

ENGINEERING RESEARCH INSTITUTE

THE MEASUREMENT OF DIFFUSIVE SEPARATION IN THE UPPER ATMOSPHERE

UNIVERSITY OF MICHIGAN - SIGNAL CORPS

UPPER ATMOSPHERE RESEARCH PROJECT

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ON

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THE MEASUREMENT OF DIFFUSIVE SEPARATION IN THE UPPER ATMOSPHERE

University of Michigan - Signal Corps
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Abstract

The experimental technique of collecting samples of the upper atmosphere in steel bottles carried aloft in rockets is described. The samples collected were analyzed primarily for helium, neon, argon, and nitrogen. The presence or lack of diffusive separation is based on the ratio of the relative abundance of the first three with respect to nitrogen in the samples to the relative abundance in ground air. The samples fall into three categories: a) Those collected in early V-2's up to 70 km. No separation was detected but the samples are in doubt because of possible contamination by ground air. b) Samples collected up to 60 km in Aerobees. These are thought to be valid samples, and the analyses show no separation up to 60 km. c) The most recent samples collected in Aerobees and one V-2 up to 100 km. In these samples separation increasing with the mass difference and with altitude was detected. However, small amounts of separation detected in ground air control bottles associated with these latter samples throw some doubt on the results. Further work is in progress.

* Members of the research group in the Department of Aeronautical Engineering which carried out this work are: F. Bartman, L. Chaney, T. Collins, H. Courtney, D. Hagelbarger, H. Hansen, L. Jones, J. King, R. Leite, V. Liu, L. Loh, R. Mills, H. Monroe, H. Neill, M. Nichols, W. Rutledge, E. Schaefer, S. Sturgeon, P. Titus, E. Turner, N. Wenk, and E. Wenzel. The research was sponsored by the Meteorological Branch of the Army Signal Corps through contracts with the Engineering Research Institute of the University of Michigan.

1. Introduction.

The Meteorological Branch of the U. S. Signal Corps has, since 1946, sponsored a program of high altitude measurements at the University of Michigan. Part of the program is sampling of the atmosphere up to 100 km. As it has worked out, the analysis of the samples obtained has been carried out almost entirely by Professor F. A. Paneth and co-workers at the University of Durham. Several of the early samples were also analyzed on similar equipment at the University of Michigan, the results confirming those of the Durham group. Analytical results of samples taken prior to May 1952 have been published by the Durham and Michigan groups. The objective of the sampling program is to assess the extent of mixing of the earth's atmosphere by measurement of the relative concentrations of the light and heavy constituents up to as high levels as possible. The procedure has been to take samples at various levels by means of rockets and subsequently to analyze these samples for helium, neon, argon, and nitrogen. All samples have been taken above White Sands, New Mexico, U. S. A. The purpose of this paper is to describe the apparatus and procedure used to obtain the samples and to summarize the results obtained to date.

2. Preparation of the Sample Bottles.

As described in section 4, early samples were recovered from the wreckage after the flight. The bottles from the early Aerobee flights were also recovered in this way, but later, parachutes were used. Thus it was necessary to use metal sample bottles. For this purpose walk-around breathing oxygen bottles (U. S. Air Force type B-2 spec. 94-40355, volume 500 cu. in.) were found satisfactory. These bottles were fabricated by welding sheet steel. The sample intake tube mentioned in section 3 was hard soldered to one end

of the bottle, and the sample extraction tube mentioned in section 4 and the evacuation tube were hard soldered to the other end. In order to measure the pressure in the evacuated bottle, a hot-wire gage consisting of a 0.003-in. diameter platinum wire 2 in. long was mounted at the extraction end. The sensitive range of this gage was 0.005 mm to 0.5 mm Hg. Fig. 1 is a schematic of an assembled sample bottle of the type used prior to December 1950. The bottles were tested for leaks by a General Electric helium leak detector (Catalog No. 6933950G3). The bottles were then sealed from the system by squeezing shut the copper evacuation tube which had been previously coated on the inside with solder. The squeezed place was then heated to fuse the solder. After sealing off, the bottles were aged for several weeks, and the pressure monitored. If the pressure rose appreciably, the bottle was not flown.

Since December 1950, a larger cross section intake tube has been used. This consisted of a straight section of copper tubing 1 in. in diameter and 10 in. long. This appears schematically in Fig. 3. The evacuation tube was incorporated in the cap of this tube as shown. Since 1 May 1952, a Phillips type ionization gage has been incorporated at the cap end of the intake tube. When the bottle is opened (as described in the next section) the Phillips gage is thrown off. The purpose of the Phillips gage is to provide a more sensitive pressure measuring device in order to detect leaks after seal-off of the evacuated bottle from the pumps.

3. Opening and Sealing of the Sample Bottles.

A detailed description of the method used to open and seal the sample bottles while still in flight has been published elsewhere.⁽¹⁾ Therefore only a brief description will be given here.

Fig. 1 shows a thin diaphragm of copper hard-soldered to the end of the sample intake tube. To open the bottle to allow the sample to flow in, a knife is driven through the thin copper diaphragm by means of a small charge of gun cotton in sylvan bellows made of brass. In later flights using the cold-weld sealer, a motor-driven knife shown in Fig. 2 was used. However, because of power requirements, etc., this was abandoned in favor of a piston type opener shown in Fig. 3. When the charge of black powder is fired, the cap is ruptured at the scored place F and thrown off. Since 1 May 1952, the cap has included the Phillips gage.

In flights prior to December 1950 the intake tube, shown in Fig. 1, was coated on the inside with a thin film of solder. By pyrotechnic means, this tube was squeezed shut and heat applied to fuse the solder. Fig. 4 is a schematic of the pyrotechnic sealer. This was developed for the sampling program by the Picatinny Arsenal. When the electric squib D is fired, the charge of black powder E drives the hammer J against the anvil H, squeezing the tube B shut. The black powder also ignites the fuse F, which in turn ignites the heating powder K, which melts the solder coating on the inside of the tube B thereby sealing it shut. In order to permit a larger intake area and to eliminate the heat required to fuse the solder, a cold-weld process was developed using gun powder to squeeze a 1-in. diameter copper tube shut effecting a cold weld. This process has been used in all rockets since December 1950. The cold-weld assembly used is shown in Fig. 5. The black powder in the capsule A is ignited by the electric squib. The gas generated in the cylinder B forces the jaws C and E closed, which pinches off and cold welds the tube D to form a seal. Recently a 2-in. tube has been successfully sealed, but this larger size has not as yet been flown.

The cold-welding technique using a small hydraulic press is also currently used in the laboratory to seal off the bottles after evacuation. A total force of 10,000 lbs. is required to seal a 1-in. tube.

4. Samples Prior to 1 December 1948

Prior to December 1948, all the sampling was carried out with the V-2 rocket. The first flight of sample bottles on a V-2 rocket took place on 29 July 1947. Since the V-2 is a large rocket (45 ft. long, weighing 26,000 lbs. loaded) an entire flight was never devoted to sampling alone. Rather, sampling bottles were installed in unused space such as in the instrument compartment (approximately 6 ft. from the tip of the rocket), the mid section, between the fuel tanks, or in the forward part of the motor section. Fig. 6 shows a typical mid-section installation with the forward fuel tank in place. The space between the fuel tanks was most frequently used because the recovery chances seemed to be greatest in this location. Because of the relatively long ducting required, it seemed necessary to continually flush the intake tube. This was done by conducting air from scoops on the side of the rocket through rubber tubing into the chamber on the right-hand end of the bottle in Fig. 1. The exhaust from this chamber was connected by a rubber hose to a reversed scoop on the skin of the rocket so that air would circulate through the intake ducting throughout the flight of the rocket.

The intake scoop for this group of samples was a 3/4-in. OD tube protruding 5 in. from the surface of the rocket. A slot 3/8 in. wide cut in the outer 2 in. of the tube constituted the intake orifice. The exit port was a 3/4 in. diameter hole in the rocket skin covered with a fairing opening to the rear. Because of the proximity of the orifices to the surface of the rocket and their location behind possible leaks in the rocket skin,

there is the real possibility of contamination by rocket-borne ground air. After the sample bottles were recovered from the wreckage, they were returned to the laboratory and pumped into soft glass storage vials. Next, the bottles were leak checked by noting the rate of rise of pressure over several days. The storage vials were later attached to the analyzers at Durham and Michigan.

The results of samples taken prior to December 1948 are summarized in Table I. The results given in Table I indicate no appreciable change in the relative amounts of the four constituents involved as compared to surface air. However, these results must be considered in the light of possible contamination due to ground air leaking from the rocket during flight. In order to eliminate this possibility the next samples were taken with Aerobee rockets in which the sample bottles were installed in the nose cone. The nose cones were completely sealed so that no air could escape from the rockets ahead of the intake scoops.

5. Samples Taken in the Period 1 December 1948 to 1 December 1950

The samples taken during this period used the Aerobee rocket with essentially the same procedure as with the V-2 rockets except that the entire apparatus was contained in the Aerobee nose cone, which was sealed against air leakage ahead of the air intake and exit scoops. Since the rocket was always traveling at supersonic speeds during the sampling, it appeared that no contamination from surface air trapped in the rocket was possible provided the rocket was not yawed by angles approaching 90° . Figs. 7 and 8 are photographs of a typical installation of this period. Fig. 9 is a photograph of the recovery of Aerobee instrumentation SC-3. This shows intake and exit scoops, the sealed joint in the nose cone, and the parachute used for recovery. Three rockets used the small scoops of Fig. 8 and two the larger ones of Fig. 9. No appreciable difference in sampling efficiency was noted.

TABLE I

Sample Bottle	Glass Vial	Rocket Number	Altitude km MSL	Date of Sampling	Analysis		Calc. Pitot Pressure mm Hg	Bottle Pressure mm Hg	Ratio	
					He	Ne				
32	1B	V-2 27	61.1-72.0	10-9-1947	0.93	1.00	1.01	0.81	0.65	0.81
32	1A	V-2 27	61.1-72.0	10-9-1947	1.00 ^{m*}	1.05 ^{m*}		0.81	0.65	0.81
45	12A	V-2 44	60.8-69.5	11-18-1948	1.04 ^{m*}	1.07 ^{m*}		1.11	0.75	0.68
35	2B	V-2 35	55.4-65.5	5-27-1948	1.01	1.00	1.00	1.76	0.75	0.43
35	2A	V-2 35	55.4-65.5	5-27-1948	0.97 ^{m*}	0.93 ^{m*}		1.76	0.75	0.43
37	3B	V-2 35	55.4-65.5	5-27-1948	0.98	0.99	1.00	1.76	0.80	0.45
37	3A	V-2 35	55.4-65.5	5-27-1948	0.98 ^{m*}	0.98 ^{m*}		1.76	0.80	0.45
42 ^b	5A	V-2 40	49.0-59.8	7-26-1948	1.05 ^{m*}	1.06 ^{m*}		1.81	1.18	0.65

a) The arrangement is in order of decreasing altitude of sampling. The helium, neon, argon contents are expressed as: (volume of gas, relative to nitrogen, in sample)/(volume of gas, relative to nitrogen in ground level air). The ratios marked with a superscript ^{m*} were obtained by the authors before the apparatus and the techniques were improved to the present state. The results with no superscripts were obtained by Paneth et al.⁽²⁾ The vial numbers refer to subdivisions of the same sample.

The analyses reported in this table were obtained by the method of selective adsorption on activated charcoal and has been described in a series of papers published by Paneth and co-workers.⁽³⁾

b) See footnote a in Table II.

c) O₂ is usually removed completely from the samples, presumably by oxidation of the copper tube surfaces.

Table II is a summary of results from this period of firings. On SC-3, radioactive CO₂ was released in the nose cone just prior to sampling in order to check on the possible contamination of samples due to air trapped in the nose cone. Analysis of a portion of the sample from bottle 77 showed that contamination due to rocket-trapped air was completely negligible, and on all subsequent flights in which this technique functioned properly the same result was obtained. It is believed that the results of Table II are representative of the atmosphere up to about 60 km. Thus no diffusive separation occurred below 60 km over White Sands, New Mexico, at the time of these firings. This seems to be in agreement with wind data⁽⁴⁾ and the long separation time required at these altitudes.^(5,6)

6. Samples Taken Since 1 December 1950

In order to extend the sampling height with the Aerobee rockets, a new design was made to eliminate the drag of the external scoops and to provide larger intake areas with shorter ducts. This design features a nose cone which is ejected prior to sampling by means of a small rocket (jato), cold-weld seals, and straight 1-in. diameter intake tubes 10 in. long. After the nose cone is removed, the intake tubes are the most forward portion of the rocket and protrude through a pressure-tight bulkhead. Fig. 10 is a schematic of this type of installation. In this figure the knife opener rather than the more recent piston opener is shown. In order to avoid contamination from a solution of gas in the glass vials, the entire metal sample bottle in each case was shipped to the Durham laboratory for analysis. Two installations of the type of Fig. 10 were also installed on V-2 59, each having its own parachute. Also on this rocket was a third installation containing one larger sample bottle, volume 2100 cu. in., which was opened and closed in the 100-km region.

TABLE II

Sample Bottle	Glass Vial	Rocket Number	Altitude km MSL	Date of Sampling	Analysis		Calc. Pitot Pressure mm Hg (closing)	Bottle Pressure mm Hg	P_b/P_p
					He	Ne			
63 ^a	19D	SC-4	54.7-58.3	7-21-1949	1.01	1.45 ^b	1.22	0.19	.16
63 ^a	19A	SC-4	54.7-58.3	7-21-1949	1.01m*	1.07m*	1.22	0.19	.16
61 ^a	15B	SC-2	53.6-57.7	6- 2-1949	1.00	1.00	1.60	1.00	.63
61 ^a	15A	SC-2	53.6-57.7	6- 2-1949	1.00m*	1.02m*	1.60	1.00	.63
77	25D	SC-3	50.4-53.3	12- 6-1949	0.98	1.00	1.32	0.80	.60
60	16A	SC-2	49.6-53.6	6- 2-1949	0.96	0.98	3.14	3.05	.97
68	21A	SC-5	42.0-45.0	9-20-1949	1.01m*	1.01m*	4.75	3.60	.76
74	28A	SC-7	41.4-44.9	12- 6-1949	1.00m*	1.02m*	5.22	3.40	.65

a) The results for these samples are not confirmed by McQueen's measurements on the isotopes of nitrogen. (1,8) Subsequent measurements on the same isotopes from sample bottles 61 and 42 by both the analytical and Nier isotope ratio instruments at the Consolidated Engineering Corporation of Pasadena, California, are in disagreement with McQueen and, within the currently estimated experimental error, show no change from surface air.

b) Unreliable because of contamination by H₂, confirmed by mass spectrometry.

Table III gives the results obtained since 1 December 1950. These samples show, for the first time, an increase in the lighter gases over the heavier gases beginning with samples taken at altitudes greater than 60 km. In order to check on such things as selective adsorption on the wall of the sample bottle, four control bottles were prepared by admitting surface air into evacuated sample bottles prepared in exactly the same way as those flown on the rockets. The results of these four control bottles are given in Table IV.

Bottles B-18-P, B-20-P, and B-21-P were prepared by breaking a small glass vial of surface air in the sample bottle after the bottle had been evacuated, baked, and leak tested in the usual way. Bottle B-17-P was prepared by sealing the bottle, opener, and bottle intake tube in an evacuated chamber, the bottle having been previously prepared in the usual way. This is illustrated in Fig. 11, which shows the intake tube and opener mounted in the base of the chamber. When the bottle opener was fired, it not only opened the bottle but also broke a small glass vial of surface air, which expanded into the chamber and through the intake tube into the sample bottle, thus simulating, with the exception of the rocket velocity, the actual sampling procedure. The results given in Table IV are not explained as yet, particularly the fact that two bottles show no deviation from surface air, and two bottles show an appreciable change. The hypothesis of selective adsorption on the walls of the bottle is being investigated. It is conceivable that the sample of four bottles in Table IV includes two which have surface conditions for which the adsorption is reversible⁽⁹⁾ and two for which this does not hold. It is hoped that the studies under way will shortly answer these questions.

TABLE III

Sample Bottle	Rocket Number	Date of Sampling	Peak Altitude km MSL	Open km	Close km	Mean km	km Open	Total Velocity km/sec	Velocity Sound km/sec	Close			Angle of Attack	Ambient Pressure mm Hg	Pitot Pressure mm Hg	Bottle Pressure mm Hg	P _b /P	Analysis (Durham)		
										Mach No.	Rayleigh Factor	Angle of Attack						He	Ne	A
C6BL	V-2 59	5-20-52	124	102.7	105.8	104.3	3.1	.62	.32	1.9	5.4	38°	22°	.00020	.0011					
C3	V-2 59	5-20-52	124	90.8	95.7	93.3	4.9	.77	.30	2.5	8.8	16°	23°	.00083	.0073	.00034	.047			0.82
C1	V-2 59	5-20-52	124	85.9	90.8	88.4	4.9	.83	.30	2.8	10.6	23°	19°	.0017	.018	.0016	.089	2.943	1.395	0.82
C5	V-2 59	5-20-52	124	66.1	73.5	69.8	7.4	1.01	.29	3.5	16.0	6°	14°	.027	.43	.021	.049	1.570	1.232	0.90
B9	SC-17	12-19-50	82.0	69.5	71.6	70.6	2.1	.52	.29	1.8	4.6			.036	.165	.021*	.145*	2.41*	1.20*	0.85*
B8	SC-17	12-19-50	82.0	67.1	69.5	68.3	2.4	.55	.30	1.8	4.9			.050	.25	.022	.088	2.02	1.18	0.89
B6	SC-17	12-19-50	82.0	64.0	67.1	65.6	3.1	.60	.30	2.0	5.6			.070	.39	.043	.11	1.44	1.08	0.93
C11B	V-2 59	5-20-52	124	58.5	66.1	62.3	7.6	1.07	.31	3.5	14.2	14°	9°	.083	1.18					
B15	SC-21	9-26-51	68.9	58.7	60.7	59.7	2.0	.43	.32	1.4	2.9	68°	41°	.17	.49	.420	.86	1.035	1.008	0.996
B13	SC-21	9-26-51	68.9	56.1	58.7	57.4	2.6	.48	.32	1.5	3.4	41°	20°	.23	.78	.70	.90	0.998	1.005	1.001

* Based on correction for leak.

The data in columns 16, 17, and 18 of Table III show the calculated pitot pressure at the time of closing, the bottle pressure measured at the time of analysis, and the ratio of the bottle pressure to pitot pressure, respectively. In all cases except B-13 and B-15, only a small fraction of pitot pressure was obtained in the sample bottles. In this connection it is important to point out that with the exception of B-13 and B-15, the exact conditions of sampling are open to question. Namely, bottle B-9 opener, which was of the type of Fig. 2, cut only a small hole instead of completely opening the bottle. There is some doubt about bottles B-6 and B-8 also on this flight. In the case of V-2 59 (all bottles with prefix C) there is question as to whether or not the nose cone was ejected prior to sampling.

TABLE IV

Control Bottle	Date	Bottle Pressure mm Hg (Michigan)	Bottle Pressure mm Hg (Durham)	Amount of Sample cc NTP (Durham)	Analysis (Durham)		
					He	Ne	A
B-17-P	10-15-52	.0185	.0189	0.151	0.994	1.00	1.00
B-18-P	9-15-52	.0165	.0165	0.134	0.997	1.003	0.99
B-20-P	12-29-52	.0202	.0150	0.12	1.25	1.03	0.92
B-21-P	12-30-52	.0174	.0162	0.13	1.36	1.08	0.92

7. Discussion

At this point, the writers believe that the samples in Table II are valid and represent the air over White Sands, New Mexico. Thus at the time of sampling, no separation is indicated under about 60 km. Since the samples in Table I may have been contaminated by surface air trapped in the rocket, the authors feel that the results in this table, particularly those above 60 km, are open to question.

The results in Table III are definitely open to question because of the results in Table IV on the control bottles and possibly because no control experiments have been performed on the process of bottle filling at supersonic rocket speeds - i.e., no experiments designed to ascertain whether the bottle filling process at supersonic speeds is selective on the basis of molecular weight have been carried out. In this connection, bottle B-17-P of Table IV, which was filled under "static" conditions through the intake tube, showed no separation.

Fig. 12 is a plot of the data of Table III on a semilog scale. If, even in the face of the above uncertainties, it is assumed that these results are actually characteristic of the atmosphere and if complete diffusive equilibrium existed, then the results shown should plot a straight line. However, since the "settling times" are greater for the heavier elements such as argon, it would be surprising if all constituents were in the same state of approach to equilibrium. The straight lines in Fig. 12 were plotted for a temperature of 427° K. According to recent measurements,⁽¹⁰⁾ the 60-80-km region over White Sands is one of decreasing temperature with increasing altitude. It would seem then that this region would be characterized by considerable turbulence and mixing rather than by the long quiescence necessary at this altitude for the amount of separation shown in Table III.

References

- 1) Loh, Neill, Nichols, and Wenzel, Rev. of Sci. Instr., 23 (1952), p. 339.
- 2) Chackett, Paneth, and Wilson, J. Atmos. and Terres. Phys., 1 (1950), p. 49.

See also
Chackett, Paneth, Reasbeck, and Wiborg, Nature, 168 (1951), p. 358.
Jones, Loh, Neill, Nichols, and Wenzel, Phys. Rev., 84 (1951), p. 846.
Paneth, F. A., J. Chem. Soc., Sept. 1952, p. 3651.
- 3) Paneth, F. A., Endeavor, XII (1953), p. 5.
- 4) Kellog and Schilling, J. of Meteor., 8 (1951), p. 222.
- 5) Epstein, P. S., Beitr. Geophys., 35 (1932), p. 153.
- 6) Lettau, H., Compendium of Meteor., Amer. Meteor. Soc., 1951, p. 320.
- 7) McQueen, J. H., Phys. Rev., 80 (1950), p. 100.
- 8) Hagelbarger, Loh, Neill, Nichols, and Wenzel, Phys. Rev., 82 (1951), p. 107.
- 9) Armbruster and Austin, J. Amer. Chem. Soc., 66 (1944), p. 159.
- 10) The Rocket Panel, Phys. Rev., 88 (1952), p. 1027.

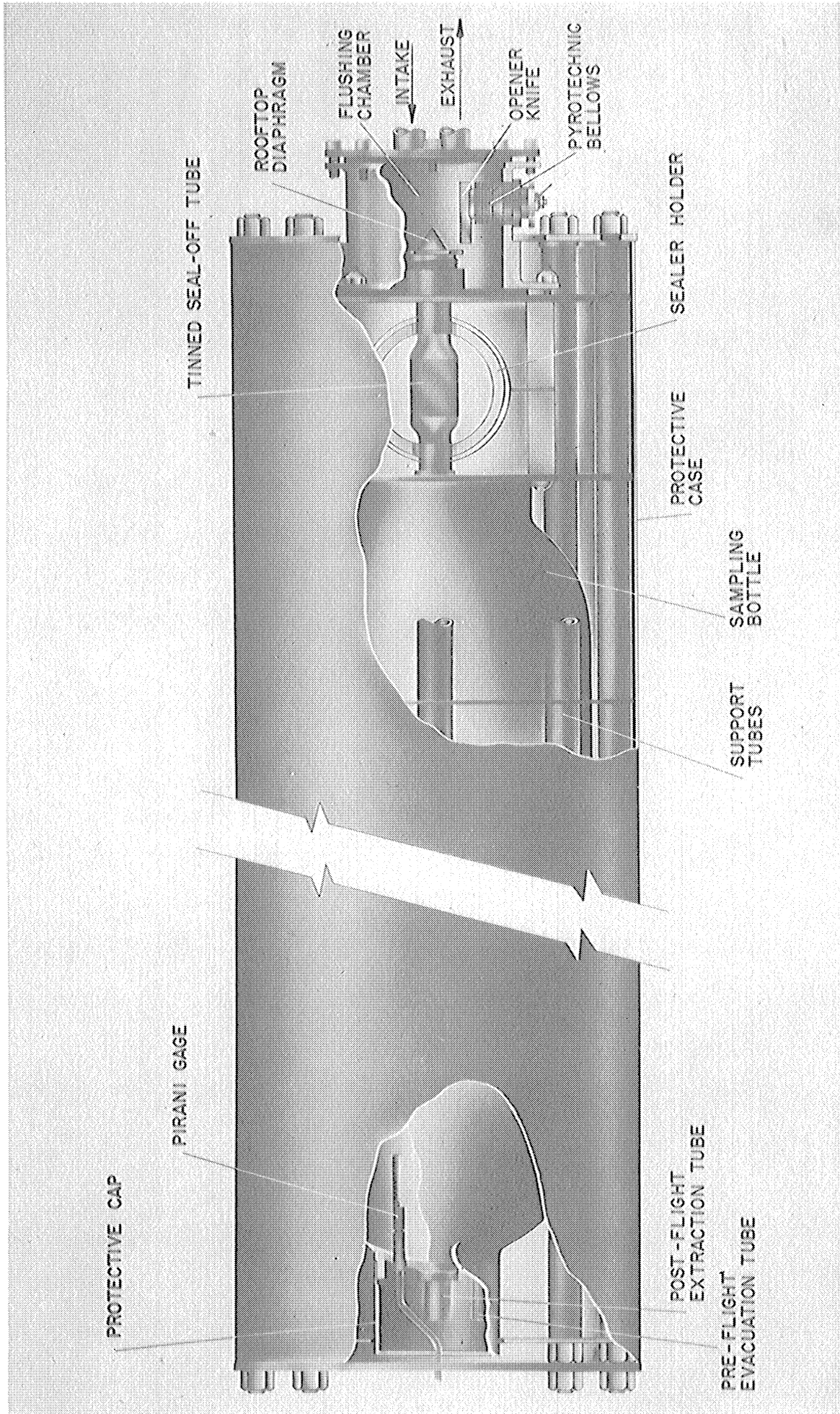


Fig. 1

Sample Bottle Used on Early V-2 and Aerobee Flights

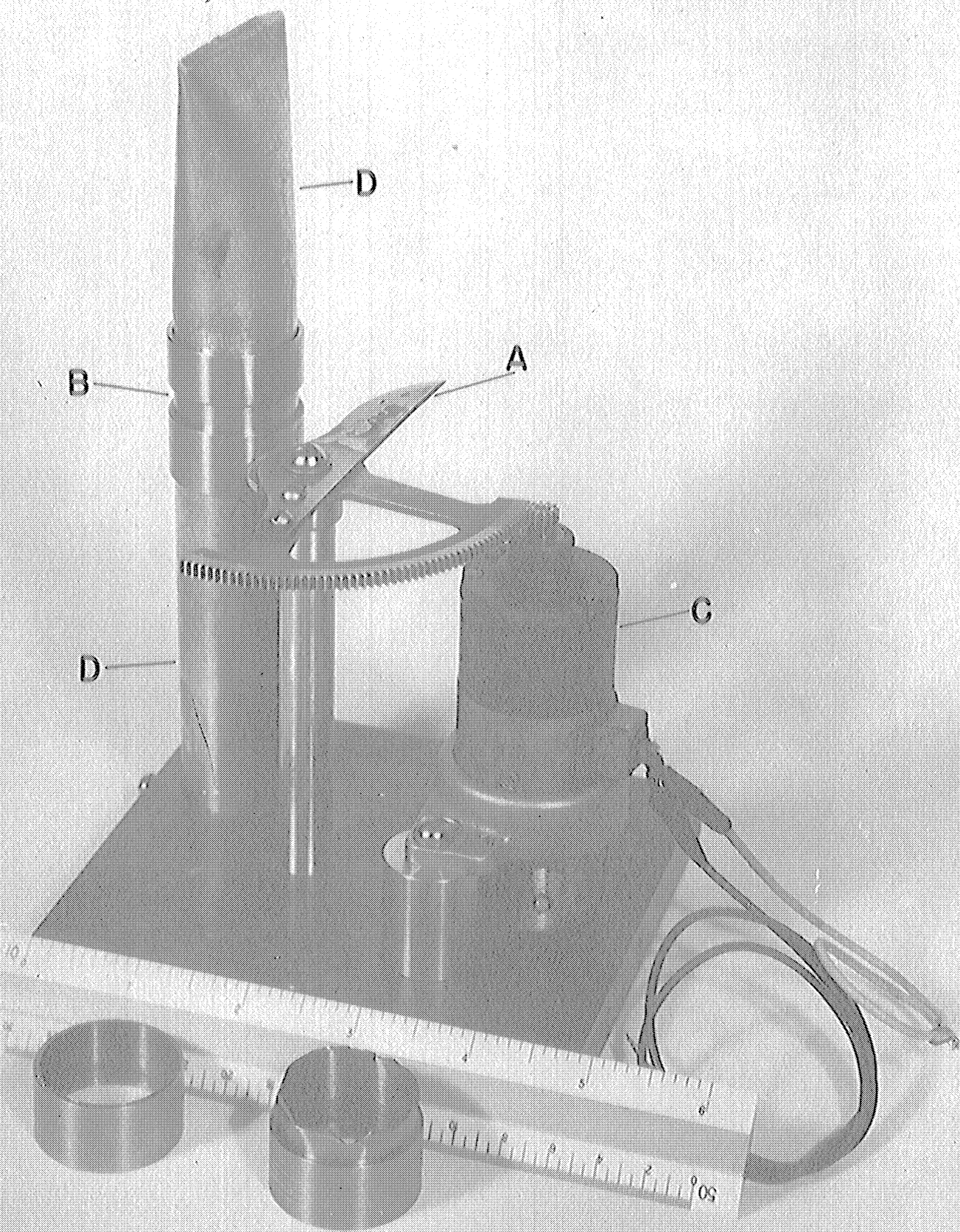


Fig. 2
Motor-Driven Knife Opener

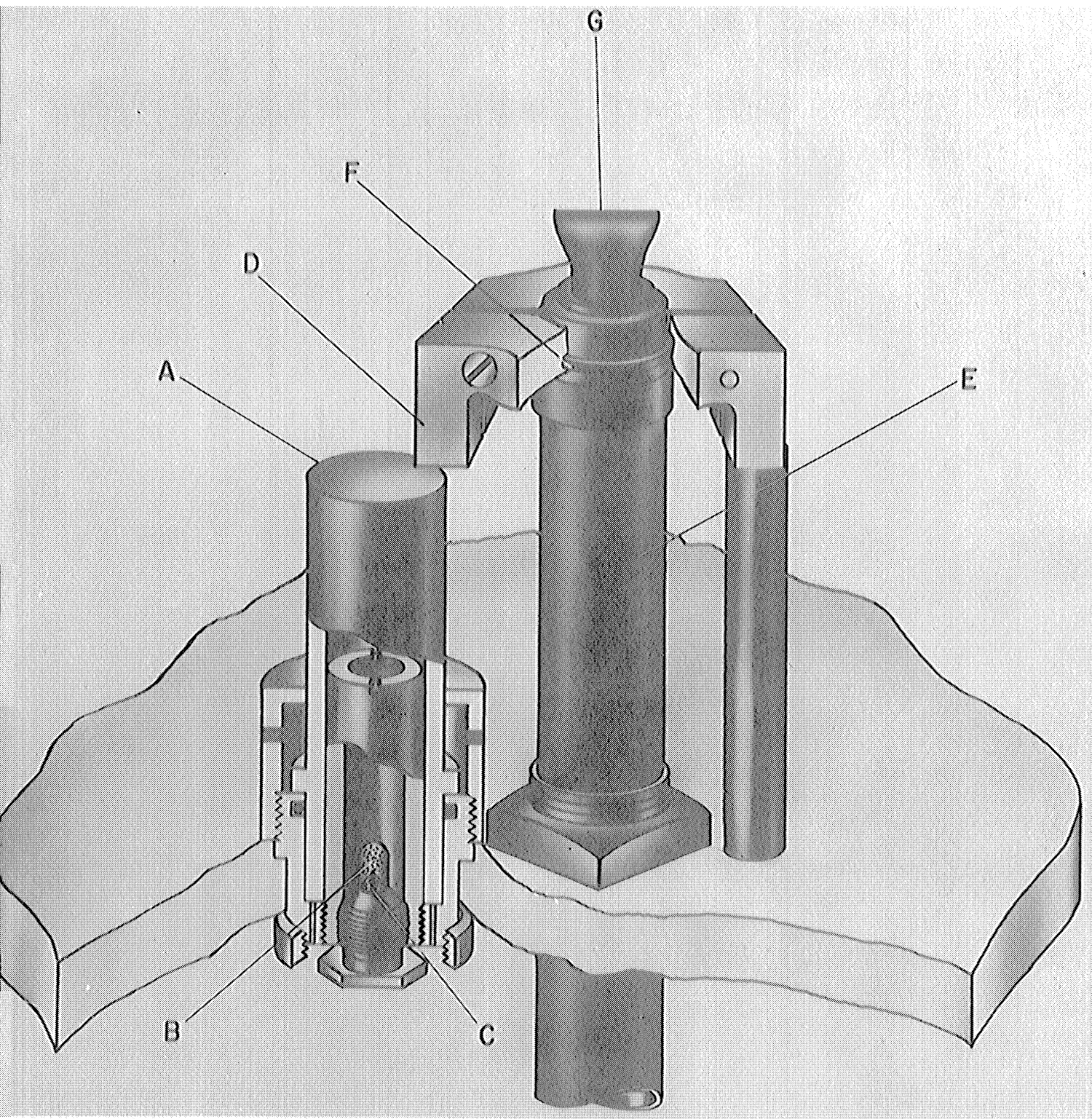


Fig. 3
Piston Opener

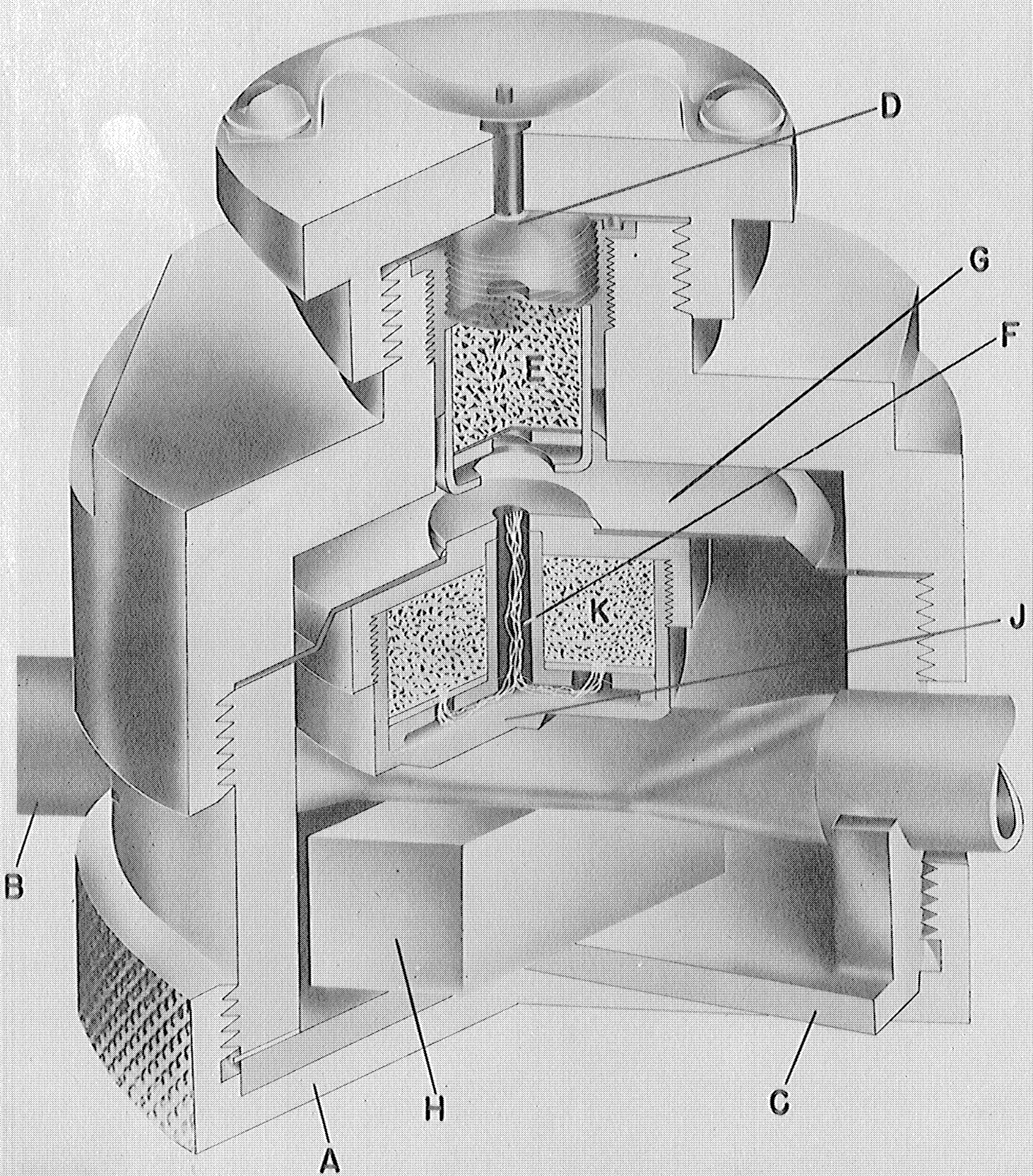


Fig. 4
Pyrotechnic Sealer

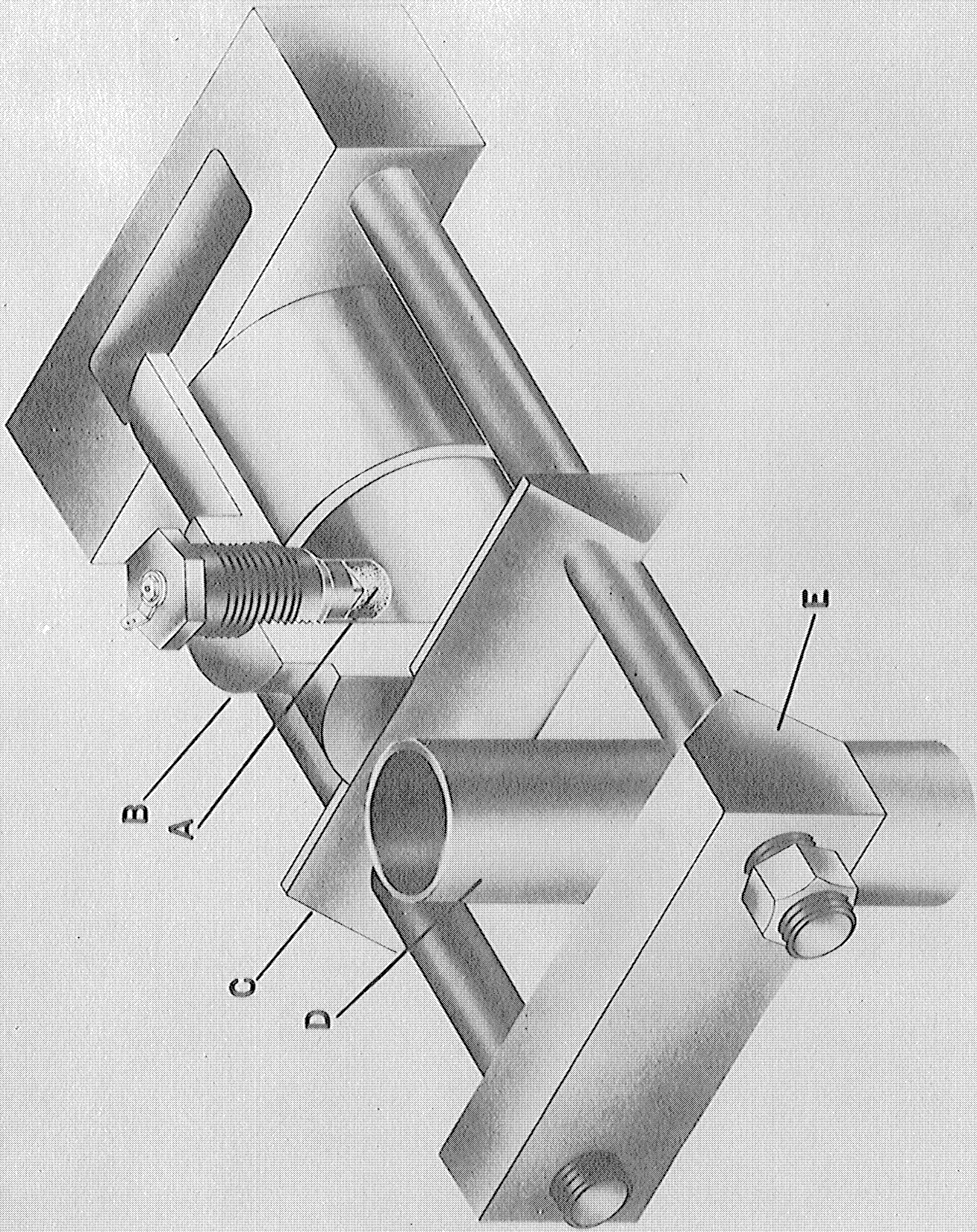


Fig. 5
Cold-Weld Sealer

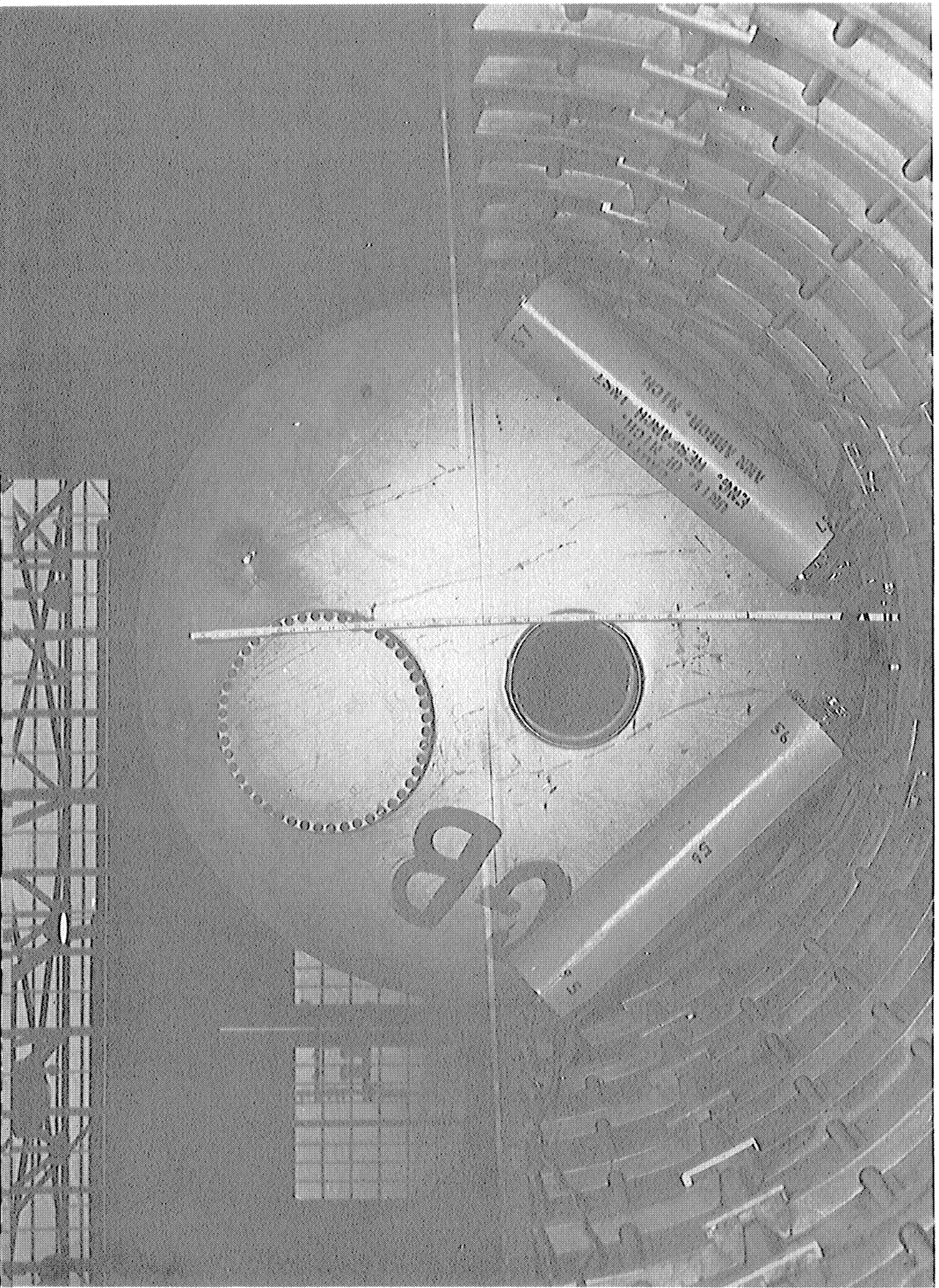


Fig. 6
Sample Bottles Mounted in Mid Section of V-2

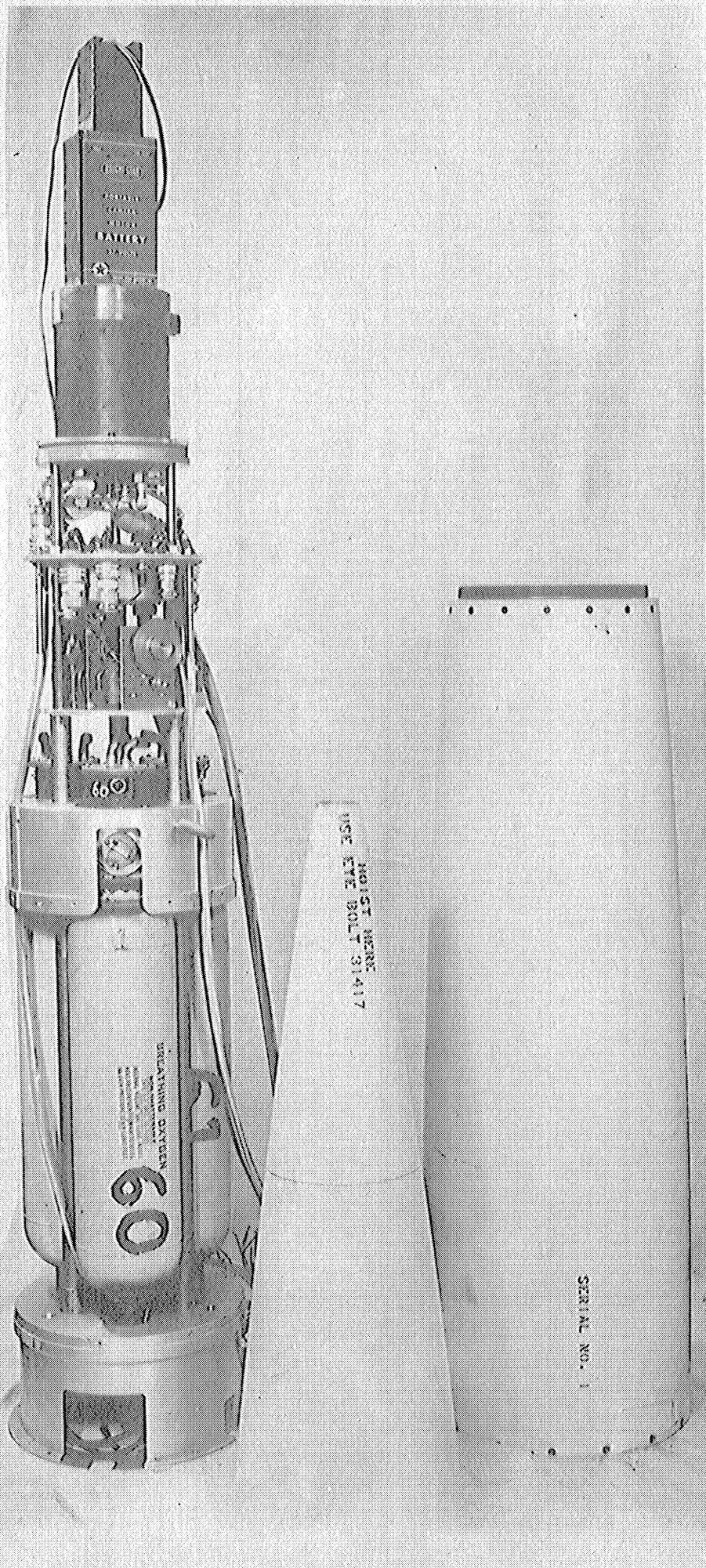


Fig. 7

Early Aerobee Bottle Installation (Scoops Not Shown)



Fig. 8

Early Bottle Aerobee Showing Small Scoops

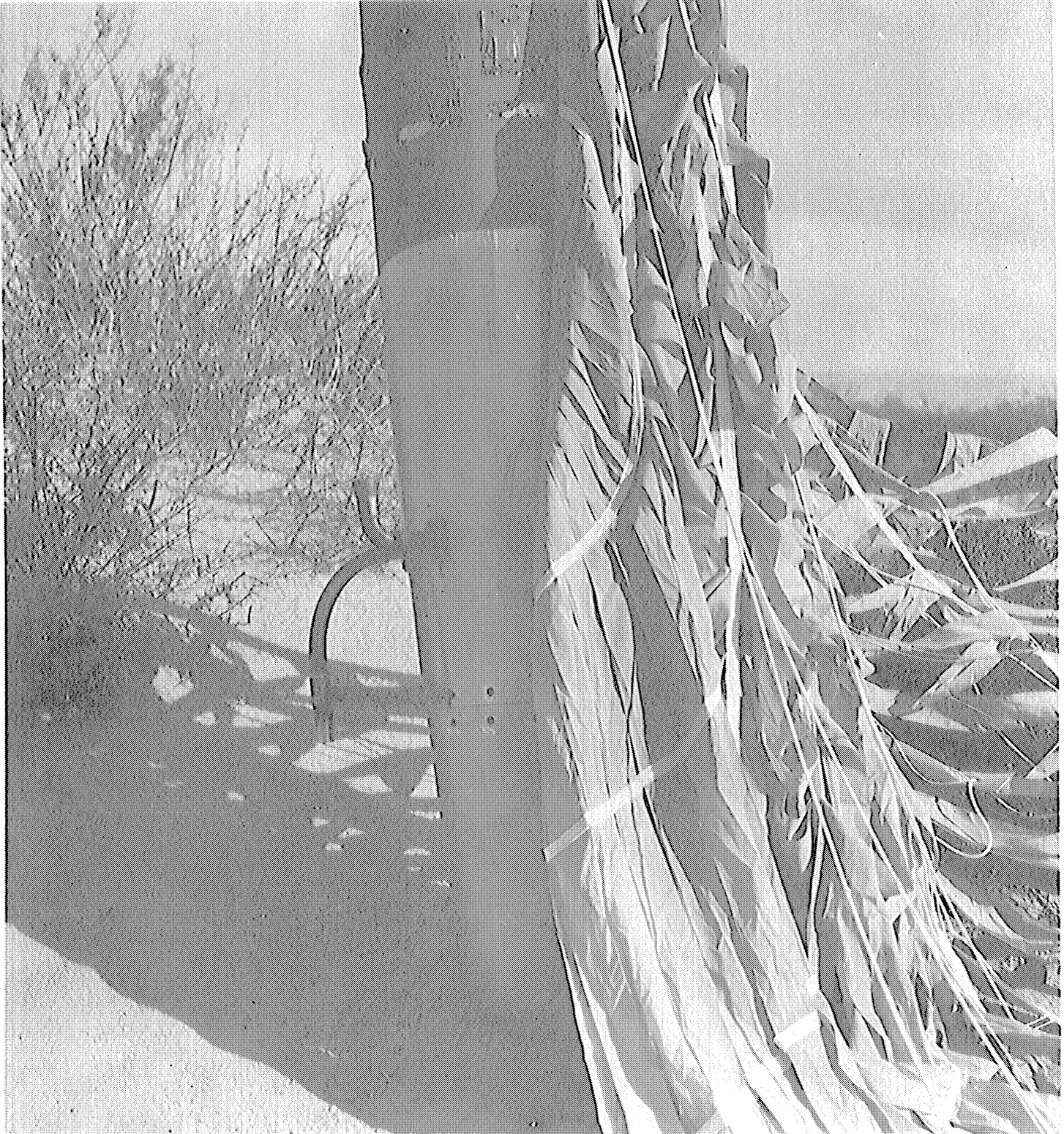


Fig. 9
Early Bottle Aerobee Recovery, Large Scoops

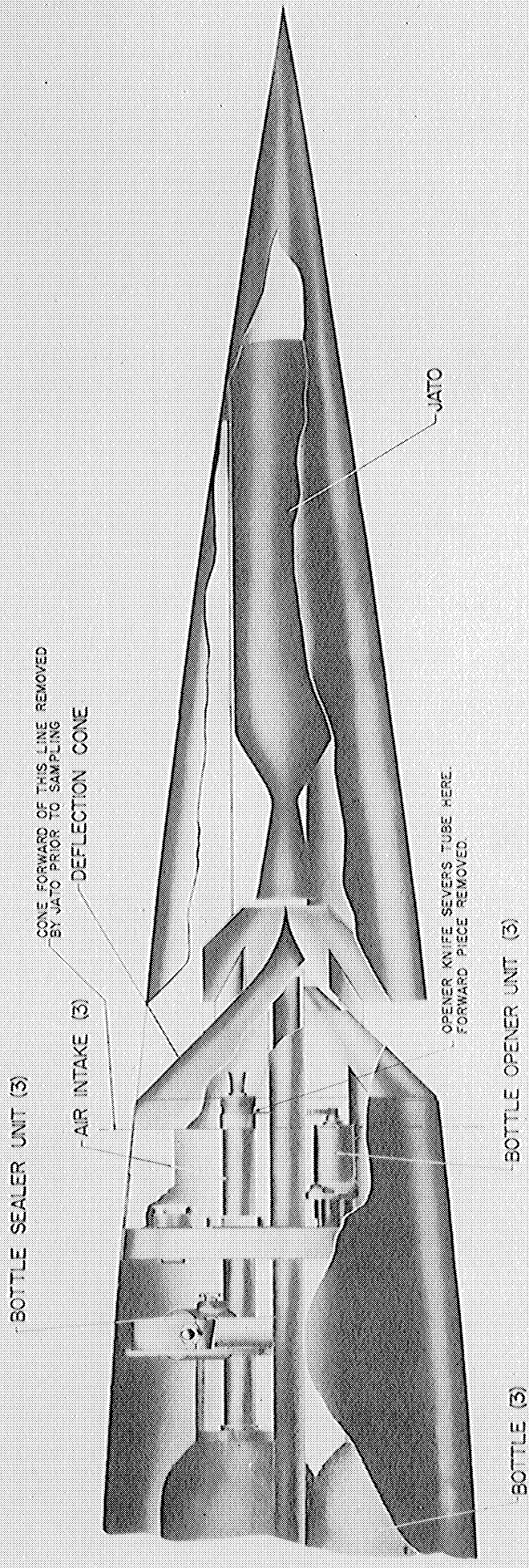


Fig. 10
Recent Aerobee Bottle Installation

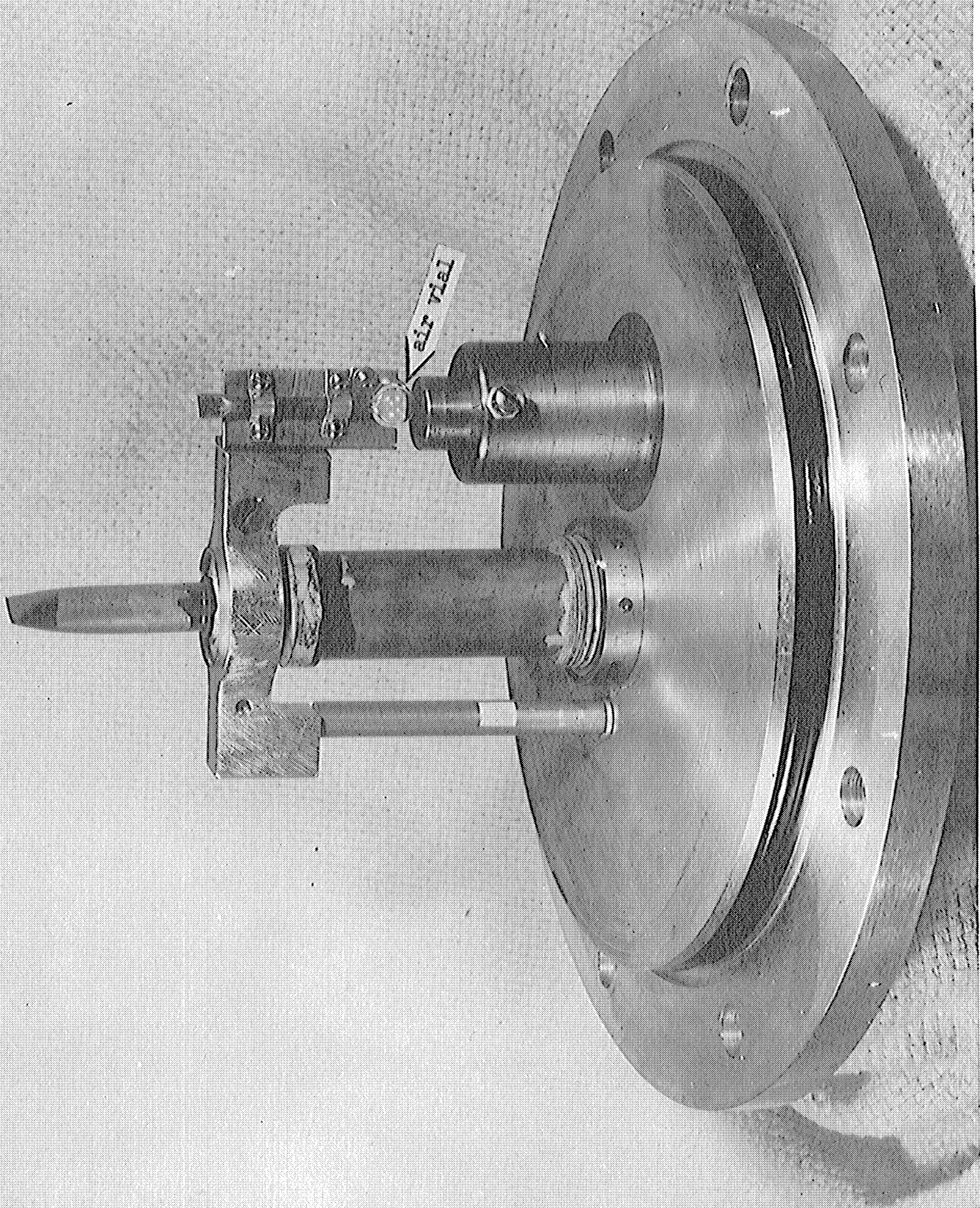


Fig. 11

Control Bottle and Ground Air Vial Mounted in Base of Chamber

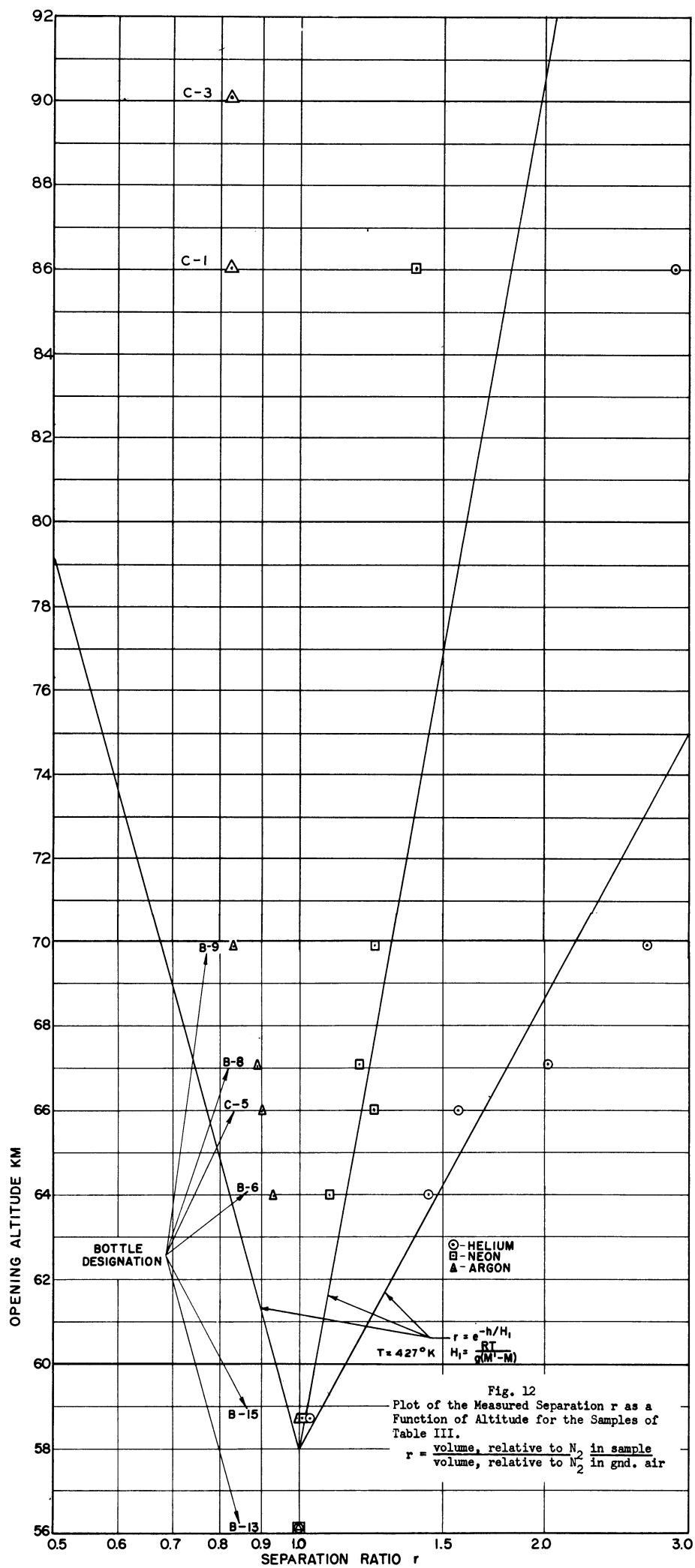


Fig. 12
 Plot of the Measured Separation r as a
 Function of Altitude for the Samples of
 Table III.
 $r = \frac{\text{volume, relative to } N_2 \text{ in sample}}{\text{volume, relative to } N_2 \text{ in gnd. air}}$

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