An Inhalant Atomizer Device and the Particle Size Distribution Measurement *

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A new atomizing instrument for inhalation experiments was designed. This new type device named "Nebulizer" which has been made glass blowing, can geneıate quite fine particles.

In addition, special techniques were developed for measurements of these fine particles. Collection method of the particle samples were carried out, by applying a principle of electrostatic precipitation. And measuring about 0.1 micron in diameter was established.

Considerably useful results has been obtained in the application of the nebulizer as an inhalant equipment for human bodies.

NOMENCLATURE

Special symbols are not used in this report.

1. INTRODUCTION

Fogs and clouds as natural phenomena and those in Wilson's cloud chamber are generated under the circumstances of adiabatic cooling of saturated vapor. The fogs of this origin are generally chracterized by the fineness and the size uniformity of their particles.

On the other hand, the mechanical methods of liquid atomization are widely applied to the fields of daily works and industries for various purposes through the installations of simple sprayers or various atomizers.

The mechanism of these sprayers or atomizers is principly based on the difference of energy of liquid vibration, collision and relative motion between liquid and air.

It is usual that the mist formed by these mechanisms is not so small and uniform in size as compared with the ones by cohesion of vapor mentioned above.

A few types such as pressure type, air-stream type and rotation type, etc. are well known as the important equipments of such kinds. Among them, the air-stream type atomizer gives the finest particles, comparatively.

In case of applying an atomizer to medical treatment to intake them in human being through respiratory tract, however, mist particles from inhalators are required to make as uniformly as possible and to control below about 5 microns in their sizes.

A new atomizer for the purpose of the stated above was devised and trially produced in glass works called "Nebulizer" **.(In this paper it will be hereafter designated as nebulizer.)

We used the nebulizer for a medical study——inhalation experiments on human being—— and obtained satisfactory results.

In the next place, in regard to the method of particle measurements——the number of particles and measuring of particle size distribution——they are prescribed in JIS Z 8808 (Methods of Measuring Dust Content in Stack Gas) and JIS Z 8810(Sampling Method of Airborne Dust by Electrostatic Dust Sampler for Gravimetry), etc.

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**The word "Atomizer" means an instrument which disperse some volume of liquid to fine particles as an atom. As against "Nebulizer" the word is derived from "nebula" means an instrument of mist generator whose particles are finer and indiscernible as a nebula.

Besides, a lot of work has been done on the measurements of liquid particles. Please refer to the bibliography given at the end of this paper.

Generally speaking, it relatively easy to measure solid fine particles and non-volatile oil particles, while special consideration is required in measuring volatile liquid particles such as mist of water or diluted aqueous solutions.

That is to say, it is necessary to collect all the particles of the mist and to keep them stable in size and shape during the measurement.

The water mist generated by the nebulizer consists of exceedingly fine particles——less than a few microns for the most part —— which makes it impossible to collect the mist by blowing it against the wall.

So, by passing the mist through a high voltage electric field, we charged the particles and drived them into a collecting film set in front of an grounded opposite electrode.

As the collecting film was coated with oil, the particles were kept stable in the oil layer after their driving into it.

In this way we succeeded in collecting the mist completely and stabilized mist particles made up to a preparation for microscopy. The particle number and the size distribution were measured with a microscope using common method.

2. MIST GENERATION BY DISPERSION METHOD

Among several types of mechanical liquid atomization, we selected the air stream type atomizer as the most appropriate in this case. In the present study we aimed at the problem of medical inhalator, so glass is considered to be the most suitable material for its transparency, sanitariness, and safety against most chemicals. Besides glass is the most handy and workable material for usual experiments.

2.1 Device of a Nebulizer

In med'cal inhalation, for example, a sedative can be taken in a patient who had an attack of asthema, a limited quantity of medicine must be effectively inhalated into the depths of the lungs (corpuscles). Consequently it is necessary for the inhalant not to generate wasteful particles more than 5 microns diameter.

The new nebulizer was devised to fulfil these requirements. Then we are to explain its developing process briefly in an orderly manner.

In an ordinary simple-type spray (Fig. 1), it often occurs that some of liquid creep out to the outside of the suction pipe and it is flying out just as large particle without atomizing. Then it givees, sometimes the pattern of water droplets as indicated in Fig. 1 (b).

It helps somewhat effective to give water repellency by treatment with silicone on the surface of pipe and nozzle. To prevent flying

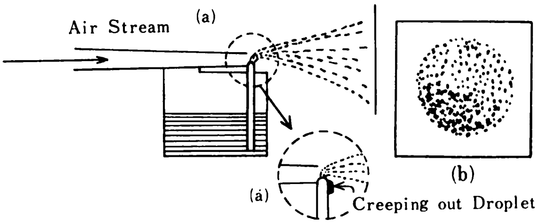

Fig. 1 Sprayer and the Pattern of Water-droplets

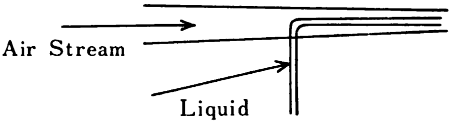

Fig. 2 Sketch of Spray Nozzle

out of the droplets completely, we can make up the formation of spray by laying the suction pipe into the center of the air stream nozzle. (see Fig. 2)

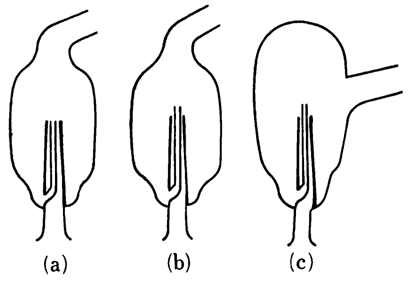

Fig. 3 Nebulizers (made of Glass)

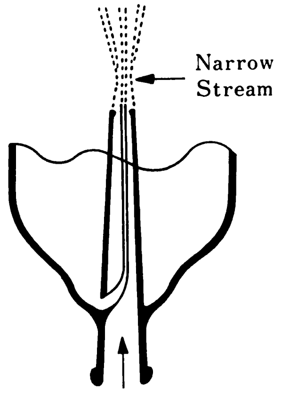

Fig. 4 Spraying Scene by Nebulizer Type(b), (c)

Shown in Fig. 3 are some of variant types of improved sprays as trially made inhalators. A mouth-piece on the bulb is put at the top in (a) and (b), and at side face in (c). In Fig. 3 part of spraying nozzle of the type (a) coincide with the sections of each tips, but of (b) and (c) the tip of inner pipe project slightly that of outer pipe. Fig. 4 shows the spraying nozzles of the type (b) and (c) magnifying its principal portion.

This reconstructed atomizer enables more effective* dispersion of liquid by means of using maximum velocity of the air stream. Moreover, there is no fear of rebounding and flying out even if generate big particles which is divided into smaller ones due to collision going straight against the spherical surface.

As a result, very fine particles alone will flow out like smoke riding on an air stream, slowly.

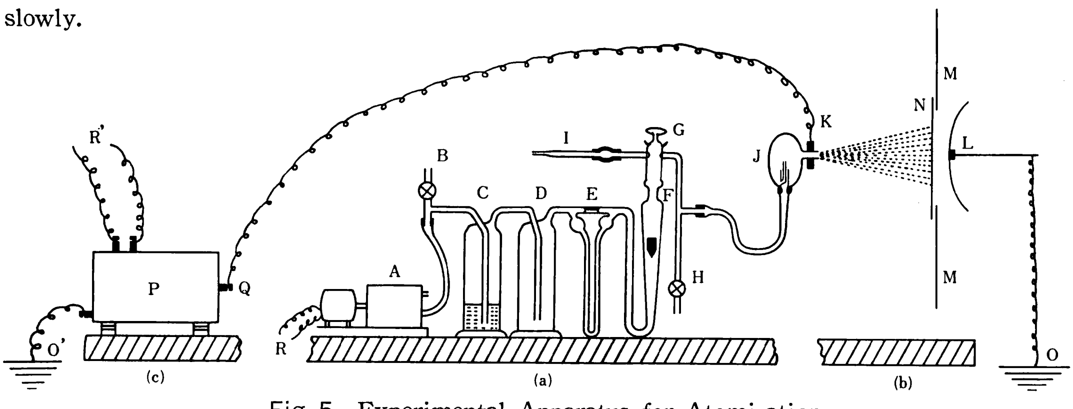

Fig. 5 Experimental Apparatus for Atomization
(a) Part of Mist Generation:- A:Air compressor, B:Two-way (glass) cock, C:Washing bottle (water), D:the same (empty), E:U-type flow meter, F:Rotameter, G:Three way cock (larger), H:Two way cock, I:Nozzle, J:Nebulizer, K:High-voltage electrode (Discharge pole).
(b) Part of Mist Collection:- L:Round cnvex-type electrode, M:Cellophane shelter, N:Mist collecting film, O:Ground.
(c) Part of high-voltage Generation:-P:High-tension generator, Q:High voltage terminal, R, R':Sources of electric power, O':Ground.

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*The mechanism whereby this construction responds to dispersions in liquid atomization effectively, is not fully understood. I do not have a mechanism except to suggest that air stream and continuously it have occurs expansive enlarging in volume of the whole fluid, gradually.

Experimental Apparatus

Generation of mist. The experimental apparatus is shown in Fig. 5
Mist is generated by the part (a) system. Air is sent forth by an air compressor A (HITACHI' s Rotary Type, RC-30, Psessure: 0.2 kg/cm^2, Air-flow: 40 l/min) by way of the gas washers which control the humidity and through the flow meters (U-type and Rotameter).

Nebulizer J (Type (c) in Fig. 3 is used) and there is a certain amount of liquid in its reservoir. The air flow is controlled with cocks for mist to be generated under the optimal conditions of the nebulizer.

2.3 Collection of mist.

This nebulizer high performance as an inhalant possesses. Then the problem is how to collect and measure the mist generated.

Because the particles are too fine and light to collide according to inertia going straight on the collecting film, and even though they are collected they die out immediately by evaporation.

As a result of many consideration, we found that the application of electrostatic precipitation gives a solution to the problem.

A metalic round ring as the high-voltage electrode K (charging voltage max. 70,000 V) is set at the nozzle to charge the mist particles. A concaved circular plate*(made of aluminium) of diameter about 15 cm is set 20 cm away as the opposite pole, being grounded.

A 50 cm square cellophane-covered glass rod flame is set up vertically 2 centimeters in front of the plate, which has in the center a round opening with a diameter of 10 cm. Ahead of the center (its opening part) collecting film N (cellophane & etc.) is fixed by uniformly coated** with a water-repellent oil(e.g. liquid paraffine, silicone oil, etc.)5～10 microns thick.

The insulation of all discharging parts (J, K, L, M, etc.) is kept by fixing their location being supported with glass stands on a wooden desk for isolation. Furthermore, all of the apparatus are set up on a wooden desk which is placed in the room center, keeping clear of all the piping.

The discharging electrode K is connected with a high-power generator P (Fig. 5 (c)). Now, we are all set to go arrangements of the experiments.

3. COUNTING AND MEASUREMENT OF PARTICLES

3.1 Arrangements of the Experiment

Regulation of the Apparatus. Starting the compressor A, adjust the cocks B, G and H. Put an adequate amount of test solution into the nebulizer, and switch on the power source for applying a high voltage on the pole. Put a stop to the cock H, and mist will come out.

Then, all particles passing through the mouth-piece are charged in the electric field of K-pole, and fly toward L-pole and collide against film N. And at the same time, they drive to the oil layer on film N and then particles smoller than below 5μ in size are buried in the oil layer (10μ in thickness) perfectly.

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*The applying of the concaved circular plate is for collecting the mist particles on the 15 cm circle area of the collecting film to unify distribution possibly.

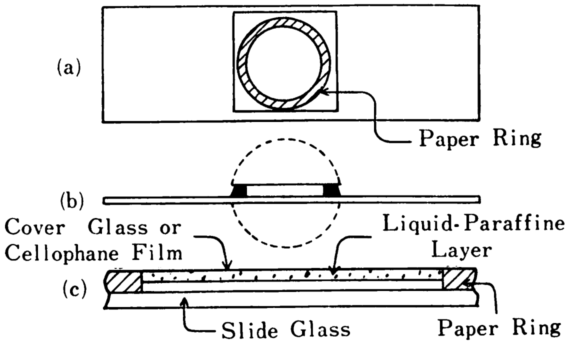
**1 cc of liquid paraffine is spreaded on the surface of 1,000 cm^2 in area by using L-shape glass rod. It is easily done by anyone growing familiar with it.

After a suitable amount* of mist was obtained, stop the generation of the mist by switching off the power source R, R' and open the cock H below.

3. 2 Preparation of Microscopic Samples.

The counting of particles and the measuring of particle size distribution are carried out with microscopy. The samples are prepared as follows:

As shown in Fig. 6 a paper ring (0. 5 mm thick)is stuck on a slide glass. On the top of it



(a):Plane Figure
(b):Lateral Figure
(c):Enlarged Figure, Middle Part of (b)

Fig. 6 Prepartion of Mist for Microscopy

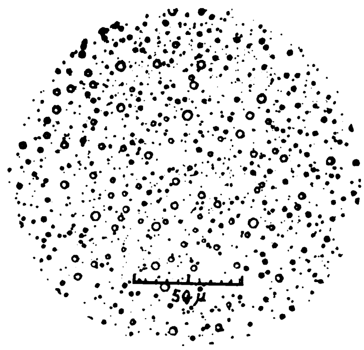


Fig. 7 Microscopic Photograph of Mist

lies a part of the collecting film with the oiled face downward, fixing it with cellophane-tape.

Microscopic samples prepared in this way will be good for quite a long time without any change in number or size of particles. In the 10-micron thick oil layer, the finer particles (smaller than 5 microns in diameter) can be floating in the complete spherical shape.

They are charged with negative and each particle's interaction force experiences no cohesion, so that their state of suspension are kept stable.

This method of preparation enables the observation of the mist particles just as they are generated by the nebulizer. Fig. 7 is one of the photgraphs of the samples.

At such a degree of magnification it is needless to concern about focal range in taking pictures.

3. 3 Measurement of Particles

The volume of air going out from the compressor is set at 16~20 1/min, considering the requirements of another (described later) experiment of human inhalation. The high voltage for charging particles is generally below 50,000V (D.C., negative pole). The maximum voltage is 70,000 V in a comparative experiment mentioned later, but it does not arrive the maximum at which the corona discharge begins.

We prepare the microscopical cover glass coated with oil, which is fixed with cellophane tape at the four corners and set on the suitable position for the collecting film N, for the preservable samples.

Particle Number. In a suitable field of view under microscope pick out at random several sections of the cross sectional graduation plate in eyepiece. Count up the particle numbers in them by using a counter. It is difficult to count the quite fine particles under 0. 1μ, so neglect them with the exception of some special cases.

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*The time of the film exposure to the mist shower determined in a preliminary experiment with a focal-plane shutter, for control to suitable concentration on the preparation films.

Particle Size. Plane diameter of particles of our samples have all spherical shape. Using a graticule, it is possible to count the number and measure the size of particles at the same time. But in the present case we have measured the same directional diameter of each particle, using an equal magnifying transparent gauge on an enlarged photographs illustrated in Fig. 8 provides typical fields of it.

A

B

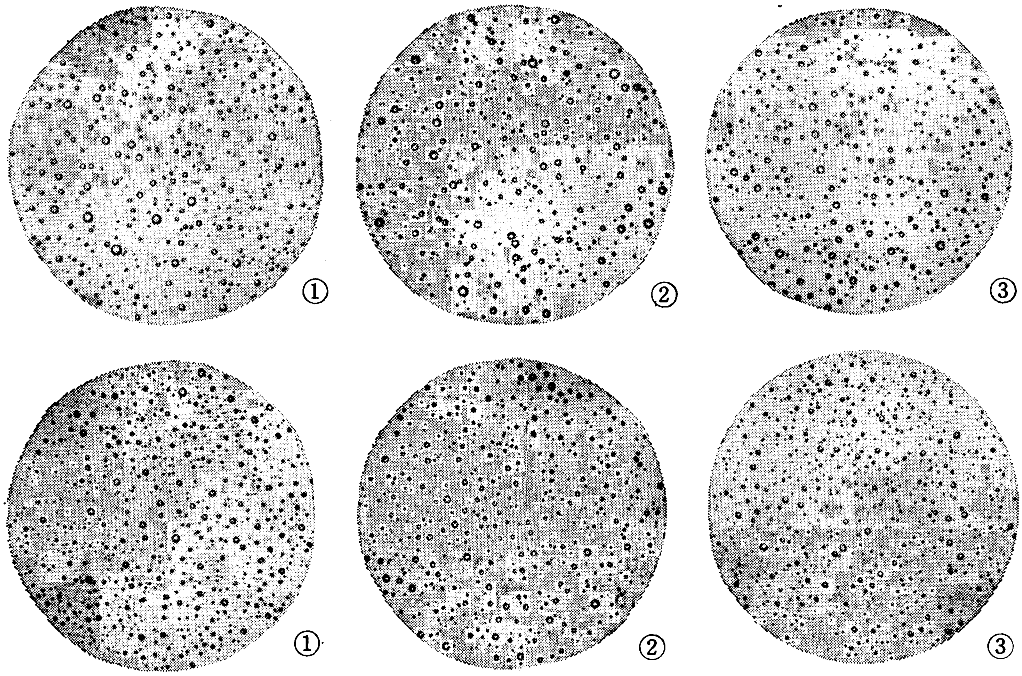


Fig. 8 Microscopic Photographs of Mists
Upper Step A: Solution of Lead Acetate
Lower Step B: Dilute Solution of Red Ink
① : Center Portion ② : 2.5cm from the Center ③ : 5.0cm from the Center

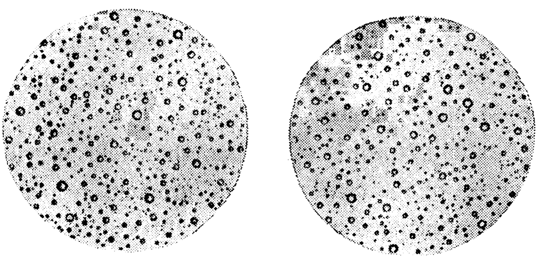
The sample solutions employed in this experiment are the following two; one is an aqueous solution of lead acetate (2 mg/ml) (A) and the other is a dilute solution of red ink (an article on the market, its pigment concentration is unknown). Their viscosities and the surface tensions were measured, respectively.

Table. I shows the data of the both solutions comparing with the datum of water. The facts

Table I Data of Viscosity and Surface Tension

Viscosity	Measured by Ostwald's Viscometer	Centipoise
	Water	1.0050
	Lead Acetate Soln.	1.0050
	Red Ink dil. Soln.	1,0375
Surface Tension	Measured by Du Nouy's Tension Balance	Dyne/cm
	Water	72.75
	Lead Acetate Soln.	72.75
	Red Ink dil. Soln.	69.47

Note: Each Value is measured at 20°C.



(a): 50,000 V (b):70,000 V
Fig. 9 Effect of Applied Voltage

become known by this data are as follows: Comparing the red-ink solution with the water and lead acetate solution, the fomer is slightly higher than the latter in viscosity and is somewhat lower in surface tension, but the diffcrences are very small in every case. Between the data on lead acetate solution and water no remarkable differences are seen.

For the purpose of clarifying the effects of mist atomization causd by applied voltage, two cases of experiment, one using 50,000 V and the other using 70,000 V, were practised on lead acetate solution. Fig. 9 shows the microscopical photographs of mists, and the difference both figures may not appear to be great on the effect of atomization by electrical charging. All other conditions except only electric voltage during the expeliment were kept similar.

4. EXPERIMENTAL RESULTS

4.1 Number of Particles

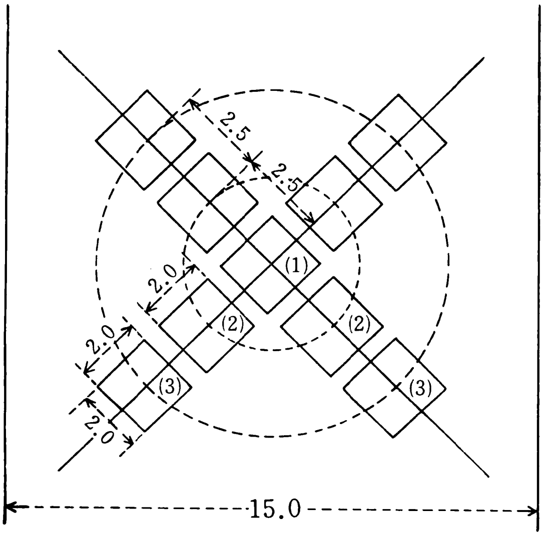
The mist particles generated by the nebulizer have a wide range of size. Some of the finer ones are beyond the resolving power of the microscope, and cannot be counted by microscopy. It may be difficult to count the number of particles accurately, even if a densitometer of photo-scattering or photo-penetrating based on the optical principles is applied.

In this study we can safely neglect the superfine particles smaller than 0.1 micron. Though, there are a great many particles smaller than 0.1 micron, their total quantity is very little comparad with the whole amount of all particles. Therefore, no care is taken for the purpose of this inhalation experiment.

4.2 Particle Size Distribution

The collected mist particles are distributed on the collection film almost uniformly, and it is evident to owing to application of the special electrode mentioned above.

For the samples of microscopic measurement, as shown in Fig. 10, 2 cm squares being cut



(1) Center Portion (2) 2.5cm from the Center
(3) 5cm from the Center
Fig. 10 Sampling Position of Micloscopy on Collecting Film (Unit is cm)

Table II Particle Size Distribution of Mists

Sort of Solns.	Range of Size(μ) / Portion	0.1~1	1~2	2~3	3~4	4~5	5~
Lead Acetate (2mg/ml)	①Central	201	63	13	9	3	0
	②Middle	200	60	30	19	6	0
	③Circum-ference	170	78	36	18	8	0
Red Ink (1% soln)	①Central	264	30	6	4	0	0
	②Middle	273	24	3	2	0	0
	③Circum-ference	276	18	6	1	0	0

centering around the points at the interval of 2.5 cm on the diagonal lines are used. Similar arrangement was applied to the microscopic cover glass.

Table. Ⅱ shows the particle size distribution of lead acetate solution and red ink solution mentioned above. The results summarized in the table indicate that no particles with a diameter of over 5 microns occurred in either solution; and particles of less than one micron in diameter are on the average 63% with lead acetate solution and as much as 90% with red ink solution.

5. CONSIDERATIONS——Summary and Application Example

5.1 Previous Studies and Special Features of Our Study

Liquid Atomization. A number of studies have been made on liquid atomization for various practical purposes. It is widely applied to, for example, household sprays, medical inhaler, spraying of agricultural chemicals, spray coating (painting), spray drying, and fuel injection an engine cylinder or a boiler, etc.

In most cases of applications mist particles are required to be made large enough to inertia. That is, the absence of the force of inertia is not an absolute requisite for most types of atomization.

The present study, on the other hand, attempting inertial motion was successful in devising a special medical nebulizer.

A lot of mechanical atomizing techniques have been developed on the basis of various principles. The typical of them are divided roughly into three types: pressure, air stream and rotation atomizations. Among them, air stream type can produce the finest particles with a relatively simple apparatus. Theoretically, the new nebulizer may come under this type. Its atomizing mechanism, however, has not been well-known in detail. This point also, has not been inquired closely this study.

5.2 Measurement of Mist. Very few attempts have been made to measure infinitesimally fine particles of mist with the diameter of one micron, although many meteological papers have dealt with on the size of water droplets.

The following condition smust be satisfied for the superfine particles to be measured:-

① Superfine particles shall be collected completely which are incapable of moving inertly.
② Mist particles shall not evaporate quickly even in an unsaturated atmosphere.
③ Collected particles shall remain stable in a spherical shape as aerosol.
④ Collectad particles shall be evenly distributed over a microscopic specimen.
⑤ In this particular study: the nebulizer shall be proved to be valid for medical inhalation.

In this study, conditions ①, ②, ③ and ④ were basically satisfied, though some problems remain to be solved. The following is an example of applied study concerning to the condition⑤.

An Application Study——Living Body Inhalation Test by Use of the NEBULIZER

Inhalation of lead into the respiratory system has drawn attention as an important problem of air pollution and occupational diseases. In this connection, an experiment was carried out on the administration of particles in the respiratory system, using the nebulizer.

This work was assisted by the colleagues of Public Health, Osaka City University.

The procedure is as follows:- The nebulizer (in Fig. 5 (a)) was set to generate the mist of lead acetate solution intermittently*, keeping time with the subjects inhaled it through the mouth (respiratory tract).

The expiration (exhalation) is led into three impingers, and the quantity of lead is measured. During the inhalation, the subject are requested not to swallow the solution remaining in the oral cavity. At the end of each inhalation, it is rinsed out (to be measured) with 100 ml of acetic acid solution with the concentration of 0.5%. Thus the actual amount of inhaled lead was to be got by drawing the amount of lead rinsed out each time. Table. Ⅲ shows the results of the inhalation experiment on seven subjects, whose aim to inhale 0.5 ml (546μg as lead) of 2 mg/ml lead acetate solution.

Table Ⅲ Resulting Data on Inhalation Experiment

Personal Subjects	Required Lead Amount for Inhalation (μg)	Lead Amount in Expiration (μg)	Remaining Lead Amount of in Oral Cavity (μg)	Real Amount of Inhaled Lead (μg)	Difference between Aimed Value (μg)	Difference Value Aimed Value (%)
H	486	17.5	3.8	464.7	− 81.3	− 14.9
N	606	33.0	9.7	563.3	+ 17.3	+ 3.2
K	738	14.5	18.7	704.8	+ 158.8	+ 29.1
A	552	18.0	20.7	513.3	− 32.7	− 6.0
I	654	21.5	9.7	622.8	+ 76.8	+ 14.1
M	769	19.6	60.0	689.4	+ 143.4	+ 26.3
B	562	18.4	43.4	500.2	− 45.8	− 8.4

Though it is varying widely due to different of lead in the oral cavity is below 4 % on the average. The problem is what percentage of the target amount (e.g., 546μg of lead in this experiment) can be inhaled with certainty.

The present experiment leaves something to be solved in the view point that the virtual inhalation rate considerably varies from subject to subject.

Lastly, the amount of the solution to be inhaled may be regurated by changing the concentration of the solution and the number of inhalations.

The NEBULIZER can be widely applied to every kind of liquid atomization.

6. CONCLUSION

To summarize the merits of our study, we can describe them as follows:-

(1) Device and making of the improved "Nebulizer" for inhalation, which prevents big particles from comming into being to be generate.

(2) Application of high power statical electric field to for successful mist collection.

(3) Adoption of peculier convex round plate as opposite electrode to avoid unbalance in density on each area of collection film.

*As above stated the flow amount of the compressor was set at 16～20 l/min. for the reason of its coincidance with the usual respiration of the subjects (i. e., the suction by equally velocity).

(4) Application of hydrophobic oil layer to preservation of shape and stability of collected mist particles.

(5) Applying to the human inhalation experiment, this equipment has been used with worthy results as we expected.

7. AFTERWORD

In recent years, a great many investigations have been made on techniques and principles of liquid atomization, in response to a growing demand for various atomizing devices in industry.

At the same time, urgent development of new methods for measuring fine particles are requested by a rapid advance of combustion techniques and the aggravation of air pollutions.

The present study has devised a new Neblizer and developed new techniques for collection and fixation of fine particles.

It is believed that they are new contributions to the theory and practice of liquid atomization.

A further study is being made on the principles and applications of the Nebulizer.

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