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Determination of Particle Size Distribution of Selected Aerosol Cosmetics. II. Cascade Impactor Use in Fluorometric and Weight-by-Difference Methods*

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Synopsis—This report describes the evaluation of two methods of analysis which can be used in conjunction with the CASCADE IMPACTOR when determining the PARTICLE SIZE DISTRIBUTION of AEROSOLS. The Cascade Impactor was employed to separate and classify areosol particles into the 0.5-, 1-, 4-, 8-, 16-, and 32-μ range. A series of determinations was carried out to ascertain the acceptability of a WEIGHT-BY-DIFFERENCE method and a FLUOROMETRIC method for determining the amount of product deposited at each stage. Two different hair spray formulations and two suspension-type aerosols were used. Findings showed that the fluorometric method produced results similar to the weight-by-difference method, with the added advantage of being less time consuming. Solids of known particle size distribution were used for those aerosols containing insoluble material. In this manner, a comparison could be made between the known particle size distribution of a material and the particle size distribution determined by both of these methods. The results indicated that this procedure was applicable for the determination of the particle size distribution of aerosols having particles in the range of 0.5 to 32 μ.

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Introduction

During the past few years increased attention has been given to the particle size distribution of aerosol products. While the importance of this property has been recognized for many years in relationship to oil-based insecticides, it has not been deemed very important for other aerosols except those intended for oral use. Although particle size alone does not determine efficacy, it does give an indication of it in such products as disinfectants, underarm deodorants and antiperspirants, feminine hygiene sprays, hair sprays, and other aerosols. A product developed for application to a given area would be troublesome if the particles were so small and light that more particles became airborne than were deposited onto the desired surface. On the other hand, a room deodorant would not be very effective or efficient if the particles settled rapidly following each application. Not only can improper particle size distribution be detrimental to the product, it can present a potential toxicity problem if the airborne particles are inhaled. The danger of inhaling mineral oil particles and the possibility of lipoid pneumonia occurring, especially in infants, is well known. Dautrebande (1–3) and Brown (4) report that particles of 1–5 μ and smaller are easily inhaled and reach the respiratory tract.

Many methods for the determination of particle size distribution are available; however, direct microscopic measurement, light scattering, microphotography, and impaction technics have found the greatest application. Several of these methods have previously been reviewed (5).

The Cascade Impactor* has been found useful in classifying particles produced from an aerosol spray in a range of from about 0.5 to 32 μ in diameter. Sciarra *et al.* (5) studied the use of the Cascade Impactor for determining the particle size distribution of several hair spray formulations and different aerosol valves. One of the problems noted in using the Cascade Impactor is the determination of the amount of material deposited upon each of the slides. In the above study, a weight-by-difference method was used and, while found to be acceptable, was time consuming and limited to those aerosols which contained a nonvolatile solid.

It is the purpose of this investigation to study the use of the Cascade Impactor for the determination of the particle size distribution of sev-

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eral aerosol products having a different particle size distribution. Particular attention has been devoted to methods which can be used to determine quantitatively the amount of material deposited on each slide. The results obtained from the weight-by-difference method and another method will be compared.

Experimental

A Cascade Impactor, Model CI-S-6, was used throughout the study. This model consists of a set of six round glass slides approximately 38 mm in diameter, fitted at each of the small jet openings. The instrument has been calibrated and the different sized particles will impact upon the glass slide as follows:

Upper slide	32 μ
Next lower	16 μ
Next lower	8 μ
Next lower	4 μ
Next lower	2 μ
Bottom slide	1 μ
Filter	0.5 μ

The impactor was assembled in the verticle position with the chamber having the largest diameter opening on top and each additional lower chamber decreasing in opening size. A glass expansion and sampling chamber was then fitted over the upper opening as shown in Fig. 1.

Each of the glass slides was previously weighed using a semimicro analytical balance. The aerosol preparation was accurately weighed and a sample of about 10 g was sprayed intermittently into the chamber. Prior to introduction of the sample, a vacuum of 17 in. of mercury was applied so that a flow rate of 12.5 l./min was obtained. This setting was used throughout the study. The aerosol container was then reweighed in order to obtain the sample weight. Vacuum was applied for an additional 5 min, after which time the vacuum was discontinued and the Cascade Impactor was disassembled. Each glass slide was then allowed to dry and, using the semimicro balance, was reweighed to obtain the weight of residue remaining at each stage. Knowing the amount of nonvolatile material present in the aerosol originally, one can calculate the amount of product deposited on each slide. This procedure was used for those samples analyzed by the weight-by-difference method.

Figure 1. Assembled Cascade Impactor fitted with sampling chamber

Fluorometric Method

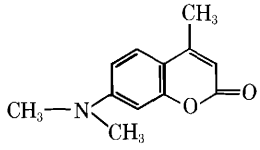
A fluorometric method was also used to determine quantitatively the amount of residue deposited on each slide. A Turner Fluorometer,* Model 111, was fitted with the proper primary and secondary filters, depending upon the nature of the fluorescent dye used as a tracer. Each of the slides from the Cascade Impactor was thoroughly washed with 5 ml of anhydrous ethanol in a 10-ml beaker in order to dissolve the residue. The sample was then transferred to a 5-ml Pyrex glass cuvette and its per cent fluorescence determined. This method was used throughout this study.

Selection of Tracer Dye

Several different fluorescent compounds were investigated. Preliminary studies indicated that Blancophor AW High Concn.† might be useful for this purpose. Chemically, this compound is 4-methyl-7-dimethyl aminocoumarin.

* G. K. Turner Associates, Inc., Palo Alto, Calif.

† GAF Corporation, Dyestuff and Chemical Division, New York.



Peak absorption occurs at approximately 366 mμ (long-wave UV range) while peak readmission of visible light occurs at between 465 and 470 mμ although there may be some variation depending upon the substrate. Solutions of Blancophor were prepared in absolute ethanol.* A solution containing 0.50% by weight of Blancophor was diluted with absolute ethanol in order to produce concentrations in the range of 0 to 0.5% by weight. The fluorometer was fitted with the filter combination shown in Table I. Five milliliters of each dilution was placed in a cuvette and the per cent fluorescence was read. These results are shown in Table I and Fig. 2 and represent the average of three determinations.

The above procedure was repeated using dilutions of Blancophor in the range of 0 to 0.05% by weight of solution using absolute ethanol as the solvent. These results are shown in Table II and Fig. 2 and represent the average of three determinations.

* Similar dilutions were prepared using SDA 40 anhyd. and no change in fluorescence was noted using the filter combinations indicated.

Table I

Fluorescence of Blancophor AW High Concn. Solutions

Blancophor Solution (Wt %)	Fluorescence Reading[a] (%)
0.50	76
0.45	75
0.40	74
0.35	70
0.30	68
0.25	65
0.20	61
0.15	54
0.10	47
0.05	36
0.025	24
0.0125	15
0.0063	12
0.0031	7
0.0016	4
0	0

[a] Primary filter, (47B + 2A); secondary filter, (2A − 12 or 15 + 1% neutral filter); range, 10×.

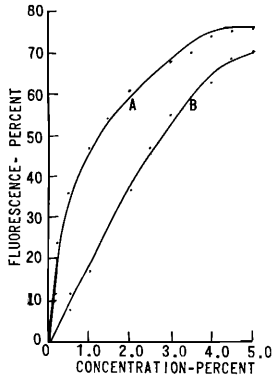


Figure 2. Effect of concentration upon fluorescence of Blancophor AW High Concn.

A. Concentration $\times 10^{-1}$
B. Concentration $\times 10^{-2}$

Table II
Fluorescence of Dilute Blancophor AW High Concn. Solutions

Blancophor Solution (Wt %)	Fluorescence Reading[a] (%)
0.050	72
0.045	69
0.040	64
0.035	60
0.030	57
0.025	49
0.020	39
0.015	30
0.010	19
0.005	10
0.0025	4
0	0

[a] Primary filter, (47B + 2A); secondary filter, (2A − 12 + 10% neutral filter); range, 1×

Preparation of Aerosol Samples

In order to determine the effectiveness of the fluorometric method and to compare the results with the weight-by-difference method, aerosols were prepared in a manner similar to that previously reported (5), except that a sufficient amount of Blancophor was added to produce a concentration of 0.05% by weight. The formulation consisted of 30%

by weight of concentrate and 70% by weight of Propellant 12/11† (50 : 50). This was packaged in an aerosol can and fitted with an aerosol valve. The particle size distribution was determined using the Cascade Impactor and the amount of residue deposited on each slide was determined by the fluorometric method of analysis. Figure 3 shows the Cascade Impactor disassembled with a view of the glass slides. The comparison of these results with those previously obtained by the weight-by-difference method is shown in Table III.

† Propellant 12 is dichlorodifluoromethane and Propellant 11 is trichloromonofluoromethane.

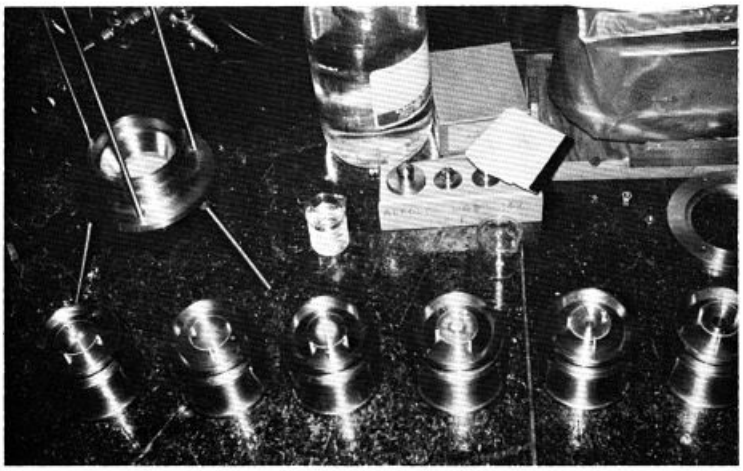


Figure 3. Cascade Impactor disassembled showing glass slides

Table III
Comparison of Cumulative Weight Per Cent Determined by Different Methods

Diameter of Particle (μ)	Hair Spray—A		Hair Spray—B	
	Weight by Difference	Fluorometric Method[a]	Weight by Difference	Fluorometric Method[a]
1	0.44	0.281	0.14	0.241
2	1.06	1.219	0.65	1.097
4	2.24	2.437	1.85	2.215
8	3.65	3.656	2.24	2.827
16	3.87	3.900	3.53	3.201
32	4.35	5.148	4.35	4.256
Over 32	100	100	100	100

[a] Average of three determinations.

Aerosols Containing Insoluble Material

Aerosols containing a solid material of known particle size were prepared. Sulfadiazine Calcomites® and Microcrystals* were employed in this phase of the study and formulated as follows:

Sulfadiazine Calcomites or Microcrystals	0.5
Span 85†	10.0
Ethanol, anhydrous	20.0
Blancophor AW High Concn.	0.064
Propellant 12/11 (50 : 50)	69.436

The sulfadiazine was placed in a mortar with the Blancophor and mixed with the Span 85. Propellant 11 was added and mixed with the powders. This slurry was then placed into an aerosol container and chilled. Propellant 12 was added by the cold process and the container was then fitted with a metered valve. By means of an oral adapter, approximately 12–15 sprays were dispensed into the sampling chamber of the Cascade Impactor. The results are shown in Tables IV and V and represent the average of three results of each powder.

Table IV

Particle Size Distribution of Sulfadiazine Calcomites Aerosol

Diameter of Particle (μ)	Mass of Collected Material (g)	Cumulative Mass (g)	Cumulative Weight Per Cent
1	...	...	...
2	0.6864	0.6864	13.61
4	1.9024	2.5888	51.70
8	2.3163	4.9051	97.95
16	0.1009	5.0060	100.00
32	...	...	...

Discussion of Results

The fluorometric method of analysis has been used for many substances which have the property of being fluorescent. Tracer studies have also been conducted by the addition of a fluorescent dye to a known system. Since the intensity of the fluorescent light emitted by a sample

* Sulfadiazine Calcomites (American Cyanamid Corp., N. Y.), 99% of the particles are less than 10μ and 95% are less than 4μ. Microcrystals, 99.7% of the particles less than 10μ and 50% less than 5μ.

† Sorbitan Sesquioleate, Atlas Chemical Industries, Wilmington, Del.

Table V
Particle Size Distribution of Sulfadiazine Microcrystals Aerosol

Diameter of Particle (μ)	Mass of Collected Material (g)	Cumulative Mass (g)	Cumulative Weight Per Cent
1	0.3803	0.3803	11.87
2	1.0309	2.0612	44.66
4	1.5309	3.5427	75.45
8	1.3127	4.8553	100.00
16	...	...	...
32	...	...	...

under constant input light intensity is directly proportional to the concentration of the fluorescent compound, these substances can be used as tracers (6). The use of one such fluorescent compound for this purpose has been shown in this paper. As can be seen in Fig. 2, the initial concentration of the material must be carefully selected so that in the range of concentration used, fluorescence becomes indicative of concentration. Concentrations of Blancophor above 0.05% were noted to produce only slight increase in fluorescence with an increase in concentration. For this reason, 0.05% was the maximum concentration of Blancophor used in this study.

Two hair spray aerosols, having a different particle size distribution, were studied by both the weight-by-difference and fluorometric methods of analysis. Table III gives a comparison of the results. It can be noted that the particle size distribution as determined by the fluorometric method tends to give slightly higher cumulative weight per cent than that given by the other method. This could possibly be due to the greater accuracy of fluorometric analysis.

To convert per cent fluorescence to cumulative weight per cent, the assumption was made that if a 5-ml sample could be recovered from the aerosol product and its fluorescence determined, the reading would be 72% (as shown in Table II and Fig. 2). By adding a sufficient amount of dye to the aerosol to produce a 0.05% by weight solution, this assumption becomes valid. The amount of dye deposited on each slide is then equal to

$$\frac{\text{Known concn. of dye in sample} \times \text{fluorescence of slide}}{\text{Fluorescence of sample}}$$

or

$$\frac{0.05 \times \text{fluorescence of slide}}{72}$$

This quantity is then indicative of the amount of spray deposited on the slide and can be used as the "mass of collected material." Since the results are given as cumulative weight per cent, it is not necessary to translate this amount into the weight of the entire spray. By multiplying the weight of aerosol sample by the concentration of dye added, one can determine amount of dye present in total aerosol sample which is then distributed over the slides in the same ratio as the particle size distribution.

Satisfactory results were produced with the suspension-type aerosols. However, since a nonvolatile material was added to the particles as a dispersing agent, a slightly different particle size distribution was obtained. Both the Microcrystals and Calcomites of Sulfadiazine produced acceptable results.

Studies are currently underway to adapt this method to a variety of other aerosol products. Since the dye used in this study is water insoluble, its use is limited to those aerosols containing alcohol and other similar solvents. Other dyes are being investigated for use with aerosols containing water.

Summary and Conclusions

Fluorometry was evaluated as a tool in determining the particle size distribution of aerosols. Various fluorescent substances were investigated for this purpose. The particle size distributions of two hair sprays having different particle size distribution were evaluated by this method and the results compared with those obtained by a weight by difference method. This method was then used to determine the particle size distribution of aerosols containing insoluble materials of known particle size.

Blancophor AW High Concn. was found to be acceptable as a tracer in this study, providing its concentration did not exceed about 0.05% by weight. The comparison of the fluorometric and the weight-by-difference methods to determine the amount of material deposited in each stage of the Cascade Impactor showed that either method could be used; however, fluorometric analysis can be considered to be of greater accuracy.

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