D. AN IMPROVED AEROSOL GENERATOR

The simplest and most commonly used technique for aerosolizing substances which are soluble in liquids, generally water, involves high velocity air-jet atomization. The resulting air borne droplets quickly dry in the ambient atmosphere leaving behind a crystallized or amorphous particulate whose size depends on the initial liquid droplet volume. Where constancy of aerosol characteristics, viz., air concentration, size, for moderately extended time periods (hours, days) is required, the method falls short in that the generator solution tends to become concentrated by the evaporative effect of the expanding jet air, the extent of concentration depending in part upon the initial volume of the solution being aerosolized. Saturation of the compressed jet air (generally 15-20 lbs/in^2) (1) merely lessens but does not obviate this effect. Compensating addition of liquid (2) is in use and an ingenious leak procedure has been suggested (3) to overcome this effect. For inhalation studies using very toxic materials, especially the radioisotopes, it is most important to use only small quantities of isotope for the aerosolization to ensure personnel safety with as little manipulation of the aerosolizer as possible during the procedure.

With these considerations in mind, a device has been developed which automatically and reliably compensates for changes in the concentration

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and volume of a solution during aerosolization with the production of air borne particulates of unchanging characteristics. It represents the redesign of a generator previously used (4) for short term inhalation exposures (1/2 to several hours) where variability in solution and thus aerosol concentration was not of great concern and the generated solutions could be replenished or replaced daily for repetitive exposures. It was, however, quite inadequate for our purposes since it required daily manipulation and was not reliable in terms of operation and aerosol constancy.

Aerosolizer Design

The redesigned aerosolizer (Fig. 1-4) consists of two lucite tubes set in a lucite base with a communicating port between them located so that solution drains from one (an air-tight reservoir tube) into the other (the generating tube) by air displacement when sufficient liquid volume has been used up by aerosolization to fall below the opening between them. The air jet passes through a #80 gauge opening drilled at a 15° angle upward from the horizontal in a plastic tube 5-6 mm in diameter (jet tube) placed in the generator tube so that the #80 gauge orifice is about 1/8" below the liquid level and the upper margin of the port which thus controls the ingress of more fluid when required.

Positioning lugs restrict movement of the air jet tube so that the distance to the impacting surface remains constant. The reservoir and generating tubes are practically isolated from each other by a set of baffles so designed and placed that air bubbles produced by the air jet cannot enter the reservoir chamber to interfere with the gravity flow of solution into the generating tube, a flow regulated as noted previously by the entrance of air into the reservoir tube when liquid level falls below the communicating port. Almost complete separation of the generating and reservoir tube is also achieved in terms of the aerosolizing solution. This is indicated by the very slight penetration into the reservoir of a dye indicator placed in the generating tube during an operational period. Whatever dye does enter may be attributable to an initial back-surge of fluid and slow diffusion.

The design and operation of the generator was based on the following considerations. Since the water content of air is dependent only on the temperature (pressure is negligibly involved) it should be possible to achieve a wide range of water content in compressed air by regulating the temperature at which saturation occurs during passage through a water-filled bubbler. The water-laden jet air pressure may be selected so that upon expansion to ambient pressure its water content is just sufficient to achieve saturation and aerosolization but not evaporation of the generating solution or condensation of air jet moisture will occur. It should be noted that the warmed jet air must not change the temperature of the solution through which it passes in order to maintain the necessary water content differential at the temperatures selected. Since real gases (but not ideal ones) tend to cool upon expansion, the possibility of temperature differential maintenance appeared to be feasible.

The cooling of expanding gases is described quantitatively by the Joule-Thompson equation and depends essentially upon the work done in overcoming intermolecular attraction. The degree of cooling is inversely proportional to the square of the compressed air temperature and directly to the pressure differential as shown in the equivalent forms below:

$$\Delta T = \frac{\left(\frac{2a}{RT} - b\right)}{C_p} \Delta P = \left(\frac{9}{128}\right) \frac{RT_c}{P_c} \left(\frac{18\, T_c^2 - 1}{T^2}\right) \Delta P$$

where ΔT and ΔP are Temperature and Pressure differentials

P_c, T_c = Critical Pressure and Temperature

R = Gas constant

T^2 = Absolute temperature

a, b = (Van der Waal's) constants

C_p = Heat capacity at constant pressure

In order to test the practical operation of the generator, an appropriate apparatus was devised to observe the cooling effect of jet air at various temperatures and pressures passing through the aerosolizer containing either air or water. These data are shown in Table I and indicate that the cooling effect of expanded water-saturated air depends upon the temperature of the compressed air stream (item D, columns 2 & 7), but not apparently or very importantly upon the airline pressures used (items A, B, C, columns 1 & 7) in either the dry or water-charged generator (column 7). In order to keep the temperature differential as low as possible to obviate generator solution heating, the lowest jet air pressure still efficient in aerosolization and compensable for the required water content should be used.

The partial pressure required in the humidified compressed air stream for compensation is indicated by the following relationship:

$$P^1_w = P_w \left(\frac{P_c}{P_a} \right)$$

where the ratio of compressed to ambient air pressures (P_c/P_a) is the volume factor of expansion, P_w is the partial pressure of water vapor at ambient temperatures, and P^1_w is partial pressure of the aerosolizing jet air. The temperature of humidification for this P^1_w is readily available from appropriate tables in standard reference books. The calculations which appear below illustrate the procedure.

Air at 20 lbs/in^2 will expand at ambient pressure to about 2.36 times its initial volume (34.7 / 14.7 lbs/in^2). The partial pressure of water vapor in the warmed expanding air approaching an assumed room temperature of 25°C (P_w = 23.75 mmHg) would have to be 56.0 mmHg (23.75 mmHg x 2.36) in order to achieve saturation after passing through the orifice. This P^1_w corresponds to a compressed air temperature of 41°C or 16.4°C above ambient. At 10 and 15 lbs/in^2, 9.1 and 13.5°C temperature differential would be required, respectively. If room temperature is assumed to be 20°C, then required temperature differentials at 20, 15, 13, and 10 lbs/in^2 are 14.6, 12, 11, and 8.6°C, respectively. The optimal conditions of aerosolization thus appear to be 13 to 15 lbs/in^2 at 20°C ambient temperature, where only slight changes in aerosol solution temperatures occur. Since aerosolizing efficiency at these pressures is good (3), a temperature differential of about 11-12°C above ambient room temperatures (20-25°C) appears to be optimal and feasible (Table I) for our purposes.

Aerosolizer Operation

The aerosol generator was charged with 5% Europium chloride solution (pH 2-3) tagged with radioactive $^{152-154}$Europium also in chloride form and operated in an environmental chamber developed especially for radioisotope inhalation studies and previously described (5). Air jet pressure was 13 lbs/in^2 and humidifying water bubbler maintained at 24 to 27°C above ambient to achieve an effective temperature differential of about 12°C. The generator was operated for 6 days, 5 hours per day. The aerosol activity concentration was determined by well-type crystal scintillation counting (NaI-thallium activated) of air samples drawn through 1" Gelman (#GA) membrane filters soon after equilibration (15 minutes) and hourly thereafter during each run. After each run the generator inlet and outlet tubes were clamped off ready for the next day's operation without further manipulation of any kind. The results are shown in

Figure 5. The solid lines indicate changes in aerosol concentration using the very first determination as the standard (100%). The time averaged level of activity indicated by the dashed line (Fig. 5) was 108% $\pm$ 8.9% (1. S. D.). While maximal variations from the average concentration were of the order of 20%, the trend appeared to be quite stable and level.

It is important to note that these values also include errors in sampling volumes and activity, as well as variations in chamber parameters (air flow, leakage, etc.).

A few preliminary tests using air pressures of 20 lbs/in^2 and humidification by passage through a water bubbler at room temperature indicated an increasing concentration of about 30% in the generator solution while rises of 40-60% were observed using non-humidified jet air.

Size Distribution Spectra

Aerosols of several of the rare earths generated by the modified aerosolizer have been used to investigate their deposition-retention characteristics in the animal lung. The size distribution of the air borne particulates was of major interest and these data were obtained in each case. The aerosol was sampled using the point-to-plane electrostatic precipitator (6) and sizing done with a Zeiss Particle Size Analyzer (TG-3) from 8 x 10 inch electron micrograph prints.

The size of the dry air borne particulates depends, in large part, upon the concentration of material in the aerosolized liquid droplet, other conditions being the same, viz., the baffling in the system, air pressure of dispersion, etc. Most of the solutions used were adjusted in molarity equivalent to that of a 5% (w/v) europium chloride solution in an attempt to obtain comparable size spectra and doses of the stable rare earths. The listing below is presented to indicate the submicronic sizes and near equivalence of all the rare earths tested in spite of differences in other physical properties.

	Mg (μ)	S. D. g
Europium chloride (1% NaCl vector)	0.075	1.67
Scandium chloride	0.130	1.70
Yttrium chloride	0.122	1.84
Cerium chloride	0.133	1.65
Europium chloride	0.145	1.83
Ytterbium chloride	0.102	2.00

Discussion

The aerosolizer described herein has been in use continually for the past several years and found to be reliable in operation and output. It should prove to be particularly valuable for long term or repetitive use where the stability of aerosol characteristics such as concentration and partial size spectrum is required. It has the advantage that the requirements for manipulation in terms of solution replenishment, etc., are minimal; the reservoir volume may be as large as experimental needs require or as small as considerations of the hazards of materials under study dictate simply by altering the relative diameters of reservoir and generating tubes but maintaining height differential. Since only 3.0 ml of solution is required in the generating tube, the solution concentration is very sensitive to air jet evaporative effects if no provision for these are made. The long term maintenance of aerosol characteristics where large volumes (ca 30-50 ml) of solution can be used as in the Lauterbach generator (1) should present little difficulty.

REFERENCES

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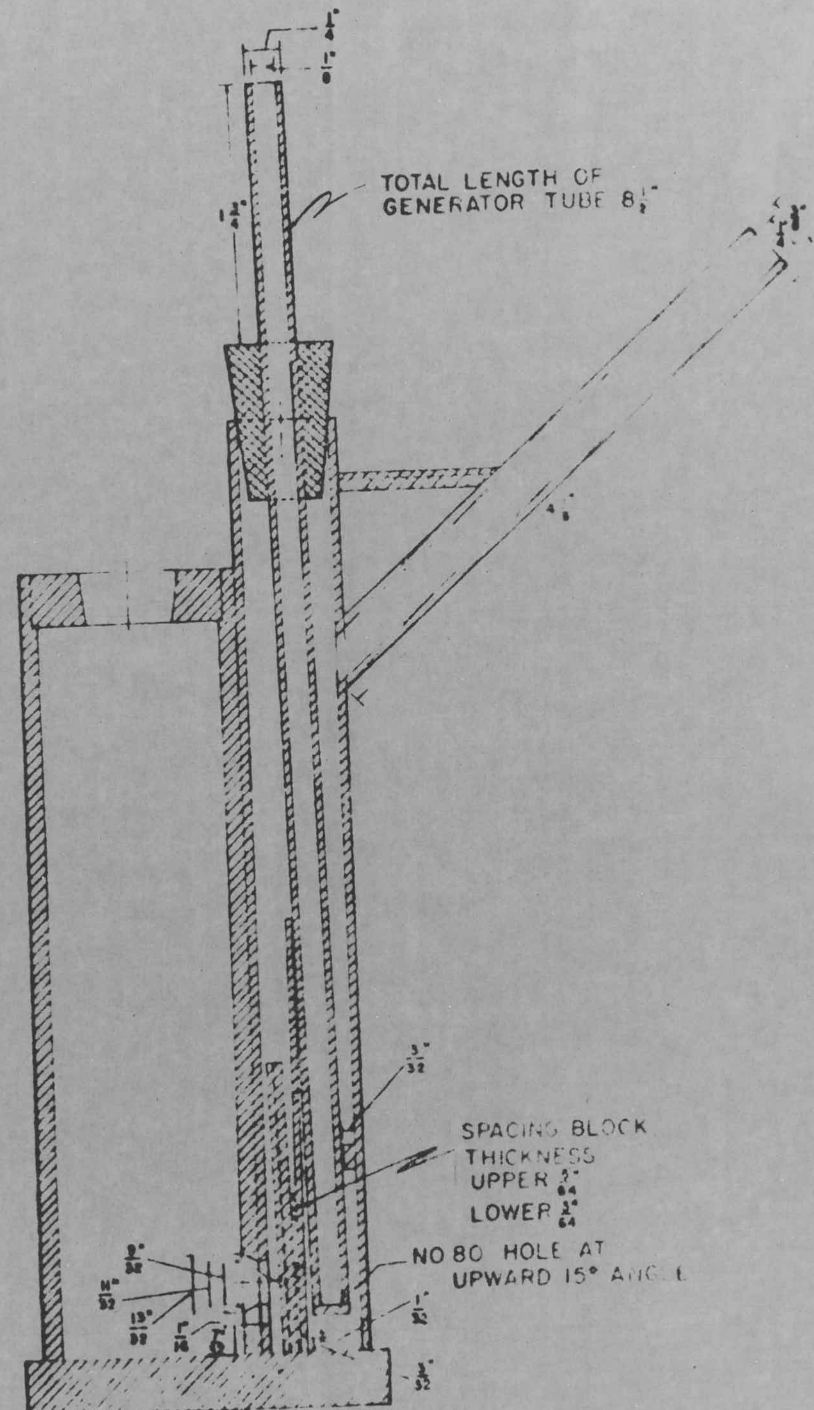
TABLE I

Cooling Effect of Air Expanding Through a #80 Gauge Orifice

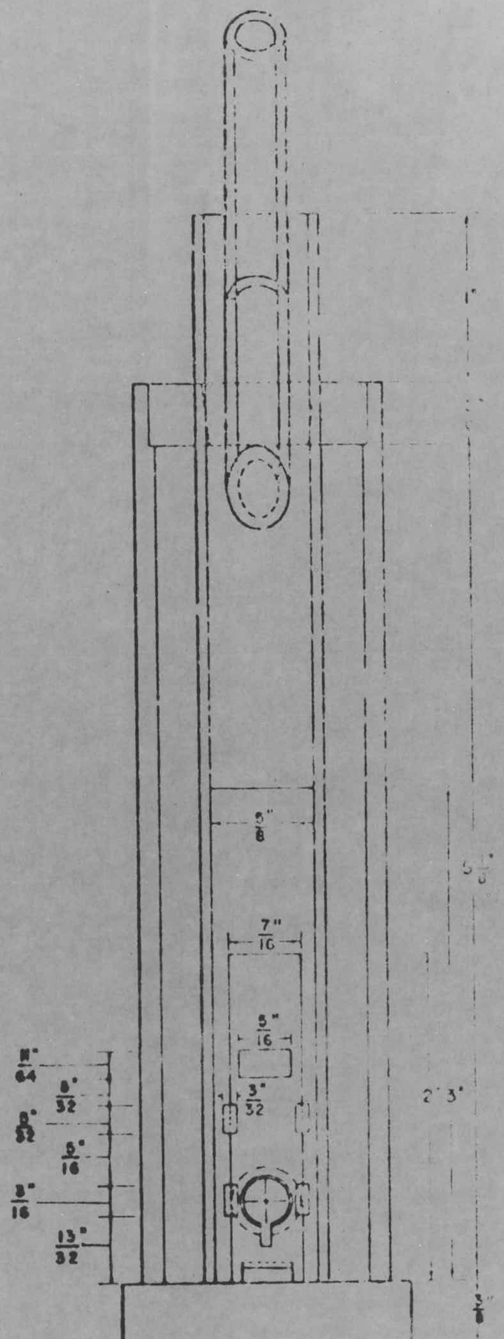
		1	2	3	4	5	6	7
Aerosolizer		Air Jet Pressure lbs/in^2	H_2O Bath Temperature °C	Room Temperature °C	Temperature Difference °C	Compressed Air Line Temperature °C	Expanded Air Jet Temp. (2-5) °C	Cooling Effect (5-6) °C
	A	10	45	25	20	36	27	9
Dry	B	15	45	25	20	35.5	27	8.5
	C	20	45	25	20	36	27	9
	D	20	39	26	13	33	27	6
	E	10	45	25	20	35	25	10
	F	15	44	25	19	36	24	12*
Water	G	15	45	25	20	35.5	24.5	11*
Charged	H	15	45	25	20	35.5	24	11.5
	I	20	45	25	20	35	24	11

FIGURE 1: DETAILS OF AEROSOL GENERATOR: LONGITUDINAL SECTION.



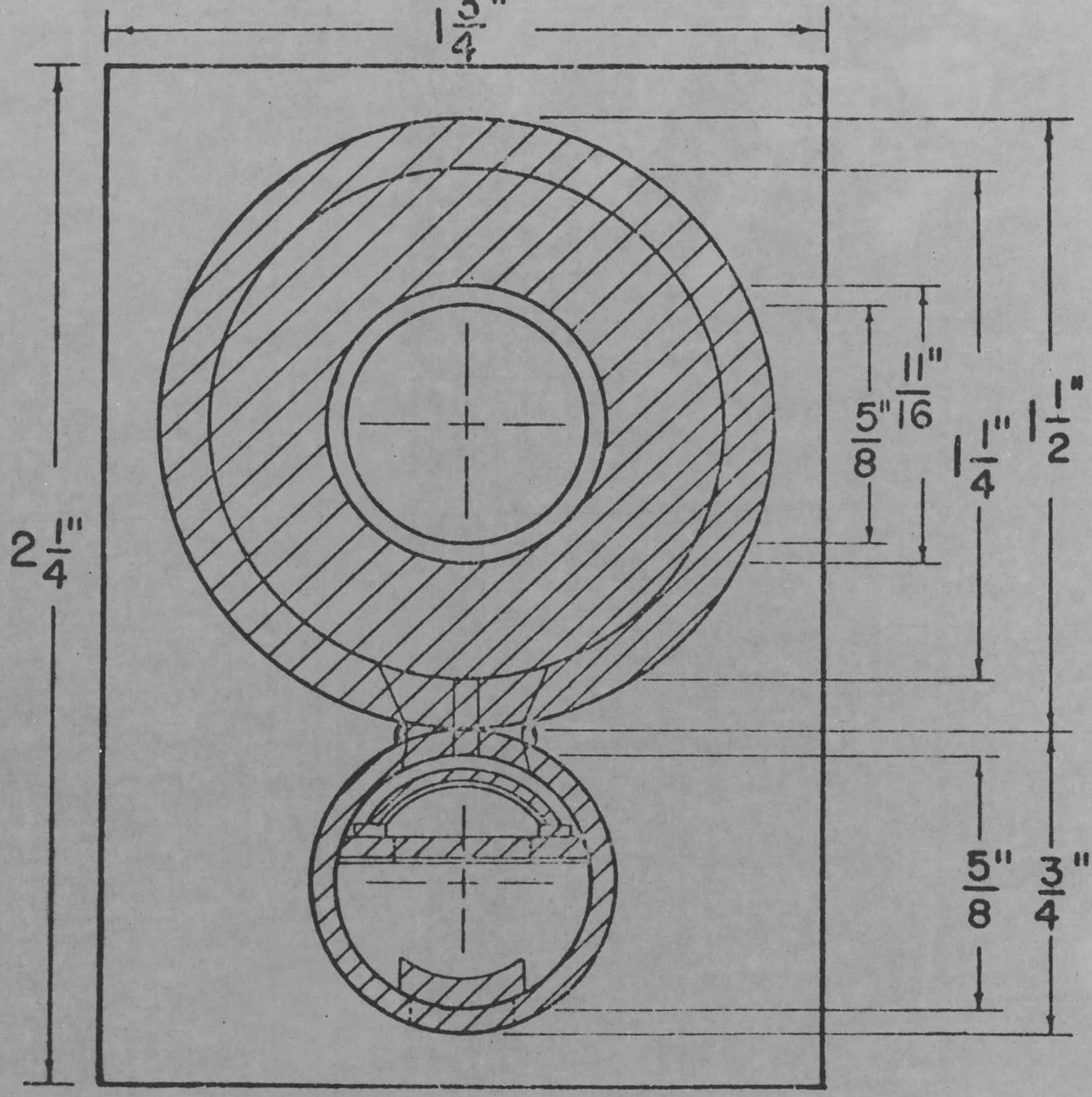
LONGITUDINAL SECTION

FIGURE 2: FRONTAL VIEW OF AEROSOL GENERATOR.



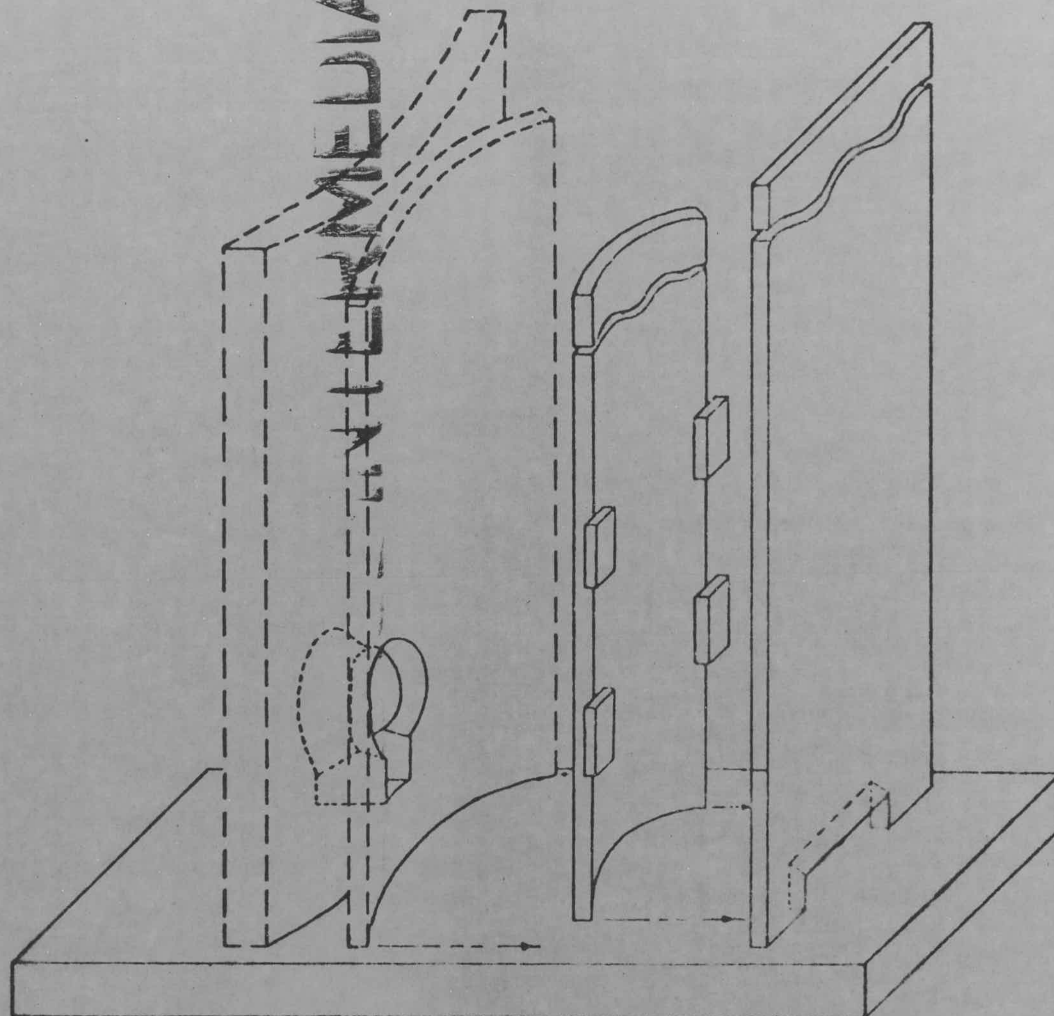
FRONTAL VIEW

FIGURE 3: CROSS SECTIONAL VIEW AT THE LEVEL OF COMMUNICATING RESERVOIR OPENING OF AEROSOL GENERATOR.



CROSS SECTION

FIGURE 4: DETAILS OF BAFFLING SYSTEM TO INSURE REGULAR FLOW OF RESERVOIR FLUID AS NEEDED FOR AEROSOLIZATION.



GENERATION AREA-
EXPANDED VIEW

FIGURE 5. AIR CONCENTRATION OF EUROPIUM CHLORIDE AEROSOLIZED AT 13 lbs/in^2 AND TEMPERATURE DIFFERENTIAL OF 24-27.5°C.