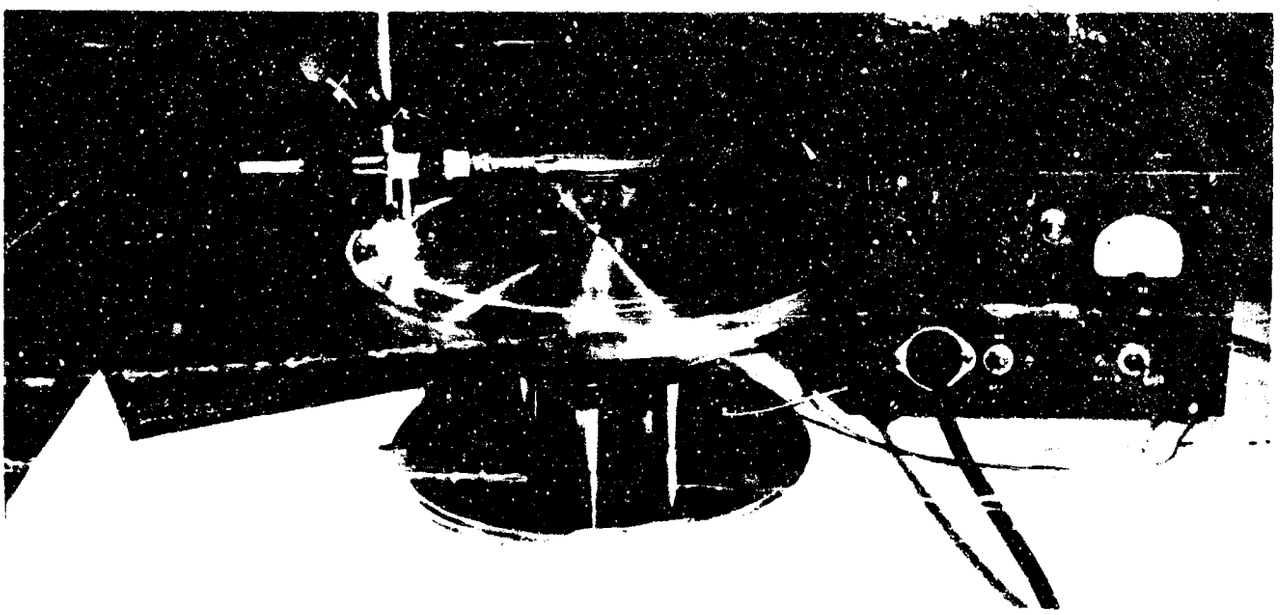


FIG. VI-4 VENTURI DISSEMINATING TUBE PICKING UP POWDER FROM A 12" DIAMETER ROTATING DISC
The powder to air ratio can be changed by altering the air flow or the rotational speed of the disc

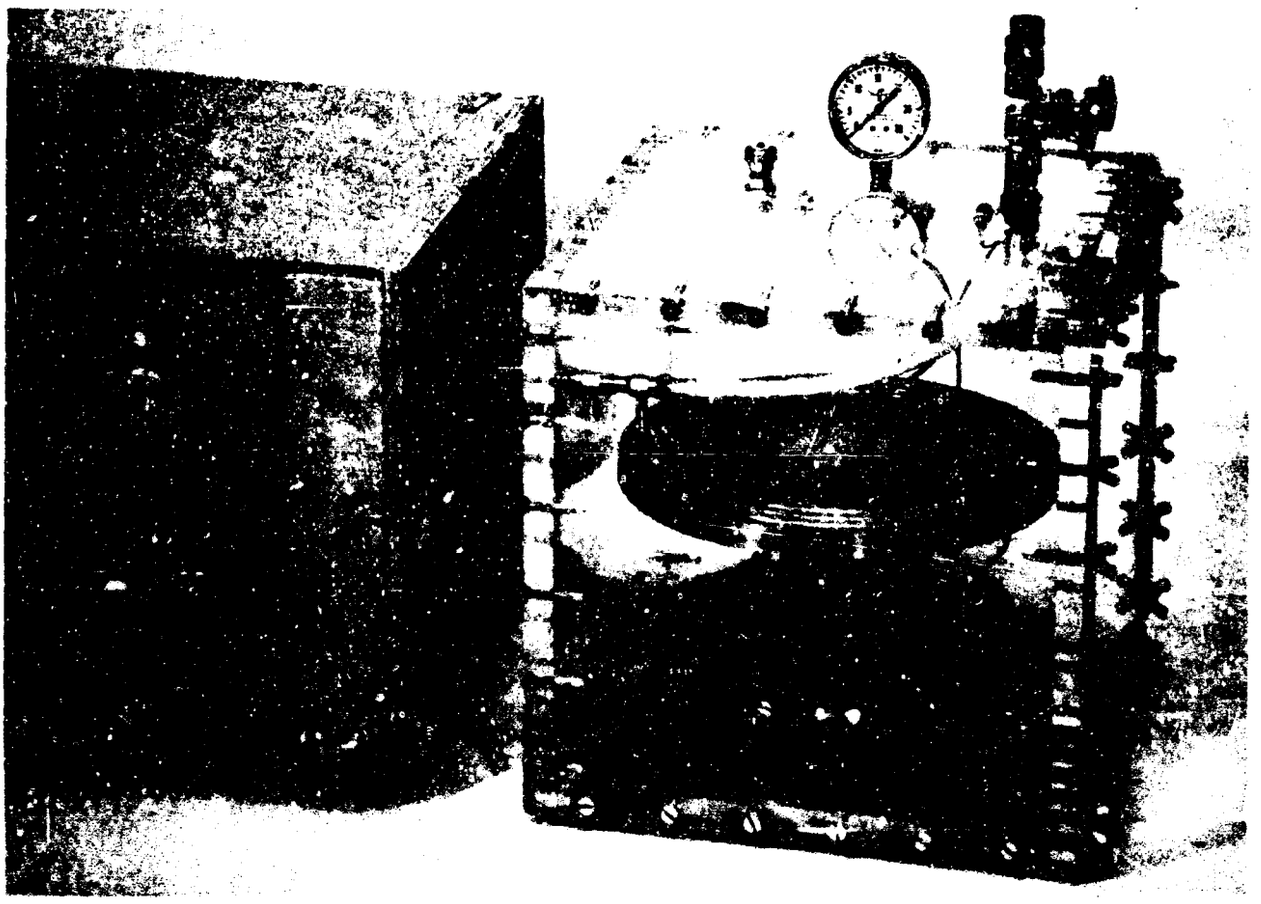


FIG. VI-5 PRESSURIZED DISSEMINATION CHAMBER WITH A 12" DIAMETER ROTATING DISC FOR METERING THE POWDER AND A VENTURI TUBE FOR DISSEMINATING IT

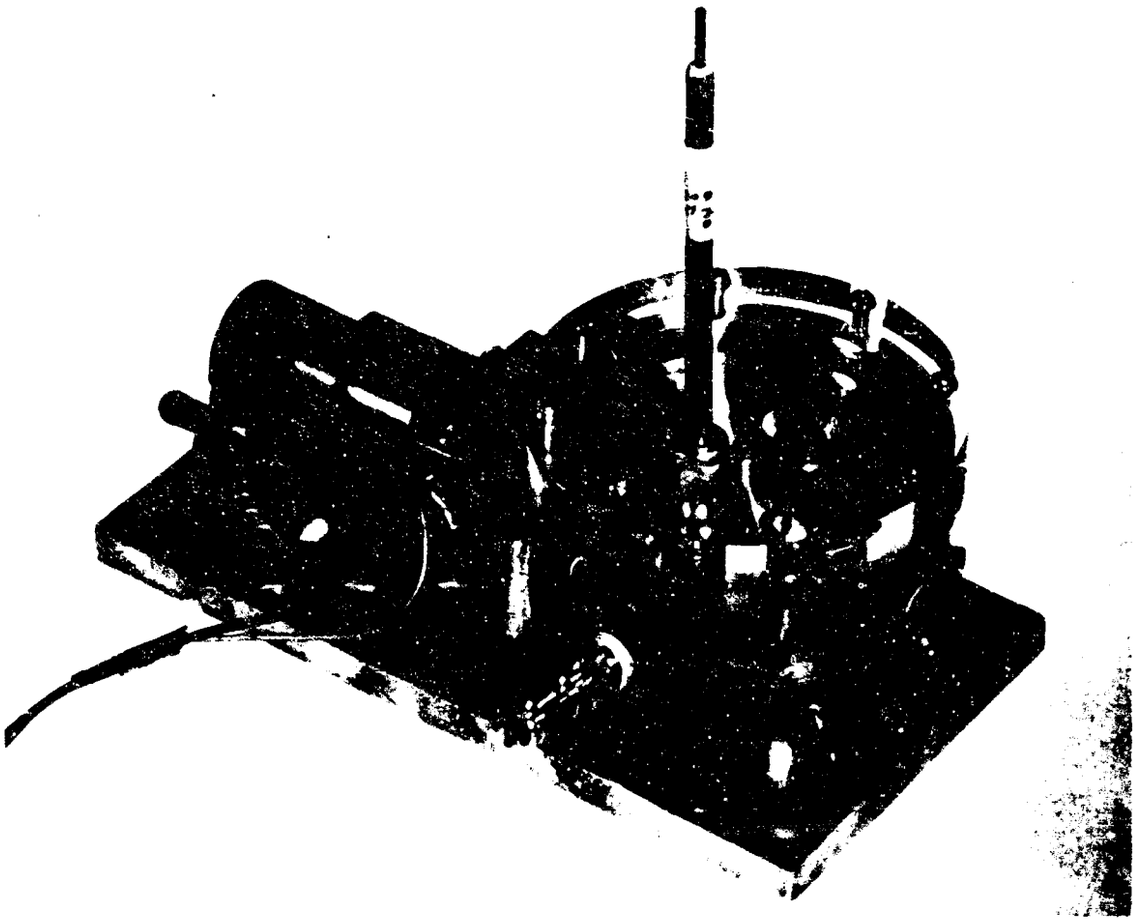


FIG. VI-6 3½ INCH DIAMETER PRESSURE CHAMBER FOR METERING AND DISSEMINATING POWDERS. The straight capillary tube shown can be removed and replaced with another tube of different configuration

The aspiration system had flexibility to permit experimenting with the different configurations of disseminating tubes. Examples of the types of tube used in experiments are shown in Figs. VI-7, VI-8, and VI-9. All the tubes were constructed from either copper or brass except in one case. The one exception is the polyethylene coiled tube shown in Fig. VI-9 which was used only to a limited extent.

The aerosol chambers used in this program varied in size and shape depending on the tests involved. The laminar flow settling chamber was most frequently used and it is described in detail in Section VII. Another chamber, a wooden box 6 feet high by 12 inches by 14 inches was also used to some extent. The inside of the box was lined with aluminum foil and electrically grounded. Figure VI-10 shows the 6 foot vertical chamber with the large pressurized powder disseminating box under it.

FIG. VI-7 VENTURI TUBE WITH TWO SUCTION PORTS FOR DISSEMINATING TWO POWDERS SIMULTANEOUSLY

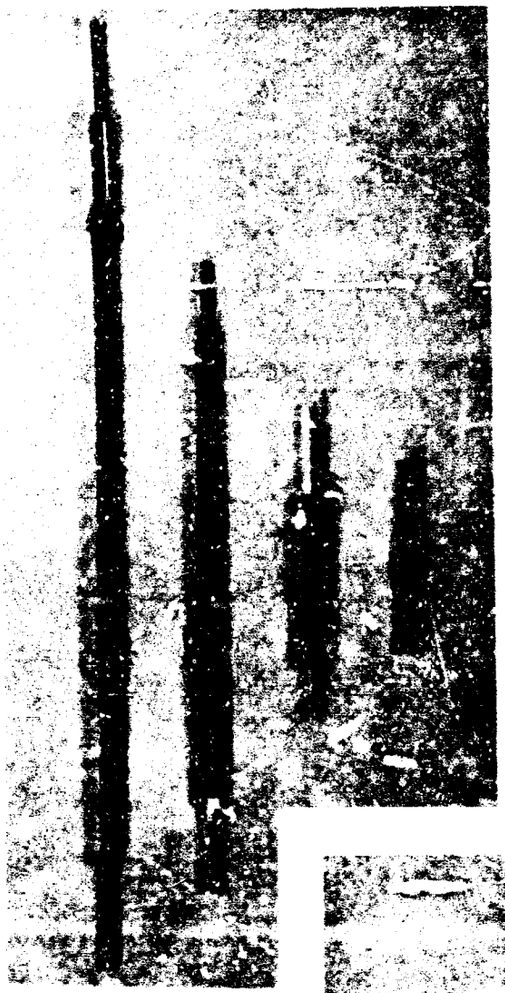


FIG. VI-8 CAPILLARY TUBES OF VARIOUS LENGTHS USED IN DISSEMINATION TESTS

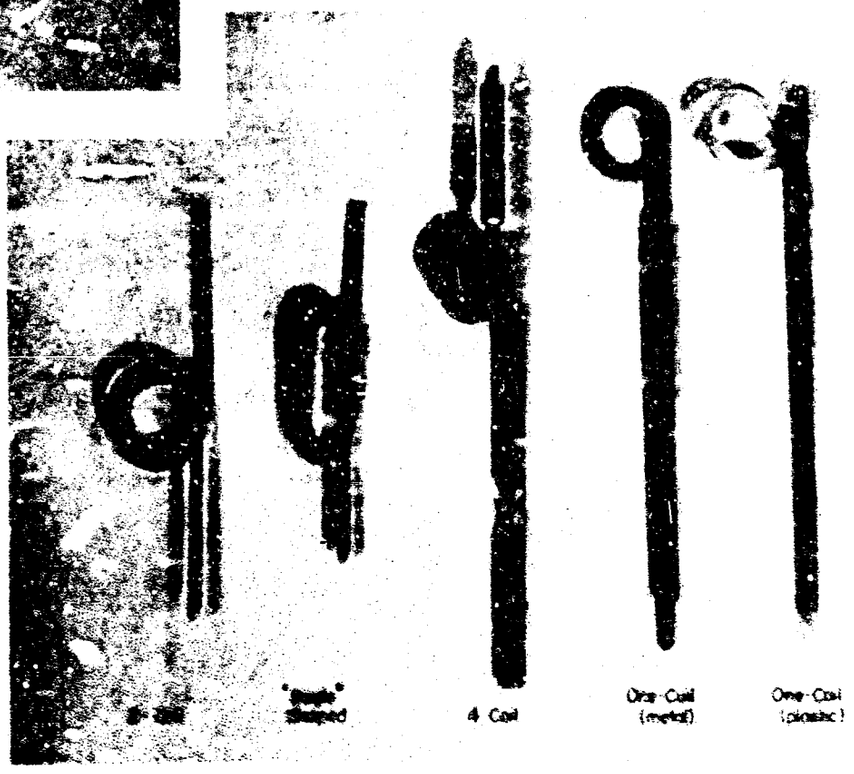


FIG VI-9 CAPILLARY TUBES COILED TO DIFFERENT CONFIGURATION. All tubes were made from brass or copper, except the tube on far right which was made from polyethylene tubing

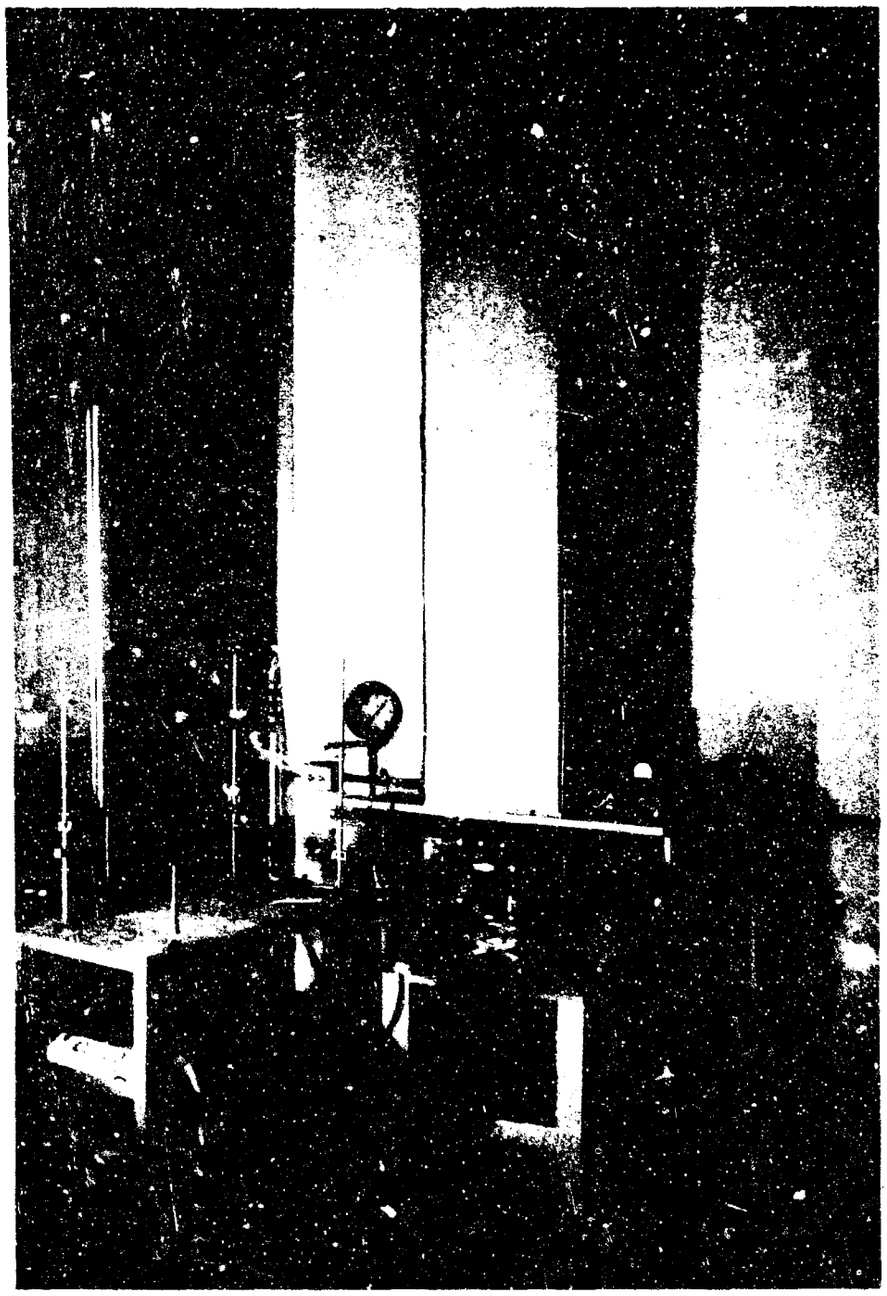


FIG. VI-10 SIX FOOT VERTICAL AEROSOL SETTLING CHAMBER WITH PRESSURIZED POWDER DISSEMINATING CHAMBER AT THE BOTTOM

VII ASSESSMENT OF AEROSOLS

A. Introduction

In the study of the dissemination of bulk powders to form aerosols it is all important to be able to measure the particle size of the aerosol and the weight fraction of the original bulk powder comprising the various size ranges of the aerosols. It is seldom possible to obtain all the desired data, except perhaps under carefully controlled and highly elaborate laboratory conditions.

At present, there seems to be no single method adaptable to all aerosolization situations. A simple and direct method is the counting and sizing of particles which have settled on microscope slides. This method provides good flexibility and choice of sampling locations. Accompanied by photomicrography, it provides a good record of results and frequently reveals information about the nature of the settled deposit that is not obtained by indirect methods. The laminar flow settling chamber described in this section allows for mass analysis of the aerosol, which, supplemented by microscopic observation of the settled particles, provides useful information on the types of particles disseminated.

B. Microscopic Counting

1. Technique

The technique for microscopic counting of particles was to view and count all particles and agglomerates within a calibrated field of the microscope eyepiece. The slide was moved such that field counts were made at random locations throughout the slide. A minimum of ten fields was counted, with the maximum number depending on the variability of the field counts. In general, 400 particles per slide were counted. From the known areas of the fields, the number of particles per mm^2 of the slide was computed.

Whenever possible, a distinction was made between an agglomerate and a primary particle and these were tabulated separately. Any two or more adhering primary particles were considered an agglomerate. The difficulty, then, occurred in the identification of primary particles. Agglomerates of glass beads, because of their spherical shape, are relatively easy to identify. On the other hand, agglomerates of saccharin or feldspar, which are irregularly shaped, were difficult to distinguish from large primary particles. For example, Figs. VII-1 and VII-2 are photomicrographs of untreated and treated feldspar respectively, which had settled on glass slides. There are obviously two large agglomerates in Fig. VII-1, but in Fig. VII-2 there are numerous small agglomerates, and even a few single particles. The difficulty lies in deciding whether the particle-agglomerates existed as a single air-borne agglomerate and scattered on the slide by impact, or whether they were small particles and agglomerates which happened to land near each other.

Therefore, assessment of aerosols by microscopic counting was limited to powders whose particles are readily identifiable as single particles or as agglomerates of particles.

The results of microscopic counting are tabulated by: the number of primary particles/mm^2, the number of agglomerates/mm^2 and the percentage of agglomerates.

The reproducibility of this assessment technique was evaluated using easily aerosolizable glass beads. Four 0.040 gm samples of glass beads with a particle size range of 5-9μ diameters, were disseminated with a gentle puff of air and allowed to fall through the 100 cm high chamber. Deposition slides were taken in the time intervals 12-60 secs and 60-300 secs, and the particles counted by three individuals. The results are tabulated in Table VII-1. The reproducibility of the individual counters in assessing a particular slide average ±14%. The reproducibility of the count--representing a measure of the overall production and evaluation of an aerosal--was calculated to be ±20%.

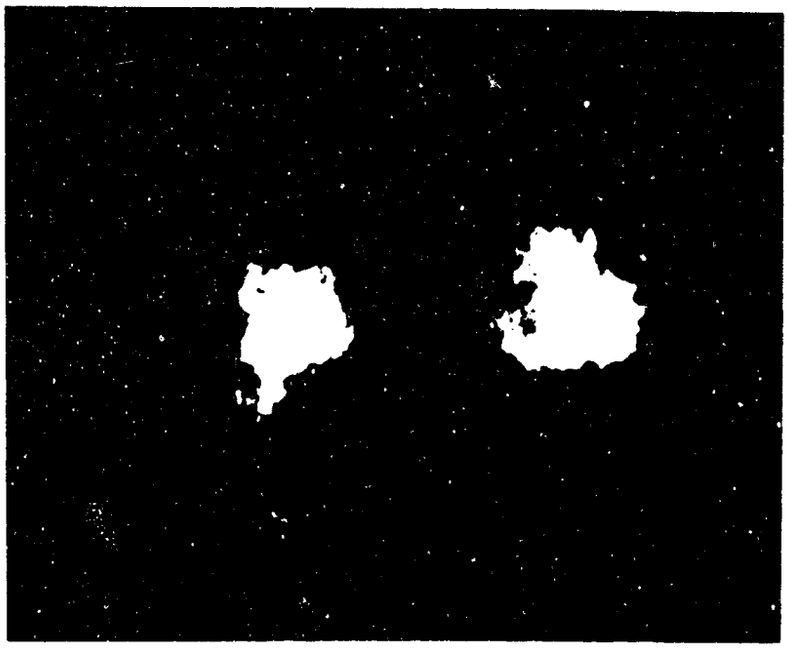


FIG. VII-1 UNTREATED FELDSPAR 0.5-50μ DIAMETER, DISSEMINATED IN THE "PUFFER" WITH THE "PUFFER" SYSTEM IN 100 cm HIGH CHAMBER

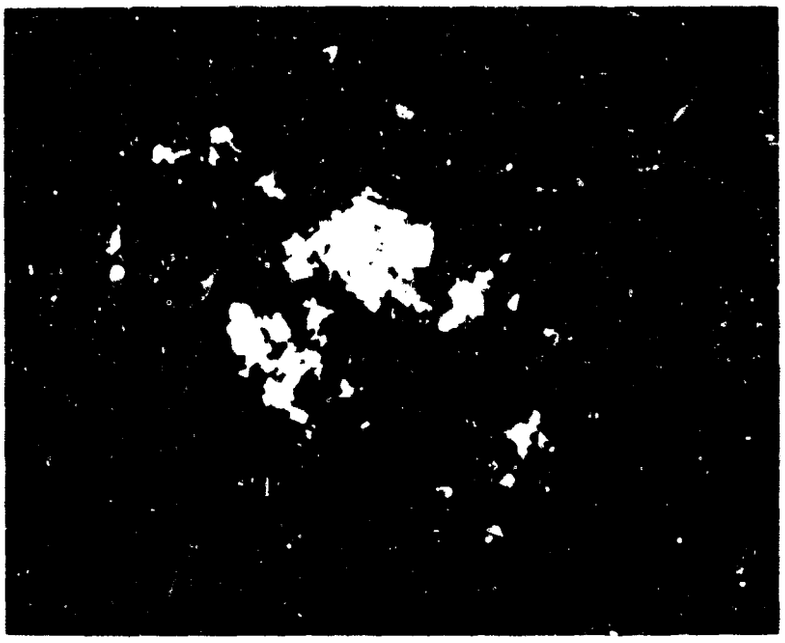


FIG. VII-2 TREATED FELDSPAR 0.5-50μ DIAMETER, DISSEMINATED AS ABOVE

Table VII-1

COUNT DATA FROM THREE COUNTERS ON FOUR REPLICATE SETS OF GLASS BED SLIDES

Set No. and Sampling Time	1st Counter			2nd Counter			3rd Counter			Averaged		
	# of Part.	# of Aggl.	% Aggl.	# of Part.	# of Aggl.	% Aggl.	# of Part.	# of Aggl.	% Aggl.	# of Part.	# of Aggl.	% Aggl.
Set No. 1 12-60 secs	482	124	26%	595	107	18%	742	121	16%	607	117	20%
Set No. 1 60-300 secs	928	198	21%	890	156	18%	1054	183	17%	958	179	19%
Set No. 2 12-60 secs	725	164	23%	602	120	20%	754	108	14%	694	128	19%
Set No. 2 60-300 secs	703	161	23%	738	120	16%	783	115	15%	741	132	18%
Set No. 3 12-60 secs	740	164	22%	843	151	18%	1087	137	13%	890	151	16%
Set No. 3 60-300 secs	1042	250	24%	1062	219	21%	1358	214	16%	1153	228	21%
Set No. 4 12-60 secs	594	122	21%	738	135	18%	800	142	18%	711	134	19%
Set No. 4 60-300 secs	1163	221	19%	1186	244	21%	1517	271	18%	1287	246	19%

2. Coincidental Settling of Particles

Another limitation of microscopic counting is that some particles always settle in juxtaposition on a slide and thus can be counted as an agglomerate, although they existed as single particles in the air stream. Thus, it is impossible by this technique to determine the extent of dissemination of presized particles in the air without some measure of the extent of coincidental settling. The effect of coincidental settling was examined using multicolored fluorescent powders, and the results gave fair agreement between observation and theory after suitable correction had been made for particle size.

The amount of overlapping of dust particles on a settling plate increases with dust concentration per unit area. For the simplest case, Irwin et al. (1949) proposed that the concentration of the primary particles on the plates can be measured by the quantity

$$\phi = \frac{\pi N D^2}{4A}$$

where N is the number of particles of diameter D (all equal size), falling at random over an area, A. ϕ is then the ratio of the sum of the areas of the particles to the area of the plates. The mean agglomerate size, is given as

$$\frac{N}{C} = \frac{4\phi}{1 - e^{-4\phi}}$$

where C is the number of agglomerates on the plate. In order to reduce the percentage error of overlapping, $100(N-C)/C$, to less than 5 percent, the number of agglomerates must be less than $0.0297\ A/D^2$.

Armitage (1949) extended the calculations to cover the more realistic cases where the particles and agglomerates are of unequal circles. However the corrections are small and for most practical applications, the estimates given by Irwin et al. are sufficient.

The above references indicated that assessment by microscopic counting is valid when the particle population is sufficiently low that the agglomerates can be distinguished from primary particles. In a recent study on aerosol reagglomeration, Knutson (1965) reported some difficulty with coincidental settling of aerosolized particles. He observed three times as many doublets and triplets as predicted by theory. The cause of this discrepancy was attributed to electrical charges on the particles. Knutson states that "an increase in the effective radius of the particles by a factor of the square root of 3 caused by particle charge would be sufficient to bring agreement between theory and experiment."

In a series of experiments similar to that of Knutson, a measure of coincidental settling was made using different colors of fluorescent zinc cadmium sulfide powder. The experiment was as follows:

An aerosol of green fluorescing zinc cadmium sulfide #3206 was first dispersed upward into a cylindrical chamber 2 feet in diameter and 7 feet high containing microscope slides at the bottom. The aerosolized particles were allowed to settle for 70 minutes at which time all the countable particles had settled. Yellow fluorescent particles #2267 were then dispersed in the same manner and allowed to settle. After complete settling the mixed-color agglomerates were counted. The results are tabulated in Table VII-2.

Some operator judgment was required to distinguish the agglomerates from single particles. Examples of the method of tabulating single particles and agglomerates are given below.

Microscope Field

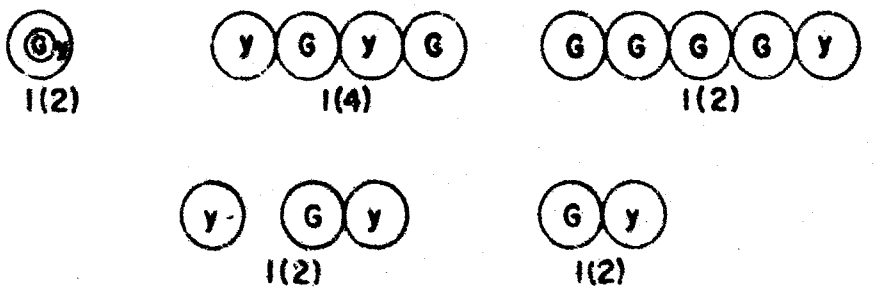


Table VII-2

DATA ON TWO-COLOR AGGLOMERATES STUDY

Test No.	Amount of Powder Aerosolized	Particles	Total Count	Average Count/ mm^2 ±	% Total
160	.1 gm Green .25 gm Yellow	Yellow	2261	640.8 ± 85.9	85.45%
		Green	312	88.4 ± 18.6	11.79%
		Aggl. (each = 1)	33	9.4 ± 5.4	
		Y in Aggl.	38	10.8 ± 6.6	1.44%
		G in Aggl.	35	9.9 ± 6.9	1.32%
161	.15 gm Green .25 gm Yellow	Yellow	2350	511.1 ± 77.5	70.39%
		Green	865	184.9 ± 18.9	25.87%
		Aggl. (each = 1)	60	14.5 ± 9.9	
		Y in Aggl.	65	15.8 ± 11.0	1.94%
		G in Aggl.	63	15.1 ± 9.7	1.89%
162	.25 gm Green .25 gm Yellow	Yellow	3116	591.8 ± 79.2	68.01%
		Green	1322	251.8 ± 28.4	28.85%
		Aggl. (each = 1)	70	13.3 ± 4.1	
		Y in Aggl.	73	13.9 ± 4.4	1.59%
		G in Aggl.	71	13.7 ± 4.4	1.55%
163	.30 gm Green .25 gm Yellow	Yellow	1666	485.4 ± 82.3	50.20%
		Green	1435	418.1 ± 85.9	43.24%
		Aggl. (each = 1)	99	28.8 ± 14.1	
		Y in Aggl.	115	33.5 ± 15.6	3.46%
		G in Aggl.	103	30.0 ± 14.9	3.10%
164	.35 gm Green .25 gm Yellow	Yellow	2256	612.7 ± 123.3	52.93%
		Green	1735	473.8 ± 62.7	40.71%
		Aggl. (each = 1)	133	36.3 ± 10.0	
		Y in Aggl.	136	37.1 ± 10.5	3.19%
		G in Aggl.	135	36.8 ± 10.8	3.17%
165	.40 Green .25 gm Yellow	Yellow	2754	799.7 ± 58.0	53.49%
		Green	1995	581.3 ± 93.6	38.75%
		Aggl. (each = 1)	192	53.9 ± 10.7	
		Y in Aggl.	201	58.6 ± 10.7	3.90%
		G in Aggl.	199	58.0 ± 10.7	3.86%

In this guide each circle represents a particle. The first number under each representation of agglomerate grouping was recorded as the number of agglomerates. The number in the parentheses is the number of particles in the agglomerate. A single bead-like chain of a single color was considered as one particle.

Providing suitable corrections are made for the size of particles, reasonably good agreement between the number of doublets found and the number calculated can be obtained as revealed by the following calculations.

After the first cloud of particles has settled, the probability that a doublet will be found between a particle of diameter D_1 from the first cloud and a particle of diameter D_2 from the second cloud will be given by

$$P_{D_1,D_2} = \frac{N_{D_1} \pi (D_1 + D_2)^2}{4A} \qquad (1)$$

where

P_{D_1,D_2} = probability of a doublet.

N_{D_1} = number of particles for cloud 1 with diameter D_1.

D_1 = particle diameter for cloud 1.

D_2 = particle diameter for cloud 2.

A = area over which particles have settled.

The number of mixed doublets found between particles of diameter D_1 and D_2 will be

$$N_{D_1,D_2} = N_{D_1} N_{D_2} \frac{\pi (D_1 + D_2)^2}{4A} \qquad (2)$$

If each cloud is made up of particles with a wide range of particle sizes the total number of mixed doublets $N_{1,2}$ will be given by

$$N_{1,2} = \frac{\pi}{4A} \int_0^\infty \int_0^\infty N_{D_1} N_{D_2} (D_1 + D_2)^2 dD_1 dD_2$$

$$= \frac{\pi N_1 N_2}{4A} \left[\overline{D_1^2} + 2\bar{D}_1\bar{D}_2 + \overline{D_2^2} \right] \qquad (3)$$

where N_1 and N_2 are the total numbers of particles from the clouds 1 and 2 respectively.

Equation 3 will overestimate the number of mixed colored doublets because there are some doublets formed of the same color, and the second color particle that settles on this doublet will in fact be a triplet rather than a doublet. This overestimation is not of great consequence. In practice, it is difficult to distinguish between single particles and doublets of the same color. Also particles of the same color vary widely in size because of incomplete dispersion. It may therefore be assumed that doublets of the same color can be counted as single particles, and Equation 3 may be used to estimate the number of mixed color agglomerates providing proper adjustment is made for the mass mean diameter and the geometric standard deviation of the particle sizes observed on the settling slide.

For the zinc-cadmium sulfide experiments described in the text, the particles per gram (PPG) of the powder were computed from the number of particles settling to the floor of the chamber divided by the weight of powder aerosolized, and the mass mean diameter (MMD) in microns computed from

$$(\text{MMD})^3 = \frac{6 \times 10^{12}}{\pi\rho(\text{PPG})}$$

where

ρ = particle density = 4 gm/cm^3 .

These diameters are given in Table VII-3. The MMD for the yellow fluorescent particles ranged from 7.8 - 9.2μ, while those from the green fluorescent particles ranged from 10.0 - 11.0μ. These diameters are much larger than the MMD of the two fluorescent particle colors determined by complete aerosolization of these materials or by complete dispersion on a microscope slide. Complete dispersion reveals an MMD 3.1μ for the yellow and 3.7μ MMD for the green particles.

A comparison between the observed and calculated number of mixed color agglomerates was made by first assuming all particles of each color to be the same size and equal to the mass mean diameter obtained from the settling experiments. The calculation was then corrected for the spread in particle size. For uniform sizes Equation 3 reduces to

$$N_{1,2} = \frac{\pi N_1 N_2 (D_1 + D_2)^2}{4A} \qquad (5)$$

The numbers of two-color agglomerates calculated and observed for each trial are shown in Table VII-3, Columns 11 and 12. With the exception of Test 162, the ratio of the calculated to the observed number of agglomerates is nearly constant ranging from 2.23 to 2.58 with a mean value of 2.39. That is, the number of agglomerates/mm^2 calculated from the mean MMD of the two materials, and a geometric standard deviation of 1.0 was 2.39 times greater than the observed number of agglomerates.

However, the mass mean diameter of the particles having a range of sizes will be larger than the surface mean diameter and a correction must therefore be made which includes the geometric standard deviation of the particle sizes on the slides. This may be measured by sizing all the particles. An alternate method is to transform the MMD to the mean square diameter by the relations:

$$\bar{D} = D_g e^{1/2(\ln \sigma_g)^2} \qquad (6)$$

$$\overline{D^2} = D_g^2 e^{2(\ln \sigma_g)^2} \qquad (7)$$

Table VII-3

CALCULATED AND OBSERVED NUMBER OF MIXED AGGLOMERATES FROM SETTLING OF YELLOW AND GREEN FLUORESCENT PARTICLES

Test No.	Weight of Powder Disseminated		No. of Particles/mm² *		Particles/Gram ($\times 10^8$)		MMD (μ)		Effective Mean Diameter† (mm^2) $\times 10^{-4}$	Number of Aggl./mm²		Ratio of $\frac{\text{calc}}{\text{obs}}$ Aggl.
	Yellow	Green	Yellow	Green	Yellow	Green	Yellow	Green		calc§	obs	
160	0.25	0.10	651.6	98.3	7.609	2.869	9.56	11.83	3.272	21.0	9.4	2.32
161	0.25	0.15	526.9	200.0	6.151	3.892	9.19	10.70	3.107	32.7	14.5	2.26
162	0.25	0.25	605.7	265 5	7.071	3.100	8.77	11.55	3.243	52.2	13.3	3.92
163	0.25	0.30	518.9	448.1	6.058	4.360	9.24	10.31	3.002	69.8	28.8	2.42
164	0.25	0.35	649.8	510.6	7.587	4.2[illegible]8	8.57	10.39	2.823	93.7	36.3	2.58
165	0.25	0.40	858.1	639.3	10.02	4.667	7.81	10.08	2.514	137.9	55.9	2.47
									(Omitting test No. 162) av.			2.39

* Includes particles in agglomerates.

† Effective mean diameter = $(\pi/4) \times 10^{-6}[MMD_Y + MMD_G]^2$.

§ The calculated number of agglomerates/mm² was based on the mean MMD of the yellow and green particles and a geometric standard deviation of 1.0.

NOTE: By assuming a geometric standard deviation of particle sizes of 2.2 for the two materials, the corrected ratio of the calculated number of agglomerates (Equation 11) to the observed number will be approximately unit.

where

$\overline{D}$ = number mean diameter

$\overline{D^2}$ = mean square diameter

D_g = geometric mean diameter

σ_g = geometric mean standard deviation.

But

$$MMD = (\overline{D^3})^{1/3} = D_g e^{3/2(\ln\sigma_g)^2} \tag{8}$$

Therefore

$$\overline{D} = (MMD)e^{-(\ln\sigma_g)^2} \tag{9}$$

$$\overline{D^2} = (MMD)^2 e^{-(\ln\sigma_g)^2} \tag{10}$$

Since the calculated MMD's for the two materials differ only slightly for the two colors, we may assume that $MMD_1 = MMD_2$ and also that $\sigma_{g_1} = \sigma_{g_2} = \sigma_g$, so that Equation (3) may be written

$$N'_{1,2} = \frac{\pi N_1 N_2 (MMD)^2}{2A}\left[e^{-(\ln\sigma_g)^2} + e^{-2(\ln\sigma_g)^2}\right] \tag{11}$$

where $N'_{1,2}$ is the corrected number of mixed-color agglomerates based on a log normal distribution having σ_g other than unity such as assumed in Equation 5.

The calculated number of agglomerates in Tablve VII-3 was calculated from Equation 5 and is equivalent to using the average value of the MMD for the two colors computed from Equation 4 from the PPG in each experiment. That is,

$$N_{1,2} = \frac{\pi N_1 N_2 (MMD)^2}{4} \tag{12}$$

The ratio of $N_{1,2}$ to the corrected value $N'_{1,2}$ is

$$\frac{N_{1,2}}{N'_{1,2}} = \frac{2}{\left[e^{-(\ln\sigma_g)^2} + e^{-2(\ln\sigma_g)^2}\right]} . \qquad (13)$$

For the ratio $N_{1,2}/N'_{1,2}$ to be 2.39, the geometric standard deviation calculated to be 2.2. A σ_g of 2.2 is not an unreasonable standard deviation, since the size of the settled particles and agglomerates on the slides ranged from about 0.5μ to 200μ. This calculation leads to the conclusion that the number observed and calculated mixed-color agglomerates can, with sufficient observations, be brought into close agreement.

C. Photography of Falling Particles

A brief investigation was made into the direct photography of falling aerosols, in an attempt to identify the particles and agglomerates while they are airborne.

The method consisted of photographing the aerosolized particles with an extended bellows camera using a stroboscopic light source to illuminate the particles. In this way, each falling particle produces a series of dots on the photograph. By knowing the magnification of the camera and the speed of the flash, a direct measure could be made of the rate of fall. Figure VII-3 is a photograph of falling p-amino-benzoic acid powder. No effort was made to obtain complete dissemination of the particles; consequently agglomerates greater than 100μ were observed. The white line in the center of the photograph is a 2.1-mm diameter rod to serve as calibration of the overall magnification. In this case, the magnification was 1.7.

With a flash rate of 83.3 flashes per second, the fall rate of particle per sec is = (Spacing between dots) (83.3)/1.7. The series of small dots indicated by A had a fall rate of 4.8 cm/sec or an aerodynamic diameter of 40μ. The line of dots indicated by B had a fall rate of 80 cm/sec or an aerodynamic diameter of 185μ. The size of dots in the photograph bears no relationship to the size of the particles. The images merely represent scattered light, and because accurate focus would be

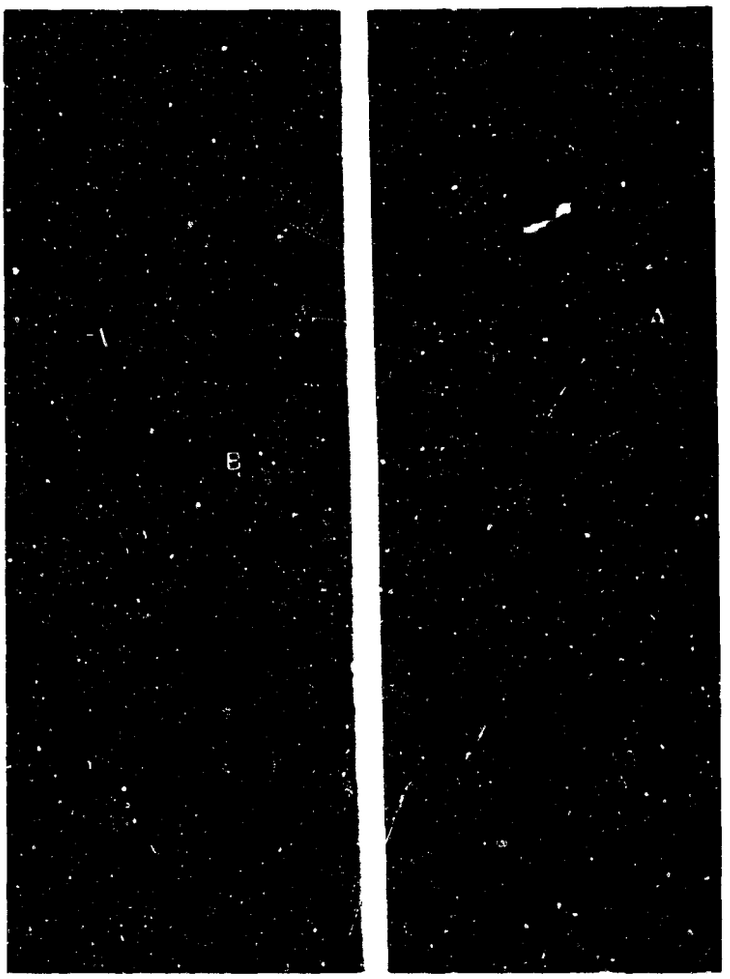


FIG. VII-3 PHOTOGRAPH OF PARTICLE TRACKS
OBTAINED BY STROBOSCOPIC PHOTOGRAPHY

only a coincidence, there is no relation between dot size and particle size. The sole criteria of aerodynamic particle size is dot spacing. It is obvious from this photograph that the aerosol contained many large agglomerates. For slower falling particles (10μ diameter) slower speeds of flashing can be used, although it would probably be difficult to record traces of particles below 10μ with the equipment available. This approach was not expanded because it did not seem practical to expend much effort to develop an assessment technique that offered limited information and which could involve elaborate equipment for the particles of interest (1-10μ).

D. Laminar Flow Settling Chamber

1. Introduction

The agglomerates resulting from dissemination of dry powders are difficult and exasperating to size, and consequently the assessment of these aerosols by agglomerates sizes is plagued with uncertainty. Even in the situations where the general dimensions of the agglomerates can be measured, the size itself does not reveal very much information. Because of the loose formation of the agglomerates, sometimes in the form of chains, it is nearly impossible to calculate the aero-dynamic properties of the particles by optical measurement of the particle sizes. To illustrate, a 10μ particle of density 1.0 gm/cc will settle at the rate of 0.3 cm per sec. A loosely bound agglomerate with an average density of 0.1 gm/cc and a diameter of 31.6μ would also settle at 0.3 cm/sec; however, the mass of the agglomerate would be 3.16 times that of a single solid particle. Therefore for most practical applications of aerosols, it is their aerodynamic properties which are of interest to the user.

For these reasons, a laminar flow settling chamber which measures the size distribution of particles and agglomerates in terms of their aerodynamic properties as they exist in the air stream, was developed as a means for assessing the aerosolization process.

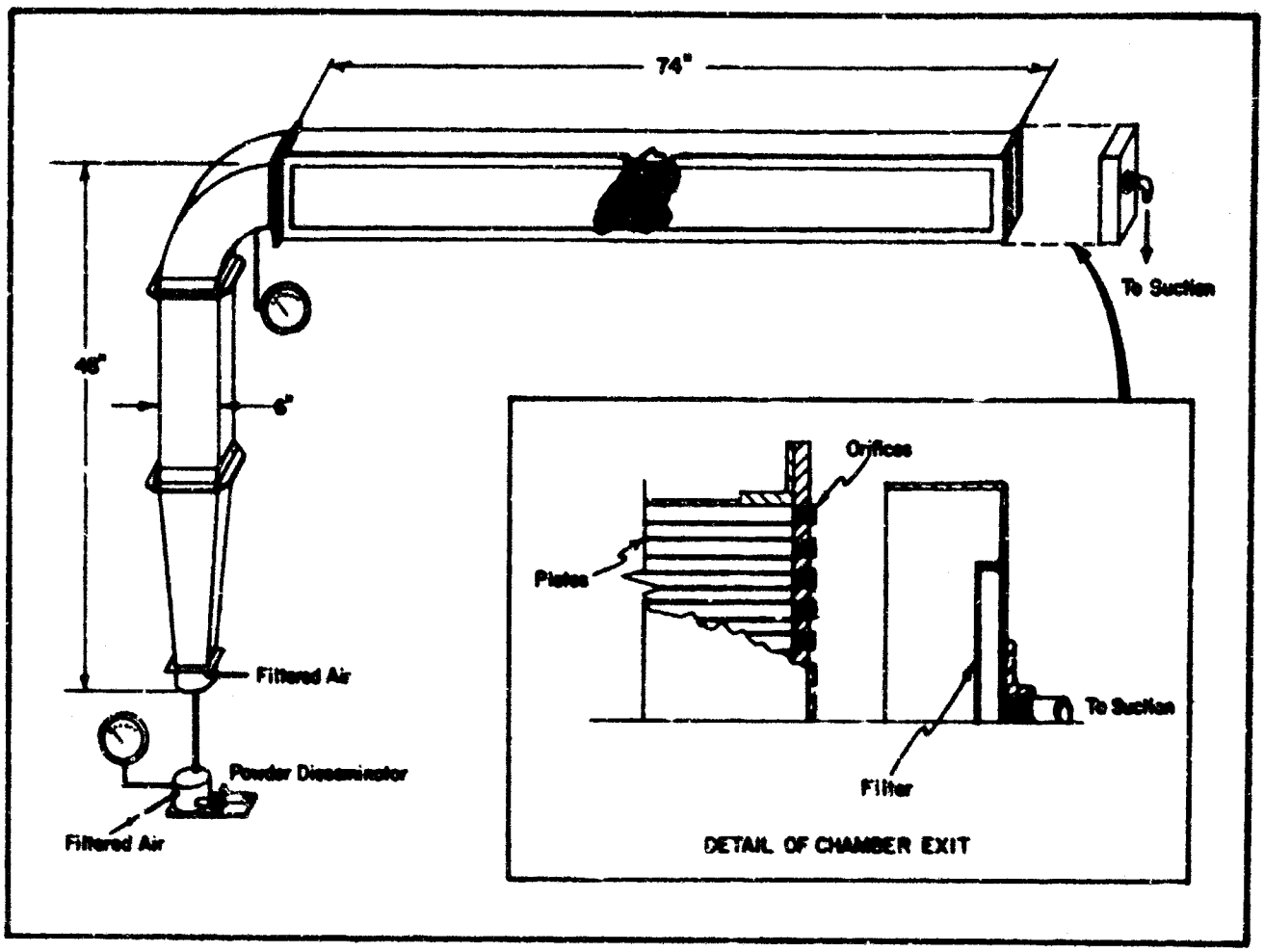
2. Description and Operation of the Chamber

The laminar flow settling chamber, in its most simple form, is a long, horizontal shallow chamber through which aerosol-laden air is passed. More complex forms have multiple number of settling plates. As the air passes through the chamber, the aerosol particles settle out, and the location where a particle settles depends on the air flow and the gravity settling rate of the aerosolized particles.

The chamber employed in this work is shown schematically in Fig. VII-4. It contained 22 settling plates which are six feet long (183 cm) and six inches wide. The vertical spacing between each plate is 0.25 inch (0.623 cm).

The duct section of the chamber for introducing the aerosol to the plates was comprised of three detachable parts. The aerosol was introduced into the chamber at the bottom of the tapered duct section, as illustrated in Fig. VII-4. The aerosol may be generated by any of the various powder disseminators. One powder disseminator, described in Section VI and shown in Fig. VI-6, was used extensively in the chamber tests. The period of aerosol generation could be varied from a few minutes to a few hours, depending on the type of powder disseminator. The air flow through the chamber was adjusted to control the particle size range over which the analysis was desired. For most of the work reported here, the air introduced with the powder was in the range of 10-33 ℓ/min. Dilution air was added such that the total flow through the chamber was in the range of 70 to 100 ℓ/min. At 100 ℓ/min, the air velocity was 7.1 cm/sec, at a Reynolds No. of 56 between plates. The air flow leaving the chamber was adjusted by means of a vacuum pump to equal the air flow entering the chamber so that the air pressure inside the chamber was essentially atmospheric.

After a test, the plates were removed individually and the material on measured sections of the plates washed off and analyzed by chemical methods. Data for particle size distribution on the plates was obtained by graphical methods from a knowledge of the weights of powder found on each section of the plates. It was found that material on all plates



VII-4 MULTI-PLATE LAMINAR FLOW SETTLING CHAMBER FOR DISSEMINATION AND ASSESSMENT OF AER

need not be analyzed since analysis of five plates gave representative results for 22 plates. The quantity of powder collected on the filter and in the duct was measured separately. For a complete mass balance, the duct section was washed and the amount of powder left there computed. If some powder were left in the aerosol generator, that too had to be analyzed. The total powder found was then equated to the weighed amount introduced into the aerosol generating system. Thin microscope cover slips were placed at various locations throughout the chamber to permit visual observation of the type of particles or agglomerates which settle.

As constructed, the chamber's overall dimensions were a compromise designed to fit space requirements and to permit ease of manipulation.

3. Principle of Operation

The principle of operation is essentially identical to that of the Oden sedimentation balance (Oden 1916), which is a batch device for measuring sediment as a function of time. The laminar flow chamber is a continuous-flow device in which sediment is measured as a function of chamber length. And while the chamber is a device for measuring distribution, it does not strictly separate particles into size range, although there is a trend in that direction.

The laminar flow settling chamber, as designed, was limited to analyses over the size range of 2 to 50 microns diameter in most cases. For smaller particles the settling velocities of the particles are so low that the length of the chamber would be exceedingly long. A filter was placed at the end of the settling plates to capture those particles which did not settle out. For particles larger than 50 microns, it was impossible to obtain a reasonable physical spread of material on the plates without using an air velocity sufficiently great to produce turbulent air flow through the chamber.

For valid operation of the chamber, three basic conditions must be fulfilled: (1) the aerosol cloud at the entrance of the plate section should be uniform so that analysis of a few plates will yield a representative sample of the aerosol; (2) the average horizontal velocity also

has to be the same in each channel and this condition was achieved by using many shallow channels of equal dimensions and restricting orifices at the outlet of each channel; (3) there should be no vertical mixing of the fluid. This last condition requires that the air flow be streamline or viscous. The low Reynolds No. of 56 assured laminar flow.

Thermal currents were considered to be of no consequence because these would not exist unless there was about 100°R temperature difference between plates. Such temperature differences would not have been possible under the conditions of use. This is shown by the following analysis.

The temperature difference ΔT needed to create full convective cells between plates b distance apart can be calculated. From Eckert and Drake (1959) these convective cells do not occur until the Rayleigh number exceeds a value of 10^3, i.e.

$$\frac{g\Delta Tb^3}{TV_g^2} \times N_{Pr} > 10^3$$

where,

g = acceleration of gravity $\approx 10^3$ cm/sec^2

T = absolute temperature $\approx 5 \times 10^2$R°

V_g = kinematic viscosity of air $\approx 1.5 \times 10^{-1}$ cm^2/sec

N_{Pr} = Prandtl number of air ≈ 1

Therefore for convective cells to occur, ΔT must be in the order of 100°R.

4. Calculation of Particle Size Distribution

The use of this method involves measurement of the overall concentration change in an aerosol as a function of distance traveled along the settling plates. Since the size distribution obtained with this device is an integral size analysis, differentiating must be made to obtain the discrete size distribution. That is, the weight of sediment at distance X down the plates consisted of (1) those particles which would have fallen out before distance X as defined by Stokes' equation assuming that

the particles began their trajectory at the very top of the shallow channel, and (2) smaller particles which have also settled out because they began their trajectory at some intermediate position in the channel, and therefore had a shorter distance to fall.

The particle size distribution is obtained by plotting the cumulative weight percent of settled material as ordinates and the corresponding length of the plates as abscissas. This results in a curve of the type shown in Fig. VII-5. As noted above, this curve gave the integrated weight percent of particles landing at a particular distance along the plate. Differentiation of the curve can be done graphically by drawing tangents to the curve at any point, and the intercept of the tangent on the ordinant gives the discrete weight percentage. For example, in Fig. II-5, a cumulative weight of 62% was found up to the 40 cm distance on the plates. A tangent is drawn to the curve and it strikes the ordinant at the 45% point. This value gave the cumulative weight percent of those particles which began their trajectory at the top of the channel and settled out by the time the aerosol reached the 40 cm mark on the plates. Thus by drawing tangents at various points on the curve, an aerosol settling distribution is obtained, which was related to the settling velocity of the aerosolized particles by the simple proportion,

$$\frac{U_t}{h} = \frac{V}{X}$$

where U_t is the terminal velocity of the dust particles, h is the vertical height of the channel, V is the linear gas velocity through the channel, and X the distance from the entrance. For a typical air velocity of 7.1 cm/sec, and a height of 0.623 cm, the settling velocity of particles landing before the 40 cm distance would be 0.114 cm/sec.

The settling velocity can be converted to aerodynamic diameter utilizing Stokes law,

$$D_p = \sqrt{\frac{18\mu U_t}{g\rho}} = \text{diameter (cm) of equivalent spherical particle.}$$

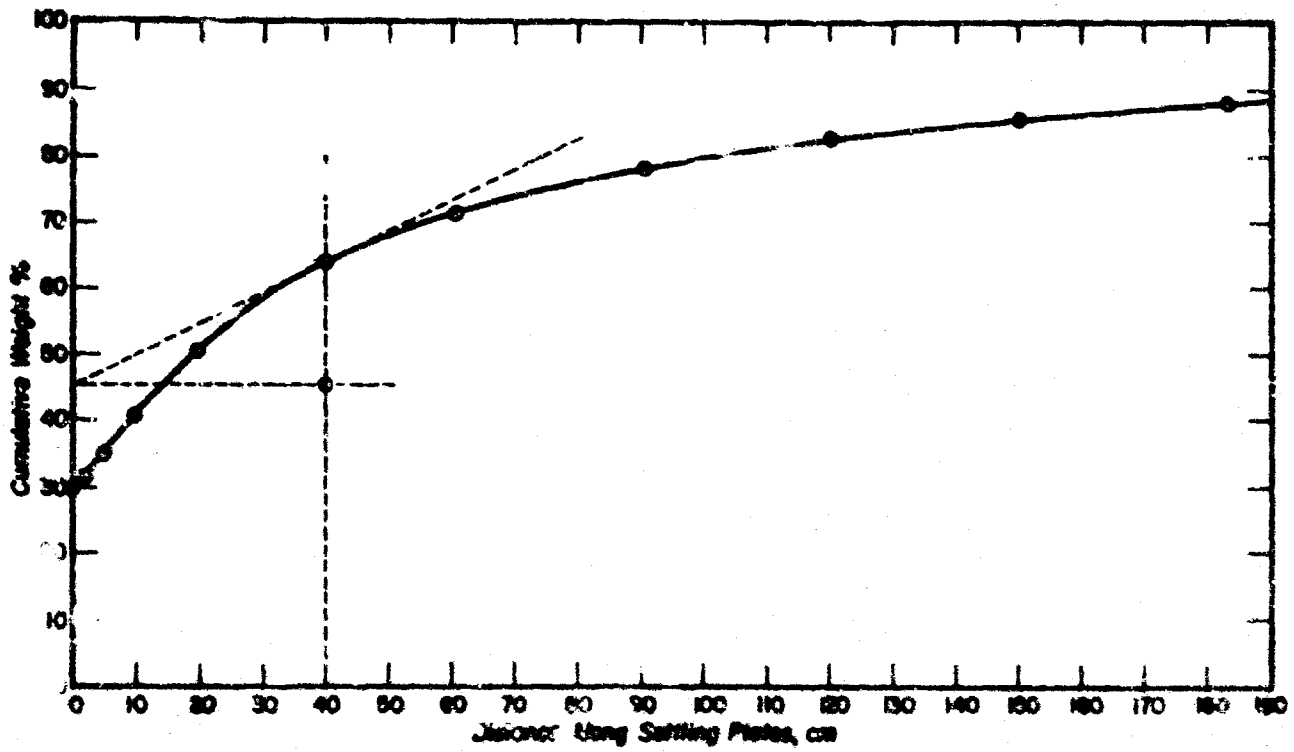


FIG. V.-* TYPICAL POWDER DEPOSITION ANALYSIS CURVE, SHOWING TANGENT METHOD FOR DIFFERENTIATING CUMULATIVE SEDIMENTATION CURVE

where

μ = gas viscosity (poises)

U = settling velocity of particle (cm/sec)

g = gravitational acceleration = 981 cm/sec^2

ρ = particle density (gms/cm^3).
(If arbitrary, the value D_2 is then an effective particle size based on this density)

For a spherical particle with a specific gravity of 1.0 this would correspond to a 6.2μ diameter particle. By plotting a number of such points it is possible to arrive at a particle size distribution.

According to Donoghue (1956), the mass determinations on the plates should be made at distances increasing by not more than $\sqrt{2}$ for very accurate work. To abide by this rule would require some 14 determinations to be done on the 183 cm length settling plate. This analysis seemed unnecessarily long and the small improvement in accuracy hardly justified the time. Therefore, for the tests reported herein, the determinations were made at distances increasing by a factor of two for a total of 10 determinations.

5. Reproducibility of Results

The reproducibility of the chamber is best illustrated with mass distribution curves from the quinine powder experiments. Quinine was selected because it could be analyzed readily by fluorometric technique. The procedure for quinine assessment was as follows:

The powder (0.1 gm) was weighed and disseminated. Air flow was maintained for five minutes after powder introduction to the chamber had ceased. Each third plate, for a total of five plates, was removed for analysis. Cotton soaked in 0.5 N H_2SO_4 made up in 20% isopropanol and 80% water vol/vol was used to wipe the quinine from the plates at marked intervals 2, 5, 10, 20, 40, 60, 90, 120, 150, and 183 cm. The filter and orifice of the chamber were cleaned by soaking them in the alcohol solution, and the duct section was cleaned by flushing the inside with 3 liters of the solution. The amount of powder remaining on the turntable was subtracted from the total quantity weighed for dissemination.

and an overall material balance was obtained by determining the quantities of quinine in solution from each section using a fluorometer for analysis. Mass balances between 80% and 120% were consistently obtained and 90% to 110% was considered satisfactory.

Figure VI-6 shows some of the duplicate analysis runs. Run #25 and Run #26 are duplicates, as were Run #35 and Run #40. These were some of the better duplicate runs, although duplicates such as Run #38 and #45 were more common. This precision of the chamber is sufficient to distinguish differences in aerosols resulting from a change in the powder, or from a change in the dissemination conditions of a powder.

6. Tests of Aerosol Settling Rates

As discussed earlier, one of the unique features of the laminar flow settling chamber was that it permitted a measurement of the effective settling velocities of the suspended particles. If the density of the airborne particles is known then the theoretical settling pattern of the particles can be predicted, using the calculation given above. Thus, the theoretical settling patterns shown in the various curves below are the maximum size of particles which will fall at the distance indicated.

Spherical particles were used for these tests because they aerosolize easily and the agglomerates are distinguishable from the primary particles. The powders used are listed below, and their particle size distributions are shown in Section IV, MATERIALS.

a. Microthene, 1-30μ, density of 0.934 gm/cc.

b. Glass beads, 1-30μ, density of 2.5 gm/cc.

c. Aluminum B-3, mmd = 3μ, density of 2.7 gm/cc.

The coiled tube and powder feeder shown in Fig. VI-6 were used in all tests. The dissemination conditions were: 0.4 gm powder disseminated at powder/air ratio of .0053 gm powder/gm air; 33ℓ/min air flow through the 0.16 cm dia. tube and 7.1 cm/sec velocity through the settling plates. Microscope cover slips were fastened to the duct sections of the chamber and at intervals of 2, 5, 10, 20, 40, 80, 120, and 183 cm. along the length of the settling plates. Microscopic observations of the nature

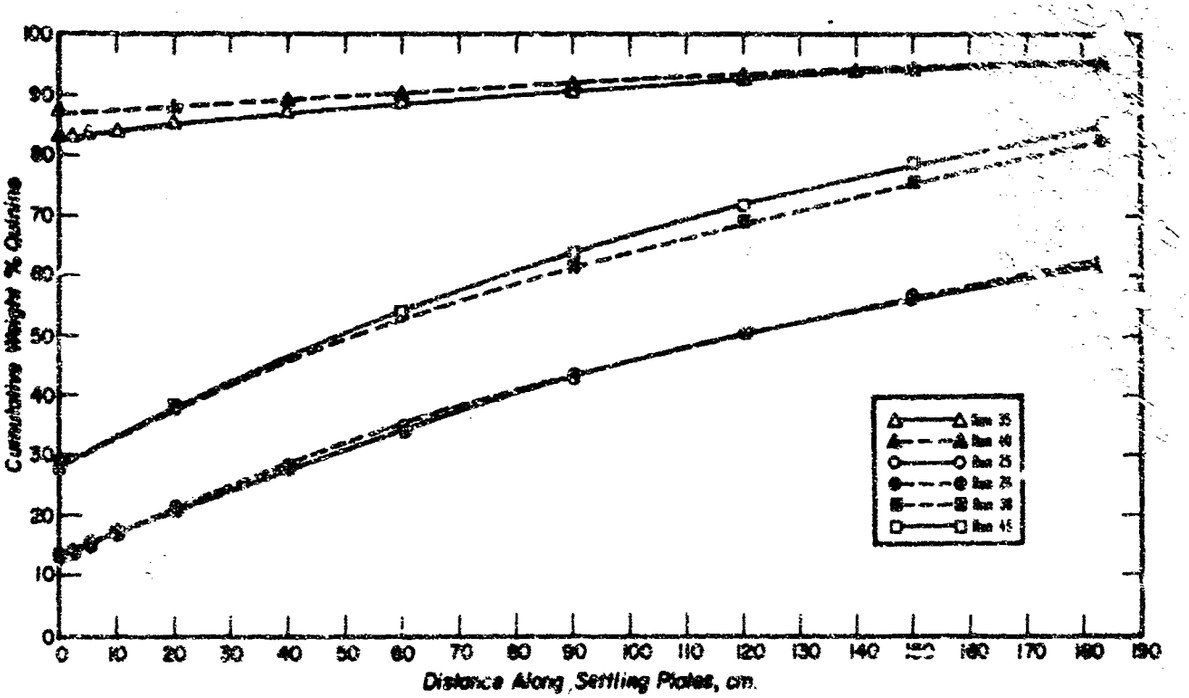


FIG. VII-6 DISSEMINATION OF QUININE INTO LAMINAR FLOW SETTLING CHAMBER. These runs show the reproducibility of duplicate runs

of the deposited powder were made from these cover slips. Approximately 150 particles on each slide were measured using a ruled ocular micrometer of the Cawood type. The microscope was calibrated with a stage micrometer and verified by sizing 3.48μ and 1.90μ polystyrene beads obtained from Dow Chemical Company. However, to compare the measured size with the theoretical size, the averaged diameter of the largest 10% of the particles on each slide was computed. For example, if 170 particles were measured, then the averaged size of the largest 17 particles was calculated and plotted in the settling curves.

a. Microthene

The theoretical and experimental settling curve for untreated Microthene is given in Fig. VII-7. It is seen that the two curves nearly coincided, thus indicating that for Microthene particles the laminar flow settling chamber was operating correctly.

A number count of the Microthene particles on the slides was also made, and by differentiating the counts along the length of the plates, the percentage of particles on the plates was found. This number came out to be 50%. The particle size distribution of the original powder showed that 99% of the particles by number is less than 40μ diameter. Since the air flow through the chamber was adjusted to deposit 40μ size particles on the settling plates, it appeared that many of the particles remained agglomerated after dissemination, and consequently did not reach the settling plates. Figure VII-8 is a photomicrograph of agglomerated Microthene particles on the wall of the duct part of the chamber. Therefore in another test, Microthene treated with 1% PQ 2340 to disperse the agglomerates, was disseminated in the same manner as above. But surprisingly, in this later test, fewer particles reached the settling plates. Yet the particles on the walls and bottom of the duct were all single particles as shown in Fig. VII-9. Obviously some force, of sufficient strength to negate the upward air flow of approximately 7 cm/sec (0.15 mph), caused the particles to be attracted to the walls of the chamber. This force was attributed to electrostatic effects.

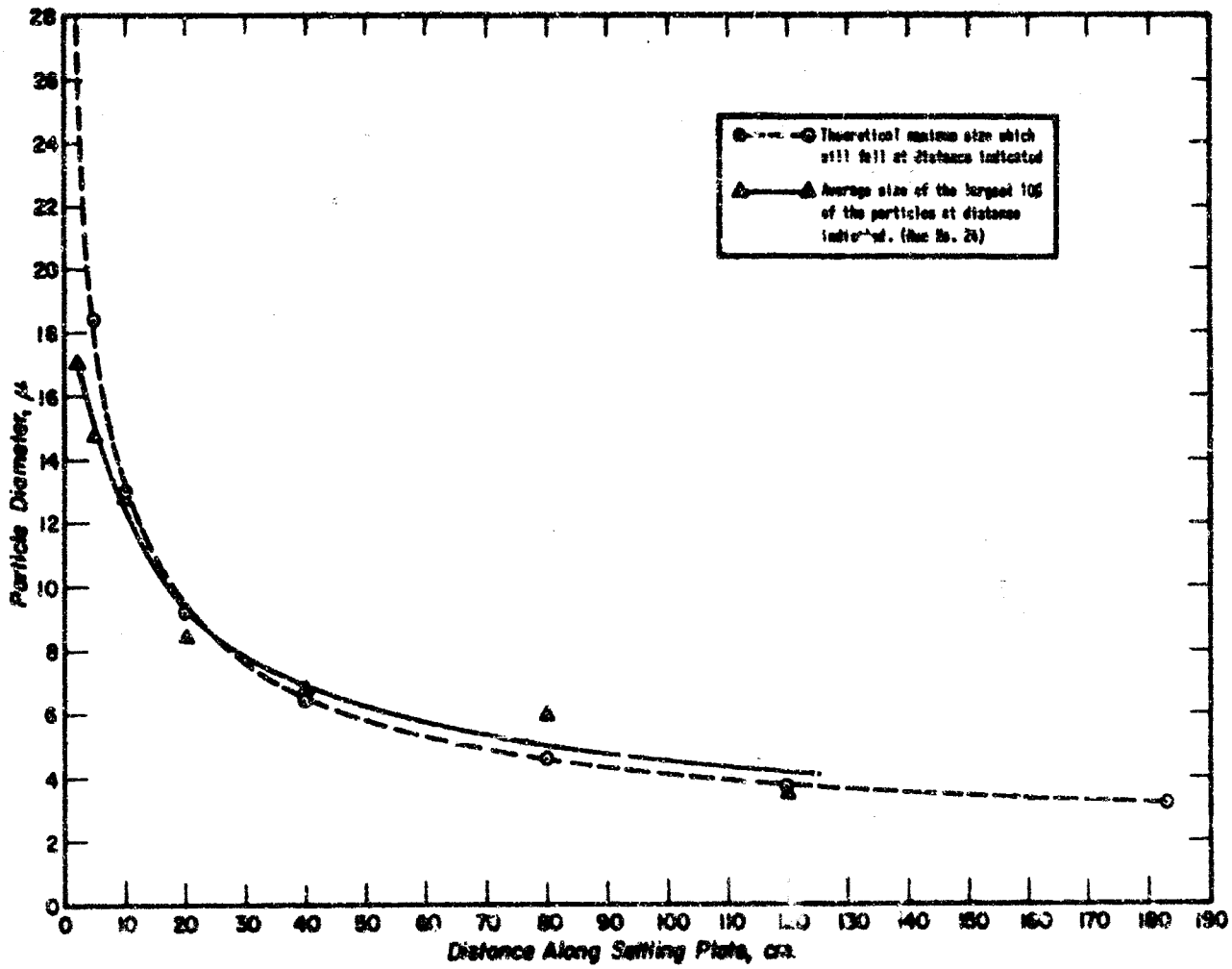


FIG. VII-7 COMPARISON OF THEORETICAL WITH EXPERIMENTAL SETTLING PATTERN FOR MICROTHENE IN LAMINAR FLOW SETTLING CHAMBER

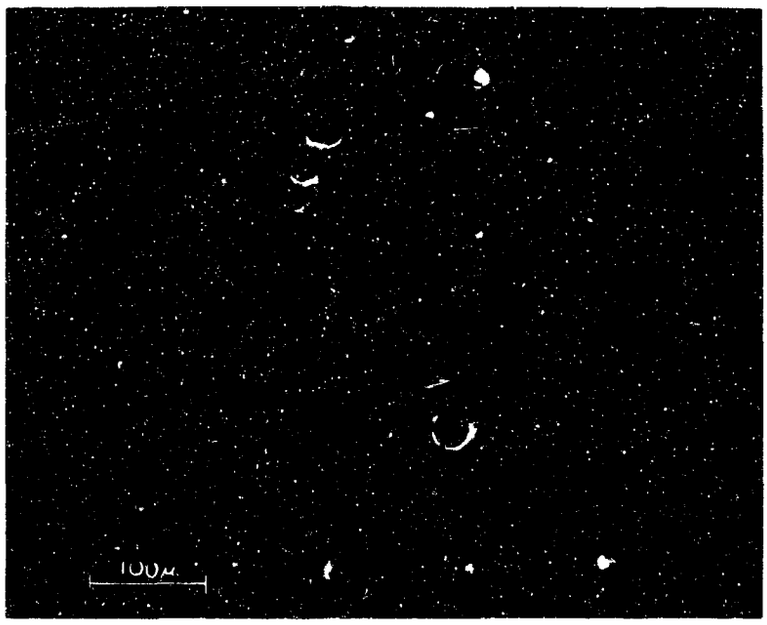


FIG. VII-8 DEPOSITION OF UNTREATED MICROTHENE PARTICLES ON DUCT SECTION OF LAMINAR FLOW SETTLING CHAMBER

FIG. VII-9 DEPOSITION OF TREATED (WITH 1% PQ 2340) MICROTHENE PARTICLES ON DUCT SECTION OF CHAMBER

b. Glass Beads, 1-30μ

Dissemination of glass beads of sizes 1-30μ was made under the same conditions as Microthene. And like the Microthene, only a small percentage reached the settling plates. Therefore it appears that the glass beads were also influenced by electrostatic forces.

c. Aluminum

Because of the metallic nature of aluminum, electrostatics should be less influential in its dissemination than with the other powders. The results of the tests are plotted in Fig. VII-10. There is some scattering of points at the beginning of the settling plates (up to about the 10 cm mark), but this is to be expected since the aerosol is progressing from a mild turbulent stage to a laminar one. However, after the initial 10 cm, the results clearly showed that the particles settled were larger than those predicted from theory. In general, the size settled was 1.0 to 1.5μ larger than the predicted size. There is no obvious explanation for the discrepancy. The air flow through the chamber was accurate to ±10%. For the experimental deposition curve to match the theoretical, the density of the particles must be approximately 0.6 gm/cc. However, the bulk density of the aluminum was carefully checked and was found to be 2.73 gm/cc. The density was also checked by immersing a sample of the powder in a 2.5 gr/cc solution, and approximately 90% of the particles settled out. It is possible that the 5% - 10% of the light density aluminum particles would be the larger sized particles, and this small percentage would be sufficient to shift the experimental curve slightly off from the theoretical.

E. Discussion

In this program emphasis was placed on the use of the laminar flow settling chamber, microscopic counting and photography of falling and settled particles. Although the principle of operation of the integrated settling chamber is not new, its use has been somewhat confined to liquid sedimentation analysis. Hewson, Raines and Punturerl (1961) were among the few to employ the technique for measuring aerosols. The theoretical

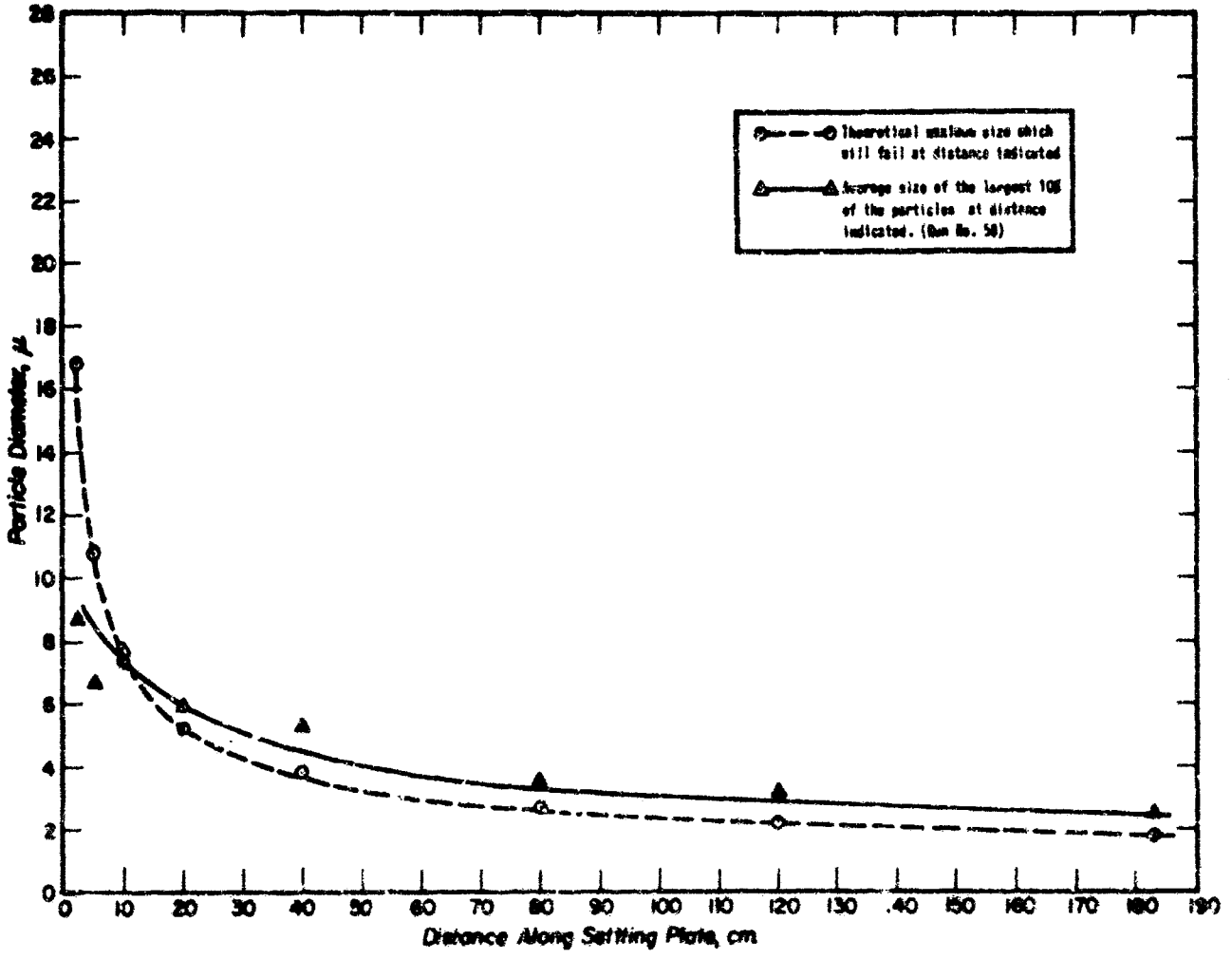


FIG. VII-10 COMPARISON OF THEORETICAL AND ACTUAL SETTLING PATTERN FOR ALUMINUM IN LAMINAR FLOW SETTLING CHAMBER

particle settling pattern, as determined with spheroidal aluminum particles, was close to but did not quite match, the experimental settling pattern. Since comparison between powder treatment is made on a relative basis, the disagreement between the theoretical and experimental is not of great significance. These methods served their purpose because the scale of operation was small and it was possible to work with reasonably representative samples of the entire aerosol population. With larger scale aerosol generation, this would have been much more difficult.

The general subject of aerosol generation and assessment covers a wide range of production rates from a few tenths of a gram to many kilograms per minute. It also includes closed chamber and field operations. The methods described above can be used generally, providing precautions are taken to assure representative sampling and to account for electrostatic effects.

Electrostatic charges may increase the coincidental settling effect. Knutson (1965), in his study on aerosol reagglomeration, noted a larger number of agglomerates than that predicted by theory. He attributed this increase to electrostatic charges on the settled particles. Owe Berg and Brunetz (1962) performed experiments which indicated that electrostatics may be the cause of the anomaly. They sprayed a mixture containing carmine, flowers of sulfur, and lycopodium onto a glass and aluminum surface. On glass, electrostatic forces made particles settle preferentially on already settled particles of the opposite polarity, forming chain-like structures. With a conductive surface such as aluminum, the particles lost their charges when they settled and hence the yellow (flowers of sulfur) and red (carmine) particles were all separated and randomly distributed. The fact that particles acquired strong charges was evident from the tests made with Microthene powder in the laminar flow settling chamber.

VIII DISSEMINATION RESULTS

A. Geometry of Disseminating Devices

There are strong shear forces adjacent to the walls of a tube, and when air-suspended agglomerates pass through the tube, these shear stresses are partially responsible for disintegrating the agglomerates into individual particles. The magnitude of these forces is discussed in Section X, and as indicated there, the shear forces are just marginal for agglomerate breakup. It was also proposed that a better mode for breakup would be found in wall impaction forces, and calculations in support of this theory are presented. In this section, the experimental results from investigation of deagglomerating forces are presented.

1. Effect of Tube Length

To investigate the effects of tube length on dissemination, a series of tests were conducted in which Microthene was disseminated through straight capillary tubes of various lengths. It was reasoned that there should be some optimum length of tube which would produce the maximum amount of deagglomeration. The tubes shown in Fig. VI-8, the six foot vertical chamber in Fig. VI-10, and the powder feeder disseminator in Fig. VI-6 were used for these tests. The aerosols were assessed by counting the number of particles engaged in agglomeration. Four series of tests at four different air flows were made. Only the data for the 20 L/min series of tests are given in Table VIII-1 since the others are similar. The results of all series are given in Fig. VIII-1.

There is an undesirable scatter of points probably due to the assessment method; however, the results indicate that tube length is of some importance and under the conditions used the optimum length appeared to be about 15-20 cm in length. The curves do not show clearly the effect of excessive tube length on deagglomeration, but they do indicate rather specifically that tubes of lengths shorter than 10 cm are not efficient disseminators.

Table VIII-1

DISSEMINATION OF MICROTRENE
THROUGH CAPILLARY TUBES OF VARIOUS LENGTHS

Type of Capillary Tube	Dissemination Conditions	Aerosol Concentration in Tube gm powder/gm air	% Particles Engaged in Agglomeration
0.19 cm I.D. 2.5 cm length	20.0 ℓ/min* 3.4 psig+	0.0116	71.5%
0.19 cm I.D. 5 cm length	20.0 ℓ/min 3.9 psig	0.0116	57.5%
0.19 cm I.D. 10 cm length	20.0 ℓ/min 6.3 psig	0.0116	33.0%
0.19 cm I.D. 15 cm length	20.0 ℓ/min 8.3 psig	0.0116	33.0%
0.19 cm I.D. 20 cm length	20.0 ℓ/min 8.7 psig	0.0116	35.0%
0.19 cm I.D. 30 cm length	20.0 ℓ/min 11.0 psig	0.0116	31.5%
0.19 cm I.D. 40 cm length	20.0 ℓ/min 15.5 psig	0.0116	41.0%

* Airflow through tube.

+ Pressure inside dissemination box.

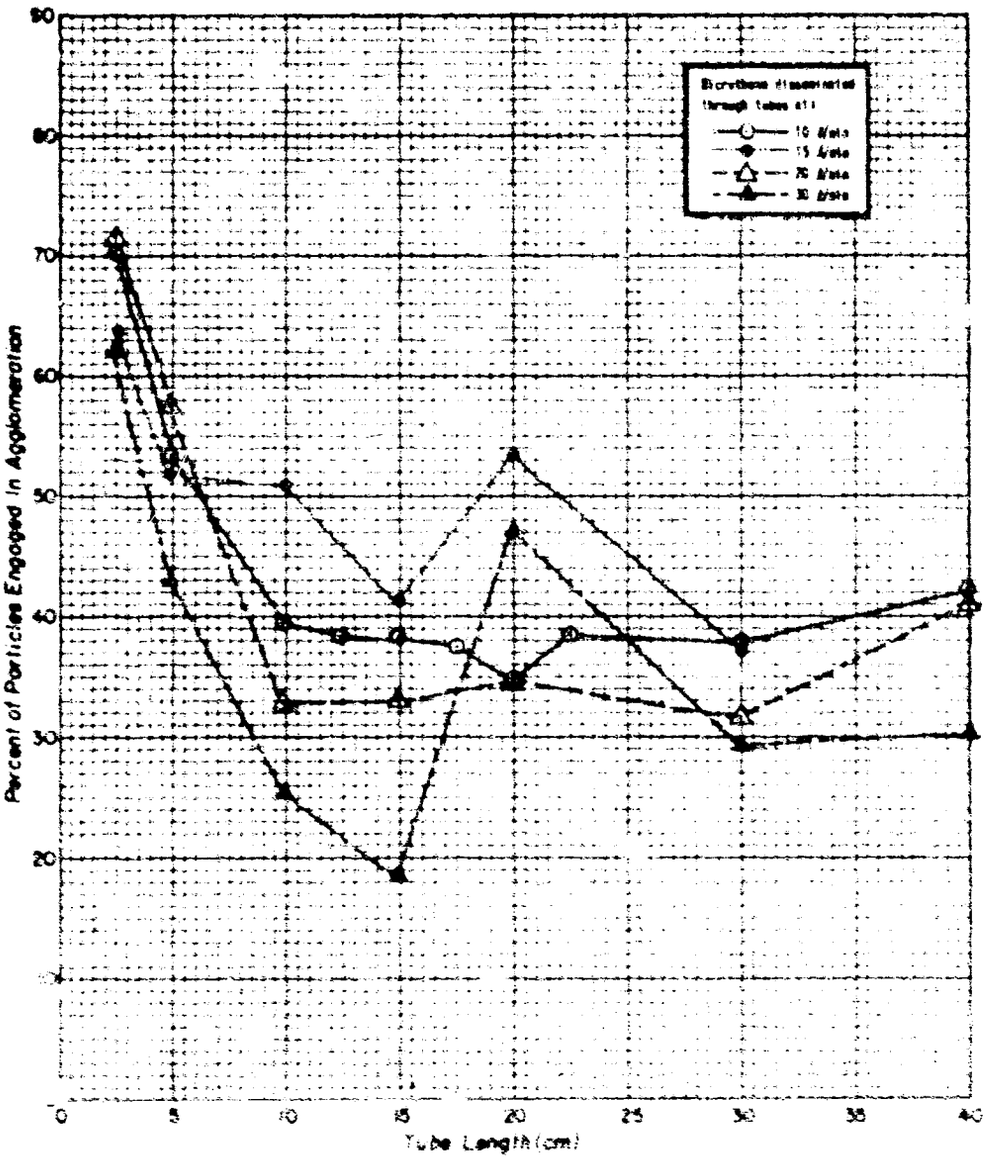


FIG. VIII-1 EFFECT OF TUBE LENGTH ON THE AGGLOMERATION OF MICROTHENE

2. Impaction Forces

Some impaction of the particles against the wall of the tube occurs in a straight tube, but there is more impaction in a curved tube. To accentuate the impaction of particles on the walls of the tube, the dissemination tube was bent into coils. These coiled tubes are shown in Fig. VI-9. Two series of tests, using aspirin and quinine powders were performed to compare the effects of straight and coiled tubes on aerosolization.

a. Aspirin

Aspirin tablets were crushed and screened to obtain 50-100 micron granules. These granules were rather firmly bound agglomerates. Since the primary particle size could not be measured by any convenient means, the agglomerate sizes were measured, and this was done by "puffing" the powder down the 100 cm settling chamber and sizing the settled agglomerates.

To test the various straight and coiled tubes, the vertical six-foot chamber and the four-inch powder disseminator were used. Dissemination conditions are given in Table VIII-2 and the results plotted in Fig. VIII-3. The particle size distribution from the "puffer" system is also plotted in Fig. VIII-2, but it should be remembered that there is considerable difference in the energies applied to the dissemination between the puffer and the aspiration system.

The results indicated that there was comparatively little difference in the aspirin aerosols resulting from the straight and coiled tubes. The "2-coils," "4-coils" and "bugle" shaped tubes gave particle size distribution curves which were within the experimental precision of the straight tube curve. However microscopic observation of particles showed that there was a small difference. The aspirin particles from the coiled tubes, Fig. VIII-3, were more rounded and less angular than those from the straight tube, Fig. VIII-4. If there is any significance to this observation, it suggests a greater collision frequency in the coiled tubes than in the straight tube. Also included is the size distribution curve from dissemination at sonic velocity, and in this case there was a significant change in the particle sizes.

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Table VIII-2

AEROSOLIZATION OF ASPIRIN
THROUGH CAPILLARY TUBES OF DIFFERENT CONFIGURATIONS

Type of Capillary Tube*	Dissemination Conditions	Aerosol Concentration in Tube gm powder/gm air	Number Median Diameter from Particle Size Distribution Curve
30 cm length straight copper tube	20 liter/min 12.5 psig	0.0130	15.0μ
30 cm length tube formed into two 3.2 cm dia loops (2 coils)	20 liter/min 13.6 psig	0.0130	16.5μ
30 cm length tube formed into four 1.6 cm dia loops (4 coils)	20 liter/min 14.6 psig	0.0130	17.5μ
30 cm length tube formed into three elliptical loops ("bugle" shaped)	20 liter/min 13.9 psig	0.0130	15.0μ

* All tubes were 0.16 cm internal diameter.

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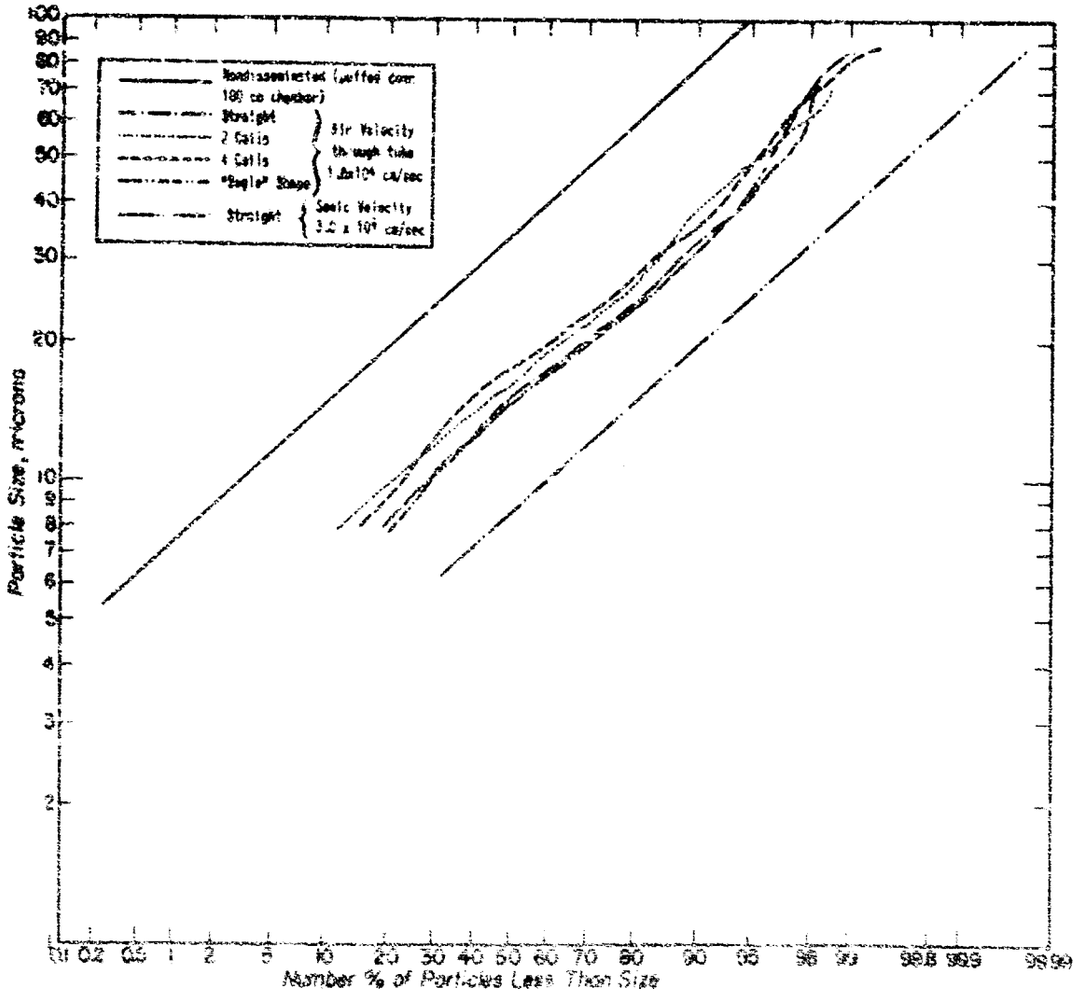

FIG. VIII-2 PARTICLE SIZE DISTRIBUTION OF ASPIRIN DISSEMINATED THROUGH 0.16 cm DIAMETER TUBES OF VARIOUS CONFIGURATIONS

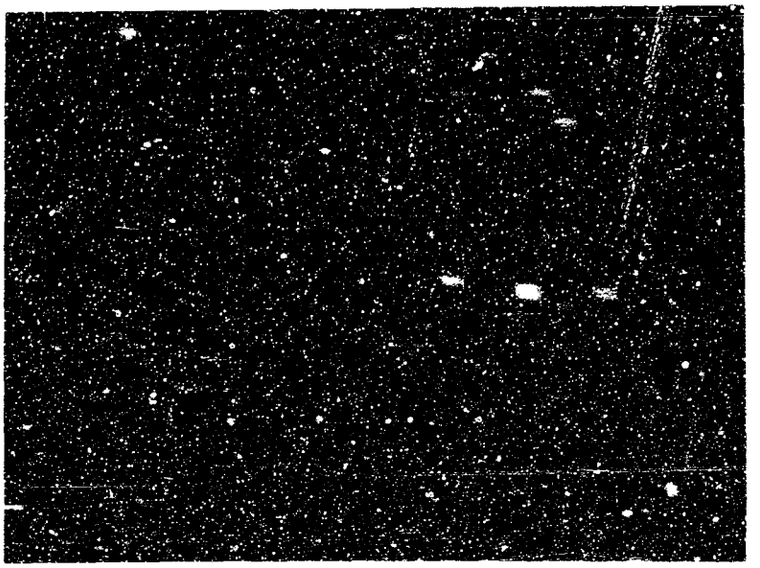

FIG. VIII-3 GRANULAR ASPIRIN DISSEMINATED THROUGH COILED TUBE (0.16 cm DIAMETER)

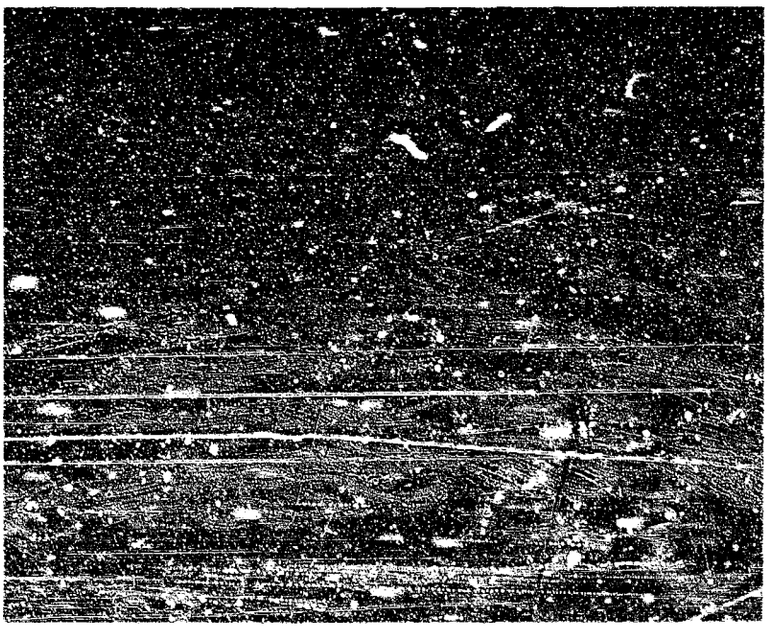

FIG. VIII-4 GRANULAR ASPIRIN DISSEMINATED THROUGH A STRAIGHT TUBE (0.16 cm DIAMETER)

b. Quinine

The second series of tests to compare the effect of straight and coiled tubes was performed some nine months after the first series. In the interval a new assessment technique, the laminar flow settling chamber was installed for aerosol evaluations. For this series, the "one-coil" tube was used. The coiled tube was patterned after the one employed by Fuchs and Selin (1964) in their study of pneumatic atomization. The internal diameter of our coiled tube was 0.16 cm and the coil formed at the end of the tube was 2.0 cm in diameter. In all of the results that follow, the "coiled tube" referred to in the text specifically refers to this particular coiled tube.

Because quinine can be analyzed easily by fluorometric means, it was selected for use in these tests. The quinine alkaloid, as received, existed in the form of agglomerates too large to be used. It was ground in an Osterizer Blender in the presence of 1% PQ 2340 to reduce the particle size. Some grinding occurred, but many large particles still existed. In one case it was desirable to remove the large particles, and this was accomplished with a double-disc separator described in Section V. The fine fraction from the separation is designated as "fine quinine."

Quinine was disseminated upward into the laminar flow settling chamber and analyzed according to the procedure given in Section VII-D-5. The dissemination conditions are given in Table VIII-3, and the data plotted in Fig. VIII-5 are the integrated mass distribution results. Figure VIII 6 shows the differentiated particle size distribution curve, obtained by drawing tangents to the curves of Fig. VIII-5.

The mass distribution curves clearly indicated that the coiled tube was a more effective disseminator than the straight tube. Good comparative tests are Runs 11 and 09 in which the PQ 2340 treated quinine was disseminated through the straight and coiled tubes, respectively, at nearly sonic velocity (2.9×10^4 cm/sec). The curved tube generated aerosol particles which are smaller than those produced by the straight tube. For example, at the 40 cm mark on the plates, Run 09 showed 80% of the particles

Table VIII-3

DISSEMINATION CONDITIONS FOR QUININE IN COMPARISON TESTS BETWEEN STRAIGHT AND COILED TUBES

Run Number, Refer to Last Symbol Two Digits	Weight of Quinine Disseminated, gms	Type of Tube Used and Flow Area in cm^2	Pressure in psi Gauge	Flow Vol. in L/min	Velocity at End of Tube in cm/sec	Reynolds Number at End of Tube	Aerosol Concentration in gm powder/gm air	Mass Mean Diameter of Particles on Plates, in Microns, Based on Their Aerodynamic Diameter
QS20-100-05	0.10	straight 0.0284	8.9	20.0	1.17×10^4	1.42×10^4	0.018	5.6
QC14-100-08	0.10	coiled 0.019	8.9	14.1	1.34×10^4	1.23×10^4	0.026	5.0
QS33-100-11	0.10	straight 0.019	23.8	33.0	2.90×10^4	2.86×10^4	0.023	4.5
QC33-100-09	0.10	coiled 0.019	30.0	32.8	2.88×10^4	3.85×10^4	0.026	3.6
QS[illegible]100-14	0.10	straight 0.019	23.8	33.0	2.90×10^4	2.86×10^4	0.026	4.2
70C33-100-12	0.10	coiled 0.019	26.8	32.2	2.83×10^4	2.80×10^4	0.026	3.4

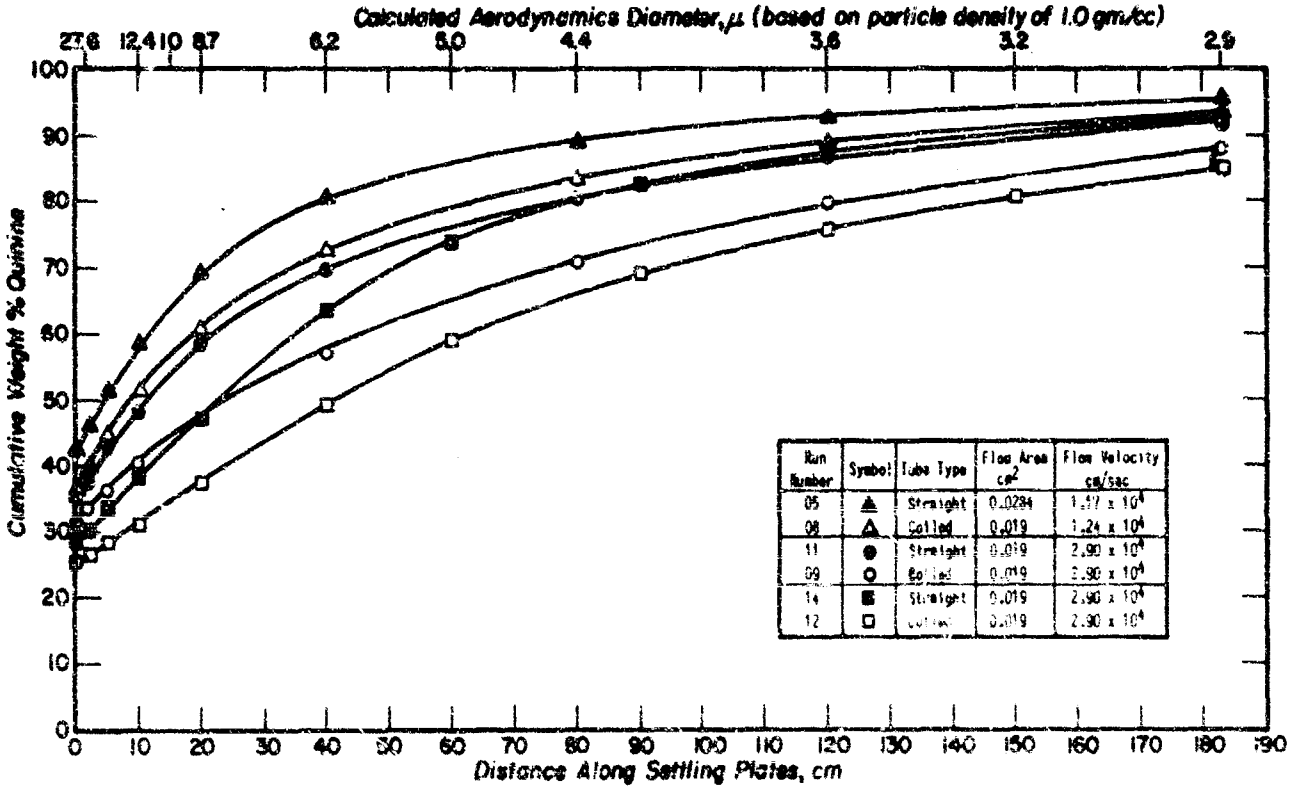

FIG. VIII-5 CUMULATIVE MASS ANALYSIS OF QUININE FOUND ON SETTLING PLATES AS A FUNCTION OF PLATE DISTANCE

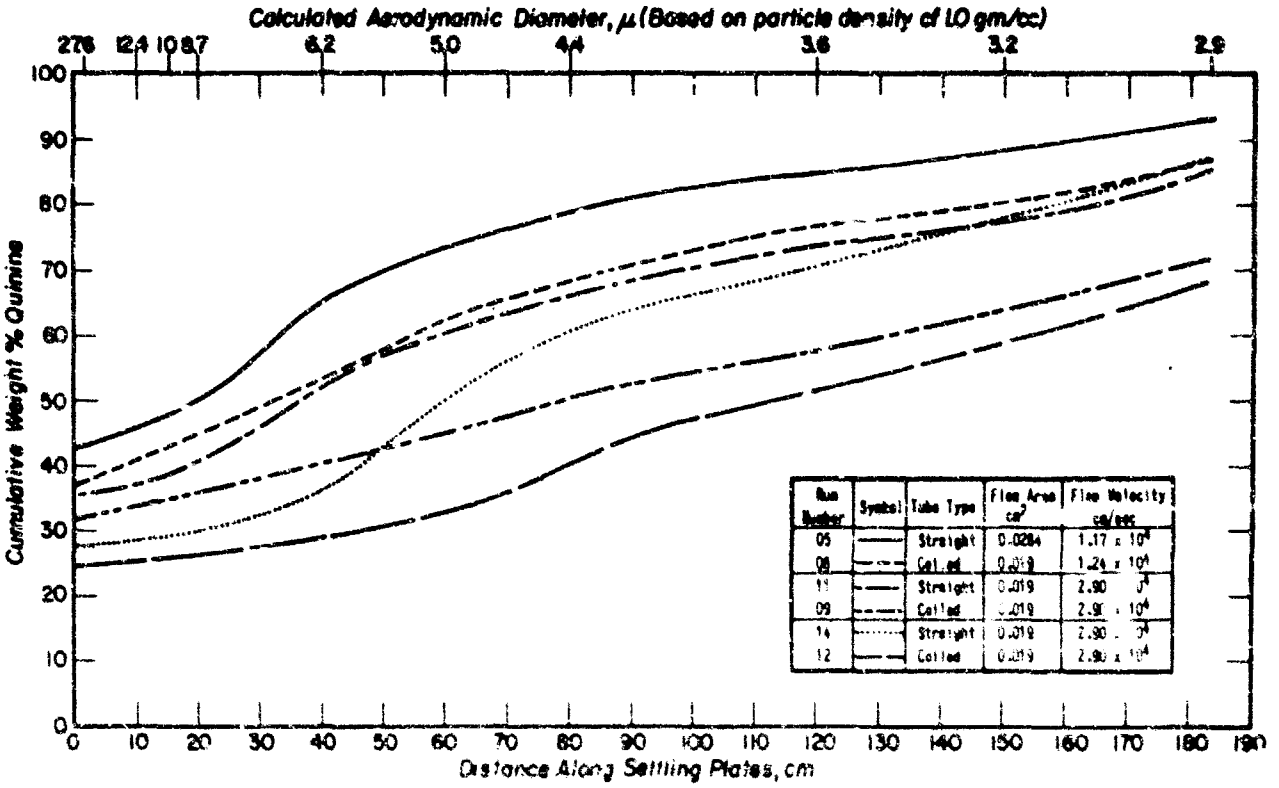

FIG. VIII-6 CUMULATIVE MASS DISTRIBUTION CURVES FOR QUININE ON SETTLING PLATES
Differentiated graphically from experimental results plotted in Figure VIII-5

had an aerodynamic diameter smaller than 6.2μ, while in Run 11, only 46% of the particles were smaller than 6.2μ.

In another comparison, Run 05 and 08, the straight tube used in Run 05 had a larger bore (0.0284 cm^2 compared to 0.019 cm^2); consequently in Run 05 it was necessary to use a higher disseminating pressure and a larger flow volume to produce the same air velocity as used in Run 08. These manipulations also resulted in a lower powder/air ratio. As indicated elsewhere in this report, a higher air pressure, a larger air velocity, and a smaller powder/air ratio contributed to better aerosolization. But dispite the favorable dissemination conditions for the straight tube run, 05, the coiled tube run, 08, gave the smaller sized aerosol.

The final comparison was made with the fine fraction of quinine, disseminated under similar conditions. The coiled tube run, Run 12, produced a much smaller aerosol than the straight tube. At the 40 cm mark, 72% of the particles had an aerodynamic diameter smaller than 6.2μ, whereas Run 14 gave only 63% smaller than 6.2μ.

Figure VIII-7 shows the actual particle size distributions for the various tests plotted on log-probability paper. The distance on the plates was translated into aerodynamic particle size, assuming a density of 1.0 gm/cc.

Another series of comparison tests clearly showed that a coiled tube was a more effective aerosol disseminator than a straight tube. The first series of tests with coiled tubes having multiple turns failed to reveal any differences between straight and coiled tubes. However, the first series was performed on a different powder, with a different assessment technique and with different tubes. It was suspected that the older assessment method was insufficiently sensitive to detect the differences, but then again the other factors--different powder and different tubes--could also contribute to the cause. Certainly the size and shape of the coil, as pointed out by Fuchs and Selin (1964), is important for optimum operation of the coiled tube.

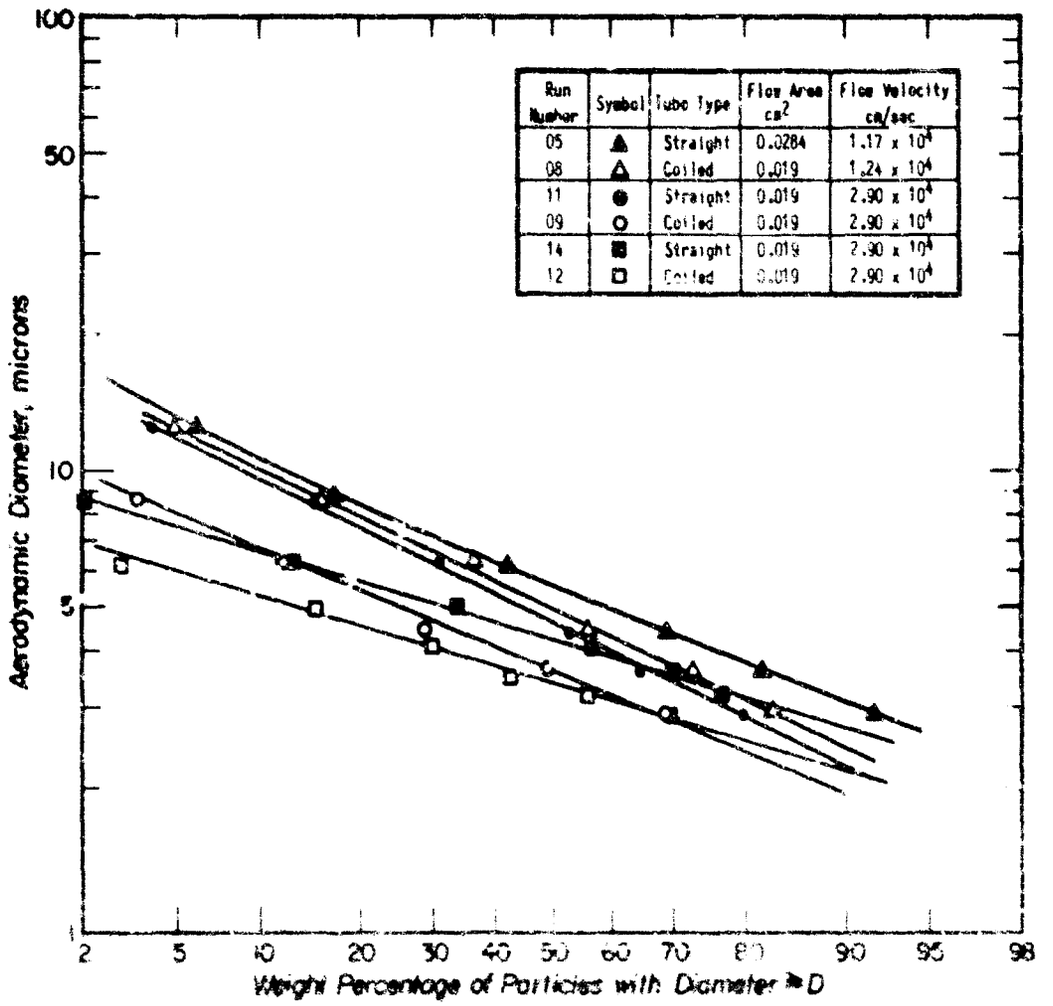

FIG. VIII-7 PARTICLE SIZE DISTRIBUTION OF QUININE ON THE SETTLING PLATES, ASSUMING PARTICLES AND AGGLOMERATES HAVE DENSITY OF 1.0 gm/cc

3. The Effect of Air Velocity on Dissemination

The first series of tests on the effect of air velocity was made with aspirin powder using the six-foot vertical chamber. A straight tube, 0.19 cm in diameter x 20 cm long was used for all these runs. The conditions for dissemination and the results of the size analysis of the aerosols are given in Table VIII-4 and Fig. VIII-8. As expected, finer aerosols were produced at higher air velocities. Some generalization between aerosolization and air velocity were obtained by plotting the number-mean-diameter vs. Reynolds number. This is shown in Fig. VIII-9. and it is seen that a linear relationship between the two exists. Thus for aspirin powder, the direction of change in size of aerosol produced can be predicted from knowledge of the disseminating velocity.

Another series of tests was made with untreated Microthene using the same dissemination set-up as above. Assessment was performed by counting the particles in agglomeration rather than by sizing. The results are shown in Table VIII-5 and Fig. VIII-10. There is a linear decrease in agglomeration with increasing Reynolds number. The Microthene powder was nearly completely disseminated (only 10% in agglomeration) at Reynolds number of 26,000. Strong electrostatic effects with Microthene were not observed in these experiments as they were in the laminar flow settling chamber (see Section VII-D-6). This may have been due, in part, to the larger cross section of the vertical section of the chamber.

To explore further the effect of air velocity, a third series of tests was made, using the identical dissemination set-up except in this case Microthene treated with 1% PQ 2340 was the experimental powder. The result, Fig. VIII-11, showed a substantial change in the agglomeration tendency of Microthene. At Reynolds number of 4000, nearly all the Microthene was deagglomerated (approximately 5% in agglomeration) and although increases in air velocity provided additional deagglomeration, the ensuing changes were small. Thus, for treated Microthene the effect of air velocity is less important than for the untreated powder. The effect of additive on aerosolizability is discussed further in Section VIII-B, but it is obvious that powder modification can substantially improve the ease of powder dissemination in the case of Microthene.

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Table VIII-4

DISSEMINATION OF ASPIRIN POWDER
THROUGH A STRAIGHT CAPILLARY TUBE AT VARIOUS FLOWS

Type of Capillary Tube	Dissemination Conditions	Reynolds Number	Velocity Flow cm/sec	Aerosol Concentration in Tube gm Powder/gm Air	Number Median Diameter Particle Size Distribution Curve
none	Puffed gently down - 12.3 cm dia x 100 cm length chamber	~2	--	--	34.0μ
Straight tube 0.19 cm dia 20 cm length	6 liter/min 1.2 psi	4,700	350	0.0127	16.2μ
Straight tube 0.19 cm dia 20 cm length	20 liter/min 8.2 psi	15,700	1,170	0.0116	13.2μ
Straight tube 0.19 cm dia 20 cm length	30 liter/min 16.0 psi	23,600	1,760	0.0222	12.0μ
Straight tube 0.19 cm dia 10 cm length	50 liter/min 30.0 psi	36,500	2,950	0.00253	8.6μ

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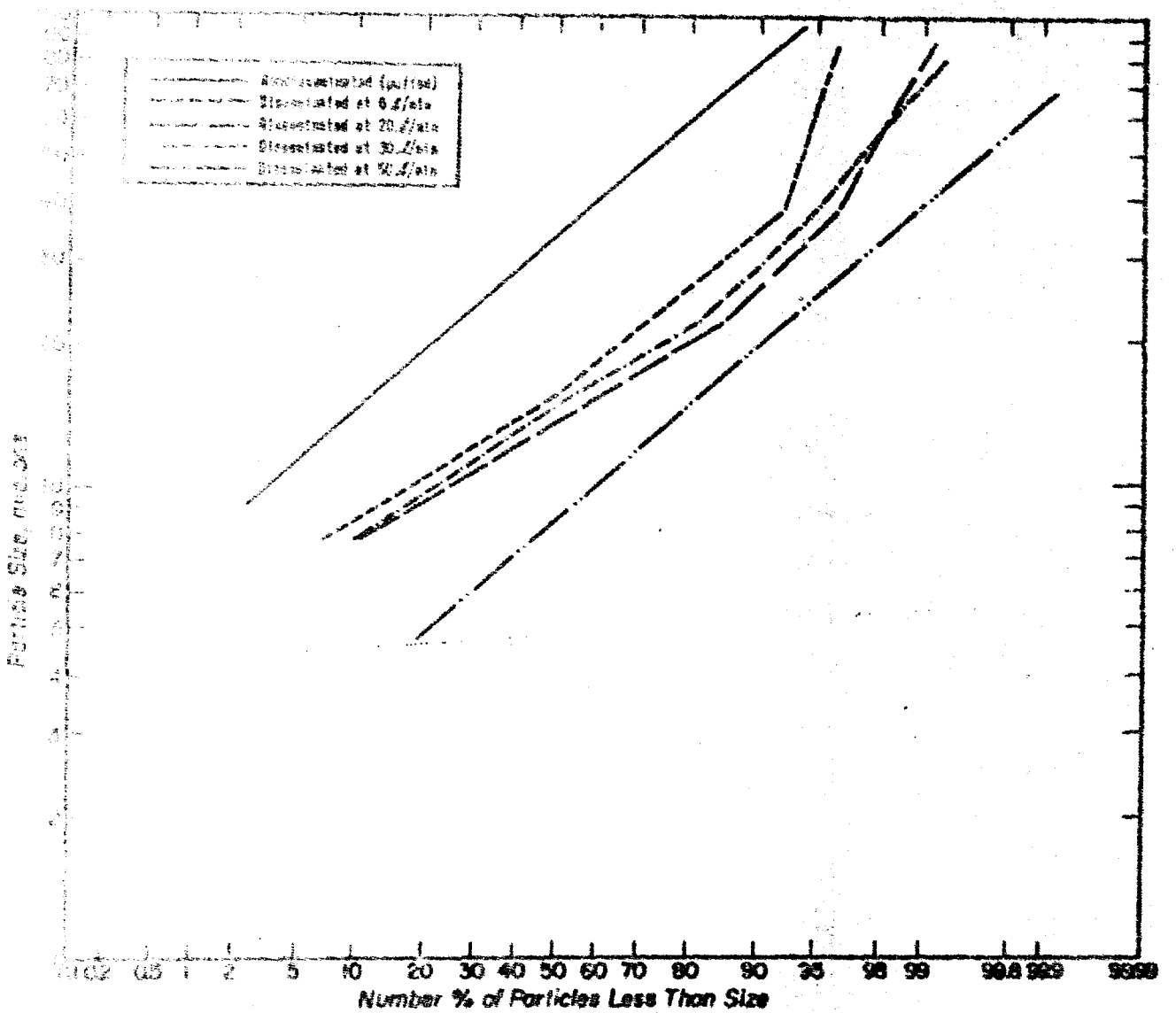

FIG VIII-3 PARTICLE SIZE DISTRIBUTION OF ASPIRIN DISSEMINATED THROUGH A 0.91 cm DIAMETER, 20 cm LENGTH TUBE AT VARIOUS AIR FLOW

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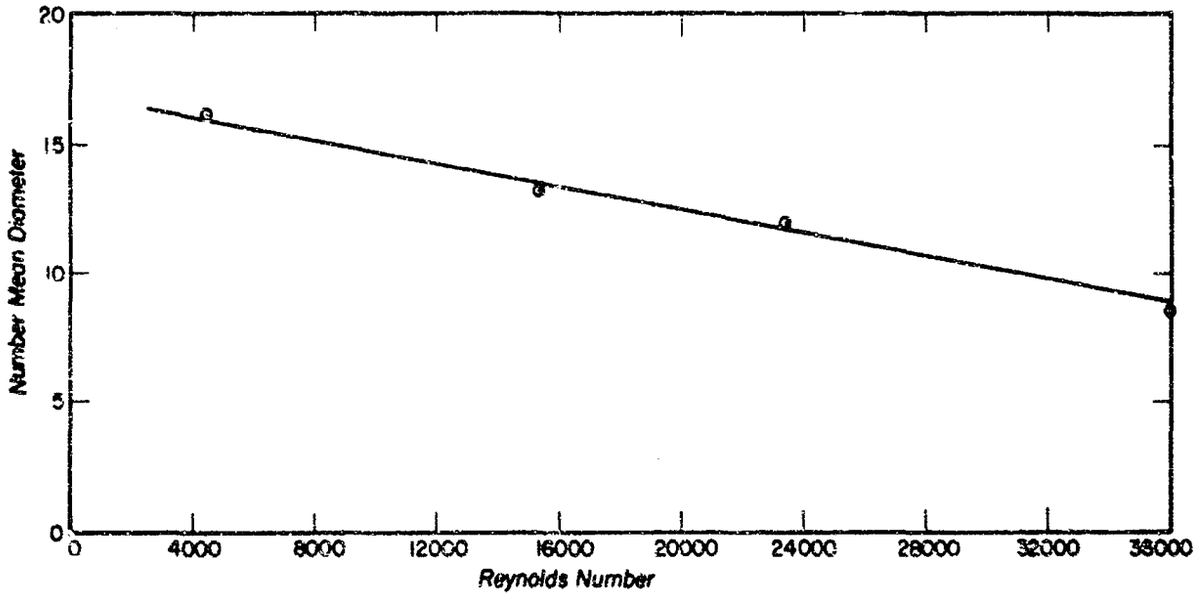

FIG. VIII-9 CORRELATION BETWEEN NUMBER MEAN DIAMETER AND REYNOLDS NUMBER FOR ASPIRIN POWDER DISSEMINATED AT VARIOUS AIR VELOCITY

Table VIII-5

DISSEMINATION OF MICROTHENE THROUGH A STRAIGHT CAPILLARY TUBE AT VARIOUS AIR FLOWS

Type of Capillary Tube	Dissemination Conditions	Aerosol Concentration in Tube gm Powder/gm Air	% Particles Engaged in Agglomeration	Reynolds Number
Straight Tube 0.19 cm dia 20 cm length	6.0 ℓ/min 1.2 psi	0.0142	43.4%	4,700
0.19 cm dia 20 cm length	13.7 ℓ/min 5.0 psi	0.0067	40.0%	10,800
0.19 cm dia 20 cm length	20.0 ℓ/min 8.1 psi	0.0132	32.1%	15,700
0.19 cm dia 20 cm length	25.0 ℓ/min 13.0 psi	0.0248	19.3%	19,700
0.19 cm dia 20 cm length	30.0 ℓ/min 16.0 psi	0.0207	16.2%	23,600

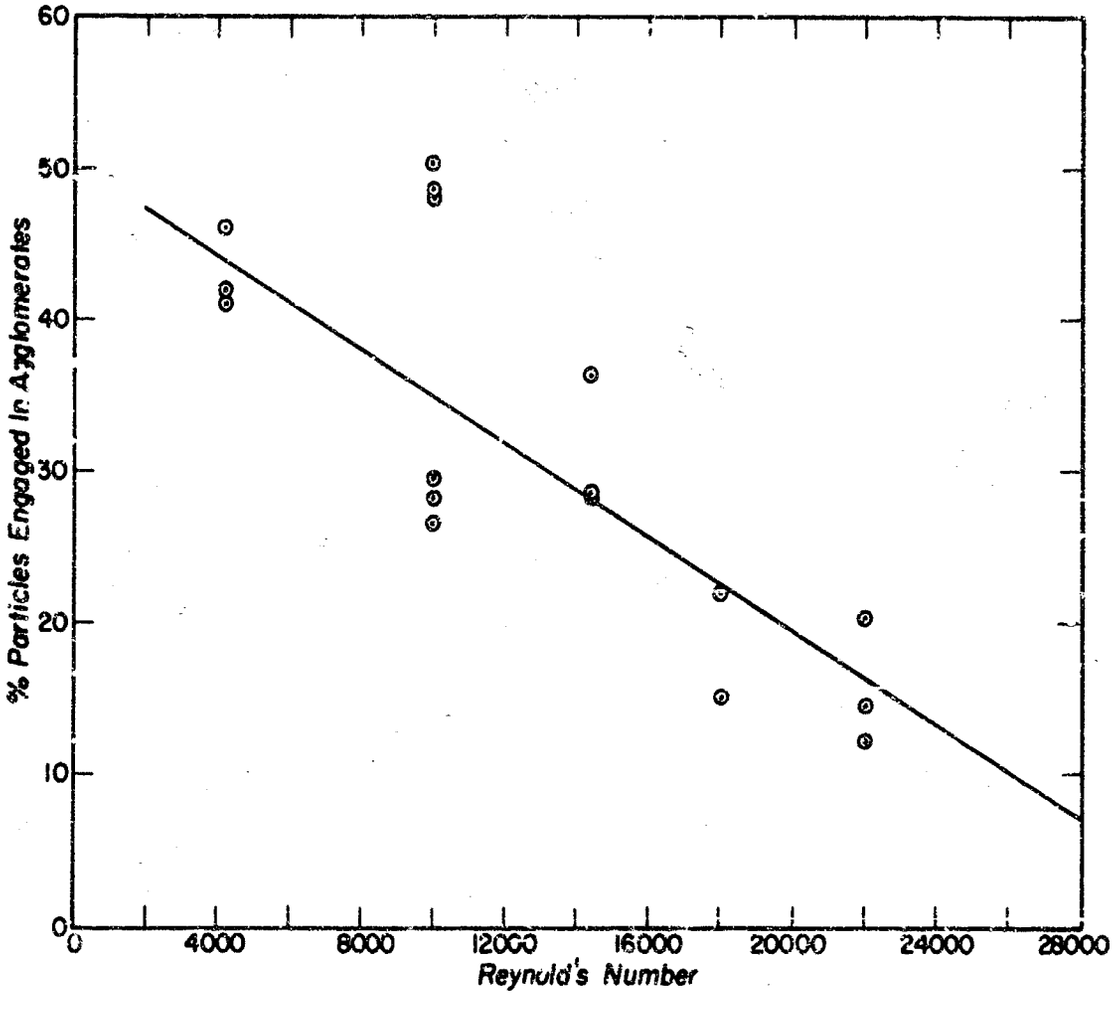

FIG. VIII-10 AGGLOMERATION OF MICROTHENE AFTER DISSEMINATION THROUGH 0.19 cm DIAMETER 20 cm LENGTH TUBE AT VARIOUS AIR VELOCITY

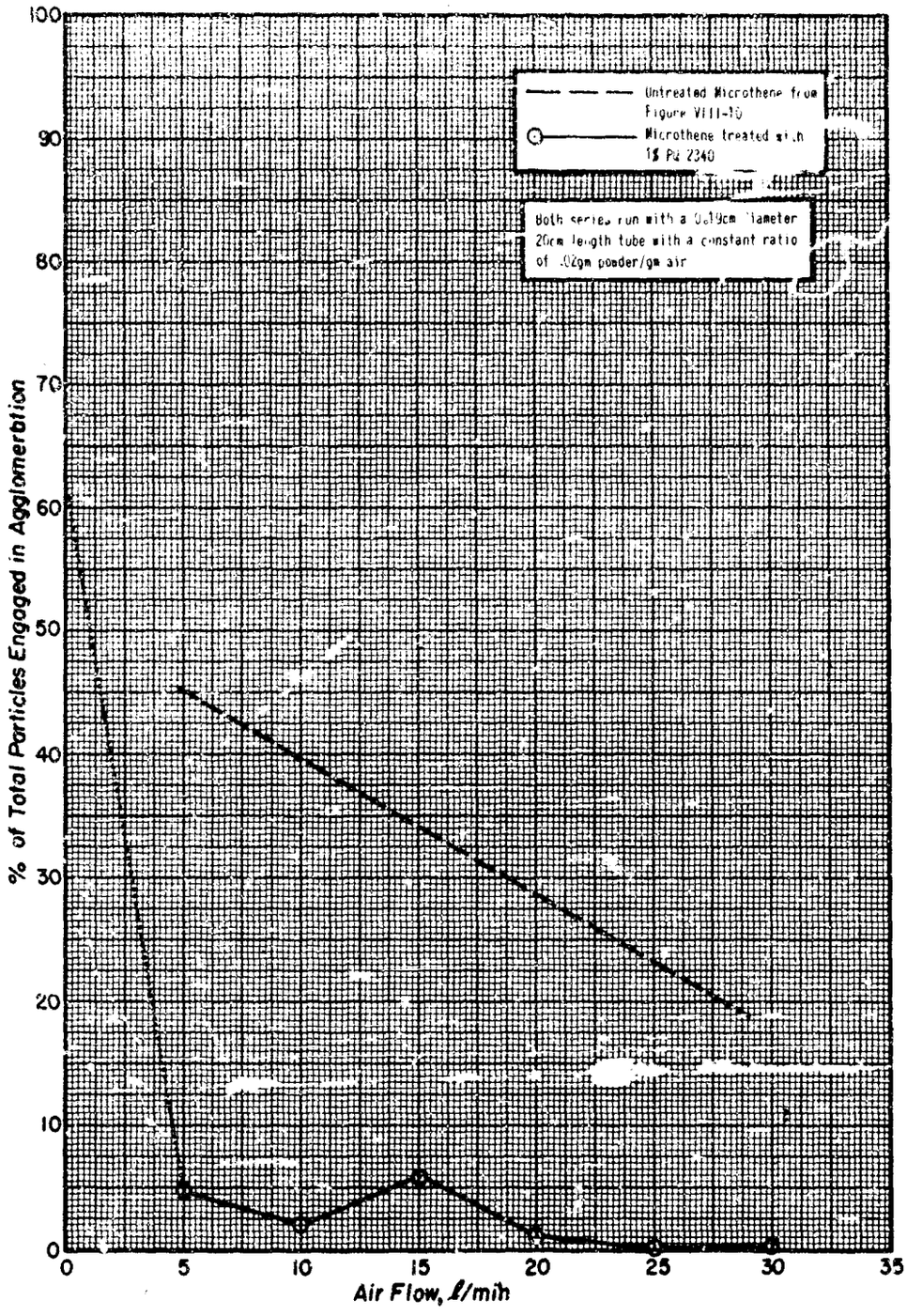

FIG. VIII-11 THE PERCENT AGGLOMERATION OF ADDITIVE-TREATED MICROTHENE AT VARIOUS AIR VELOCITY

4. Effect of Powder/Air Ratio Used in Dissemination

The first series of tests on the effect of powder/air ratio was made with Microthene in the six-foot vertical chamber. A straight tube, 0.19 cm in diameter x 20 cm length was used. The powder/air ratio was varied from 0.166 gm powder/gm air to 0.00128 gm powder/gm air. The data are given in Table VIII-6 and plotted in Fig. VIII-12. The lowest number of Microthene particles engaged in agglomeration was obtained with the smallest powder/air ratio. At 0.00128 gm powder/gm air, only 15% of the particles were agglomerated. There is a steady increase in agglomeration with increasing powder/air ratio in the interval from 0.001 to 0.01 gm powder/gm air. At greater ratios the fraction agglomerated remained steady, and there was little increase in Microthene agglomeration in the range 0.01 to 0.1 gm powder/gm air.

Tests were also made with quinine powder on the effect of powder/air ratio, The powder was disseminated through the single-coil tube and assessed with the laminar flow settling chamber. The differentiated mass distribution curves shown in Fig. VIII-13 for Run 18 and Run 20 revealed

Table VIII-6

DISSEMINATION OF MICROTHENE THROUGH CAPILLARY TUBES AT VARIOUS AEROSOL CONCENTRATIONS

Type of Capillary Tube	Dissemination Conditions	Aerosol Concentration in Tube gm Powder/gm Air	% Particles Engaged in Agglomeration	Reynolds Number
0.19 cm dia 20 cm length	30.0 ℓ/min 16.5 psi	0.00128	14.9%	23,600
0.19 cm dia 20 cm length	15.0 ℓ/min 5.4 psi	0.00256	30.8%	11,800
0.19 cm dia 20 cm length	15.0 ℓ/min 5.4 psi	0.0106	47.9%	11,800
0.19 cm dia 20 cm length	15.0 ℓ/min 5.4 psi	0.0412	48.6%	11,800
0.19 cm dia 20 cm length	15.0 ℓ/min 5.4 psi	0.0822	46.8%	11,800
0.19 cm dia 20 cm length	7.3 ℓ/min 1.7 psi	0.166	54.8%	5,750

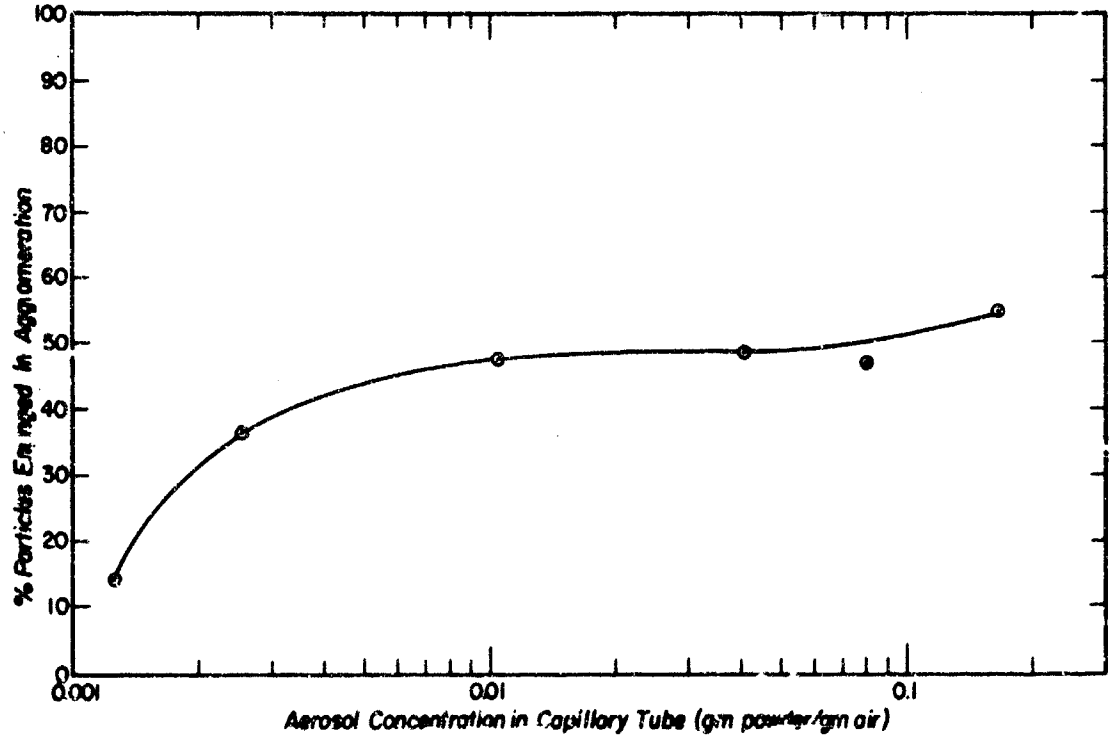

FIG. VIII-12 AGGLOMERATION OF MICROTHENE AFTER DISSEMINATION THROUGH 0.19 cm DIAMETER, 20 cm LENGTH TUBE AT VARIOUS AEROSOL CONCENTRATION (GM POWDER/GM AIR)

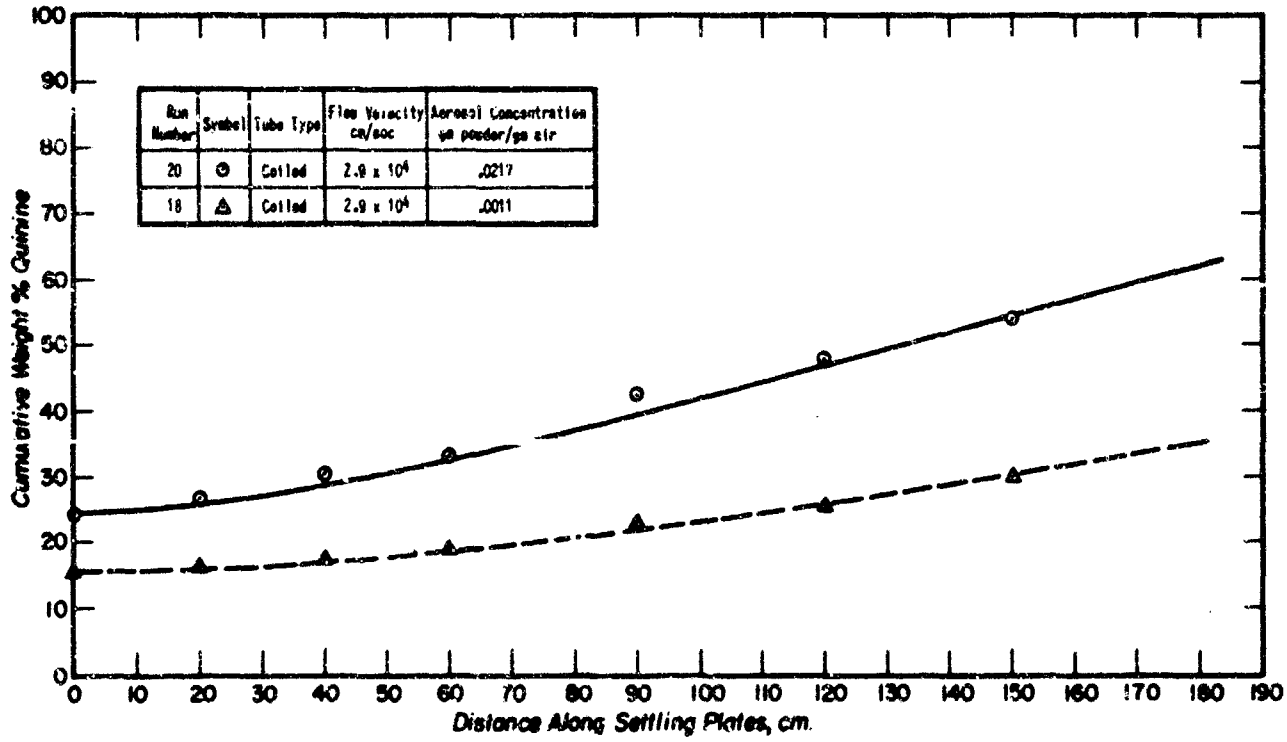

FIG. VIII-13 DIFFERENTIATED MASS [illegible] CURVES FOR QUININE ON THE SETTLING PLATES OF THE LAMINAR FLOW CHAMBER. The effect of powder/air ratio on the dissemination of quinine is shown

an obvious advantage to lower powder/air ratios. At a ratio of 0.001 gm powder/gm air, Run 18, the aerosol contained 84% of the particles having aerodynamic diameters equal to or less than 10μ, whereas with a ratio of 0.0217 gm powder/gm air, only 74% of the particles were equal to or less than 10μ. The data again indicates that small powder/air ratios favor more complete dissemination. The obvious conclusion is that reagglomeration of the aerosolized particles is a significant process, and must be given serious consideration in the design of any dissemination tests. Further discussion of the mechanics of the reagglomeration process is given in Section X-D.

B. Effect of Powder Modification on Aerosolization

There were several objectives for the study of powder modification, such as flowability, bulk density, storage, etc., but the primary objective was to improve the aerosolizability of a powder.

1. Effect of Additives

It is common practice in industry to use millimicron-size particles to modify the physical and chemical properties of bulk powders. These particles have been defined by various investigators as free-flow agents, powder-conditioning agents, additives, anti-caking agents, fluidizing agents, etc. To avoid confusion, the various agents are all categorized as additives throughout this report.

For some powders, the effect of additives is dramatic, not only in the flow properties of the powders, but also in their aerosolizabilities. Figures VIII-14, VIII-15 and VIII-16 of untreated and treated Microthene, saccharin and feldspar respectively show qualitatively how additives can affect aerosolizabilities. The powders were disseminated by the "puffer" and allowed to settle in the 100 cm chamber.

More quantitative data on the effect of additives are given below.

Samples of saccharin treated with 1% Estersil additive and with 1% PQ 2340 were disseminated with the "puffer" in the 100 cm settling chamber. Samples were collected on microscope slides during the intervals

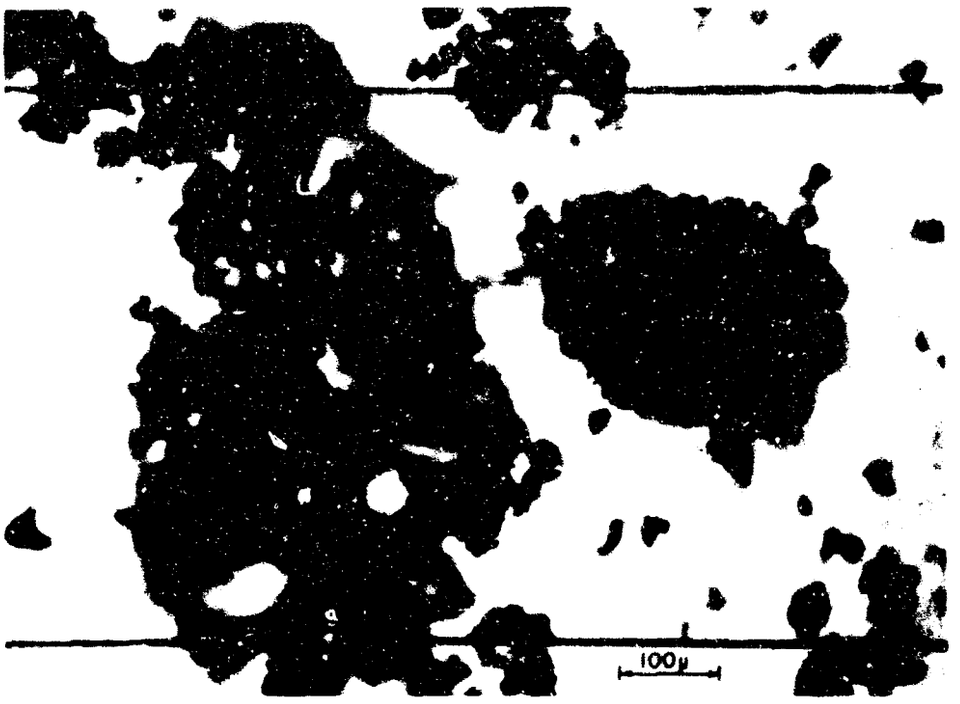

FIG. VIII-14a UNTREATED MICROTHENE DISSEMINATED BY GENTLE PUFF OF AIR, AND ALLOWED TO SETTLE ONTO SLIDES

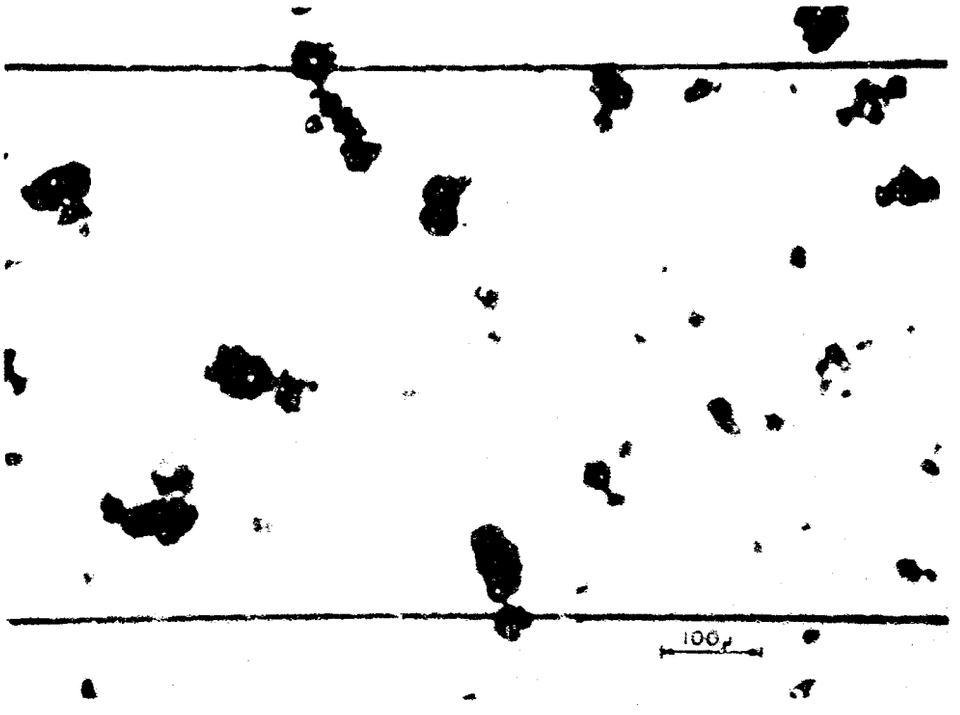

FIG. VIII-14b MICROTHENE, TREATED WITH 5% ADDITIVE, AND DISSEMINATED AS ABOVE

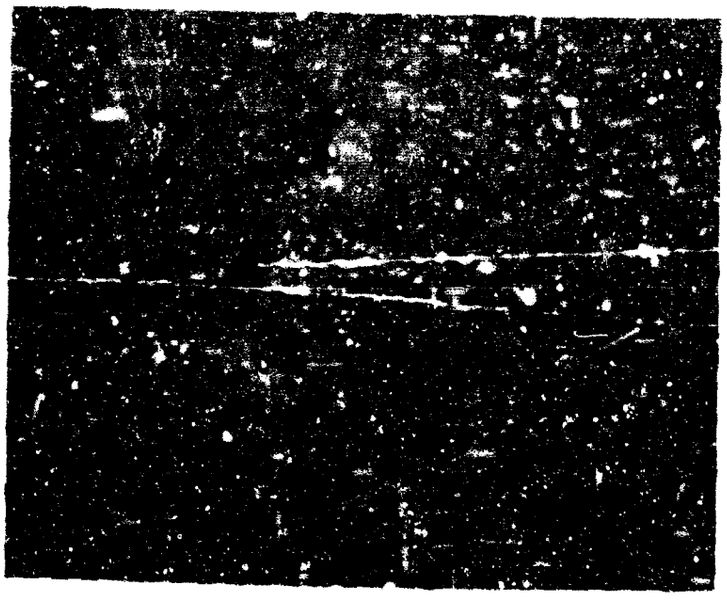

FIG. VIII-15a UNTREATED SACCHARIN, DISSEMINATED IN THE "PUFFER" SYSTEM

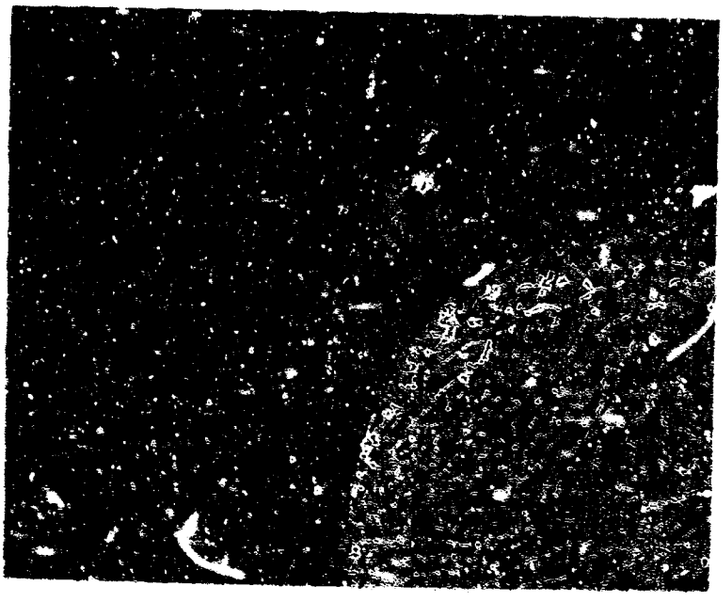

FIG. VIII-15b SACCHARIN, TREATED WITH 1% PQ 2040, DISSEMINATED AS ABOVE

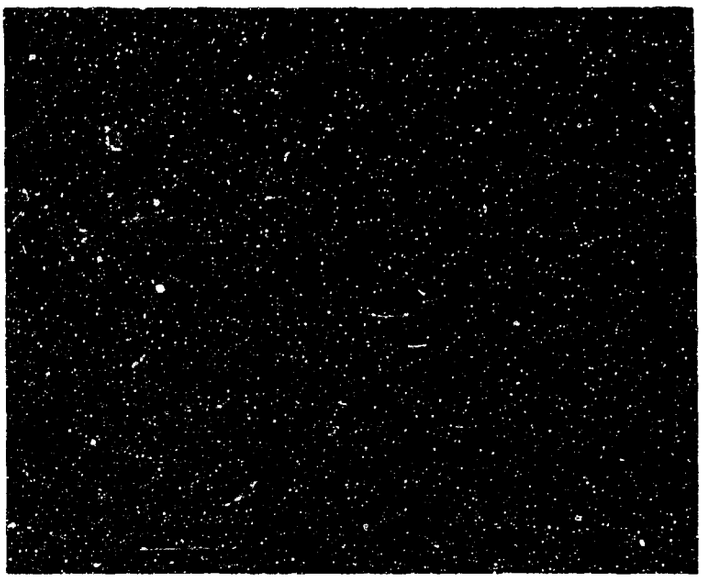

FIG. VIII-16a UNTREATED FELDSPAR, DISSEMINATED IN "PUFFER" SYSTEM

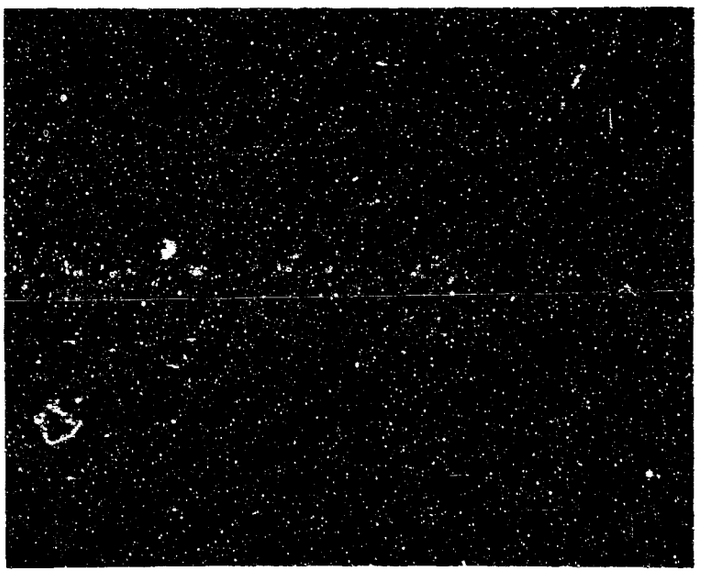

FIG. VIII-16b FELDSPAR TREATED WITH 1% PQ 2340, DISSEMINATED AS ABOVE

12-60 seconds, 60-300 seconds, and 300-600 seconds from the time of dissemination. The number of particles and agglomerates per square millimeter on the slides was then counted.

The data in Table VIII-7 show that saccharin treated with Estersil and with PQ 2340 produced 763 and 1050 particles per mm^2 respectively, while the untreated particles yielded only 253 particles per mm^2. For the same weight of powder disseminated the larger number of particles per square millimeter obtained with the treated powder was taken to be indicative of better dissemination than the smaller number obtained with untreated powder. Thus the data indicate a three and fourfold increase in the number of particles aerosolized at low energy.

Table VII

DISSEMINATION OF SACCHARIN TREATED AND UNTREATED AT LOW ENERGY

Slide No.	Sample	Time (secs)	Number of Particles/mm^2	Total Number Particles/mm^2 for Time 12-600 secs
175	Untreated saccharin	12-60	102	
176	Untreated saccharin	60-300	102	253
177	Untreated saccharin	300-600	49	
179	Saccharin w. 1% Estersil	12-60	249	
180	Saccharin w. 1% Estersil	60-300	413	763
181	Saccharin w. 1% Estersil	300-600	101	
184	Saccharin w. 1% PQ 2340	12-60	358	
185	Saccharin w. 1% PQ 2340	60-300	402	1050
186	Saccharin w. 1% PQ 2340	300-600	390	

The method of rating the powder by counting the number of settled particles did not properly weight large agglomerates that settled first and had a large mass compared to individual particles. In another series of tests similar to the above, powders were used which could be analyzed for their mass. Rhodamine B and quinine powders were selected because they were readily available and because they could be detected in small quantities by fluorometric analysis.

The selections of the additives were arbitrary, although the ones selected had been effective in reducing the agglomaration of other powders. Rhodamine B was tested with 5% of Cab-O-Sil and with 5% PQ 2340 to compare a hydrophilic additive with a hydrophobic one (PQ 2340). Quinine was treated with 5% Organ-O-Sil. Dissemination of the powders was performed as above. Settled particles on the microscope slides were dissolved and the mass determined with a fluorimeter. The results are tabulated in Table VIII-8.

Table VIII-8

DISSEMINATION OF RHODAMINE AND QUININE AT LOW ENERGY

Powder	Sampling Time Seconds	Weight of Dye Deposited on Slide (microgram/cm^2)	Weight Percent of Dye Deposited on Slide, Normalized	Weight Percent Deposited in Time Interval 12-600 secs
Rhodamine Untreated	0-12	15.5	49%	51%
	12-60	6.9	22%	
	60-300	7.2	23%	
	300-600	1.9	6%	
Rhodamine & 5% Cab-O-Sil	0-12	7.2	15%	85%
	12-60	15.0	30%	
	60-300	22.7	46%	
	300-600	4.3	9%	
Rhodamine & 5% PQ 2340	0-12	12.3	10%	90%
	12-60	37.5	29%	
	60-300	67.0	52%	
	300-600	11.2	9%	
Quinine Untreated	0-12	88.2	70%	30%
	12-60	26.7	21%	
	60-300	8.0	7%	
	300-600	2.7	2%	
Quinine & 5% Organ-O-Sil	0-12	20.6	30%	70%
	12-60	40.0	58%	
	60-300	5.9	9%	
	300-600	2.4	3%	

The quantity of powder found in the time intervals 0-12, 12-60, 60-300 and 300-600 secs are tabulated in columns three and four. Column four gives the mass analysis, normalized. It is seen that for the

untreated powders, 50% of the rhodamine B and 30% of the quinine remained airborne after the first 12 seconds. For the treated powders, 85-90% of the rhodamine and 70% of the quinine were airborne after 12 seconds, thus indicating that the additives were effective in causing more particles to remain airborne for a longer period of time.

In Section V-D, measurement of the bulk properties of powders such as angle of repose, adhesion and viscosimetry indicated that there was an optimum amount of additive (approximately 1%) which produced a desired effect. Therefore, it was not unreasonable to hope that a similar relationship could be found between the amount of additive and the aerosolizability.

To explore the above possibility, Microthene containing 0.0%, 0.1%, 0.4%, 0.7%, 1.0%, 1.5% and 3.0% PQ 2340 was disseminated using the puffer system. The particles and agglomerates were allowed to fall onto glass slides, and the slides assessed by counting the number of particles in agglomerates. These curves are shown in Fig. VIII-17. The curves fell into the distinct groups representing: 1) highly agglomerated powder, occurring with untreated powder, 2) medium agglomerated powder, occupied by the 0.1% and 3.0% additive powders, and 3) the less agglomerated powder, occupied by the 1.0%, 1.5%, 0.4% and 0.7% powders. The data showed that small and large amounts of additive (0.1% and 3.0%) provided some reduction in the agglomeration of untreated Microthene But the greatest reduction occurred with amounts near the 1% level. For example, if a line is drawn across the curve at the 10-particle size agglomerates, and if the total number of particles engaged in agglomerates of 10 or less particles is plotted against the percent additive, a curve peaking at 1.0% additive is obtained as in Fig. VIII-18. A different curve could have been obtained by drawing a line at the 30-particle or 70-particle agglomerates level in Fig. VIII-17, but in general, the curves indicate that Microthene containing approximately 1% PQ 2340 was the least agglomerated mixture when disseminated at low energy. This was approximately the same concentration of additive that produced the smallest angle of repose as described in Section V-D.

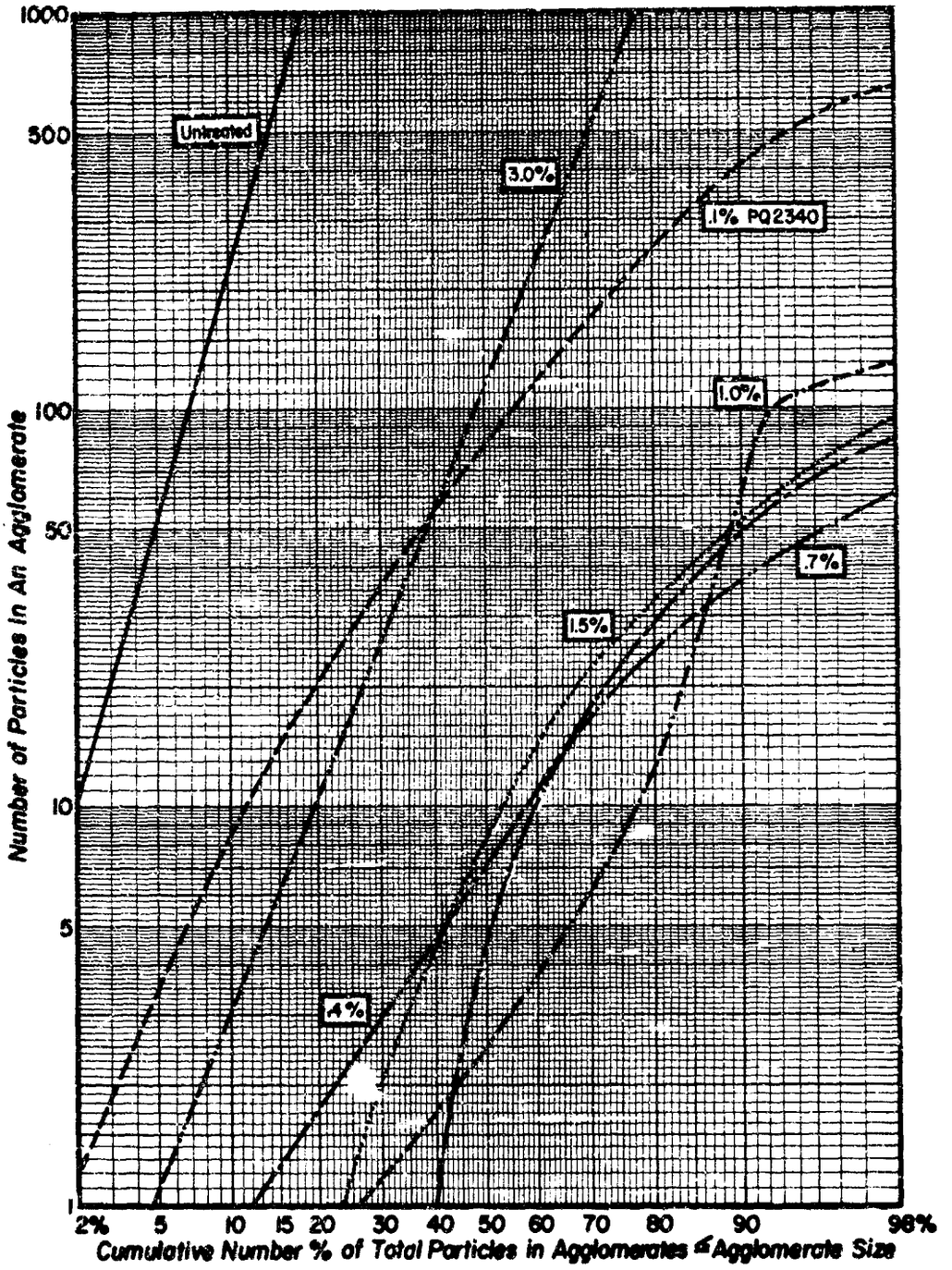

FIG. VIII-17 CURVES SHOWING SIZES OF AGGLOMERATES OF MICROTHENE FOR DIFFERENT CONCENTRATIONS OF PQ 2340 DISSEMINATED AT LOW ENERGY

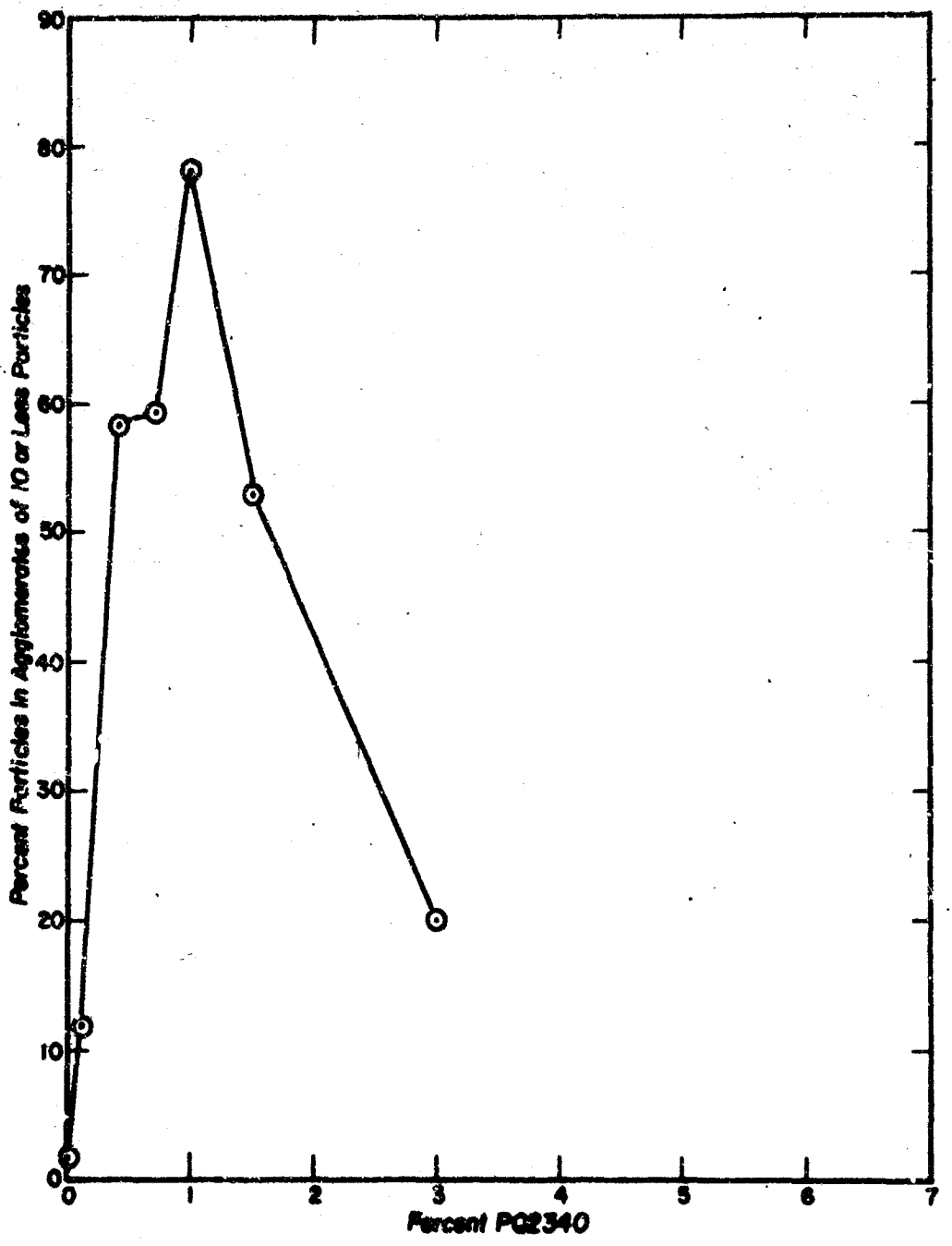

FIG. VIII-18 CURVE RELATING PERCENT AGGLOMERATES AND PERCENT ADDITIVE, FOR 10-PARTICLE SIZE AGGLOMERATES. Data taken from Fig. VIII-17

To explore the effect of higher dissemination energy on the deagglomeration of additives, Microthene containing 0.0%, 0.1%, 0.5%, 1.0%, 3.0%, 5.0% and 7.0% PQ 2340 was disseminated at an air velocity of 11,700 cm/sec through a 0.19 cm diameter, 20 cm long brass tube into the six-foot vertical settling box.

Slides were laid on the bottom of the chamber to collect the particles and agglomerates. From these slides, the number of particles engaged in agglomeration were counted and the results plotted in Fig. VIII-19. It can be seen that the optimum concentration of additive lies around 0.5%-1.0%. This is almost the same concentration of additive that produced the least agglomerated powders when disseminated with the low energy puffer system. It is likely that the higher energy used for dissemination in these later tests obscured the agglomeration tendency of the mixtures containing 5%-7% additive by completely deagglomerating them.

It had been suggested that small turbulent air cells would assist in breaking up small agglomerates in the dissemination tube, and a possible method of generating small turbulent cells was to disseminate large heavy particles in an air stream along with the powder.

Mixtures of Microthene containing 10%, 30%, 50% and 95% by weight tungsten spheres (60 micron MMD; density 19.3 gm/cc) were disseminated upward through a 0.19 diameter tube at 20 liter/min. The percentage of particles engaged in agglomeration was determined by counting the number of Microthene particles attached to each other. Figure VIII-20 shows the results of this study. The percentage of agglomeration varied only slightly with the amount of tungsten added, from 32% to 43%. Untreated Microthene had 35% to 45% of the particles engaged in agglomeration under identical flow rates; therefore, the effect of the tungsten on the deagglomeration of Microthene was minimal.

The 10% tungsten-90% Microthene mixture was also disseminated at various air flows, 10 liter/min, 20 liter/min, and 30 liter/min. The results shown in Fig. VIII-21 were not sufficiently conclusive to proceed further with this approach.

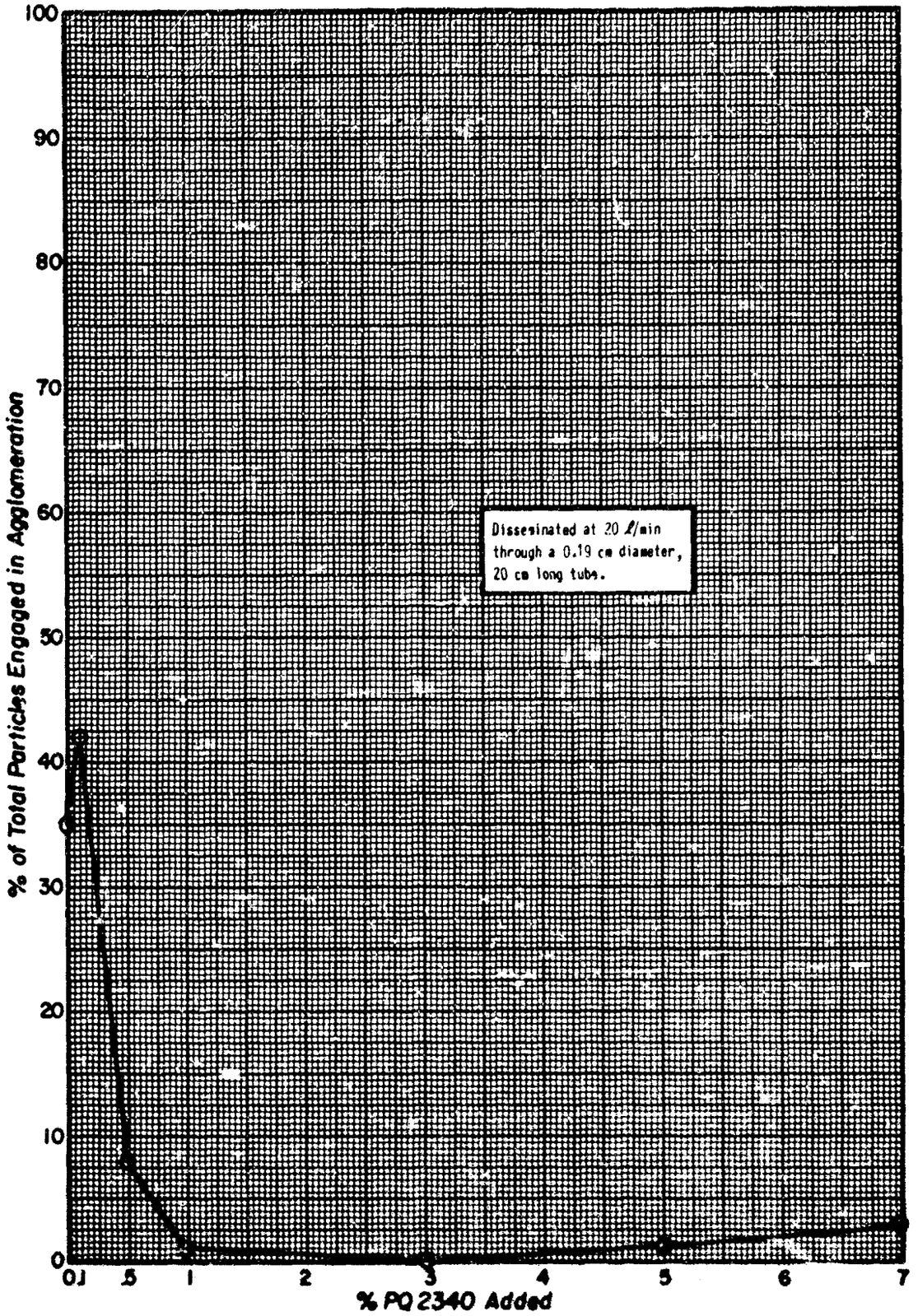

FIG. VIII-19 THE AGGLOMERATION TENDENCY OF MICROTHENE TREATED WITH VARIOUS PERCENTAGE OF PQ 2340 ADDITIVE AND DISSEMINATED BY A RELATIVELY HIGH ENERGY SYSTEM

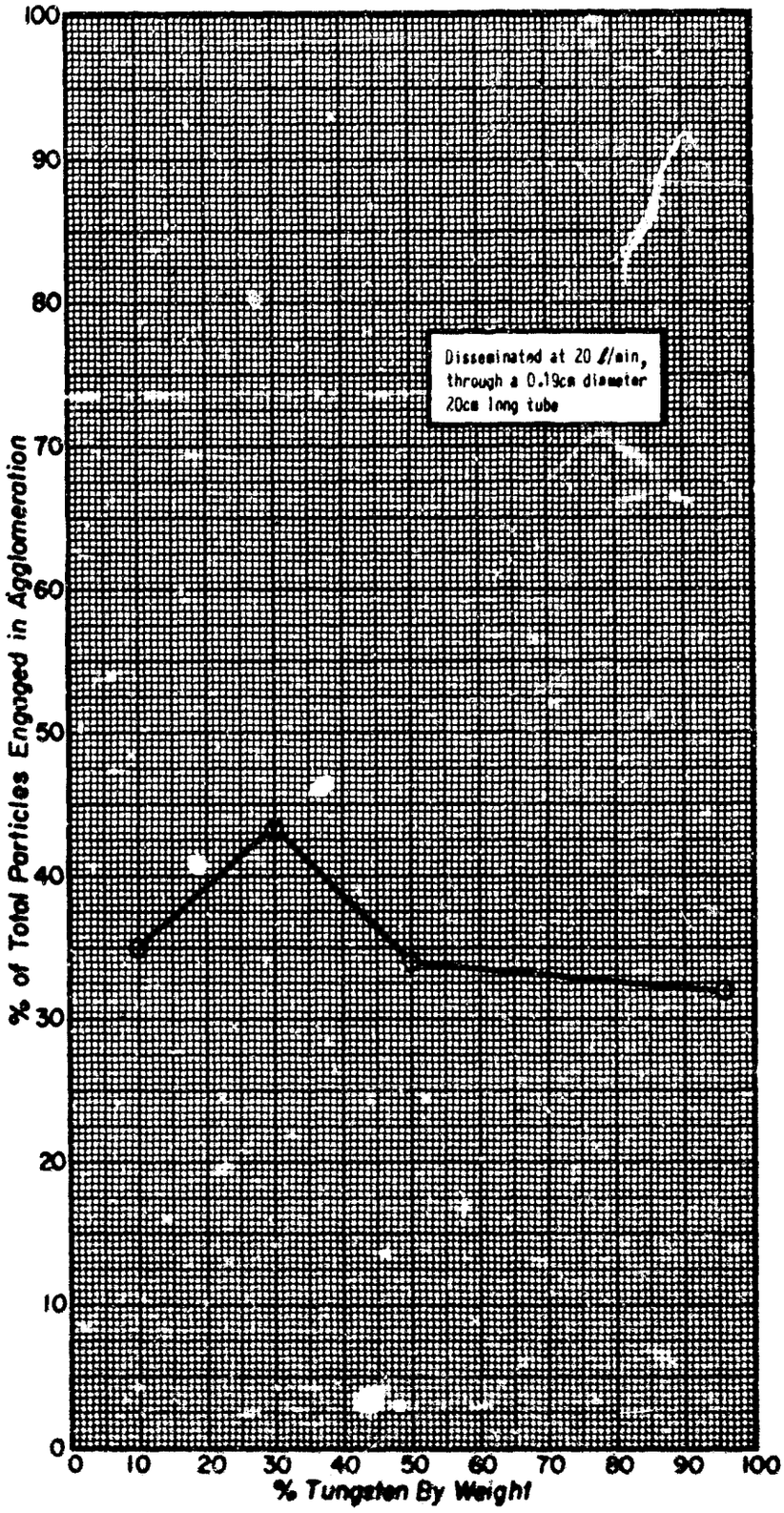

FIG. VIII-20 THE EFFECT OF VARIOUS CONCENTRATION TUNGSTEN PARTICLES (60μ DIA) ON THE DEAGGLOMERATION OF MICROTHENE

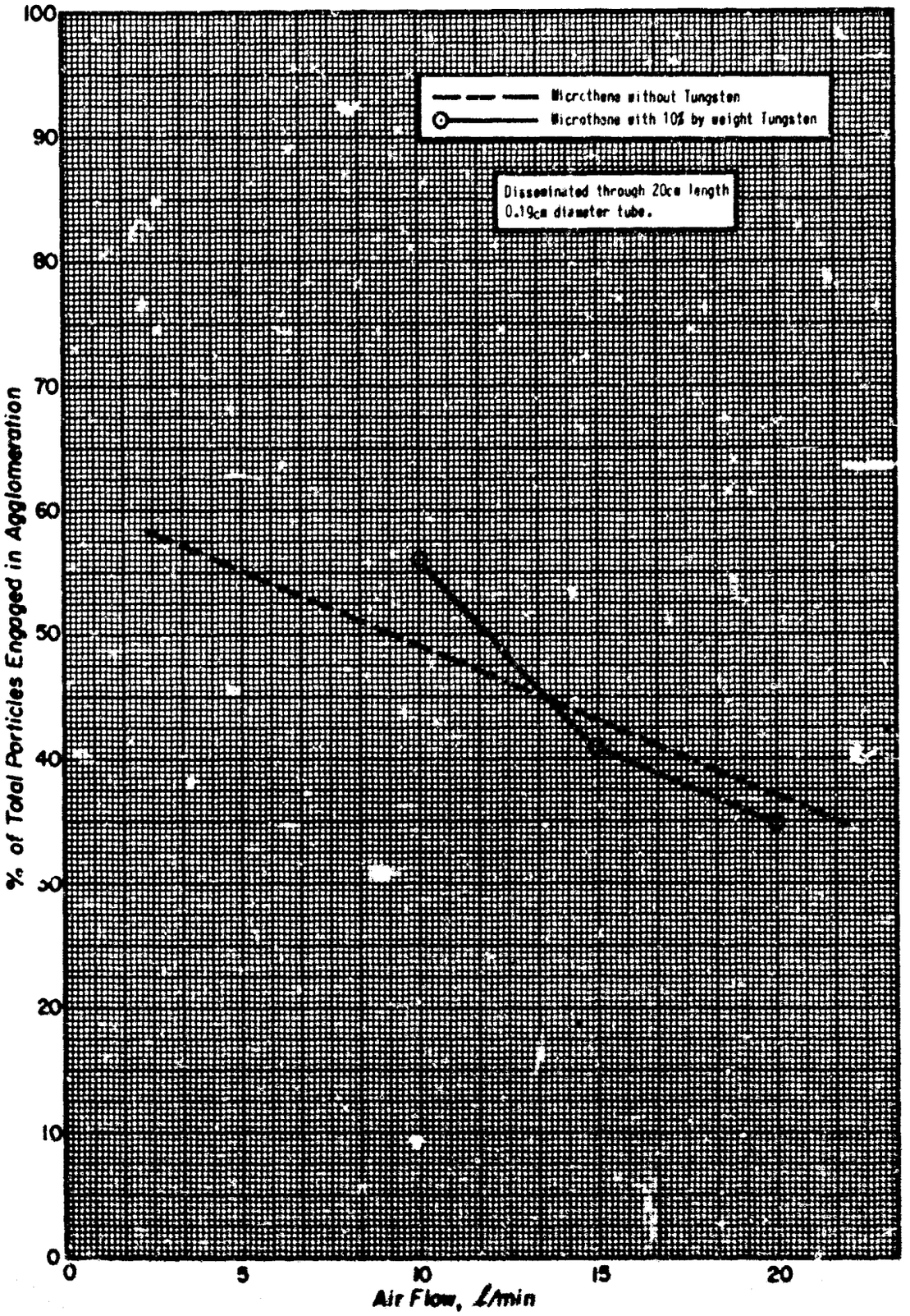

FIG VIII-21 AGGLOMERATION OF MICROTHENE CONTAINING 10% TUNGSTEN SPHERES WHEN DISSEMINATED AT VARIOUS AIR FLOWS

2. Comparison of Additives

The aerosolizability of saccharin was tested with three additives to obtain some comparison between the effects of different additives. The additives are all millimicron size silica particles, and they have different physical and surface properties. The additive, PQ 2340 was chosen because it was the most negative of the silica additives tested, as determined by electrostatic tests in Section IX, and Organ-O-Sil was selected because it was the most positive. Cab-O-Sil was tested because it is hydrophillic, in contrast to the other two which are hydrophobic, and because it has been employed extensively by other investigators. All the treated powders contained 1% additive.

The saccharin mixtures were disseminated with the puffer system in the 100 cm chamber. During the assessment, it was found that the physical shape of saccharin made it difficult to distinguish a multi-particle agglomerate from a large primary particle. Consequently, for these tests, no distinction was made between an agglomerate and a single particle, and all agglomerates were counted as single particles. Since equal weights of the powders were disseminated, the greater the number of particles found on the deposition slide, the better the aerosol.

The results are tabulated in Table VIII-9 below. In terms of the number of particles counted, all the powder with additive gave a better aerosolizability than powder without. The improvement in aerosolizability was obvious from merely observing the particle-agglomerates. Figures VIII-22, VIII-23, VIII-24, and VIII-25, are photomicrographs of the untreated, Cab-O-Sil treated, Organ-O-Sil treated and PQ 2340 treated particles. From these pictures, the PQ 2340 treated powder yielded the largest number of particles/mm^2, and consequently, the smallest size particles. These results showed the types of additives which are most

Table VIII-9

COUNT DATA FOR SACCHARIN TREATED WITH SEVERAL SILICA ADDITIVES

Slide No.	Saccharin Treatment	Counts Particles/mm^2
140	As received	112
164	1% Cab-O-Sil	132
160	1% Organ-O-Sil	190
168	1% PQ 2340	453

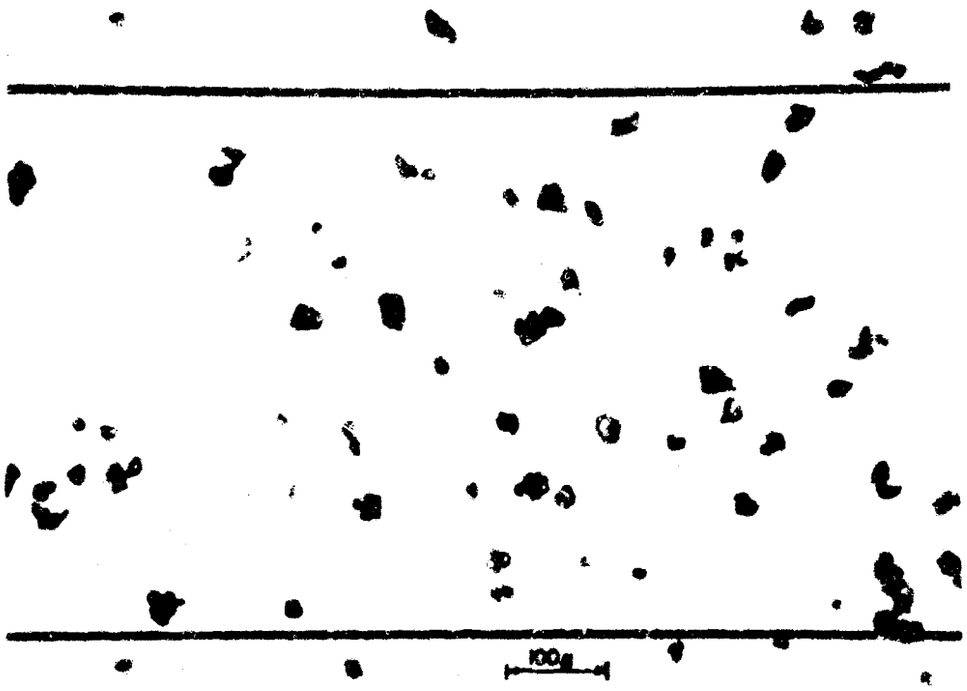

FIG. VIII-22 AEROSOLIZED PARTICLES OF SACCHARIN, UNTREATED

FIG. VIII-23 AEROSOLIZED PARTICLES OF SACCHARIN TREATED WITH 1% CAB-O-SIL

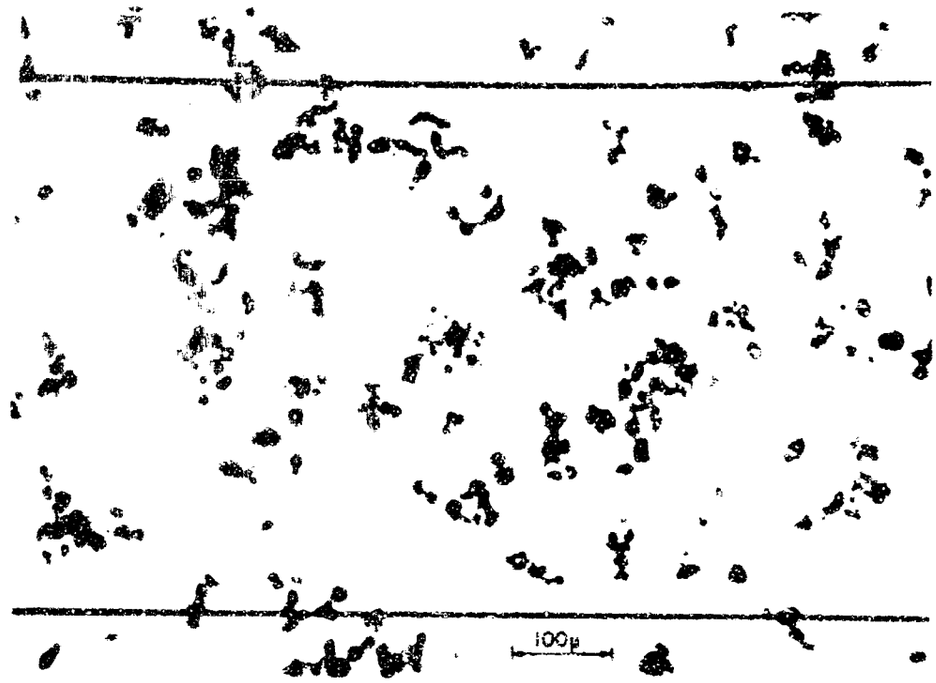

FIG. VIII-24 AEROSOLIZED PARTICLES OF SACCHARIN TREATED WITH 1% ORGAN-O-SIL

FIG. VIII-25 AEROSOLIZED PARTICLES OF SACCHARIN TREATED WITH 1% P.G. NO. 2340

beneficial with saccharin. It does not follow that the best additive for saccharin would be the best for some other material.

3. Effect of Particle Size and Shape

Theory dictates that the stress required to separate fine particles is greater than that required to separate coarse ones. A few simple experiments were designed to verify the theory. This was done by selecting particles of narrow size ranges and subjecting them to dissemination under identical conditions and then observing the relative aerosolization of each size. This was accomplished with spherical glass beads which had been classified into five size ranges: greater than 18μ, 18-12μ, 12-9μ, 9-5μ, and less than 5μ. The double-disc air classifier described in Section V was used for classifying the beads.

At large dissemination energy, all the beads, including the smallest fraction, were nearly 100% aerosolized. To observe the agglomeration tendency it was necessary to use low energy for dissemination. Thus the beads were disseminated using the "puffer" system and the 100 cm settling chamber. The extent of agglomeration for the five fractions of beads are given in Table VIII-10 below. The data indicated an increase in agglomeration with decreasing particle size. In this assessment technique, each agglomerate irrespective of number of beads attached, was counted as one agglomerate, but identification of agglomerates by number without a description of agglomerate size does not reveal the full extent of agglomeration. For example, the agglomerates in fractions greater than 18μ and 18-12μ consisted mainly of two adhering particles, whereas agglomerates in fractions less than 5μ and 9-5μ were made up of three or five or even 20 particles. Consequently, the weight fraction of agglomerates in the smaller particle sizes was actually greater than that tabulated. Nevertheless, the

Table VIII-10

DEGREE OF AGGLOMERATION
OF FIVE FRACTIONS OF GLASS BEADS

Sample of Glass Beads	% Agglomerates
Fraction "I," >18μ	15%
Fraction "H," 18-12μ	19%
Fraction "F," 12-9μ	22%
Fraction "D," 9-5μ	23%
Fraction "B," <5μ	32%

results showed that particle sizes, even in a very narrow size range of 5μ to 18μ, played an important part in its agglomeration tendency.

To explore the effect of particle shape, a comparison between irregularly shaped quartz powder and glass beads was made. The quartz powder was classified into the same Stokes class sizes using the same machine setting as was used for glass beads. The same weights of quartz powder and glass beads were disseminated under identical conditions. The results in Table VIII-11 showed that quartz powder produced fewer agglomerates (approx. 10%) than the glass beads (approx. 42%). For identical weights, the quartz powder yielded 847 particles/mm^2 to the bead's 81 particles/mm^2, a factor of more than ten. If the corresponding size fractions of quartz and beads contained equivalent numbers of particles, then the data indicated that the quartz powder is the more easily aerosolizable powder. The two materials have the same specific density, thus the difference in aerosolizability may be attributed to the particle shape,

Table VIII-11

DISSEMINATION OF QUARTZ POWDER AND GLASS BEADS
TO COMPARE THE EFFECT OF PARTICLE SHAPE ON AEROSOLIZABILITY

Sample	No. of Particles per mm^2	Agglomerates per mm^2	% Agglomerate
Quartz 9-5μ dia.	341	46	13%
Quartz 12-8μ dia.	266	34	11%
Quartz 18-12μ dia.	240	12	5%
	Total 847	Total 92	Averaged 10%
Glass Beads 9-5μ dia.	35	19	54%
Glass Beads 12-8μ dia.	30	9	23%
Glass Beads 18-12μ dia.	16	6	27%
	Total 81	Total 34	Averaged 42%

assuming that their intrinsic properties are identical. These results are to be expected if it is assumed that there are fewer points of contact in the bulk powder between irregular shaped particles than there are for spherical particles.

4. Effect of Chemical and Physical Modifications

This section reveals the effect on aerosolization of chemical and physical treatment of quinine. In some instances the chemical treatment is merely the incorporation of an additive, but when this occurs simultaneously with a change in the physical state of the powder, i.e. change in particle size, then it is difficult to determine which of the two changes is responsible for the differences in aerosolization. In most cases, the two are intertwined such that both treatments are partially responsible.

All the tests in this section were made with the laminar flow settling chamber. Modified and unmodified quinine was the test powder, and the single-coil tube was used for dissemination.

In one of the first chemical and physical treatments, quinine with 1% PQ 2340 added was blended in an Osterizer Blender for six minutes. Some grinding of the particles should have occurred, but at the same time, the heat generated by the grinding could have caused some fusion of the particles. An estimate of the particle size distribution of this treated powder is given in Fig. IV-4. The size distribution was obtained by microscopic sizing after first dispersing the powder in oil, and working the dispersion with a wooden stick. The number mean diameter was 1.8 microns. In comparison the "as received" powder, dispersed and sized by the same procedure, gave a nmd of 1.6μ, Fig. IV-2. However, it should be mentioned that the particle size of quinine is sensitive to the amount of dispersion performed on the microscope slide.

A comparison of the mass distribution curves between the untreated quinine and the PQ 2340 treated powder is shown in Fig. VIII-26. Since the untreated powder gave a lower mass distribution curve, it was concluded that the treatment of blending PQ 2340 with quinine was detrimental to the aerosolizability of the powder. Mass analysis showed that 78% of the

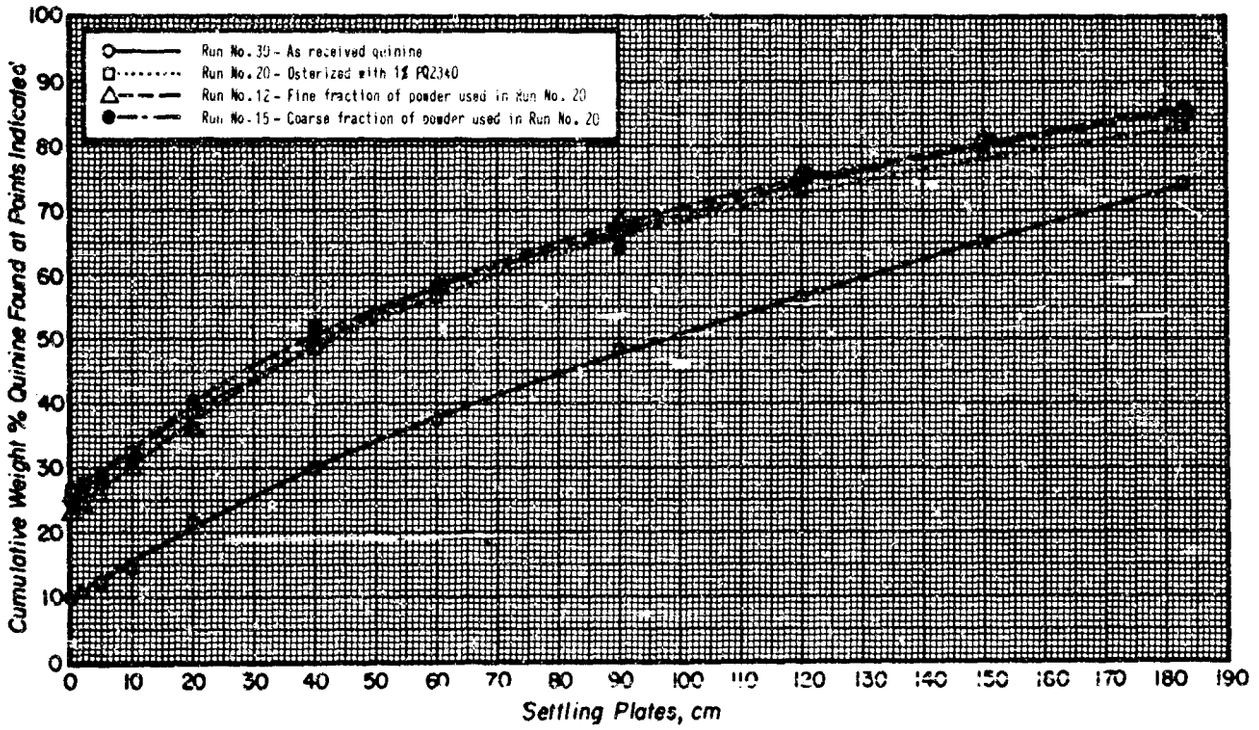

FIG. VIII-26 MASS DISTRIBUTION CURVES OF QUININE DISSEMINATED INTO LAMINAR FLOW SETTLING CHAMBER TO COMPARE THE EFFECTS OF CHEMICAL AND PHYSICAL MODIFICATION OF THE POWDER

untreated powder aerosolized to particles less than 10μ aerodynamic diameter, whereas with the osterized powder, only 65% were equal to or less than 10μ diameters.

On the same figure are mass distribution curves from a fine and coarse fraction which had been separated from the 1% PQ 2340 treated powder using the double-disc separator. Surprisingly, both the coarse and fine fractions gave similar distribution curves, and both curves matched that obtained from the unfractionated PQ 2340 treated powder.

The physical sizes of the fine and coarse fractions were vastly different as shown below. To explain the phenomenon, either the agglomerates from both materials break down to the same ultimate particle sizes, or the size of the settled agglomerate is controlled by a reagglomeration mechanism.

Figure VIII-27 and VIII-28 are photomicrographs of the fine and coarse fractions, respectively, dispersed in oil. The fine fractions showed well dispersed rectangular particles, but it would not be unexpected nor difficult for the large agglomerate of the coarse fraction to break down into similar sized particles in the dissemination process. The fine fraction dispersed easily in oil and size analysis of the material gave a nmd of 5.7μ. However a measure of the nmd of the coarse fraction was not feasible because the primary particle size was dependent on the amount of work devoted to dispersing the agglomerates.

To observe the compositions of the agglomerates formed by the fine and the coarse fractions, the powders were disseminated (under conditions comparable to Runs 12 and 15) upward into a vertical chamber and allowed to settle on slides to avoid the classification of sizes that occurs in the horizontal chamber. Photomicrographs were made of the settled agglomerates, Fig. VIII-29a and Fig. VIII-30a, of the fine and coarse fractions respectively. Figures VIII-29b and VIII-30b are the same settled particles but dispersed in oil. The nmd of the dispersed fine particles was 2.1μ as compared with 5.7μ in the non-disseminated material. This decrease in particle size strongly suggests that grinding of quinine occurred in the coiled tube, even with particles as small as the fine fraction.

FIG. VIII-27 FINE FRACTION OF QUININE WHICH HAD BEEN TREATED WITH 1% PQ 2340
Nondisseminated; dispersed with oil

FIG. VIII-28 COARSE FRACTION OF QUININE WHICH HAD BEEN TREATED WITH 1% PQ 2340
Nondisseminated; dispersed in oil

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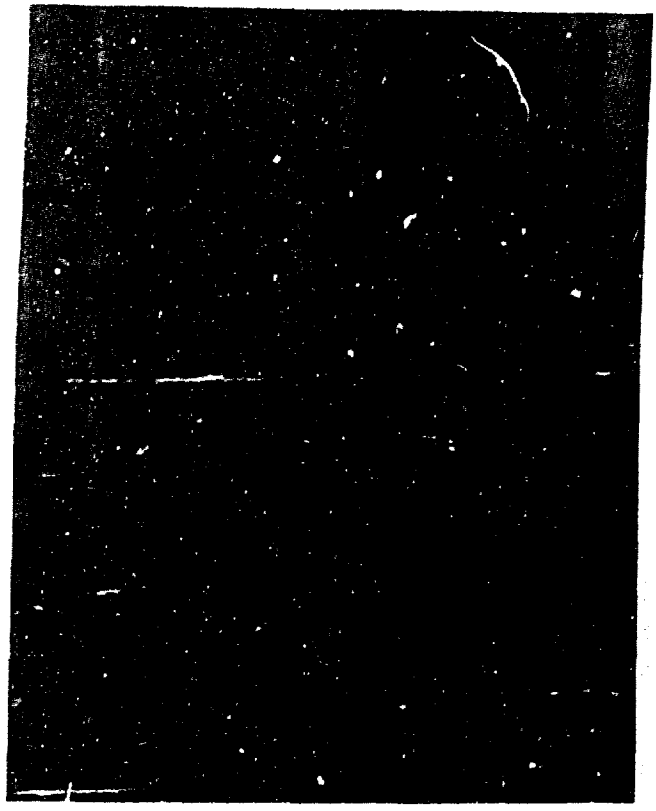

FIG. VIII-29a AGGLOMERATES OF QUININE FROM FINE FRACTION AFTER DISSEMINATION THROUGH COILED TUBE

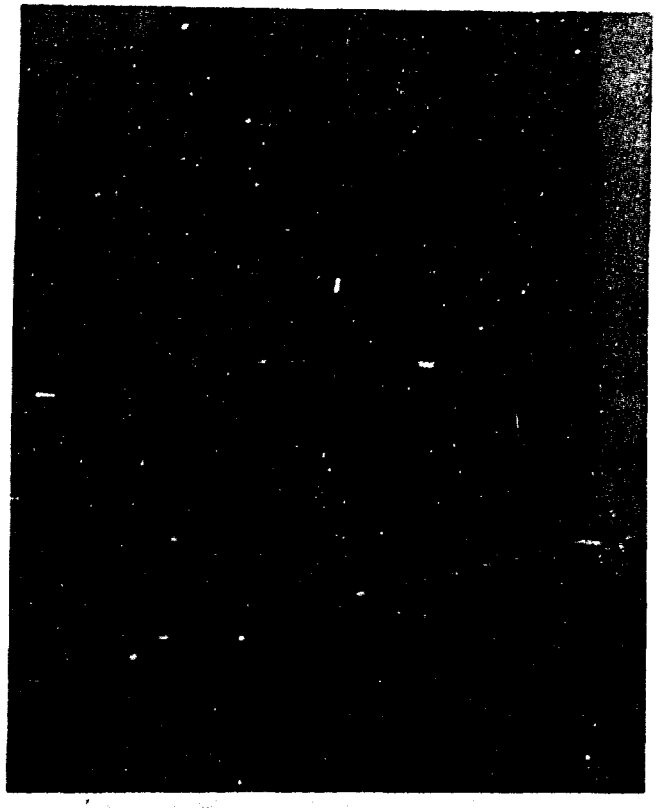

FIG. VIII-30a AGGLOMERATES OF QUININE FROM COARSE FRACTION AFTER DISSEMINATION THROUGH COILED TUBE

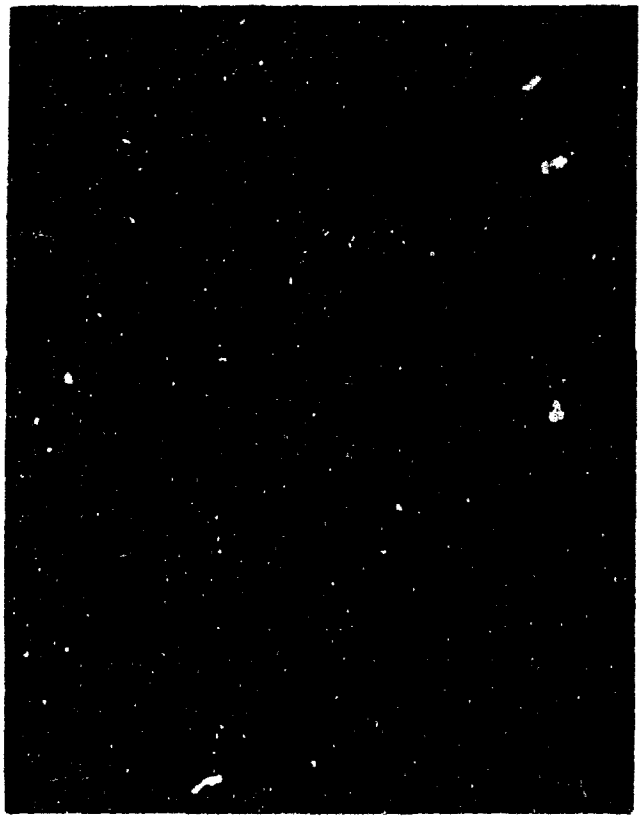

FIG. VIII-29b AGGLOMERATES OF QUININE (FROM FIG. VIII-29a) DISPERSED IN OIL

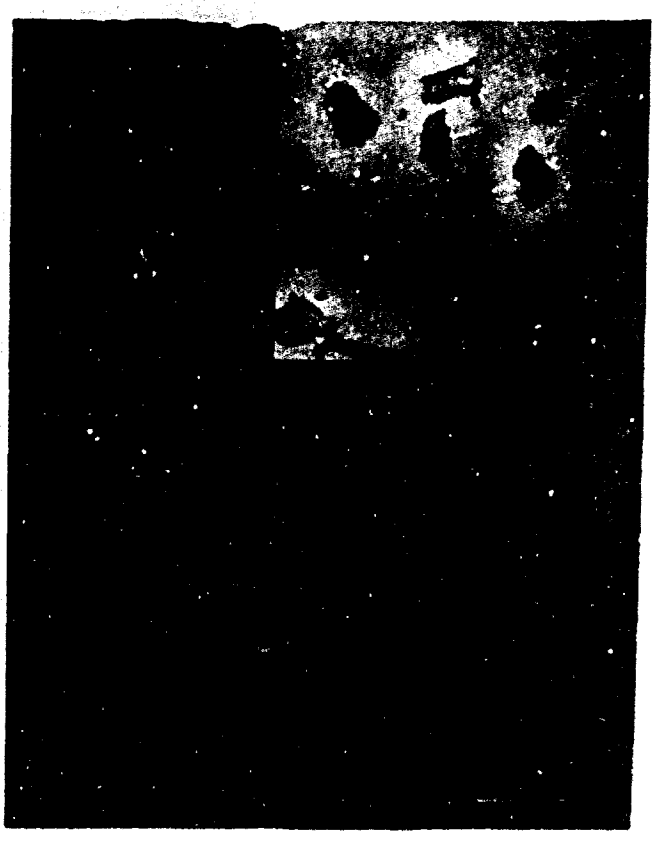

FIG. VIII-30b AGGLOMERATES OF QUININE (FROM FIG. VIII-30a) DISPERSED IN OIL

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In contrast to the effect of blending quinine with PQ 2340, which was detrimental to the aerosolization process, another chemical-physical treatment served to improve the aerosolizability of quinine. In this treatment, quinine was ground in a ball-mill for eight hours using petroleum ether as the suspending medium. One sample contained only petroleum ether in the medium, but in an accompanying sample, 1% hexamethyldisilizane and 4% Cab-O-Sil was added to the petroleum ether. The two materials were added because they were found by Rauner et al. (1966) to be effective in improving the aerosolizability of some organic simulants.

The dissemination results of the two ground quinine powders are shown in Fig. VIII-31. Both powders produced small agglomerates on the settled plates. There was little difference between the two products which suggested that the hexamethyldisilizane and Cab-O-Sil had no effects. However, the ball milling in petroleum ether was beneficial as both powders aerosolized better than the "as received" powder (Run 30), and considerably better than the PQ 2340 blended powder (Run 20).

A third treatment involved the use of n-butyl amine. Its use was suggested from the favorable results obtained by Nash et al. (1963) when they used it to reduce the agglomeration tendency of Carbo-Wax powders. Since the quinine is soluble in the amine, the treatment was limited to exposing the quinine to the amine vapors for approximately eight hours.

The result of the amine treated powder is also shown in Fig. VIII-31, and the data show that the treatment was not beneficial. The mass distribution curve showed it to be more aerosolizable than the PQ 2340 blended sample, but less than the "as received" or the ball-milled samples.

To illustrate the structures and sizes of agglomerates which land at the various spots on the settling plates, photomicrographs were taken at distances of 2 cm, 40 cm, 80 cm and 183 cm along the settling plate for Run 37, the ball-milled treated powder. The photographs are shown in Figs. VIII-32a, b, c, and d, respectively.

A distinctive feature of all quinine agglomerates is their loose structure and chain-like (not necessarily straight) appearance. The optical diameter of the particles on any section of the plate was

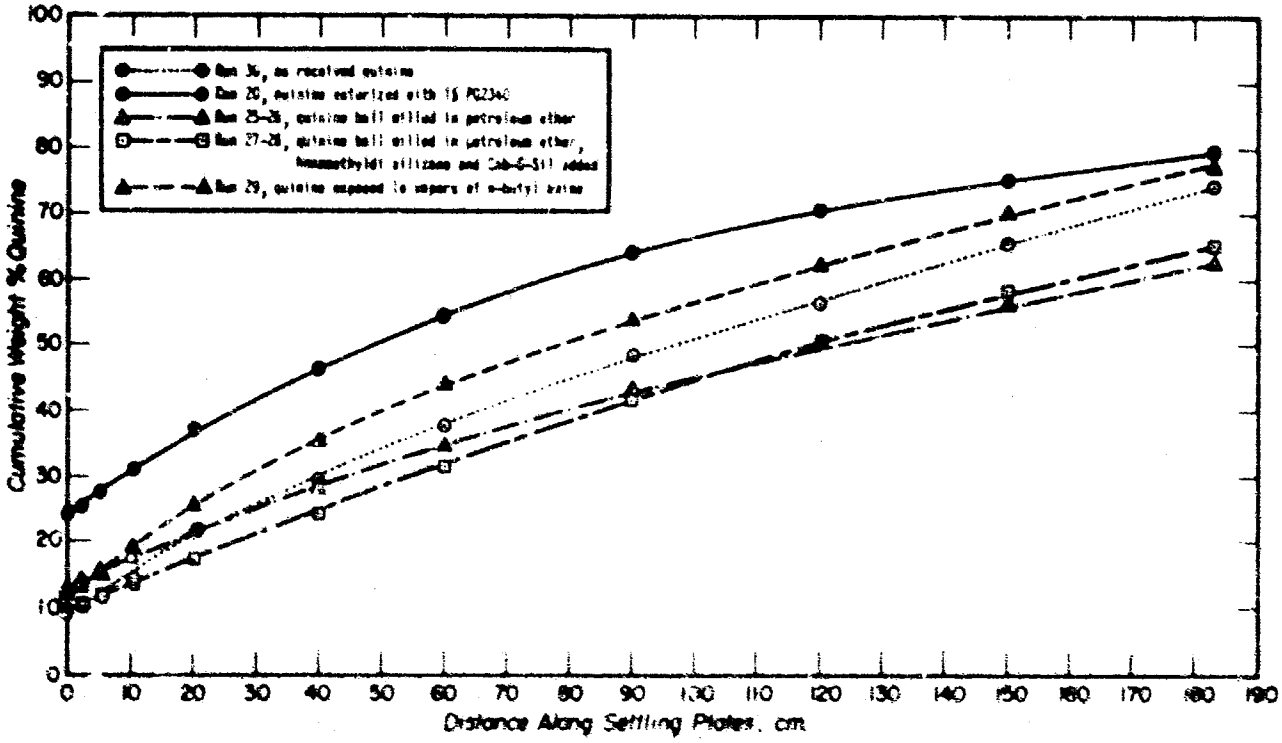

FIG VIII-31 MASS DISTRIBUTION CURVE FOR QUININE DISSEMINATED INTO LAMINAR FLOW SETTLING CHAMBER TO COMPARE THE EFFECT OF CHEMICAL AND PHYSICAL TREATMENT ON THE POWDER

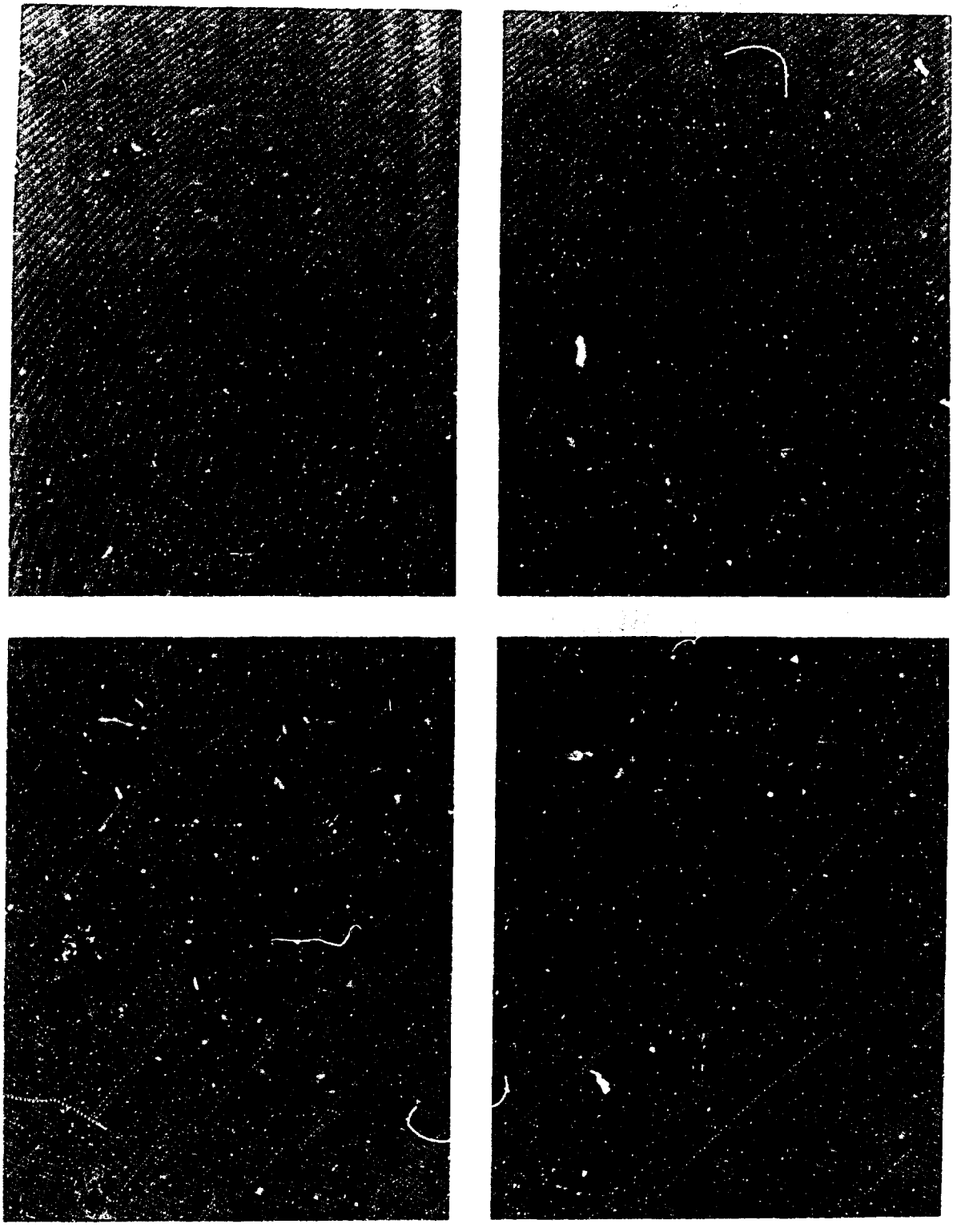

FIG. VIII-32 AGGLOMERATES OF QUININE AS SETTLED ON THE PLATES AT THE DISTANCES INDICATED

A — Plate length 2 cm, largest aerodynamic diameter calculated 28μ, based on density of 1.0 gm/cc

B — Plate length 40 cm, largest aerodynamic diameter calculated 6.2μ, based on density of 1.0 gm/cc

C — Plate length 80 cm, largest aerodynamic diameter calculated 4.4μ, based on density of 1.0 gm/cc

D — Plate length 183 cm, largest aerodynamic diameter calculated 2.9μ, based on density of 1.0 gm/cc

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considerably larger than that which would have existed for a spherical particle of density 1.0 gm/cc settling at the same position. The apparent density of the agglomerates is, therefore, less than 1.0 gm/cc.

A few measurements of the optical diameters of the agglomerates were made, and from this the apparent densities of the agglomerates were found to be approximately 0.15 gm/cc. Thus, it seems that successful dissemination is obtained not only by decreasing the particle size, but also by significantly decreasing the agglomerate density.

Because of the difference in densities, an agglomerate of density 0.15 gm/cc must increase its mass 2.5 times to maintain the same settling velocity as a single particle of density 1.0 gm/cc. Since settling velocity is proportional to density times the square of the diameter, the ratio of the mass of an agglomerate of diameter D_1 and density ρ_1, that has the same settling rate as a single particle of diameter D_2 and density ρ_2, to the mass of the latter is equal to the ratio $(\rho_2/\rho_1)^{1/2}$.

The implication of the above results is that the size of the primary particle subjected to dissemination may not be of the same significance as originally believed. Particles larger than 10 microns will settle slowly if they can be made to exist in sufficiently low density agglomerates. Control, not elimination, of agglomeration may be the most desirable approach to dissemination of powders into particles of suitable aerodynamic properties.

C. Discussion

For powders disseminated in a capillary tube, there seems to be an optimum length for the best separation of particles. Shorter lengths are not as effective as the optimum and little or no benefit results from use of longer tubes.

The length of the tube for optimum deagglomeration can be approximated from a calculation of the stopping distances of aerosols moving in still air. The distance required to accelerate a particle from rest to the same velocity as the moving air is the same distance that the particle would move if ejected at that velocity into a still atmosphere and allowed to

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come to a stop by friction from air. This stopping distance S_d, may be calculated by $S_d \approx V_o \rho d^2 / 18\mu$ where V_o is the initial velocity of the particle relative to air, d is the diameter of a particle of density ρ, and μ is the viscosity of air. As an example, a 10μ particle of density 1.0 gm/cc will require approximately 10 cm of tube length to be accelerated to 3×10^4 cm/sec (nearly the speed of sound). This value is in good agreement with the 10-15 cm length found in Section VIII-A for the optimum tube length for maximum deagglomeration. When the particle has attained the same velocity as that of air, the impact and shear forces arising from differential flow between particle and air have reached a minimum, but the impact forces resulting from turbulent collision with the wall will have reached a maximum. As shown in Section X, the impact forces will be of several orders of magnitude greater than the shear forces. Therefore, the maximum deagglomeration will occur when the tube lengths are sufficiently long to permit the particles to reach the velocity of the air, and thus provide for maximum overall impact forces.

It is of interest to note that Orr et al. (1957), in their investigation with capillary tubes, found "that increasing tube length at constant Reynolds number and (tube) diameter increases the deagglomeration" of 8.7μ Serratia marcescens powder. However, they only experimented with tube lengths between 1.7 cm and 7.6 cm.

The separation of particles (in the 1-10μ range) can be accomplished best by taking advantage of the stresses imposed by strong impact. The superiority of impact forces for breakup of agglomerates has been cited in other studies, notably those of Aeroprojects (1956), Fuchs and Selin (1964) and Fitz et al. (1955). In this respect a curved tube disseminator, which accentuates the strength of impact forces, is superior to a straight tube one.

The increase in aerosolization with increase in air velocity has been observed in other studies utilizing capillary tubes for dissemination. Orr et al. (1957) stated that the mass-mean-diameter of the aerosolized particles decreases with increasing Reynolds number. Fuchs & Selin (1964) showed a linear relationship between particle diameter and dissemination pressure.

Improved dissemination of some powders can be obtained by incorporating an additive, but all powders have not been tractable to such treatment. Some powders are easily disseminated without treatment and in these instances the beneficial effect of the additive is not revealed except under conditions of low energy application. The proper pairing of powder and additive is important as there seems to be some specific interactions between the two. A literature review on the action of additives is given in Appendix B.

Reagglomeration of particles occurs and is a factor that must be considered. It has been one factor that has prevented complete separation of the smaller particles into individual airborne identities. Small ratios of powder to air favor a more complete dissemination, because fewer particles (unit volume) are available for reagglomeration.

Physical and perhaps chemical surface treatment can be used to improve the aerosolizability of some powders, and properly accomplished it should be applicable to all raw powders. In this program ball-milling quinine in petroleum either produced a powder which was more aerosolizabl than any of the other quinine powders. In two recent studies on anti-agglomeration treatments, Rauner et al. (1964) and Lerman and Bartsch (1965) were successful in improving the aerosolizability of CW agents by chemical and physical modifications.

In summary, improvement in aerosolization can be accomplished best by subjecting the powder to the proper chemical-physical treatment and then aerosolizing it with an impactor-type disseminator.

IX ELECTROSTATIC EFFECTS

A. Introduction

Throughout the course of this program there was ample evidence that electrostatic effects are important in the dissemination of powders and were complicating factors in their assessment.

The description of the phenomena which produce the charging of the particles was covered in Chapter XII of SRI Special Technical Report No. 2 (Poppoff, 1965). A companion program by Stanford Research Institute was designed to study the fundamental aspects of electrostatic charges on aerosols. As a consequence, our program was limited to observations of a few of the electrical effects that might influence results.

One of the first evidences of electrostatic effects was revealed early in the program during the dissemination of Microthene. Rather odd-shaped agglomerates appeared which suggested electrostatic attraction between particles, as shown in Fig. IX-1. Quinine and saccharin disseminated to form light, fluffy chain-like agglomerates, again suggesting that electrostatic forces were bringing about reagglomeration. The chain-like agglomerates are not unusual; Dalla Valle et al. (1954) among others had observed them, and they concluded that "the aggregates of charged particles showed a pronounced tendency to be oriented in chain-like patterns, indicating polarization."

Another manifestation of electrostatic effects occurred during dissemination of treated Microthene in the laminar flow settling chamber. Under the conditions of the experiment, practically all of the particles were attracted to the walls of the chamber, thus rendering the chamber rather useless for assessing this type of material.

Experiments were conducted to demonstrate that the electrical properties of powders can be modified by additives and that the introduction of gas ions into the aerosol stream can be made to modify the resulting aerosol. The results of these tests are reported below.

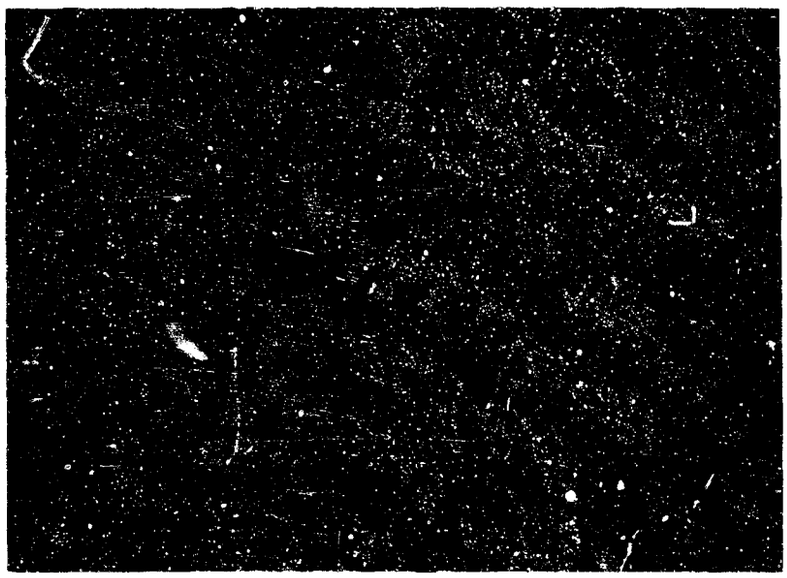

FIG. IX-1 MICROTHENE DISSEMINATED AT HIGH AEROSOL CONCENTRATION SHOWING THE CHAIN-LIKE AGGLOMERATION

B. Electrostatic Measurements of Powders and Additives

1. Measurements with a Parallel-Plate Charge Analyzer

Initial tests were made to determine some of the fundamental electrical properties of the powders and additives which were used in this program. For example, it was desirable to know the type of charges on a powder when it is being metered out from the bulk powder container. This measurement was made with a parallel-plate electrostatic charge analyzer, shown in Fig. IX-2a and 2b. It is a simple apparatus and it has been employed by various investigators, notably Nash et al. (1963) and Owe Berg et al. (1963).

The cell in which the observations were made consisted of a glass tube 10 cm in diameter and 13.8 cm high equipped with plexiglass cover and bottom. Two brass plates evenly spaced 4.9 cm apart were connected to a variable high voltage supply. Powder was fed from a small vibratory feeder into a funnel at the top of the cell from which it descended to the midpoint between the two plates.

In the photograph, the vibratory tube conveyor was constructed of plexiglass and the funnel was copper. These can be substituted by other materials for observing the effects of materials of construction on the triboelectric effect. The funnel was connected to ground and the positive and negative electrodes could be varied from zero to ±7500 volts with respect to ground. To permit a more quantitative measurement of the powder, small boxes were placed under the electrodes to catch the powder, and were weighed to obtain the percentage of positive, neutral and negative particles.

Results obtained using this apparatus for powders and additives are shown in Tables IX-1 and IX-2 respectively.

Table IX-1 showed that some powders were charged one-sided, while others were equally charged. Microthene and saccharin, for example, contained many more negative-charged particles than positive-charged ones. On the other hand, quartz and aluminum contained almost equal proportions of positive and negative particles. In all cases, the majority of the powder was neutral.

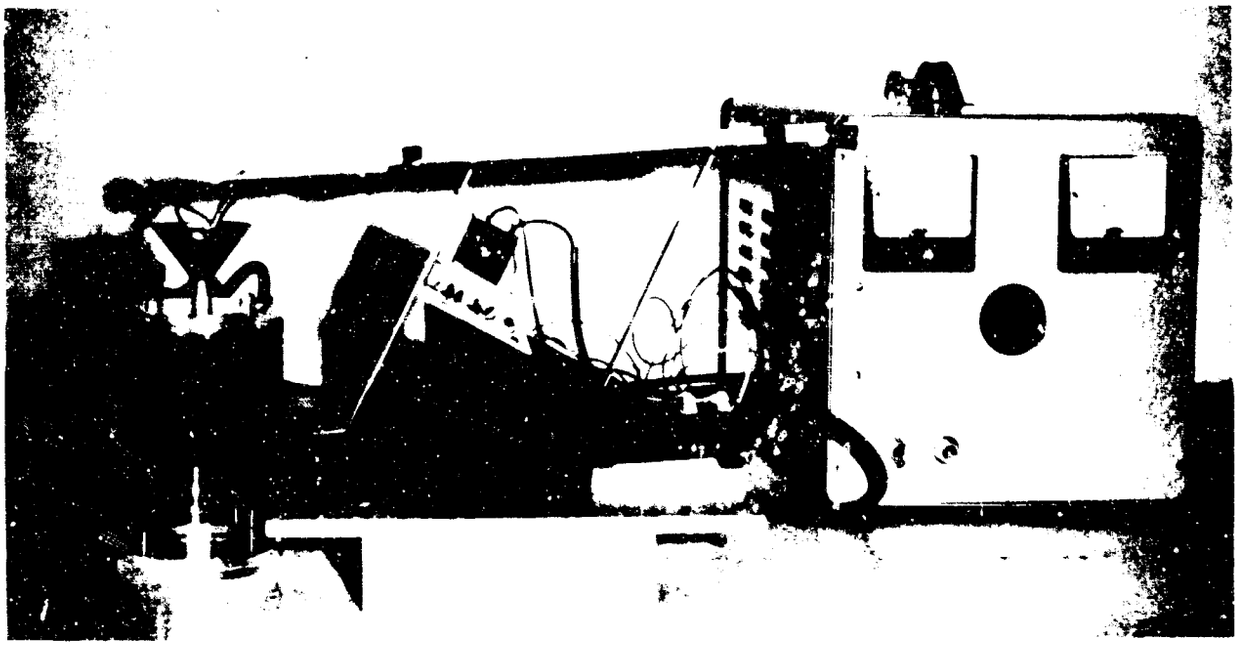

FIG. IX-2a GENERAL APPARATUS FOR MEASURING ELECTRIC CHARGES ON FALLING POWDERS

FIG. IX-2b PARALLEL-PLATE ELECTROSTATIC CHARGE ANALYZER FOR POWDERS

Table IX-1

ELECTROSTATIC ANALYSIS OF POWDERS

Powder	% Deposited in Negative Electrode Box	% Deposited in Neutral Box	% Deposited in Positive Electrode Box
Saccharin	2%	52%	46%
Microthene	2%	67%	31%
Glass Beads (1-30μ)	13%	65%	22%
Quartz	9%	75%	16%
Aluminum	9%	81%	10%

Table IX-2

ELECTROSTATIC ANALYSIS OF ADDITIVES

Trade Name	Primary Particle Size and Surface Area	Average % Deposited at Negative Electrode	Average % Deposited in Neutral Box	Average % Deposited at Positive Electrode
PQ 2340	0.013μ 150-175 m^2/gm	3%	12%	85%
Cab-O-Sil M-5	0.015μ 175-225 m^2/gm	2%	18%	80%
Valron Estersil	0.01-0.03μ 200 m^2/gm	1%	26%	73%
Hydrophobic Silica GS	0.01μ 200-300 m^2/gm	16%	24%	60%
Quso F-22	0.021μ 325 m^2/gm	27%	36%	37%
PQ 2326	0.013μ 150-175 m^2/gm	6%	67%	27%
Organ-O-Sil S-5	0.015μ 175 m^2/gm	46%	41%	13%

The additives analysis, Table IX-2, showed variations in their charges. Like the powders, most of the additives contained more negative-charged particles than positive-charged ones, but unlike the powders, the negative particles dominated the charges, i.e., 80% negative particles vs 2% positive particles for PQ 2340, Cab-O-Sil. The only additive which yielded a majority of positive particles was Organ-O-Sil, and it gave a 46% positive and 13% negative ratio. One additive, Quso F-22 gave approximately equal percentages of positive and negative particles.

In addition to the differences in electric charges, the individual additives exhibited a characteristic behavior of their own. Cab-O-Sil, for example, built up in very thick layers on the electrodes whereas the particles of Quso F-22 jumped rapidly from one electrode to the other several times during their descent. The Organ-O-Sil was unique in that it coated not only the front but also the back sides of the electrodes. The powder also spread throughout the chamber, coating the walls with a thin, but tenacious layer.

Electrostatic analysis of saccharin treated with 1% of several of the additives is given in Table IX-3 below.

Table IX-3

ELECTROSTATIC ANALYSIS OF SACCHARIN TREATED WITH SEVERAL ADDITIVES

Powder	% Deposited at Negative Pole	% Deposited in Neutral Box	% Deposited at Positive Pole
Saccharin, untreated	2%	52%	46%
Saccharin w. 1% Cab-O-Sil	16%	74%	10%
Saccharin w. 1% PQ 2340	16%	59%	25%
Saccharin w. 1% Organ-O-Sil	40%	26%	34%
Saccharin w. 1% Quso F-22	3%	77%	20%
Saccharin w. 1% Esterail	28%	18%	44%

Except for Quso F-22, the additives served to unskew the charge distribution, giving nearly equal amounts of positive and negative charged particles. The Quso F-22 did not alter the charge pattern appreciably from that of the untreated powder. There does not seem to be any correlation

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between the charges on the individual additives and powders, and the resulting charge on the mixtures of the two.

Another series of powder-additive combination tests is given in Table IX-4. With saccharin, two non-silica additives, magnesia and carbon black, served to neutralize the powder. Thus as in the other tests, additives tended to equalize the amount of positive and negative particles, but did not always make them more neutral.

Table IX-4

ELECTROSTATIC ANALYSIS OF SEVERAL POWDERS AND ADDITIVES COMBINATION

Powder	Additive 1%	% Deposited in Negative Electrode Box	% Deposited in Neutral Box	% Deposited in Positive Electrode Box
Saccharin	none	2%	52%	46%
	PQ 2340	24%	40%	36%
	Cab-O-Sil	15%	61%	24%
	Magnesia	3%	92%	5%
	Carbon Black	2%	91%	7%
Glass Beads (1-30μ)	none	9%	69%	22%
	PQ 2340	17%	61%	22%
	Cab-O-Sil	17%	50%	33%
	Magnesia	20%	43%	37%
	Carbon Black	19%	56%	35%
Quartz	none	8%	74%	18%
	PQ 2340	5%	85%	10%
	Cab-O-Sil	18%	64%	18%
	Magnesia	7%	86%	7%
	Carbon Black	11%	82%	7%
Aluminum	none	9%	81%	10%
	PQ 2340	7%	84%	9%
	Cab-O-Sil	7%	83%	10%
	Magnesia	11%	81%	8%
	Carbon Black	12%	86%	2%

With glass beads and quartz powder, the same general trends were observed. Aluminum powder is neutral to begin with, and no changes occurred there due to the additives.

In a recent study on the electrostatic effects of a powder (Carbo-Wax) and an additive (Cab-O-Sil), Nash et al. (1965) suggested that

neutralization of the mixture occurred because of the attraction of the negative charged Cab-O-Sil to the positive charged Carbo-Wax. Although this mechanism could happen, the data in Table IX-3 and IX-4 indicate that a general shifting of charges of a powder-additive mixture will occur, irrespective of the electrostatic charges on the individual powders and additives.

A brief exploratory test was made to determine whether this electrical effect was an inherent property of the powder or in some way reflected the method of handling. A sample of untreated saccharin powder was electrostatically separated into those portions which were attracted to the positively and negatively charged electrodes respectively. The powder was removed from each electrode and again allowed to settle into the space between the two electrodes. Results shown in Table IX-5 below reveal that fractionated powder behaved essentially the same as the powder which had not been electrostatically separated.

Table IX-5

ANALYSIS OF ELECTROSTATICALLY SEPARATED SACCHARIN

Powder	Fraction	% Deposited in Negative Box	% Deposited in Center Box	% Deposited in Positive Box
Untreated Saccharin	Unfractionated	2%	40%	58%
	Powder removed from positive electrode	2%	31%	67%
Saccharin Containing 1% Organ-O-Sil	Unfractionated	40%	25%	35%
	Powder removed from positive electrode	35%	25%	40%
	Powder removed from negative electrode	46%	15%	39%

The procedure was repeated with saccharin treated with 1% Organ-O-Sil. In this case, also, there was essentially no change in the behavior of the powders between those which had been previously fractionated and those which had not been. It appears that the charges on the saccharin, treated

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and untreated, are not fixed. It is not evident how the fractionated powder re-orients its charges to form the charge distribution of the original powder, except that the charge distribution observed was controlled by triboelectrification in the vibratory feed and funnel system.

2. Measurements with a Metronics Electrometer

The first series of electrostatic measurements gave data sufficiently sensitive to establish general trends. However, it was desirable to expand the measurements by determining the magnitude of the charges on the bulk powders. In the second series of tests, a sensitive electrometer developed by Metronics Associates was used to measure the type and magnitude of charges carried by a powder. The instrument is shown in Fig. IX-3. Full scale deflection, either positive or negative, corresponds to about 100 picocoulombs (10^{-10} coulombs) of charge on the probe.

The measuring technique was also changed by forcing the powder through a 140 mesh metallic screen rather than allowing it to slide over a flat metal surface which is the situation with the vibrating feeder. The probability of contact charging is greatly increased by the greater surface area of the wire screen and the closer proximity of the wire to all the particles passing through. The experimental procedure consisted of weighing a piece of aluminum foil which was then placed on the flat probe. The powder to be tested was put in a wire screen which was tapped or brushed causing the powder to fall on the aluminum foil. Enough powder was processed to cause the meter to read full scale, and this was repeated several times. Measurements were made with Microthene treated with PQ 2340.

The data for Microthene is shown in Table IX-6 below.

Table IX-6

ELECTROSTATIC MEASUREMENTS OF MICROTHENE TREATED WITH PQ 2340

1% PQ 2340 in Mixture	Net Charge	Grams Needed to Register Full Scale (100 picocoulombs)	Charge-to Mass Ratio, nano-coulombs/gm
none	+	0.0940 gm	1.06
0.06%	+	0.1282 gm	0.78
0.075%	+	0.5824 gm	0.17
0.085%	+	0.4030 gm	0.25
0.100%	-	0.1282 gm	0.78
1.0%	-	0.1445 gm	0.69
100.0%	-	0.001 gm	10.0

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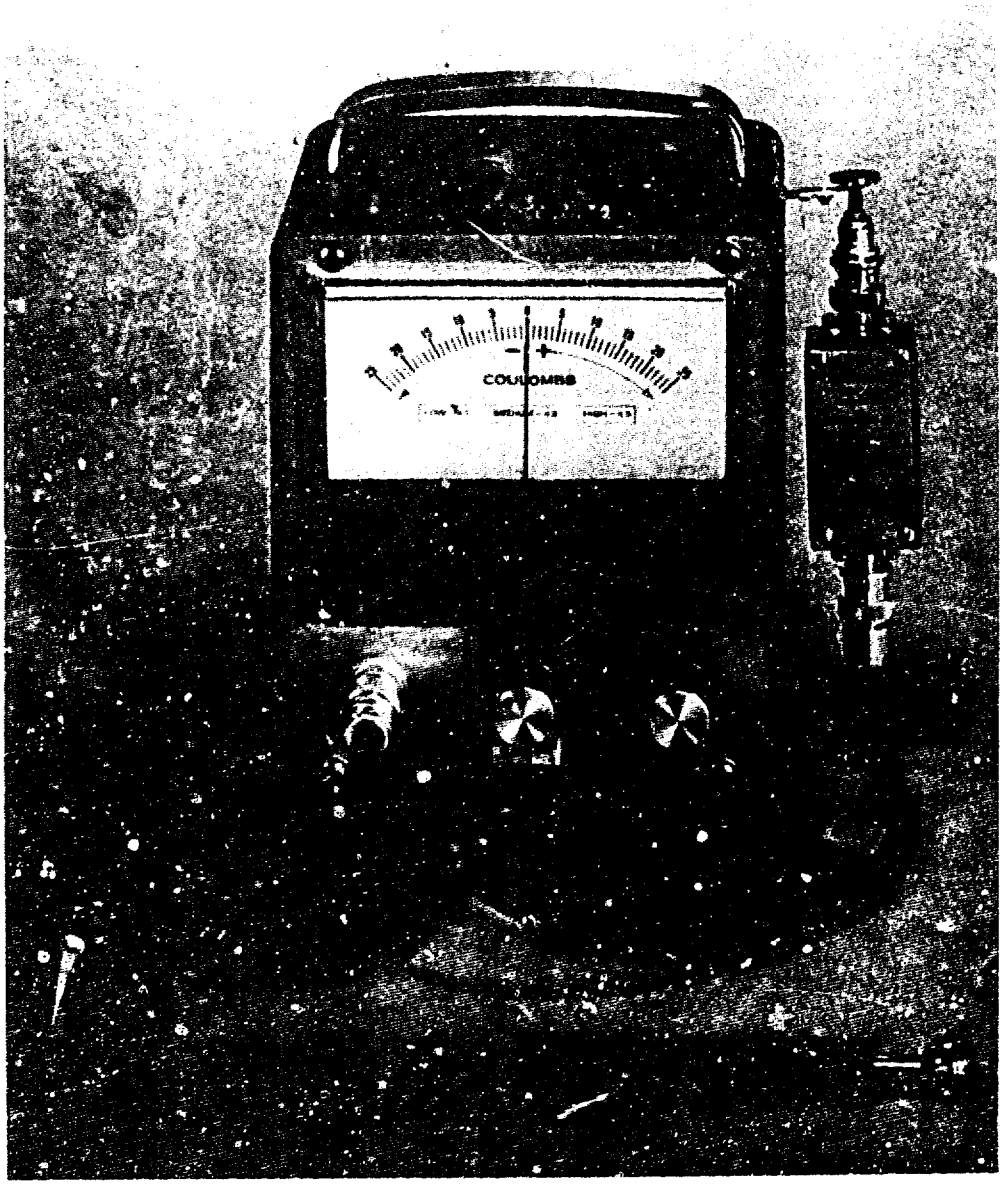

FIG. IX-3 METRONICS ASSOCIATES' ELECTROMETER FOR MEASURING THE TYPE AND MAGNITUDE OF ELECTRIC CHARGES

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Untreated Microthene exhibited a positive charge while Microthene with 1% PQ 2340 showed a negative one. PQ 2340 alone was negative. The values obtained with the 0.075% PQ 2340 content indicated that these percentages may be the transition range between charges. A significantly larger amount of powder was needed to cause the meter to register full scale with these mixtures and the meter fluctuated between positive and negative readings.

C. Effects of Electrostatics in the Dissemination of Powders

Measurements of the electrostatic charges on the bulk powders was revealing in that they provided some background knowledge of the type and magnitude of charges carried by the powders. The ultimate objective, however, is to correlate the electrostatic properties to the aerosolizability of a powder. For example, the light fluffy chain type of agglomerates formed by quine suggests that electrostatic forces are operating. Such forces should be altered by the presence of gas ions injected into the air stream containing the particles leaving the disseminating tube. In preparation for the dissemination of powders, under the influence of electrostatics, a few preliminary measurements were made to see how electrostatics might be applied.

1. Preliminary Measurements of Ion Charging.

The first measurements were made to determine if electric charges could be induced into powders and additives. A 26-inch U.S. Radium Co. Linear Ion Generator (radioactive type) was used for bombarding the powder or additive with ions, and the high voltage parallel plate analyzer was used for quantitative measurement. The ion generator could be caused to emit either positive, negative or bi-polar ions at various intensities. The powders in a grounded aluminum cup were placed three inches under the generator for charging and then hand fed to the electrostatic analyzer with a plastic spoon in small, 10-20 mg quantities.

Three additives, PQ 2340, Cab-O-Sil, and Organ-O-Sil, were exposed to the ion generator at maximum intensities. The data are plotted in Fig. IX-4. They show that the charge distribution patterns for PQ 2340 and Cab-O-Sil were no different with the generator off or on negative ion

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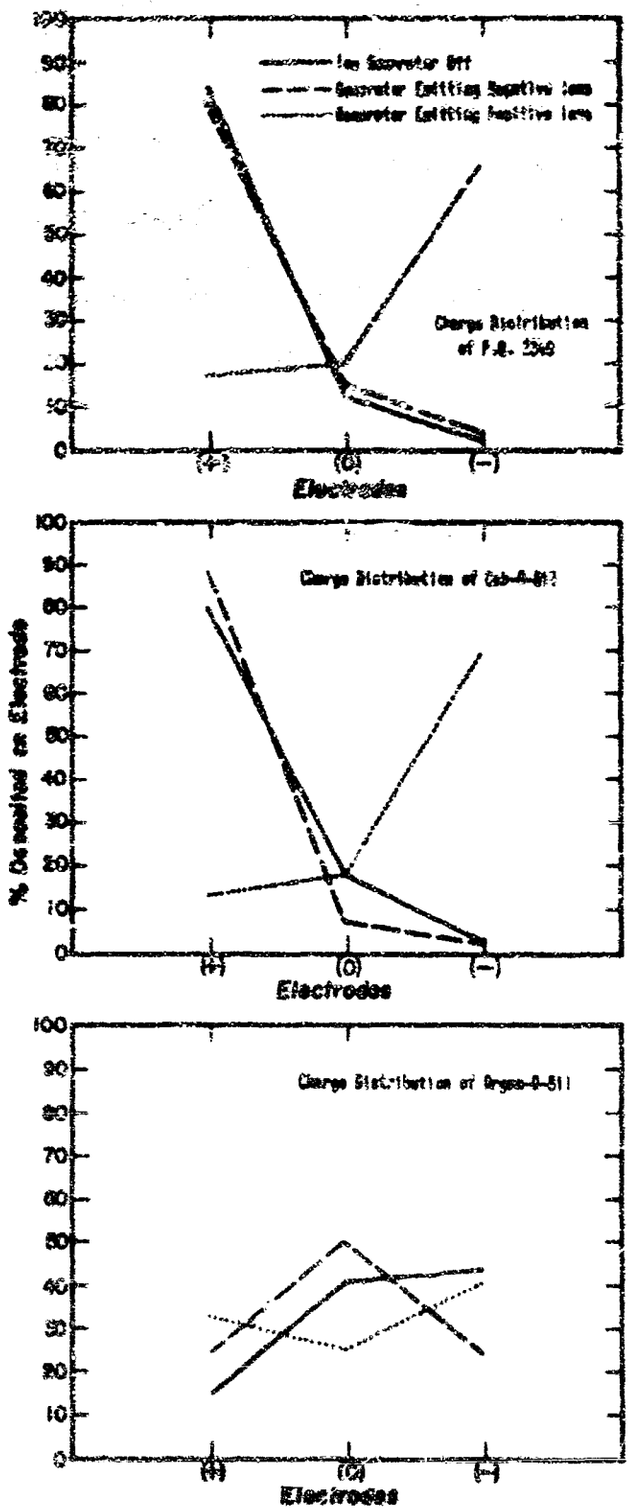

FIG. IX-4 EFFECT OF ION BOMBARDMENT ON ELECTROSTATIC CHARGE DISTRIBUTION OF SEVERAL SILICA ADDITIVES

output. With the generator emitting positive ions, however, the charge distribution for PQ 2340 and Cab-O-Sil showed a reversal in the distribution between the negatively and the positively charged plates. For example, the amount of powder deposited on the positive electrode decreased from 80-90% when the generator was off or emitting negative ions to 10-20% when the generator was emitting positive ions. Thus, these two additives responded strongly to positive ion emissions. In contrast, Organ-O-Sil responded only slightly to the effect of the ion bombardment, whether positive or negative.

The effect of the ion bombardment on the electrostatic properties of saccharin was also investigated, and the results plotted in Fig. IX-5. Since the saccharin was predominantly negative, the effect of the negative ion output did not alter the charge distribution. This duplicates the results observed with the PQ 2340 and Cab-O-Sil additives. However, directing positive ions onto the powder completely reversed the deposition on the positive and negative electrodes, such that 49% of the powder was positively charged, 49% neutral and 2% negatively charged.

The effect of adding 1% Organ-O-Sil to saccharin was to completely nullify the effect of the ion bombardment, so that the powder distribution was nearly the same whether the ion generator was emitting positive or negative ions.

The magnitude of the charges on saccharin powder was also measured using a Faraday cage type of arrangement. Measurements were made on treated and untreated powder, with and without ion bombardment. The saccharin powder was transferred to the Faraday cage manually in scoops of 25-50 mg quantities until a charge of 5×10^{-9} coulombs was built up. The charge was discharged and the procedure repeated until a 1.0 gm quantity has been put through. Reproducibility was about ±20%.

The results are tabulated in Table IX-7. The untreated saccharin carried a residual negative charge of 3.4×10^{-8} coulombs/gm. After treating with 1% Organ-O-Sil, the saccharin registered no charges, indicating that either the particles contained no charges or the particles contained charges which were equal in quantity and magnitude. Previous

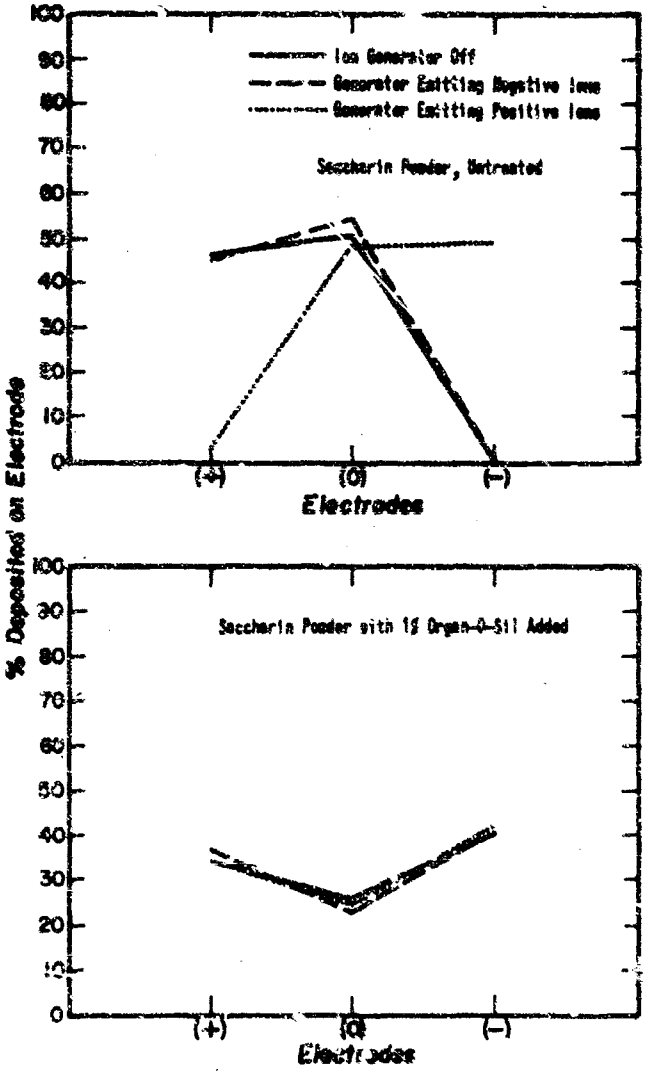

FIG. IX-5 EFFECT OF ION BOMBARDMENT ON ELECTROSTATIC CHARGE DISTRIBUTION OF SACCHARIN, TREATED AND UNTREATED

analysis of the treated saccharin with the parallel-plate analyzer indicates that the latter assumption was more probable.

Table IX-7

MAGNITUDE OF ELECTRIC CHARGE ON SACCHARIN POWDER

Powder	Radioactive Ion Generator	Polarity of Charge Being Registered on Electrometer	Charge, Coulombs/gm	Averaged Charge, Coulombs/gm
Saccharin Untreated	off	negative	33×10^{-9} 27×10^{-9} 31×10^{-9} 46×10^{-9}	$(-)\ 3.4 \times 10^{-8}$
Saccharin with 1% Organ-O-Sil	off	none	none	none
Saccharin Untreated	ion generator emitting negative ions	negative	35×10^{-9} 40×10^{-9}	$(-)\ 3.8 \times 10^{-8}$
Saccharin Untreated	ion generator emitting positive ions	positive	44×10^{-9} 29×10^{-9} 25×10^{-9} 31×10^{-9}	$(+)\ 3.2 \times 10^{-8}$
Saccharin with 1% Organ-O-Sil	ion generator emitting positive ions	positive	6×10^{-9} 9×10^{-9} 9×10^{-9} 4×10^{-9}	$(+)\ 0.7 \times 10^{-8}$
Saccharin with 1% Organ-O-Sil	ion generator emitting negative ions	negative	10×10^{-9} 13×10^{-9} 13×10^{-9} 11×10^{-9}	$(-)\ 1.2 \times 10^{-8}$

Next the powders were placed under ion bombardment to determine its effect on the electric charges. Exposure of the untreated powder to negative ions from the ion source gave approximately the same charge as the untreated powder, or $(-)\ 3.8 \times 10^{-8}$ coulomb/gm. However, when the emitting ions were switched from negative to positive, the saccharin acquired a positive charge, amounting to approximately $(+)\ 3.2 \times 10^{-8}$ coulomb/gm, which is almost equal in magnitude to that of the untreated powder.

Treatment with Organ-O-Sil served to reduce the capability of saccharin to acquire charges from the ion source. This was previously shown in Fig. IX-5, and the conclusion is supported again in these tests. The charges acquired by the treated powder were 1/3 to 1/5 of that absorbed by the untreated saccharin.

It may be of interest to note that Shaffer (1962) measured a charge of (-) 2×10^{-8} coulombs/gm for untreated saccharin which had been worked a little with a spatula. This figure is in good agreement with the (-) 3.4×10^{-8} coulomb/gm obtained in these tests.

The above observations indicate that the electrostatic charges on additives and powders can be changed and that a low activity ion source (approximately 2×10^{-9} coulombs/sec) is sufficient to change the sign of the charge. However, the addition of an additive to a powder served to minimize the effectiveness of the ion generator to charge the mixture. Thus in support of other data reported herein, the effect of an additive can be to neutralize the electric charges, and at the same time to inhibit the ability of the powder to acquire a charge.

2. Dissemination of Powders With Ion Charging

As indicated above, the radioactive type ion generator was a low level ion source. In addition it was a bulky piece of equipment not readily adaptable to dissemination testing. Consequently a new ion generator, of the high voltage variety was fabricated for dissemination tests. The generator used was a modification of one designed by Whitby et al. (1965). It is shown in Fig. IX-6.

Figure IX-7 is a schematic drawing of the method used to determine the condition for maximum ion output from the ion generator. A 510 V battery was connected to one of the plates and the high side of a Keithley Electrometer, and a 22 megohm resistor was placed between the other plate and the ground. An air flow of 62 ℓ/min was used for adjusting the needle setting. The ion output was most steady when the needle was at 0.035" away from the 1/16" dia. exit hole, and so this setting was used for all ion generator tests. The applied voltage for all tests was 6000 volts. A quantitative measure of the ion output was not considered necessary

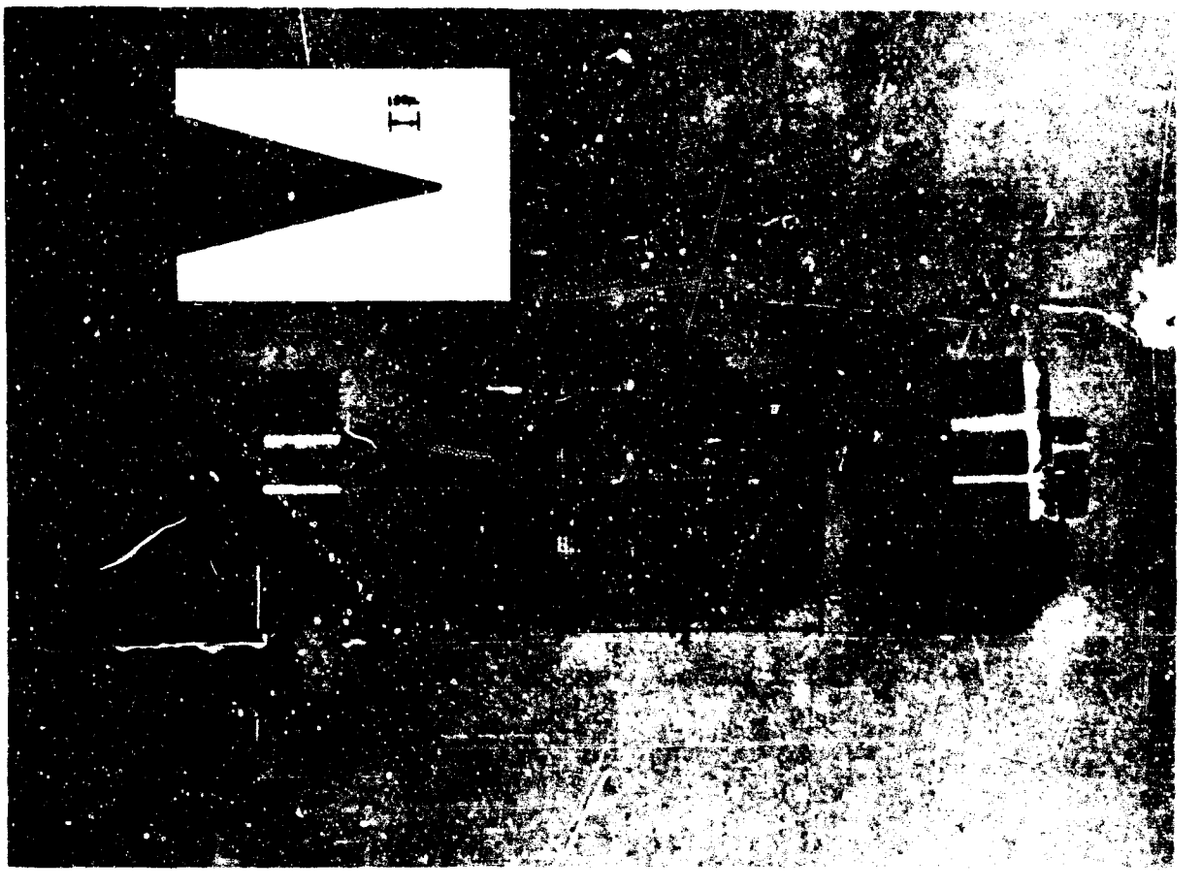

FIG. IX-6 CORONA-DISCHARGE TYPE ION GENERATOR

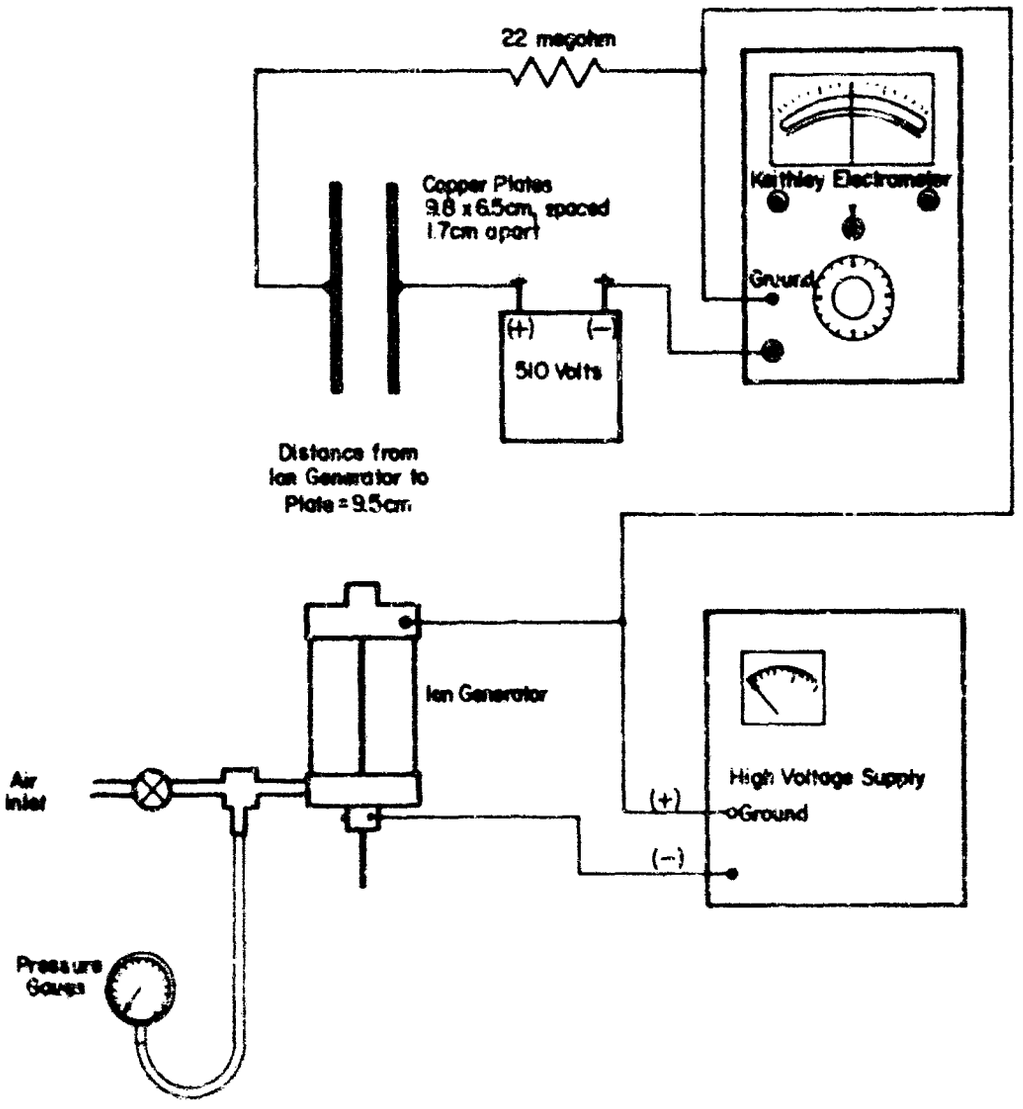

FIG. IX-7 SCHEMATIC DIAGRAM OF METHOD FOR DETERMINING ION OUTPUT OF CORONA-DISCHARGE ION GENERATOR

because the amperage registered on the Electrometer is a function of (1) the size of the plates used, (2) the gap between the plates, (3) the air flow through the ion generator and (4) the distance the ion generator is located from the measuring plates.

The most convenient means for introducing the ions into the dissemination chamber would have been to pipe them in. However experiments with various pieces of metallic and plastic pipes, some as short as one inch, revealed that the ions could not be piped at all. Consequently it was necessary to redesign the powder inlet to the laminar flow chamber such that the powder and ions are introduced side-by-side. This arrangement is shown in Fig. IX-8. A few dissemination tests using quinine were made with the ion generator attached, but no voltage applied. The mass distribution pattern duplicated that obtained with the previous method of powder introduction, thus indicating that there was no significant change caused by the mechanical rearrangement for introducing the powder into the chamber.

The dissemination results for quinine osterized with 1% PQ 2340 are plotted in Fig. IX-9. Runs #35 and #40 were made with the high voltage electrode being negative and attached to the needle, and the positive electrode (ground) attached to the exit-plate. Since the two runs were in excellent agreement, only the average is plotted. For Run #41, the polarity was reversed with the positive electrode attached to the needle. This run matches the #35-40 run, thus indicating that a negative charge or a positive charge on the needle gave similar effects. As obvious from the curves, most of the quinine (85% or so) from either of the unipolar runs remained in the duct, whereas in the conventional situation, with no ions (Run #38-43) only 30% remained in the duct and the rest of the quinine fell onto the plates and into the filter. The first conclusion was that the unipolar ions had a tremendous detrimental effect on the aerosolizability of the quinine powder. But microscopic observation of the settled particles, Fig. IX-10, showed a large number of single particles smaller than 10μ. Since all particles smaller than 50μ should be carried up by the air stream, it must be concluded that the ions in the neighboring air stream caused the particles to be attracted to the

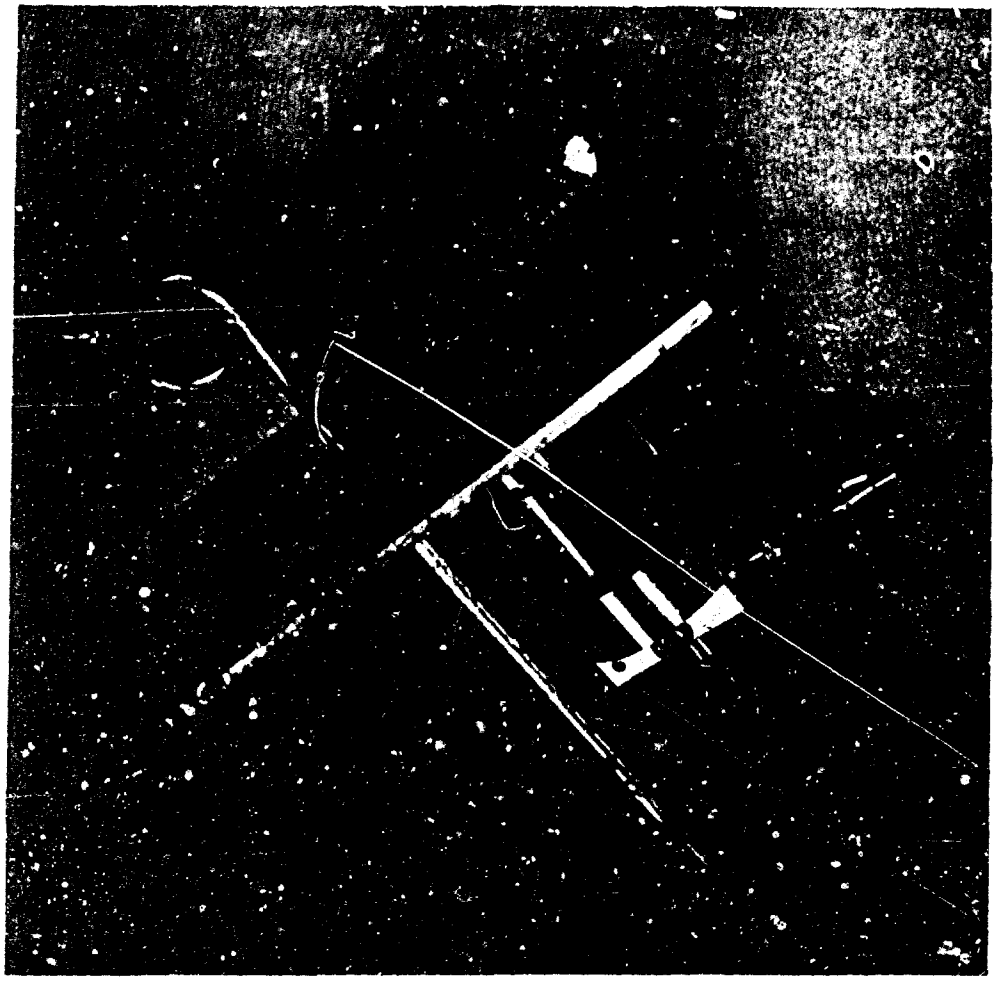

FIG. IX-8 BOTTOM PLATE OF DUCT SHOWING THE ION GENERATOR AND POWDER DISSEMINATION TUBE SIDE BY SIDE

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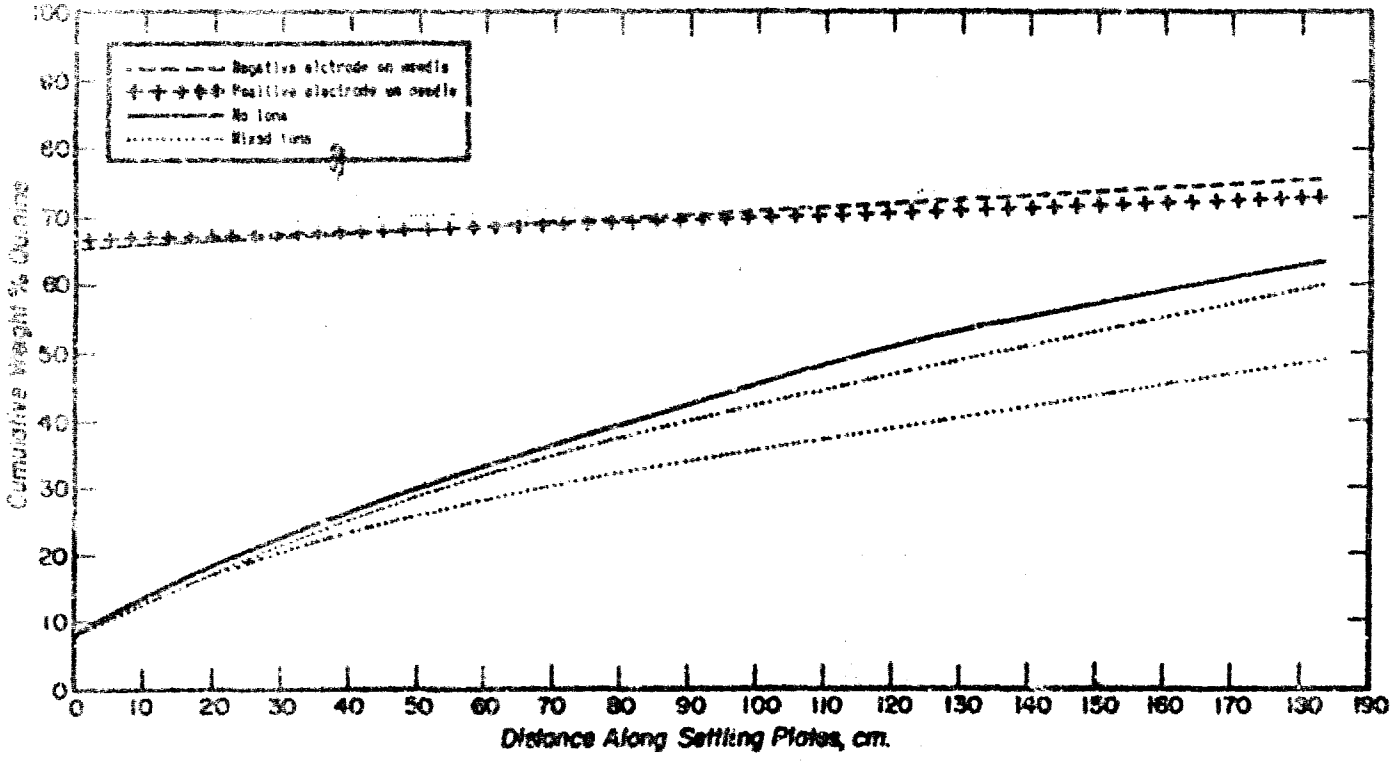

FIG. IX-9 NONDIFFERENTIATED MASS DISTRIBUTION CURVES FOR QUININE SHOWING THE EFFECT OF ION CHARGING OF THE AIR STREAM DURING DISSEMINATION

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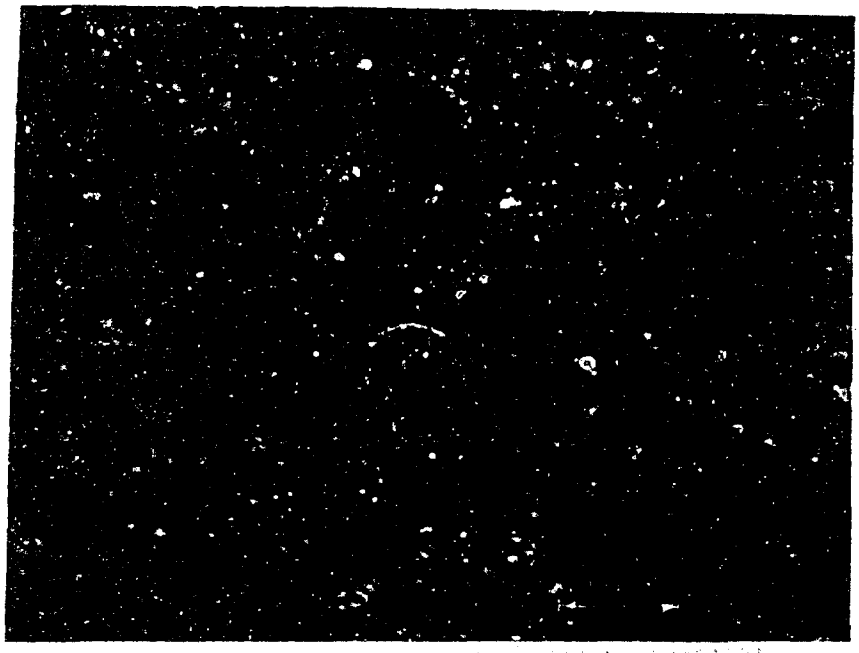

FIG. IX-10 QUININE PARTICLES ON SIDE OF DUCT RUN #45
Conventional run, no ions generated

FIG. IX-11 QUININE PARTICLES ON SIDE OF DUCT RUN #40
Negative ions generated in the neighboring air stream

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wall of the duct. Ordinarily these particles would be carried up by the air stream and deposited on the settling plates. Figure IX-10 can be compared to Fig. IX-11 to illustrate the difference in particle deposition on the wall between an "ion" and a "no ion" run.

This difference illustrates that the behavior of an aerosol of solid particles can be altered by the introduction of gas ions. It is not safe to conclude which aerosol, i.e., with or without ion introduction, is the better one. The aerosol produced in the presence of ions had more single particles, but their aerodynamic properties could not be determined because of their rapid migration to the walls.

Also plotted in Fig. IX-9 are quinine runs in which "mixed ions" (produced by high frequency alternating current) were generated. This run gave a mass distribution pattern only slightly different from the no-ions run, and no definite conclusions can be made. Microscopic examination of the slides from the "mixed ions" and "no ions" runs revealed that the deposited particle-agglomerates from both runs were similar.

D. Discussion

A large number of measurements were made on the electrostatic properties of the bulk powder, and it was found that most of the powders tested carried a majority of negative charged particles. Most of the additives were also strongly negatively charged. Despite their original charge distribution, the effect of mixing an additive with a powder resulted in a neutral powder mixture. The neutralization of a powder with an additive has been observed in other studies, e.g., Nash et al. (1965), Woffinden (1964).

Correlations between the electrostatic properties of the bulk powder and its aerosolizability were not evident. In fact, there are indications which suggest that the electrical properties of a powder in the bulk stage are considerably different from the particle properties in the aerosolized state. Shaffer (1962) and Owe Berg (1963) noted large differences in the electric charges between powders which were at rest, and those which had been disseminated.

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The electrostatic properties of a powder can be altered, either by additives or by ion charging. The influence of ion charging on the aerosolization of quinine was quite evident in this program, but its effect has not been as dramatic in other studies.

In a recent study on the dissemination of solid and liquid agents, Whitnah et al. (1965) employed a corona-discharge type ion generator (similar to the Whitby model) to study the effect of injecting ions into an aerosol cloud. The material used was powdered sugar with a cornstarch additive. The powder was injected downward from the top of a one cubic meter chamber while the ionized air was injected upward from the bottom of the chamber. Positive ions were injected into the aerosol cloud (no runs were made with negative or mixed ions).

Their results showed that the positive ions did affect aerosol behavior. However, the change was small, and Whitnah et al. would not conclude that the ions actually did suppress the reagglomeration of the aerosol. Because of changes in their light scatter analysis data with aerosol dilution, they concluded that "considerable reagglomeration could have taken place during the settling phase of any aerosol runs reported here and escaped detection." It is probable that a more pronounced difference in aerosol behavior would have occurred if the ions were injected closer to the disperser outlet. Experience in this laboratory with similar ion generators showed that the ions decay very rapidly with distance. It is quite possible that the aerosol near the disperser did not really "feel" the effect of the ion generator.

In his discussion on agglomeration of aerosols, Woffinden (1964) concluded that collisions between aerosol particles "are caused by long-range electrostatic forces, and that agglomeration (adhesion) is caused by short-range electrostatic forces." He indicates that random collisions are extremely rare, as compared to collisions resulting from electrostatic forces. He did not present data to support his conclusions, but in Section X of this report, reasoning is presented which strongly suggests that electrostatic attraction is the most plausible mechanism for the reagglomeration of the aerosolized particles.

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X* DISCUSSION ON DISSEMINATION AND REAGGLOMERATION

A. Introduction

The beginning and end of the aerosolization process is easily stated. The starting material is a bulk powder of solid particles 1 to 10 microns in diameter, and it is to be disseminated into an aerosol cloud containing 10^{-5} to 10^{-6} gm powder/cc of air. This concentration requires that the final cloud should contain 10^{6} to 10^{7} particles/cm^{3} of 1-micron size or 10^{3} to 10^{4} particles/cm^{3} of 10-micron size.

A dry presized powder was specified for this program because it was postulated that less force would be required to separate single particles than to comminute bulk material and that an aerosol of an easily controlled size distribution would be generated. Unfortunately, when particles of sizes below 10-micron are disseminated, the aerosol particles are usually agglomerates of the primary powder particles.

Cloud mass concentration depends primarily on total energy dissipation. In any dissemination processes almost all of the energy expended is used to dilute the suspended particles into an aerosol cloud. Various aspects of concentration and energy dissipation are given in another report (Ranz, 1967), but since they are basically independent of particle size, they will not be discussed here.

The size of suspended particulates depends primarily on the forces and stresses which cause breakup of the powder agglomerates. Since the powder must be diluted with at least a small amount of air before the particles can be said to be suspended, breakup is intrinsically connected with the dilution process, marking its beginning However, large scale dilution and cloud formation are essentially a separate process, and it

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* This section is a revision and amplification of material contributed in memoranda by W. E. Ranz.

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is not of immediate concern here. The objective of this program was to seek quantitative information about capabilities and limits in the breakup of agglomerates and the initial dilution of the particles into a suspension.

In this section theoretical considerations are shown that explain most of the phenomena observed in the dissemination of dry powders.

Aspiration of a powder through a straight tube will produce some individual particles in the 5-micron range by impact with the tube wall. But a spiral tube or other device that assures impaction of all particles with a solid surface is considerably superior for air suspension of single particles.

Reagglomeration of particles occurs except at very dilute aerosol concentrations. The mechanism for reagglomeration cannot be explained by 1) contact between particles during turbulent flow through the tube, 2) contact between particles in the air jet at the exit of the tube, or 3) contact during the settling of the suspended particles. The calculations show that electrostatic charges on the suspended particles is most likely explanation for reagglomeration.

B. Mechanics of Breakup of Agglomerates

1. Yield stresses

Each particle in a powder bed of uniformly sized particles touches other particles at from three to nine contact points, with the lower value being the more probable. The smallest particles will have a minimum of three contact points, but larger particles, because they are surrounded by smaller particles, will be touched at more points. For particles of sizes less than several hundred micron, the force needed to separate each contact point is variously estimated and measured to be from 10^{-2} to 1 dyne. A literature review on adhesive force measurements of powders and particles is given in Appendix B. This contact force was found experimentally to be proportional to particle size,

To simplify the theoretical considerations, the discussion on the breakup of agglomerates is limited to one contact point between two particles. Also, the change in binding forces caused by an additive is not considered here. The existence of an additive has been shown elsewhere in this report to reduce the binding forces. Although the effect is observed in the dissemination process, there are no quantitative data on the magnitude of force reduction and no agreement in the literature on the mechanism by which additives operate. In what follows it is assumed that no additive effects have been introduced.

It is convenient to begin the discussion by considering the binding force to be a van der Waal force. A simplified equation for van der Waal's forces, given in Poppoff (1965), p. XII-39, is that the force between two spheres of diameter D_p is $F = KD_p/2$, where K is a constant associated with the surface energy of the spheres. For most organic materials K is of the order 200 dyne/cm. For 1-micron particles this force is 10^{-2} dyne; for 10 micron particles the force is 10^{-1} dyne. These forces can be translated to apparent yield stresses by dividing them by the projected area of the particle, that is, $10^{-2}/(10^{-4})^2 = 10^6$ dyne/cm^2 for 1-micron particles, and $10^{-1}/10^{-3})^2 = 10^5$ dyne/cm^2 for 10-micron particles. The apparent yield stress of final breakup increases with decreasing particle size. Indeed, for 1-micron particles, it reaches values of the order of 10^{-2} times the yield strength of particle material itself. In a sense one can look upon 1-micron particles in contact as welded back together again over an area equal to about 10^{-2} times their projected area, or bonded by a 10^{-10} cm^2 patch of solid, structured material on the particle surfaces.

When applied to close-packed, multi-particle agglomerates, this method of estimating yield stress will continue to give the same order of magnitude for the stress as that for the final, two-particle agglomerate. With more probable open packings, the yield stress would be less by factors of two or three. However, one should expect the apparent yield stress to continue to be primarily a function of particle size, rather than agglomerate size.

For small particles the making and breaking of contact forces causes powders to flow and take on some of the characteristics of liquids. Powders show apparent "viscosities" and "surface energies," and it is reasonable to expect that they may disseminate only as well as liquids with equivalent mechanical properties.

It is useful to compare the contact yield stresses with the surface tension stress or "survival stress" of liquid drops, $4\sigma/D_p$, where σ is the surface tension and D_p the drop diameter. For organic solvents with a surface tension of the order of 25 dyne/cm, the yield stress is 10^6 dyne/cm^2 for 1-micron drops and 10^5 dyne/cm^2 for 10-micron drops. On this basis of comparison, it should be as difficult to disseminate single 1-micron solid particles as it is to obtain 1-micron sprays (nearly impossible), and to generate single 10-micron particles as it is to obtain 10-micron sprays (always difficult).

2. Gas-dynamic Stresses in a Suspension of Agglomerates

By definition of pneumatic dissemination, the applied stresses causing agglomerate breakup is of gas-dynamic origin. Therefore final breakup of the smallest agglomerates occur in a suspension of agglomerates. But the stresses arising in the flow of ordinary disseminating devices are rarely large enough to achieve a suspension of single particles with sizes less than 10-micron. The maximum possible aerodynamic breakup stress will be the stagnation pressure of air at some maximum relative velocity between air and powder. The value of this stress is given by $\rho_a V^2/2g_c$ where ρ_a is the density of air, V is the relative velocity, and g_c is the proportionality constant (1 gm cm/dyne sec^2). For the practical maximum velocity of 3×10^4 cm/sec (sonic velocity) in a gas disseminator, the largest breakup stress is $10^6/2$ dyne/cm^2, just sufficient to give a few 2-micron agglomerates in the small end of the suspended size distribution. For the more applicable situation in which the breakup stresses are working on already accelerated agglomerates, Ranz (1967) calculated that the values are from 10^{-1} to 10^{-3} times less than the maximum given above.

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3. Collision or Impact Stresses

If a suspended particle or agglomerate strikes a wall or another particle at relative velocity V, it will be subject sometime during the collision to breakup stresses at least as large as the plastic stress, $\rho_p V^2/2g_c$, which would occur if the particle deformed like an ideal fluid. Here ρ_p is the density of particle material. Thus an agglomerate striking a hard solid surface at sonic velocity (3×10^4 cm/sec) will be subject to a breakup stress of about 10^9 dyne/cm^2 if the particle material has a density of 2 gm/cc. Not only is this a sufficient stress to break 1-micron contact forces, it is large enough to cause further grinding of single particles. Actually, collisions of agglomerates at only one-tenth of sonic velocity should eventually produce a suspension of 1-micron particles.

Thus, an aerosol of single solid particles with sizes less than 10-microns can be obtained by a fluid regrinding during suspension. And in some situations, further comminution of presized particles may have to be avoided by limiting the maximum stress.

The comminution of single particles on collision is not a simple matter. Early studies (i.e. Fitz et al. 1955) indicated that the primary breaks occur because of a concentration of shock waves reflected from the particle surface farthest away from the impact region. More recent research by McFarland (1967) shows that there is no sharp breakup stress at which particles divided into pieces. There is rather a wide range of applied stresses over which larger and larger fragments are chipped from the primary particle. However, somewhere in the middle of the range of apparent impact stresses where breakup is observed to occur, the material yield strength value is also found.

C. Dissemination Methods Using Collision Principle

Dissemination of suspended agglomerates into single, micron-sized particles has been accomplished in several ways using the collision principle. Fuchs and Selin (1964) for example, found a vast improvement in the aerosol when the powder was injected through a spiral tube; Baddorf et al. (1965) found that a cone impactor was a highly efficient

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disseminator. In some devices, breakup by centrifugation of agglomerates against a wall is combined with classification where smaller particles escape in an outlet from the vortex, as done by Perkins et al. (1952).

Grinding mills which produce 1-micron particles also appeared to depend on the mechanism of wall-particle or particle-particle collision. For example, in the Spinco mill described by Tanner (1957), 10-micron particles given an angular velocity of order $10^4/2$ cm/sec by a turbulent Couette flow are centrifuged against a stationary outer wall at a radial velocity of order 10^3 cm/sec. They traverse axially the 0.05 cm gap between rotor and stator in 10^{-3} sec, and several collisions are assured. Disruption may be enhanced by spinning off the walls, but rotational velocities imparted by main flow vorticity, rotor velocity multiplied by particle size and divided by gap width, are not sufficient for breakup. This mechanism for particle breakup is not the same as that advocated by Tanner, but the values of the force causing breakup is of the same order of magnitude.

Gross analysis of these devices can be made by relatively simple models. Consider first wall collisions caused by turbulent flow at velocity V along a wall. If the agglomerates move randomly and independently of the gas, then, according to Levich (1962), the impact stress can be as high as $\rho_p VV'/2g_c$ where V' is the root-mean-square fluctuating velocity. For $V = 3 \times 10^4$ cm/sec, $V' = 1.5 \times 10^3$ cm (turbulence intensity of five per cent), and $\rho_p = 2$ gm/cm^3, the maximum breakup stress can be as large as 5×10^7 dyne/cm^2.

Of equal importance to the model is the collision frequency. This frequency will be no larger than $nV'/6$ where n is the average number density of agglomerates in the flow over the surface. If it is assumed that each hit breaks an original agglomerate into particles and that the agglomerate remains turbulently mixed in a direction normal to the impact surface, then the differential changes in the number density in a tube, in the axial direction z, is $-\pi D_t^2 V dn/4 = (nV'/6)\pi D_t dz$ where D_t is the diameter of the tube. For $n = n_o$ at $z = 0$, $n/n_o = \exp(-4V'z/6VD)$ at length z in a tube. Thus, after $60D_t$ of tube length in a turbulent flow

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of 5% intensity ($V'/V = 5 \times 10^{-2}$), the number density of agglomerates will be $n = n_o e^{[-4(5 \times 10^{-2})(60/6)]} = n_o / e^2$. Thus a significant decrease in agglomerate concentration will occur.

An impaction bend, such as the coiled tube described elsewhere in this report, causes wall collisions in a more positive way. If an agglomerate follows the main flow at velocity V in a bend, it will be subjected to a centrifugal acceleration of order V^2/r where r is the radius of the bend. From Stoke's law, the radial velocity, U_r would be

$$U_r = \frac{V^2}{r}\left(\frac{D_p^2 \rho_p}{18\mu_g}\right) = V\left(\frac{D_p^2 \rho_p V}{18\mu_g r}\right) .$$

Particles centrifuged from the flow would hit the wall at velocity V if the impaction parameter, $\rho_p D_p^2 V/18\mu_g r$, has a value of unit or larger. In the present case a 5-micron particle or agglomerate of density 1.0 gm/cc will have an impaction parameter of 2.3 if V is sonic velocity and if the radius of the bend is 1.0 cm. Therefore it is expected that the particles will hit the wall at a velocity nearly equal to V. The fact that the flow is turbulent would not change this conclusion.

Nearly all of the particles with D_p above a certain "cut size" will hit the wall of a single bend or an impact anvil. According to this criterion, at sonic velocity of air the radius of curvature has to be less than 0.10 cm to impact 1-micron particles (of density 2.0 gm/cc) and 1.0 cm to impact 3-micron size particles (see Ranz, 1956).

The maximum impact stress experienced by a particle hitting the wall at a velocity of 3×10^4 cm/sec would be of the order 10^9 dynes/cm^2, and it would experience this breaking stress perhaps four times in a 360° spiral tube.

From the foregoing it may be concluded that the breakup of agglomerates smaller than about 20 microns requires a multiplicity of impacts with solid surfaces to break them into primary particles.

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D. Mechanical Reagglomeration of Disseminated Particles

In the formation of any aerosol cloud, considerable attention must be given to the particle concentration in the suspension. At large particle concentration (greater than 10^4 particles/cc), reagglomeration of the aerosolized particles becomes important. Experimental data from this program, as well as from investigations carried out by Knutson (1956), and Dalla Valle et al. (1954), indicated that reagglomeration is a real phenomena. For example, the large fluffy agglomerates of quinine shown in Fig. VII-29 dispersed readily into many discrete one-micron sized particles by merely adding a drop of oil to the deposit. Since grinding of the quinine particles occurred during dissemination, it is obvious that the formation of the fluffy agglomerate could only have occurred from reagglomeration of the one-micron sized particles. Other tests with saccharin powder also point to a strong reagglomeration tendency at sufficient high aerosol concentration. In the following discussion, the possibility of reagglomeration by physical contact is found to be an insufficient explanation of the phenomena.

1. Turbulent Collisions in the Flow Tube

For flow in the tube it is assumed that the particles are breaking when they hit the wall but are reagglomerating in the bulk flow along the center of the tube. At an air velocity of $V = 2.9 \times 10^4$ cm/sec through the 0.16 cm diameter tube, the Reynolds number of the tube flow is 2.8×10^4, for which the friction factor of a smooth tube is $f = 6 \times 10^{-3}$. The friction factor f can be interpreted in terms of volumetric mechanical energy dissipation rate ϵ by the identity, $\epsilon = (2\rho_g V^3/g_c D)f$. For air at the above conditions, the energy dissipation rate becomes $\epsilon = [(2)(1.1 \times 10^{-3})(2.9 \times 10^4)^3/(1)(0.16)](6 \times 10^{-3}) \approx 2 \times 10^9$ ergs/sec cm^3.

According to Levich's equation (Levich 1962) the random velocity of particles is

$$v' \simeq (g_c \epsilon \rho_p D_p / \rho_g^2)^{1/3} = \left[3 \times 10^9 \times 1 \times 5 \times 10^{-4}/(1.1 \times 10^{-3})\right]^{1/3}$$

$$= 10^4 \text{ cm/sec}$$

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for five-micron particles. In the same reference the volumetric collision frequency, N_c is given as $N_c = (\pi/r)n^2V'D_p^3$. Thus for a particle concentration of 4.5×10^5 particles/cm^3 in the tube,

$$N_c = (\pi/4)(4.5 \times 10^5)^2(10^4)(5 \times 10^{-4})^2$$

$$10^8 \text{ collisions/cm}^3 \text{ sec} \quad .$$

Thus, $N_c/n \cong 10^3$ collisions per particle per second are expected, but the particles are in the 20 cm long tube less than 10^{-3} sec. The result of the calculation, therefore, is that each 5-micron particle will collide about one time with another particle in passing through the tube. The same model predicts $nV'\pi D_t^2/4$ collisions with the wall of the tube per unit time and $nV\pi D_t^2/4$ particles per unit time going through the tube. Superficially, each particle collides with the wall $(4L/6D_t)(V'/V) \approx 30$ times in going through a straight tube, but the central particles may not have time to migrate to the wall during passage through the tube. Thus the calculation can explain breakup, but not reagglomeration.

3. Collisions at Tube Exit

A gross approximation of the number of bouncing collisions each hard aerosol particle will undergo with other particles like itself in the turbulent mixing field of a free jet of any high velocity as it travels from a distance x_o in front of the orifice of diameter D_o to any axial distance x downstream is given by Ranz (1967) as:

$$\approx 28\left(\frac{\rho_{pso}}{\rho_p}\right)\left(\frac{\rho_{go}}{\rho_g}\right)^{1/2}\left(\frac{\rho_p}{\rho_g}\right)^{1/3}\frac{D_o}{D_p^{2/3}x_o^{1/3}}\left[1-\left(\frac{x_o}{x}\right)^{1/3}\right]$$

x_o is somewhat arbitrary, but it should be at least the distance at which breakup no longer occurs. For the dissemination conditions below:

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ρ_{pso} = 3×10^{-3} gm/cm^3, particulate density of cloud jet at orifice,

ρ_p = 1 gm/cm^3, density of particulate material in particle.

ρ_g = 1.1×10^{-3}, air or gas density in surroundings.

$\rho_{go} \approx \rho_g + \rho_{pso}\ \rho_g$, density of cloud jet at orifice.

D_t = 0.16 cm, orifice or initial jet diameter.

D_p = 5×10^{-4} cm, particle size, assumed uniform.

x_o = 0.30 cm, distance beyond which breakup no longer occurs.

$x \rightarrow \infty$

The number of collisions, N_c, calculates to be 0.30, indicating that each particle has only a 30% chance of a single collision. Even if this value is in error by a factor of 10^{-2}, it still does not explain the reagglomeration of the large fluffy agglomerates of quinine.

5. Collisions in the Settling Chamber

In the slow flow before the settling plates, differences in settling velocities can bring particles into contact. The mean free path for straight fall, that is, the distance one can "see" in the aerosol, is equal to $1/n\pi D_p^2$. In the settling plate section of the chamber, the aerosol concentration is estimated to be 1.5×10^5 particles/cc. Therefore, for 5-micron particles, the mean free path is estimated to be 8 cm. If differences in settling velocity were of the order of the terminal velocity of a single 5-micron particle, that is, of the order of 10^{-1} cm/sec, then it would take several minutes for each particle to average one collision. Since the particles are in this slow flow region for only half a minute, simple differences in settling velocity cannot explain the observed reagglomeration.

E. Electrostatic Reagglomeration

The role of electrostatic attraction can be treated in a number of ways, and elaborate models and calculations are possible. For example, one could work in terms of an increased "impaction efficiency" when

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particles pass one another, or in terms of an increased "impaction cross section" or decreased "mean free path." One should note, however, that mechanical collision is so far from explaining the observed coagulation that electrostatic attraction alone must provide an explanation, if there is an explanation for reagglomeration.

In the critical review on electrostatic phenomena, Lapple (see Poppoff 1965, Section XII) worked out a chart in which the charge on a particle, expressed as the specific surface gradient Σ_{ps} (volts/micron) is related to the coagulation rate for various aerosol concentrations. This chart is reproduced in Fig. X-1.

Data on the specific surface gradient, Σ_{ps}, of aerosolized quinine are not available. However, some data on saccharin are available, and since saccharin aerosolizes similarly to quinine, the discussion below should be applicable to both quinine and saccharin. Owe Berg and Flood (1963) measured the electronic charges on saccharin which had been pneumatically disseminated through a hypodermic needle at air flow of 1.5×10^3 cm/sec, and found the charge to be approximately 4 coulombs/Kg or a specific particle surface gradient of ≈0.5 volts/micron. Since Σ_{ps} increases with increasing air flow, values of 1.0 volts/μ for saccharin would not be unlikely for sonic dissemination. Referring to Fig. X-1, for an aerosol concentration of 1×10^{-5} gm/cc (in the settling plate section) and an Σ_{ps} of 1.0 volts/μ, the rate of decrease of concentration is approximately 100% in 10^{-1} sec. Even if this value is off by several orders of magnitude, the coagulation rate due to electrostatics is sufficiently high to account for the reagglomeration phenomena.

In a recent study on aerosol reagglomeration Knutson (1965) found that the observed reagglomeration coefficient for saccharin due to differential settling of an aerosol was approximately 2.3×10^{-5} cc/sec. However, the theoretical agglomeration coefficient was only 1.3×10^{-6} cc/sec, a factor of 17 lower than the observed. Even after making allowances for increased agglomeration because of the existence of a polydispersed aerosol (rather than a monodispersed one) and for the presence of turbulence, the experimental agglomeration coefficient was still

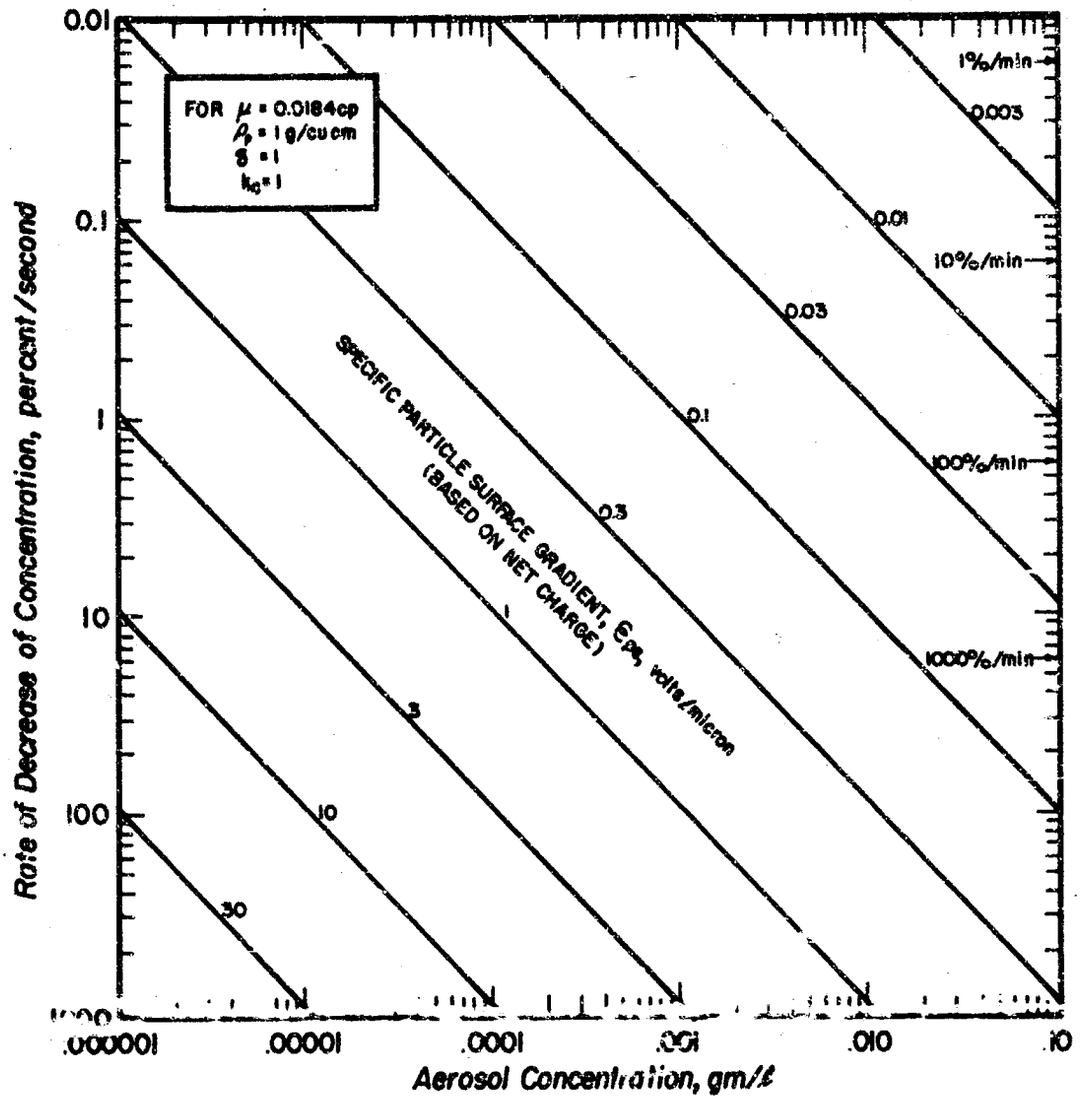

FIG. X-1 RATE OF COAGULATION OF A CHARGED CLOUD AS A FUNCTION OF AEROSOL CONCENTRATION (from Lapple, printed in Poppoff, 1965, p. XII-11)

higher than the theoretical by a factor of 10. He concluded that electric charges on the particles probably were the cause of the discrepancy. If the data of Owe Berg and Flood (1963) for the electric charge of saccharin is extrapolated, then a specific surface gradient of approximately 0.7 volts/micron is obtained. Then for an aerosol concentration in the one cubic meter chamber of 0.0005 gm/ℓ, the rate of decrease in aerosol concentration (from Fig. X-1) would be of the order of 50% per second. This number would be sufficient to account for the higher reagglomeration coefficient observed by Knutson (1965).

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Appendix A

LITERATURE REVIEW ON ADHESIVE FORCE OF POWDERS AND PARTICLES AND THE EFFECT OF ADDITIVES ON ADHESION

Appendix A

LITERATURE REVIEW ON ADHESIVE FORCE OF POWDERS AND PARTICLES AND THE EFFECT OF ADDITIVES ON ADHESION

A. Introduction

In connection with bulk properties measurements, a literature survey was made on the adhesive forces of powders and particles. Adhesive force measurements are considered important because they are most likely the correlation between a bulk powder characteristic and its aerosolizability. The aerosolization process is basically one of separating particles; therefore it is logical to expect particles whose adhesiveness is reduced, to aerosolize more easily than particles which are tenacious.

The review covers (1) the methods for measuring adhesive forces, (2) the values obtained by the different methods and (3) the mechanism by which additives operated to reduce the adhesive forces between particles.

B. Literature Reviews

There have been a number of recent literature reviews on the adhesive forces of particles and powders. Morgan (1961) prepared a review on the Adhesion and Cohesion of Fine Particles. The areas covered by him included (1) measurements on powders, (2) long-range forces, (3) adsorbed layers, (4) electrostatic charges, and (5) the nature of the contact. In his literature search on the Mechanism of Adhesion Between Solid Aerosol Particles, Owe Berg (1961) discussed (1) adhesion between large solid bodies, (2) adhesion between powder particles (3) coalescence of solid bodies and particles (4) flowability and agglomeration in powders (5) effect of electrostatic charge and (6) applications to aerosols. Many of the same subjects were reviewed by Corn (1961a) in his review on the Adhesion of Solid Particles to Solid Surfaces. A somewhat more detailed review is given in Corn's (1961b) Ph.D. thesis. Akin to general subject of adhesion of particles is the Ph.D. thesis of Larsen (1957) on Fundamental Studies of

Particle Dynamics and Adhesion. A good review on the methods of Adhesion Measurement Involving Small Particles is presented by Boehme et al. (1962).

Although the above are the major reviews noted on adhesion of particles, there are many smaller, but nevertheless comprehensive, reports on particular aspects of adhesion. These references can be found by scanning the reviews above. Some references may also be found in the Bibliography on Pneumatic Dissemination of Dry Powders, which is included as Appendix C of this report.

C. Methods for Measuring Adhesive Forces

Boehme et al. (1962) gives a good review of the various methods for measuring adhesion force. They described them as (1) the weighing method, (2) the pendulum method, (3) the aerodynamic method (4) the centrifuge method and (5) the incline plane method. The weighing method involved measuring the force required to separate particles from each other or from a substrate. The pendulum method is similar to the weighing method except the gravitational force of the object being measured becomes the separating force. The substrate to which the suspended particle adheres is rotated until the particle falls off. In the aerodynamic method, an air stream is directed at a particle and the force required to move or dislodge a particle is computed. The centrifuge method employs centrifugal forces to remove particles from a substrate. The incline method differs from the other four in that the adhesive force of a bulk powder mass is measured rather than the forces of individual particles. The method involves tilting a powder bed until the powder mass slides off due to the force of gravity.

Benarie (1961) employed a Couette viscometer to measure the cohesive forces within a powder mass, but his so-called "Cohesion factor" was not directly translatable to cohesion force (in dynes). His experiments were very similar to the viscosity measurements made in this program.

From the discussions and reviews, it appeared that the centrifuge method was the most reliable method for measuring adhesive force. Boehme et al. (1962) considered it as the most accurate one for measuring small

particles 2μ to 50μ. The aerodynamic method assumed a certain air velocity profile in the calculation of forces acting on the particle; however, this assumption is difficult to verify experimentally. Measurements via the weighing or pendulum method are limited to large particles generally larger than 25μ. Although the tilting plane method has been widely used, there is considerable debate regarding its validity. Because the method was currently being used in this program, detailed review of the method is given below.

1. Cremer, Conrad and Kraus (1952)

The tilting plane method was developed by Cremer et al. (1952), who proposed that the adhesive force of a powder bed can be determined by plotting mg cos θ versus mg sin θ where m is the mass of powder on a plate sliding off at the angle θ when the plate is tilted from the horizon, and g is the acceleration due to gravity. By increasing the mass of powder each time, a number of points are obtained through which a line can be drawn. The slope of the line is the coefficient of friction for the powder mass, while the intercept of the line with the ordinate is the adhesive force.

Apparently the reproducibility of the technique was good, and they were able to predict the particle sizes from knowledge of the adhesive forces of the powder masses.

2. Orr, Dallavalle, Stonecypher and Corbett (1957)

In their study of agglomeration and deagglomeration of solid particles, Orr et al. (1957) duplicated Cremer's method for measuring adhesive force, but obtained a rather large spread in the data. They tried to improve the technique by several innovations, but the reproducibility was not significantly better. Orr et al. defined the measurement as shear force studies rather than adhesive force.

3. Nash, Zeller and Lerter (1960a, 1963)

Nash et al. (1960) employed Cremer's method for saccharin, untreated, and saccharin, treated with 1% Cab-O-Sil. They experienced difficulty in coating the plate with a layer of powder permanently affixed to the surface.

The difficulty was in obtaining a uniform coating. Therefore, they employed a polished aluminum plate and consequently the measurement may not be the adhesive force between the powder particles, but rather between the particles and the aluminum plate. For unstated reasons they did not continue the study, and the only data presented was that given for saccharin. Untreated saccharin gave an "adhesive power" of 100 dynes while the Cab-O-Sil treated sample gave approximately 60 dynes.

4. Patat and Schmid (1960)

Patat and Schmid (1960) made a detailed experimental investigation into Cremer's method and concluded that:

a. The scattering of results was large compared with the variations in the so-called adhesive force.

b. Cremer's claim that the adhesion between powder and substrate as measured by their method implies that the powder mass is practically rigid. Residual particles were observed on the inclined plane after the measurement, in contradiction to the implicit assumption of a rigid powder-mass.

c. The "adhesive force" is markedly reduced if spherical particles are used instead of irregular-shaped ones. This points to a "ball-bearing" effect which was assumed to be absent.

d. Negative "adhesive forces" are measured: a fact which is inconsistent with the physics of the system.

5. Whitnah (1961)

In their study on the dissemination of solid and liquid BW agents, Whitnah (1961) employed Cremer's method to determine the coefficient of friction between talc and Sm powders on aluminum, glass, Teflon and stainless steel bases. However, they encountered difficulties which they described as: "For small masses of powder, there was no angle at which the mass would slide off (up to 90°). For larger masses, the powder would break away from the mass in varying amounts and slide off. The entire mass of powder would seldom slide off at the same time." He solved the problem by compacting the powder (with approximately 250 psi of pressure) and then putting the compacts on the tilt table. The reproducibility was improved by this modification. Deviation of the angle of slide

(the reproducibility) varied from 1.2° for talc on Teflon base to 8.3° for Sn on aluminum base. Although he did not consider the [illegible] to nullify the method for measuring coefficient of friction, there is reason to believe that it does.

6. Owe Berg, Hunkins and Stansbury (1963)

Using silica sand 40 to 200μ, Owe Berg et al. found that the force of adhesion between particles is proportional to the particle surface area. Because they were interested in the effect of humidity, the measurements were performed in a controlled humidity box using mechanical manipulators. Although the expressed that some element of judgment on the part of the operator is involved in the technique, they showed that the shapes of the curves obtained by two operators were essentially the same. Consequently for the material measured, it appeared that they considered the method valid and reproducible.

7. Baerns (1966)

Baerns employed Cremer's method to measure the interparticle adhesive force of spherical iron, spherical nickel, glass balloons, and irregular-shaped alumina particles. Adhesive forces for glass balloons smaller than 115μ could not be obtained due to nonreproducible results. However, he did not indicate any difficulty with the other materials, nor did he indicate any difficulty with the procedure. He calculated the adhesive force of the individual particles by estimating the number of contact points between the sliding particles and the nonsliding particles. This number can be estimated by dividing the area of the powder bed by the cross-sectional area of one particle, or roughly, the area of the bed divided by the $(\text{mass mean diameter})^2$.

In his review on the various methods for measuring adhesive forces of small particles, Boehme et al. (1962) concluded that Cremer's method is not an objective method for measuring adhesive forces between a mass of particles and a substrate because of the insufficiently explored relationship between adhesion and friction and the complicated nature of the sliding process. Cremer's method is valid for a powder mass which is practically

rigid. However, the lack of rigidity in many powders affects the results and nullifies their correlation with results obtained from other systems.

D. Discussion of Results

The adhesive forces given by Cremer et al. are given in Table A-1 below. They are the adhesive forces for the powder bed as a whole. Careful scrutiny of their paper revealed that the powder mass was placed on a smooth substrate, such that the adhesive force may be between the powder particles and the substrate, rather than between powder particles.

Table A-1

RESULTS FROM CREMER et al. 1952

Powder	Size	Substrate, cm^2	Adhesive Force (dynes)	Adhesive Tension
Magnesite	200-300μ	Glass Plate (2.84)	110	0.97
	150-200μ	↓	160	0.99
	88-150μ	↓	235	0.99
	75-88μ	↓	330	0.95
	60-75μ	↓	480	1.14
Magnesite	200-300μ	Pressed Magnesite Plate (3.48)	100	0.72
	150-200μ	↓	140	0.70
	88-150μ	↓	210	0.72
	75-88μ	↓	310	0.72
	60-75μ	↓	360	0.70
Magnesite	200-300μ	Nickel Plate (3.83)	105	0.69
	150-200μ	↓	150	0.69
	88-150μ	↓	220	0.69
	75-88μ	↓	320	0.68
	60-75μ	↓	390	0.69
Iron Powder	150-200μ	Nickel Plate (3.83)	15	0.07
	100-150μ	↓	20	0.06
	60-100μ	↓	30	0.06
	60μ	↓	110	--
Molybdenum	11μ	Molybdenum Plate (4.0)	1400	0.32
Powder	2.5μ	↓	6100	0.38

Cremer et al. found that the adhesive force of a powder mass was inversely proportional to the particle diameter. They also noted some other relationships which have not been widely quoted, but which are quite interesting. They found that (1) for a mixture of an adhesive and a

nonadhesive powder, the adhesive force was directly proportional to the amount of the adhesive powder present in the mixture. (2) There is an adhesive tension which is a constant for a particular powder/base system, irrespective of the size range of the powder (see Column 5, Table A-1). (3) A mixture of two adhesive powders behaves similarly to a single powder in that the adhesive force varied in inverse proportion to the average particle size of the mixture. (4) In powders which contain very fine particles (less than 1μ) the adhesive force measured is that due to the fine particles, since it is these which touch the plate and determine the adhesion.

The adhesive forces measured varied from 10 dynes for iron powder on glass base to 6100 for molybdenum powder on a molybdenum plate.

Baerns, using Cremer's method but modified by gluing on a fixed layer of powder, found that the adhesive force was directly proportional to the particle size. The contradiction was temporarily exasperating, but on closer analysis there was no real discrepancy. The data given by Cremer was the adhesive force of a powder mass whereas Baerns gave the "interparticles" adhesive force. In some articles the adhesive forces were not clearly defined, and on hurried reading, the two forces can be confused.

The results of Baerns (1966) are given in Table A-2 below. If the mathematical formulation of Baerns was used to calculate the interparticle

Table A-2

RESULTS FROM BAERNS (1966)

Material	Particle Dia.	Adhesive Force (dynes)
Iron, spherical	1.6	2.5×10^{-5}
	3.3	5.5×10^{-5}
	12.0	6.5×10^{-4}
Nickel, spherical	18	1.5×10^{-3}
	81	1.7×10^{-2}
	137	1.8×10^{-2}
Alumina, irregular shape	11	7.7×10^{-4}
	13	7.8×10^{-4}
	27	3.1×10^{-3}
	68	2.0×10^{-2}
	96	5.2×10

adhesive forces, then Cremer's particle-to-particle forces would be in the region 10^{-2} to 10^{-4} dynes, which is similar to Baerns.

Baerns also differentiated between the experimentally determined "interparticle force" and the "effective interparticle force" and concluded that they were not equal. The distinction between the two is not clear. To quote Baerns, "the interparticle adhesive force determined the stability of the channels which, in turn, established the fluidizability of a particulate material. The effective interparticle force is related to the adhesive force of a particle, but it is also influenced by other effects such as the inhomogeneity of the (fluid) bed structure."

Nash et al. gave the adhesive force of the powder bed. However, since they did not mention the area of the powder bed, it was impossible to compute the adhesive force of the particles.

As mentioned earlier, Orr et al. found that the adhesion of most powders was so low that the results were inconsistent, and no general conclusions were made regarding the shear forces. However, in a later paper, Kordecki and Orr (1960), using a centrifuge method for removing particles from surfaces, found an increase in adhesion with increasing particle size.

Boehme et al. (1962), also using a centrifuge method, measured the adhesive forces of starch, iron and iron oxide powders. For starch particles on a starch substrate, the adhesive force (0.02 to 2 dynes) were independent of the particle size in the size range 7-21μ. For 2μ to 4μ iron or iron oxide particles, the adhesive force (0.01 to 0.02 dynes) was roughly proportional to the particle size. However, because of the limited size range investigated, Boehme may have overstated the relationship. By oxidation and reduction of the iron particles, smooth and wrinkled surfaces were obtained. They contended that smooth surfaces allowed for larger areas of contact than wrinkled surfaces and hence the measured adhesive forces between smooth surfaces were a factor of 10 higher than forces between wrinkled surfaces.

Corn (1961b) employed a quartz cantilever microbalance for adhesion measurements, and concluded that the adhesion increase directly with

particle size for quartz and Pyrex particles 25-88μ diameter. Adhesive forces were in the range 0.01 to 1.5 dynes. Using glass plates of varying roughness, Corn observed a decrease in adhesion with increased surface roughness. He also compared the adhesive forces between fibers with round and spherical ends, and found the spherical-end fiber to have a slightly higher adhesive force.

Larsen (1958) using air jets for dislodging the particles, indicated an increase in adhesion with particle size. However, he observed an increase in adhesion with surface roughness, which is contrary to the findings of other investigators. In his review Corn (1961) attributed the anomaly in Larsen's results to electrostatic charges.

Morgan (1961) reported that Dawes (1952) obtained adhesive forces which were inversely proportional to the square of the particle size.

Investigators using the centrifuge method have obtained adhesive forces of the same size, in the range 0.01 to 1.0 dynes. However, since particles of the same material, and under identical conditions have shown a variation in adhesive force of two or three, it is not surprising to expect a wide range of values.

If the force required to separate two particles was 0.1 dyne, then the energy expended to separate the particles to a distance of 0.1μ (1000Å) apart would be 0.1 dyne $\times$ 0.1 $\times 10^{-4}$ cm = 1 $\times 10^{-6}$ ergs. This value is in line with the estimate given in the critical literature review, in which the binding energy for solid structure adhesion was approximately 5×10^{-6} ergs.

In general, then, the adhesive force of a powder mass varies inversely to the particle diameter; the adhesive force between particles varies directly with the particle size; and adhesive force between objects decreases with surface roughness. These correlations are fairly well established and supported by all the investigations.

Except for the work by Corn on the adhesion of round and spherical-end fibers, there was almost no data on the effect of particle shape on adhesion. No correlation between particle size distribution and adhesive

force was found. However, it is generally agreed that the adhesive forces would be greatly reduced if the powder contained only smooth, spherical particles of a uniform size since such a system would have minimum number of particle-to-particle contact points. Based on this hypothesis Lerman and Bartsch (1965) prepared smooth, spherical particles from irregularly shaped ones. Aerosolization tests of the original and treated powders showed that the treated powder contained fewer agglomerates, and this, the authors concluded, was due to the change in the shape and size distribution of the particles.

E. Mechanism of Additives and Their Effect on Adhesive Force

A considerable amount of time was devoted to the use of additives for powder modification. In support of this work, a review of the literature was made on the mechanism by which additives operate. If the mechanism is known, then the effect of an additive on a powder can be predicted.

There was some evidence (primarily from electron microscopy) which indicated that the additive functioned by attaching itself around the base particles. Nash et al. (1965), Craik (1958) and Whitnah (1964) have shown electron photomicrographs of powders, treated and untreated, which reveal a coating of additive particles around the base particles. There is, however, no evidence to elucidate the mechanism by which additive alters the physical and/or chemical properties of base particles.

For the system Cab-O-Sil (0.014) - Carbowax powder (11μ MMD), Nash et al. (1965) indicated that the coating occurs by attraction between the negatively charged Cab-O-Sil particles and the positively charged Carbowax 6000 particles. They showed, by means of electron photographs, how the smooth surface of the carbowax particles is made rough by the coating of the tiny irregularly shaped Cab-O-Sil particles. They suggested that the change in the flow properties is caused by the Cab-O-Sil layer acting "as a layer of tiny ball bearings, permitting the Carbowax 6000 particles to slide over each other more easily...." If the Cab-O-Sil particles act as ball bearings, then it would appear that the flow properties of a 2% addition or a 3% addition, would be equal to the 1% mixture. Instead they found that excessive Cab-O-Sil particles (more than that required

to give one complete coating) was detrimental to the flow. The ball bearing concept also implies that the Cab-O-Sil particles are lightly attached to the Carbowax ones, and that there is no preferred orientation of the silica particles. It would be revealing to determine whether or not the Cab-O-Sil particles are strongly attached to the Carbowax particles.

In his review on flow properties of dry powders Crosby (1960) states, "reduction of surface roughness and alleviations of stickiness are accomplished...by the use of 'lubricants' or 'glidants,' as they are sometimes called." He suggested that the role played by the fineness may be that of filling in the surface impressions of the larger particles. The smoother the surfaces of the particles become, the better the flowability of the small size classes." Although the relationship between smooth surface and flowability is obvious for macroscopic bodies, the concept may not be true for microscopic particles. There has not been any exhaustive experiments which showed that smooth particles in the 1-10μ range flow better than irregularly shaped ones (excluding needle-shaped or other unusual shaped particles). Thus Crosby's hypothesis, that flowability is increased by the fines filling the impressions of the base particles, thus producing a smoother surface, is based more on thoughts than on facts.

In their study on deagglomerating agents, Owe Berg, Gernish and Flood (1963) contended that an additive works by forming an agglomerate of the order of 25μ in size with particles of the base powders. The bonds holding these agglomerates are comparatively weak and are readily broken up by impaction, friction, etc., which occur in powder flow, and therefore the agglomerates do not prevent free flow of the powder. On the other hand, the strength of these bonds are strong enough to hold the agglomerates together in the aerosol state. They did not state the mechanism and manner by which the additive adheres to the base particles, although they did state that the additive causes a change in the electrostatic charge distribution so that there was equal abundances of positive and negative particles. Actually they did not directly observe the 25μ size agglomerates, but deduced it from calculations on the size of agglomerates supported by a vertical air stream flowing at a certain velocity. On landing

(or impact) the 25μ size agglomerates broke up and thus they were not able to directly observe these large agglomerates. Their conclusion, based on calculation and indirect observation, is at best a working hypothesis. The formation of 25μ size agglomerates from treatment with the "right" additive has not been observed in this program.

Haddorf et al. (1965) in their treatment with additives to prevent agglomeration, found that certain additives were helpful in increasing the aerosolizability of powders, but the evidence was too widely scattered to state any definite correlation. Their conclusions re the mechanism of surface additives were similar to Owe Berg et al. (1963). They stated, "there is evidence both in our observations and those of other investigators (referring to Berg et al.) that the so-called "anti-agglomerants" act by causing the formation of small uniform agglomerants which roll over one another and thus produce "visibly" free-flowing powder, yet do not reduce overall energy requirements for deagglomeration." There was, however, no data (other than observations) to support the above statement.

A few investigators, e.g. Eisner (1960), Lane (1961), found that waterproofing certain powders with a hydrophobic coating served to reduce the adhesiveness of the powder bed. In this situation the coating repels the adsorption of water vapor, thus minimizing adhesion due to water surface tension.

Another mechanism by which surface additives operate is given by Craik (1958) and Craik and Miller (1958). He investigated the transformation of starch from a sticky, difficult-to-flow powder to a free flowing one by the addition of magnesium oxide (~0.05 dia). Using electron microscopy he showed that the starch grains were covered with a uniform pattern of protuberances representing the magnesium oxide particles. He stated, "the effect of the magnesium oxide on the flowing properties does not appear to conform to the usual principles of solid lubrication. A solid lubricant, such as talc, depends for its effect on its layer-lattice structure, with very low bond strength between the layers which thus slip easily over each other. Magnesium oxide has an unrelated cubic structure and would not be expected to act as a solid lubricant." Also the reduction

in the flowability, which occurs when more than a certain amount is added (1%), would not be expected to occur as a result of lubrication. Surface roughness is not a major factor governing the flow of the starch since the adsorption produces a rougher surface, which should give an opposite effect to that which is observed.

To explain the phenomenon, Craik postulated that the layer of magnesium oxide particles imposed a gap of approximately 1000 Å between the starch grains, such that the van der Waals forces become greatly reduced, probably to insignificance. It is considered that the corresponding forces between the magnesium oxide particles are less effective owing to the particles having smaller areas of true contact; this would explain the reduced adhesion. Another reason for the reduction in adhesion may be due to the orientation of the oxide particles. "If the starch grains have on their surface a small unsaturated valency density, this would cause the magnesium oxide particles to be oriented on the surface in such a way as to present sheets of valency charge of a uniform sign to the exterior, and thus they would have the effect of greatly increasing the charge density and the mutual repulsion of the surfaces."

In his study on the adhesive force of iron and iron oxide particles (obtained by oxidation-reduction of the same particles) Boehme et al. (1962) concluded that smooth surfaces (iron particles) allowed for large areas of contact and hence the adhesive forces between smooth surfaces were a factor of 10 higher than the forces between wrinkled surfaces (iron oxide particles). Using glass plates of varying roughness Corn (1961) in his adhesive force study also observed a decrease in adhesion with increased surface roughness. Although these studies were limited, they do indicate some data to support the hypothesis that rough particle surfaces are less adhesive to each other than smooth ones. In contrast there has been almost no data which might indicate that smooth surfaces are less adhesive.

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Appendix B

BIBLIOGRAPHY ON PNEUMATIC DISSEMINATION OF DRY POWDERS

Appendix B

BIBLIOGRAPHY ON PNEUMATIC DISSEMINATION OF DRY POWDERS

The first phase of this program on pneumatic dissemination of dry powders was a literature survey of the subject. The information uncovered was reported in Chapter IX of Poppoff (1965), Aerosol Dissemination Processes - A Critical Review, Volume I. From then to the present (April, 1967) the literature was continuously searched, both for new and old references pertaining to the subject. The bibliography presented here includes the new references as well as the old. In addition other references, which are not directly related to the subject but which are sufficiently useful as background information, are also included.

Efforts were made to obtain copies of these articles which were of special interest to the subject. These articles are on file at Metronics Associates, and they are marked with an asterisk (*) in the bibliography.

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