INHALED PARTICLES
IV

(IN TWO PARTS)

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ON THE DEPOSITION OF UNIPOLARLY CHARGED PARTICLES IN THE HUMAN RESPIRATORY TRACT

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Abstract—The effect of unipolar electrostatic charges on the total deposition efficiency in the human respiratory tract has been investigated.

Monodisperse carnauba wax particles of diameter between 0.3 and 1.1 μm were charged, in a corona discharge apparatus, with a number of elementary charges ranging from about 30 to 110 per particle, with a narrow distribution. The experiments were performed with volunteers breathing through the mouth at 12 resp/min at constant flow rate, without pauses.

The increase in deposition was of the order of 15 to 30%, relative to neutral but otherwise unchanged monodisperse aerosol. The increase depended on the charges carried by each particle, but was independent of the concentration. Therefore it may be concluded that the variation can be ascribed to the effect of electrostatic attraction between the particle charge and the image charge on the airway wall.

INTRODUCTION

Electrical charge is a very important parameter in aerosol physics, as it may influence the particle behaviour in suspension and in deposition (FUCHS, 1964; WHITBY and LIU, 1966).

For example, it has been noted that the electrical charge influences the efficiency of fibrous filters: deposition is increased both by the charges carried by the particles and by the charges on the fibres (DAVIES, 1973; MERCER, 1973, p. 122). The fact is confirmed by the decrease of the efficiency when the filters are exposed to radioactive sources, which can neutralize the charges on the fibres and, at least partially, those carried by the aerosol (CAGNETTI and ROSSI, 1971).

Therefore, it is of interest in connection with some practical problems in industrial hygiene to determine the effect of the electrical charge carried by an aerosol on its deposition efficiency in the human respiratory tract. At the moment the literature on this subject is scarce, and the only data existing indicate a minor influence of the electrostatic phenomena (MERCER, 1973, pp. 7–8). However, it has been remarked that some differences in aerosol behaviour may be explained by the electrical charges carried by the particles.

Preliminary experiments carried out in our laboratory showed clearly that the deposition of particles in human airways was influenced by the electrical charge of the particles. This result suggested the present investigation, aimed at producing more accurate data in order to quantify the phenomenon and to explain the interaction mechanisms.

EXPERIMENTAL APPARATUS

Our experiments were performed with monodisperse aerosol of carnauba wax generated by condensation (PRODI, 1972). These particles were solid spheres at human body temperature; the median diameter was adjustable in the range 0.2 to 2.0 μm with a geometric standard deviation between 1.03 and 1.10. An electron photomicrograph of 0.65 μm carnauba wax particles is given in Fig. 1.

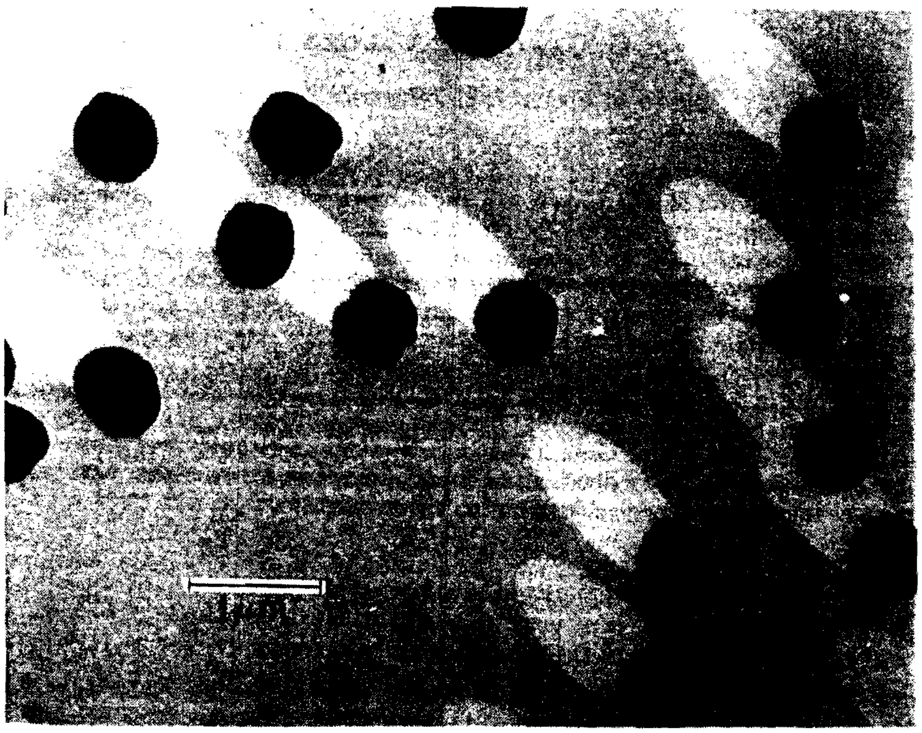

FIG. 1. Electron photomicrograph of carnauba wax particles.

The particle dimensions and concentration were stable: the number of particles per unit volume at the outlet of the dilution stage was constant within $\pm 3\%$ and the size was also constant within the mentioned standard deviation for a long period (several hours). Nevertheless, the aerosol characteristics were frequently checked during the operations by means of an electrical mobility analyser (GIACOMELLI MALTONI *et al.*, 1973); it permitted determination of the median diameter with 2% accuracy and the distribution.

The aerosol thus generated was statistically neutral with an absolute average charge between 2 and 3 elementary charges per particle.

Carnauba wax has not been proved toxic and it is not hygroscopic; hence the particle size is constant along the human respiratory tract.

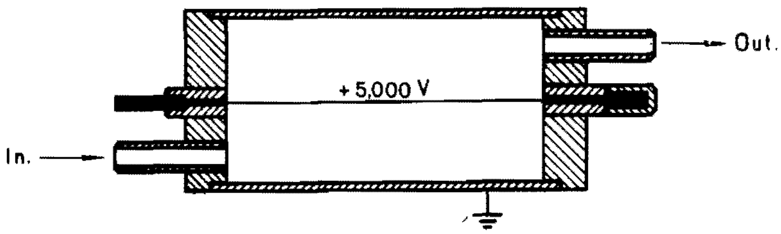

FIG. 2. Schematic diagram of the charger.

For our experiments on the deposition efficiency versus the electrical charge carried by the particles, the aerosol was unipolarly charged in a corona discharge apparatus. Figure 2 gives a schematic diagram of the charger. The internal diameter of the cylinder is 33 mm and the tungsten wire diameter is 50 μm. The voltage of the axial wire was set at +5000 V with respect to the cylindrical outer electrode, therefore the aerosol was positively charged.

Under given conditions the charges acquired by the particle depended on their size and would be adjusted by varying the aerosol flow through the charger. Particles deposited on the electrodes tended to decrease the aerosol charge which stabilized around a constant value in approximately an hour. The average charge carried by the particles and its distribution were determined by means of the electrical mobility analyser with a lower voltage.

For 0.65 μm aerosol we could easily vary the average charge carried by each particle in the range from 30 to 110 elementary charges with a half-width at half-maximum of

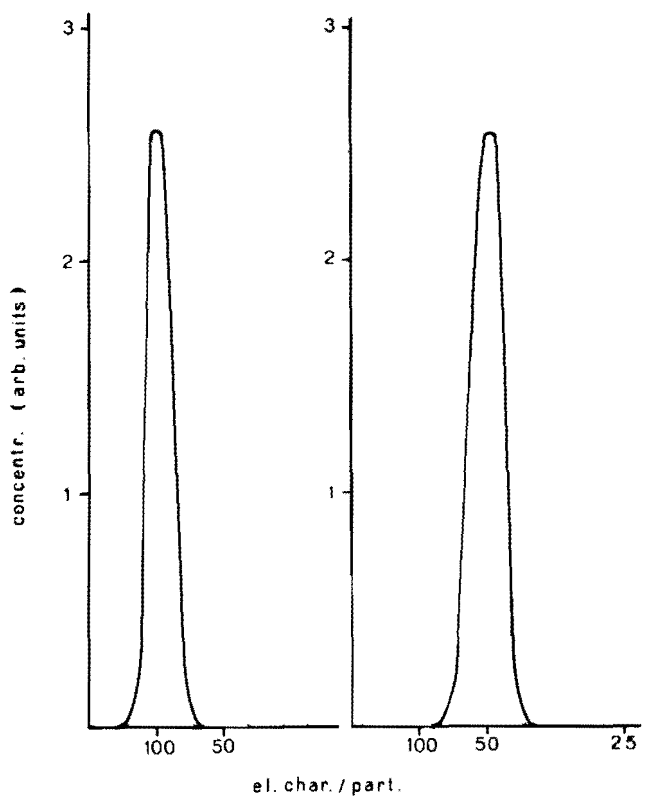

FIG. 3. Patterns of electrical charge distribution on 0.65 μm aerosol.

15 to 18%. In Fig. 3 two patterns of electrical charge distribution on 0.65 μm aerosol are given.

The apparatus used for the inhalation tests was a modification of the one used for total deposition experiments (GIACOMELLI MALTONI *et al.*, 1972). It is schematically shown in Fig. 4. Of the aerosol generated 60 to 120 l/h passed through the charger A

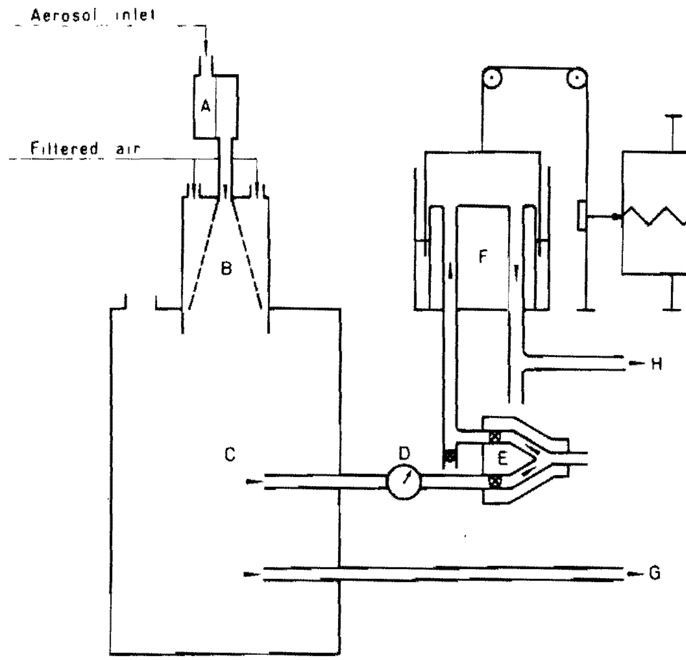

FIG. 4. Schematic diagram of the inhalation apparatus. A—charger. B—dilution stage. C—mixing stage. D—Wright respirometer. E—two-way valve. F—spirometer. G and H—light-scattering cells.

(which was on line even when no voltage was applied). A dilution stage B supplied about 3000 l/h of filtered air to obtain the wanted concentration of aerosol. A mixing stage C sent the aerosol to be inhaled to a Wright respirometer D (supplied by the British Oxygen Corp.). The volunteer inhaled the aerosol through a two-way valve E operated by a timer which alternately connected the mouthpiece to stage C (inhalation) and to a wet spirometer with a 5-litre bell, F (exhalation). This last component provided a compensation volume, measured the exhalation and controlled the whole respiratory cycle. The particle concentration in the inhaled and exhaled air was continuously monitored by two light-scattering cells G and H. The cells' response was compared and normalized before each single measurement then rechecked afterwards.

All the components of our equipment were metallic, electrically connected to one another and grounded.

EXPERIMENTAL RESULTS

The volunteers, four healthy adult males, were trained to breathe through the mouth with 1000 cm^3 tidal volume and at 12 respirations per minute, with a constant flow rate both during inhalation and exhalation, without pauses.

A first set of measurements was performed at a fixed concentration and electrical charge carried by the aerosol for three particle sizes. It was agreed to undertake experiments with particle sizes where the predominant effects were Brownian diffusion (0.33 μm) and sedimentation (1.10 μm). An intermediate size (0.65 μm) was also investigated.

Our experimental conditions are illustrated in Table 1, where the aerosol concentration and the elementary charges per particle are given for each size.

TABLE 1. CHARACTERISTICS OF THE AEROSOL USED IN THE FIRST SET OF EXPERIMENTS

Particle diameter (μm)	Neutral particles	Charged particles	
	Concentration (part./cm^3)	Concentration (part./cm^3)	Elem. char./part.
0.33	2×10^5	1×10^5	29
0.65	6×10^4	4×10^4	45
1.10	4×10^4	2×10^4	95

Initially for each subject many runs (10 to 14) were performed with 0.65 μm aerosol in different days and hours. The aim of this procedure was to evaluate the variability due to our experimental apparatus and to the biological variability of each volunteer. The results are shown in Table 2 where the deposition efficiency of neutral and charged particles and its standard deviation are given along with the number of runs for each subject.

TABLE 2. DEPOSITION EFFICIENCY (%) FOR NEUTRAL AND CHARGED AEROSOL OF 0.65 μm DIAMETER

Subject	Neutral particles		Charged particles (45 elem. char./part.)		Deposition increase
	No. runs	Deposition efficiency	No. runs	Deposition efficiency	(%)
F.M.	10	21.7 ± 1.4	12	28.1 ± 1.4	30
D.Z.T.	10	23.0 ± 1.3	12	29.8 ± 1.4	30
M.C.	12	28.8 ± 1.8	12	34.0 ± 1.2	18
B.G.	10	23.8 ± 1.0	14	30.7 ± 1.1	29

The relative increase in deposition efficiency for charged particles is also reported. The global standard deviation averaged over all the data of deposition efficiency is 1.3. The standard deviations are small and constant enough to enable us to perform our measurements at 0.33 μm and 1.10 μm with a smaller number of runs (5 for each set of parameters and subject).

In Tables 3 and 4 the results at 0.33 and 1.10 μm diameter are summarized.

TABLE 3. DEPOSITION EFFICIENCY (%) FOR NEUTRAL AND CHARGED AEROSOL OF 0.33 μm DIAMETER

Subject	Neutral particles	Charged particles (29 elem. char./part.)	Deposition increase (%)
F.M.	25.8 ± 1.5	30.7 ± 1.9	19
D.Z.T.	25.5 ± 1.2	32.6 ± 1.2	28
M.C.	28.7 ± 0.6	34.0 ± 0.9	18
B.G.	27.7 ± 0.7	34.4 ± 0.8	24

The variance analysis of the data reported in Tables 2, 3 and 4 shows that the differences in deposition efficiency of charged and neutral particles are statistically very highly significant (confidence limit <1%). The relative deposition increase is 15 to 30% which, although not of major relevance, cannot be neglected.

Supplementary investigation is still needed to point out the mechanisms responsible for the deposition increase of a charged aerosol along the human respiratory tract. Two hypotheses could be proposed: one based on the effect of the electrostatic forces between the particle charge and its image on the walls of the respiratory tract; a second one based on the electrostatic expansion of the aerosol cloud due to the unipolar charges of the particles.

TABLE 4. DEPOSITION EFFICIENCY (%) FOR NEUTRAL AND CHARGED AEROSOL OF 1.10 μm DIAMETER

Subject	Neutral particles	Charged particles (95 elem. char./part.)	Deposition increase (%)
F.M.	27.6 ± 1.1	32.1 ± 1.3	16
D.Z.T.	28.8 ± 2.6	36.3 ± 1.3	26
M.C.	33.5 ± 0.6	38.6 ± 1.6	15
B.G.	29.4 ± 1.6	34.4 ± 1.4	17

If the effect of the second hypothesis were predominant the deposition efficiency should depend on the aerosol concentration and increase with it, while in the first hypothesis the efficiency should keep constant.

In Table 5 the deposition efficiency is given at three different concentrations of 0.65 μm particles carrying the same charge (45 elem. char./part.).

It is evident that the particle concentration is not responsible for the deposition increase of a charged aerosol: these last data are practically coincident with those reported in Table 2.

So far it is apparent that the electrostatic induction of the single particles predominates over the electrostatic cloud expansion.

TABLE 5. DEPOSITION EFFICIENCY (%) FOR 0.65 μm CHARGED AEROSOL AT DIFFERENT CONCENTRATIONS

Subject	Charged particles (45 elem. char./part.)		
	C1 (1.8×10^4 part./cm^3)	C2 (4×10^4 part./cm^3)	C3 (8×10^4 part./cm^3)
M.C.	33.8	33.8	35.2
B.G.	30.5	30.8	30.9

On the other hand, the deposition efficiency increases with the number of elementary charges carried by each particle. In Fig. 5. such behaviour is shown for subject B.F. inhaling 0.75 μm aerosol charged, from time to time, with 35 to 105 elementary charges. The particle concentration could not be kept constant, but decreased with increasing particle charge from 5×10^4 to 1.5×10^4 part/cm^3: this had the effect to keep the absolute charge concentration (elementary charges per unit volume) practically constant. As particle concentration proved not relevant for deposition efficiency, the behaviour of Fig. 5 can be ascribed only to the increase of the charge carried by each particle.

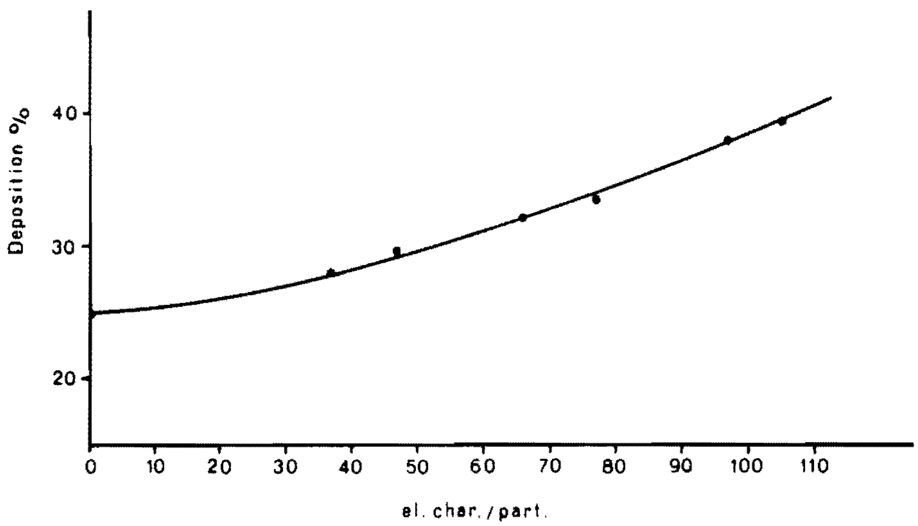

FIG. 5. Deposition efficiency *vs.* elementary charges carried by 0.75 μm aerosol.

CONCLUSIONS

Our data indicate that the deposition increase of a charged aerosol along the respiratory tract is to be ascribed to the electrostatic effect of the charges induced on the respiratory walls by the charges carried by each aerosol particle.

Such effect should also be active when bipolar aerosols containing both positively and negatively charged particles are inhaled; these are particularly frequent in many instances when aerosols are generated by mechanical disruption, like atomization and grinding. Consequently the particles may carry a very large number of elementary charges: then deposition efficiency may be greatly affected by the charge distribution.

It could be of importance to study in detail the effect of the charge on the regional deposition, which may bear relevant consequences for industrial hygiene.

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DISCUSSION

S. E. DEVIR: In my opinion, based on my own studies (*J. Coll. Sci.* 1967, **23**, 80) and those of other workers, the final conclusion on this controversial problem is that, in practice, electric charge has a negligible effect on the deposition of particles in the lungs. In industrial or natural processes, any highly charged particles that may be formed are quickly neutralized or reduced to a few electron charges. In any case, the effect of image forces on the respiratory wall will be nullified in the case of symmetrical bipolar charged aerosols.

Prof. PRODI: I agree that unipolar aerosols are very rarely encountered in practice, but bipolar aerosols containing both positively and negatively charged particles are not uncommon. The deposition mechanism we have described will be effective for such particles when the distance from the wall is small compared with the distance between particles. Perhaps Dr Devir is thinking of individual particles with bipolar charges (dipoles); the image forces will, of course, be small for such particles.

I agree also that highly charged particles will lose their charge, but the time required for this in natural conditions may be too long to allow the charge to be neglected.

D. C. F. MUIR: Is ozone produced in your apparatus in sufficient quantity to affect the lung airway?

Prof. PRODI: The corona equipment works on pure nitrogen gas and then afterwards it is diluted 20–50 times so there is no such effect.

S. J. ROTHENBERG: According to Fig. 4, the exhaled aerosol concentration is measured after the aerosol concentration has passed through the spirometer. Do you not think errors might arise due both to aerosol loss, and to phase differences between the spirometer and concentration records caused by the time taken for the aerosol leaving the mouth to reach the Tyndall cube? Is there any reason why the Tyndall cube was not placed immediately after the valves (and before the spirometer) in your flow line?

Prof. PRODI: We tried to minimize the dead spaces and so we decided to divide as soon as possible the exhaled air from the inhaled air. I would not expect that this could cause any effect on this work because this was just a comparison between the so-called neutral particles and charged particles; but the effect of the equipment is cancelled out because the cells are calibrated before and after each run with the same aerosol and so the response is normalized.

W. H. A. BEVERLEY: Does the deposition rate alter with inhalation of *negatively* charged particles? If so, does it increase or decrease?

Prof. PRODI: We have not yet run a careful set of experiments with negative particles. Preliminary experiments indicate a similar behaviour, but I would not expect anything different.