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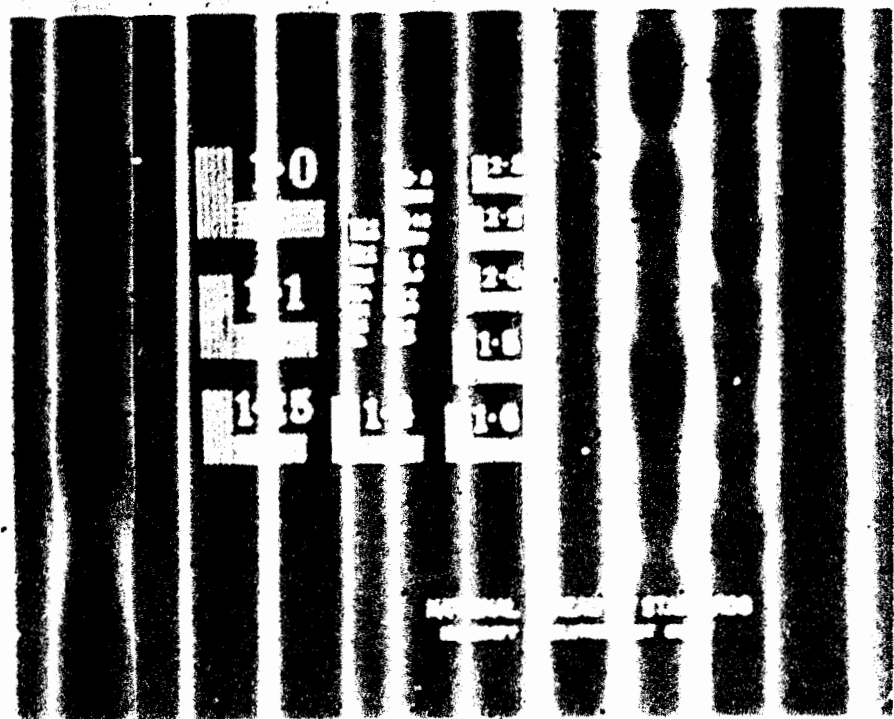
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RESEARCH NOTE 24

THE PREPARATION AND INJECTION OF TEFLON/GAS
DISPERSIONS FOR SHOCK TUBE INVESTIGATIONS

by Robert R. Holden and W. J. Hooker

HELIODYNE CORPORATION
Los Angeles, California 90064

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ABSTRACT

A narrative account is presented of the problems associated with the development of the powder injection shock tube facility at Heliodyne Corporation. Techniques for handling and controlling the contamination level of submicron-size powders are described. A discussion is presented of the various stages in the evolution of the presently used powder injection system. Methods for measuring powder concentration distribution are discussed, and the results of independent measurements are shown.

TABLE OF CONTENTS

Section		Page
	ABSTRACT	i
1.	INTRODUCTION	1
2.	MATERIAL CONTAMINATION	3
3.	POWDER-INJECTION TECHNIQUES	7
4.	POWDER DISTRIBUTION DETERMINED FROM WEIGHT MEASUREMENTS	17
5.	TEFLON POWDER DISTRIBUTION DEDUCED FROM CF_2 ABSORPTION	21
6.	CONCLUSIONS	29
	REFERENCES	31

1. INTRODUCTION

Fundamental to the use of the SAPAG (Shock Accelerated Particles and Gases) facility^{1,2,3} for the study of the optical and electromagnetic properties of ablation product/air mixtures is the requirement that submicron-size particles of selected ablation materials be fabricated, handled and injected uniformly and reproducibly into the shock tube. The size of the particles to be injected is determined by the requirement that they burn up in the shocked flow in a time that is short compared to the available experimental testing time. Calculations⁴ of the burnup rates of small particles and gases have shown that the particle sizes required for conventional shock tube facilities are in the micron and submicron range for materials whose thermodynamic properties vary from those of low-temperature ablators, such as teflon, to very high-temperature ablators, such as graphite. Numerous materials of commercial interest are available in this size range, and the techniques for producing laboratory quantities of materials not available commercially, have been documented elsewhere.⁵ Not so well documented, however, are the arts and science of handling finely divided particulate matter so as to avoid contamination, to weigh them in very small amounts, and to disperse them uniformly throughout a gas in a container such as a shock tube. In this Note are described some of the elements of the learning curve acquired at Heliodyne Corporation in the process of developing the powder-injection shock tube currently being used. The developments reported herein, while primarily directed at the handling of teflon powders, are generally applicable to any material.

2. MATERIAL CONTAMINATION

In a normal shock tube experiment, i. e., one involving gaseous compounds only, the problem of material purification is largely one of removing impurities present in the parent material, as purchased. Numerous adsorption, fractionation and change-of-phase techniques are available to perform such purification. Once purified, a well trapped shock tube system introduces no further contamination to the test gases. While gases of commercial interest are generally available with contamination levels in the low ppm, it is found that materials that have a practical application as ablation heat shield materials are produced with tolerances that are comparatively wide because of the bulk methods used to produce them, and the lack of more stringent tolerances required by the majority of these applications. Since the problem of preparing solid materials of controlled high purity is beyond the scope of the current program, our approach has been to obtain materials as pure as are readily available, and to use all practical precautions in maintaining the materials in their original state of purity. This requires information on the contamination levels in the bulk material, and the material when processed into fine, particulate matter.

When any material is reduced to particles of submicron size, the specific surface area of the material increases greatly, and the resulting powders take on some of the properties of a molecular sieve, i. e., they have the ability to adsorb gases and vapors. In addition, the particles of the selected material can also agglomerate with like particles or with particles of other contaminant materials. Therefore, if a finely ground particulate material

is exposed even to a normal room atmosphere, this material will be contaminated with moisture, dust, smoke, etc., to an extent that is a complicated function of the material itself. Since such an exposure will produce an uncontrolled and non-reproducible contamination level, the materials used in this program are stored and handled only under dry-box conditions.

The dry-box used for these purposes, shown in Fig. 1, maintains the material in a controlled inert atmosphere with a contamination level of less than 3 ppm of water vapor and less than 1 ppm of oxygen. While the dry-box may be used to maintain stored materials at the desired level of purity, it can also be used to a certain extent to clean up materials that have been exposed to an atmosphere from which it adsorbed gases. We have dusted thin layers of submicron-size material on metal slides and have weighed them under dry-box conditions. Subsequently, we have taken these slides and exposed them to a normal room atmosphere for 24-hour periods. Upon placing the exposed slides back in the dry-box and weighing immediately, it was found that the slides had accumulated a substantial amount of adsorbed materials. However, upon remaining in the dry-box for another 24-hour period, it was found that the sample slide had returned to its original weight. We have performed this test a number of times and have found that the dry-box is an efficient device for out-gassing finely-divided particulate matter.

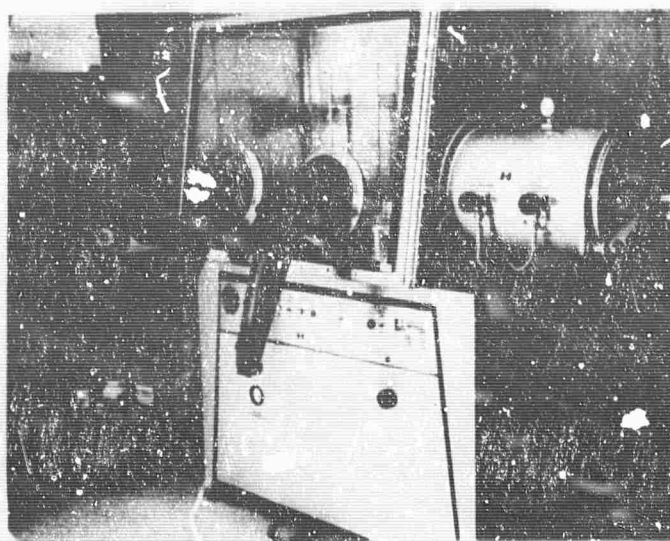


Fig. 1 Inert atmosphere dry box which is used to minimize contamination of fine powders. The gas used in the dry box is nitrogen from which the oxygen is removed by catalytic reaction with hydrogen, and water vapor is removed by adsorption on a molecular sieve. The typical level of contamination in this dry box is less than 3 ppm water as read on an electrolytic moisture detector. The oxygen level is sufficiently low that a tungsten filament bulb can be maintained incandescent for days unprotected without failure.

3. POWDER-INJECTION TECHNIQUES

Figure 2 is a schematic of the original shock tube, which was mounted vertically. The original powder injector consisted of an oscillating sieve mounted directly above the vertical shock tube. This sieve, upon being mechanically driven, would sift the desired material down through the driven section of the shock tube. The particles, falling under the action of gravity, would attain a velocity limited by local viscous forces, which could be calculated from Stokes' law. Typically, a particle 10 microns in radius, would fall at a rate of approximately 2 centimeters per second. Particles of this size would, therefore, take approximately 4 minutes to fall the length of the 6-meter driven section. However, since particles in the micron and submicron range are needed for complete ablation under normal shock tube conditions, it was seen that this method of introducing the powder to the tube would require too great a period of time for complete dispersion throughout the tube.

The second powder injection system tried consisted of an injection tube which is shown in the bottom insert in Fig. 2. This small volume tube was loaded with the desired powdered material and test gas to such a pressure that upon being injected into the already evacuated driven section of the shock tube, produced a final pressure which was the desired initial driven pressure. With this system, the violence with which the powders were injected into the driven section of the shock tube was such that a substantial portion of the injected material impinged on the wall opposite the injector and remained fixed there. In order to overcome the difficulties with these two methods, a compromise system was tried.

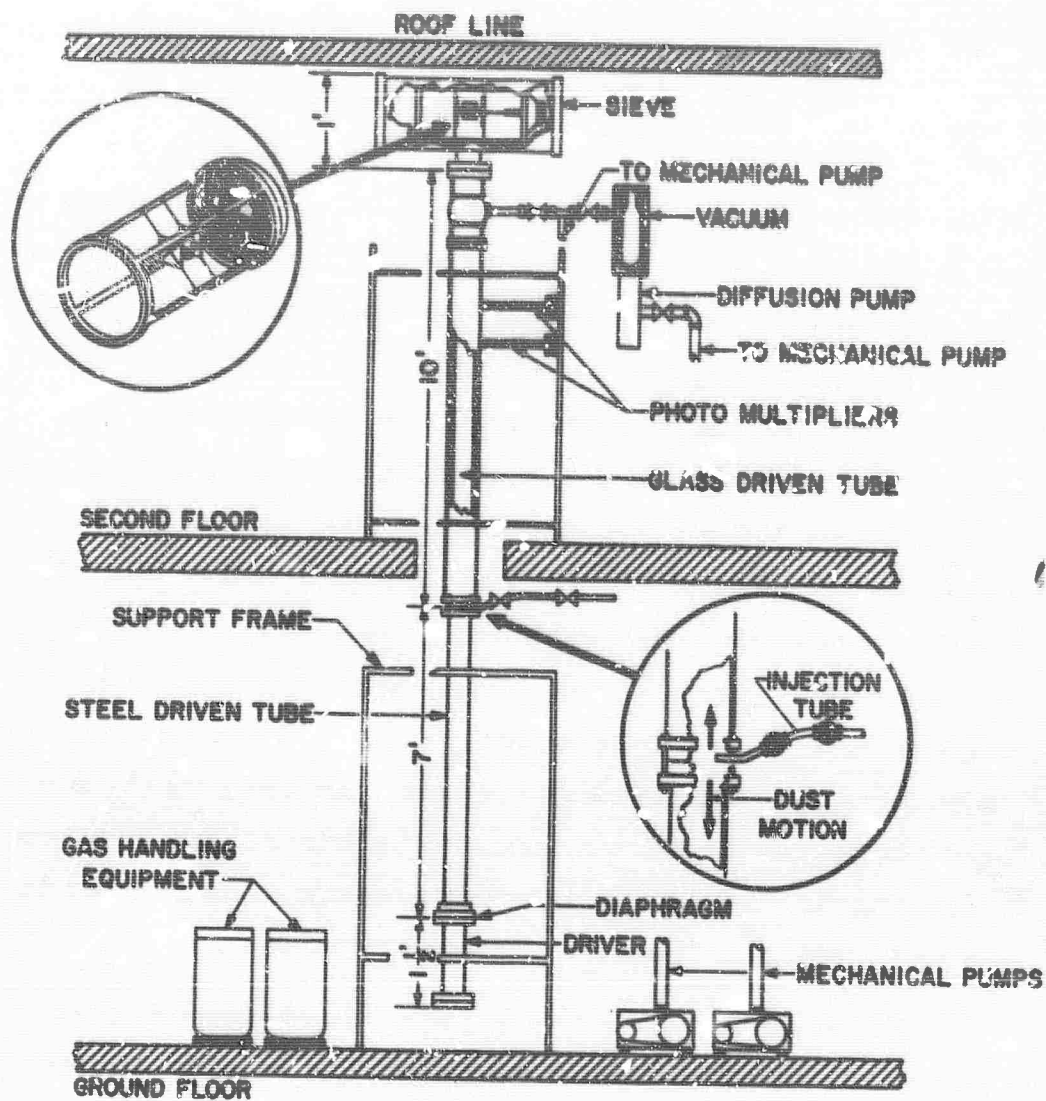


Fig. 2 Schematic of the Vertical Shock Tube. All gas handling equipment and mechanical pumps are on the ground floor. Electronic and spectroscopic devices are located on second floor.

The third system had the basic features of the original oscillating sieve and was, indeed, mounted in the same tank assembly as shown in Fig. 2. However, a valve was inserted between this tank and the end of the driven section of the shock tube. In addition, an electrically driven fan was inserted in the oscillating sieve chamber as shown in Fig. 3. In operation, powder was loaded in the sieve in the open atmosphere and then the entire shock tube was sealed off from the atmosphere. The valve connecting the injector tank to the shock tube was opened and the entire system was evacuated with the pumps. When the desired ultimate vacuum was attained, the valve connecting the shock tube to the injector tank was closed, and the injector tank was pre-pressurized to a value which would produce the desired initial driven pressure when connected to the shock tube. The sieve and fan were then set into motion, in order to produce a uniform cloud of powder and gas in the injector tank. When the mixture appeared uniform, the motion of the sieve and the fan was stopped, the diffusion pump valve was closed, and the valve connecting the injector tank to the driven section of the shock tube was opened, thereby injecting the entire mixture into the shock tube. After a period of elapsed time during which any very large particles present would have dropped to the bottom of the shock tube, the tube was fired and the measurement was made. Although this method of injection did not provide the uniformity of powder distribution in the driven section of the shock tube that was desired, it did show, however, that there was no fundamental reason why this form of injection could not be used with the shock tube in a horizontal position. Since the operation of the shock tube in the vertical position was extremely cumbersome from the point

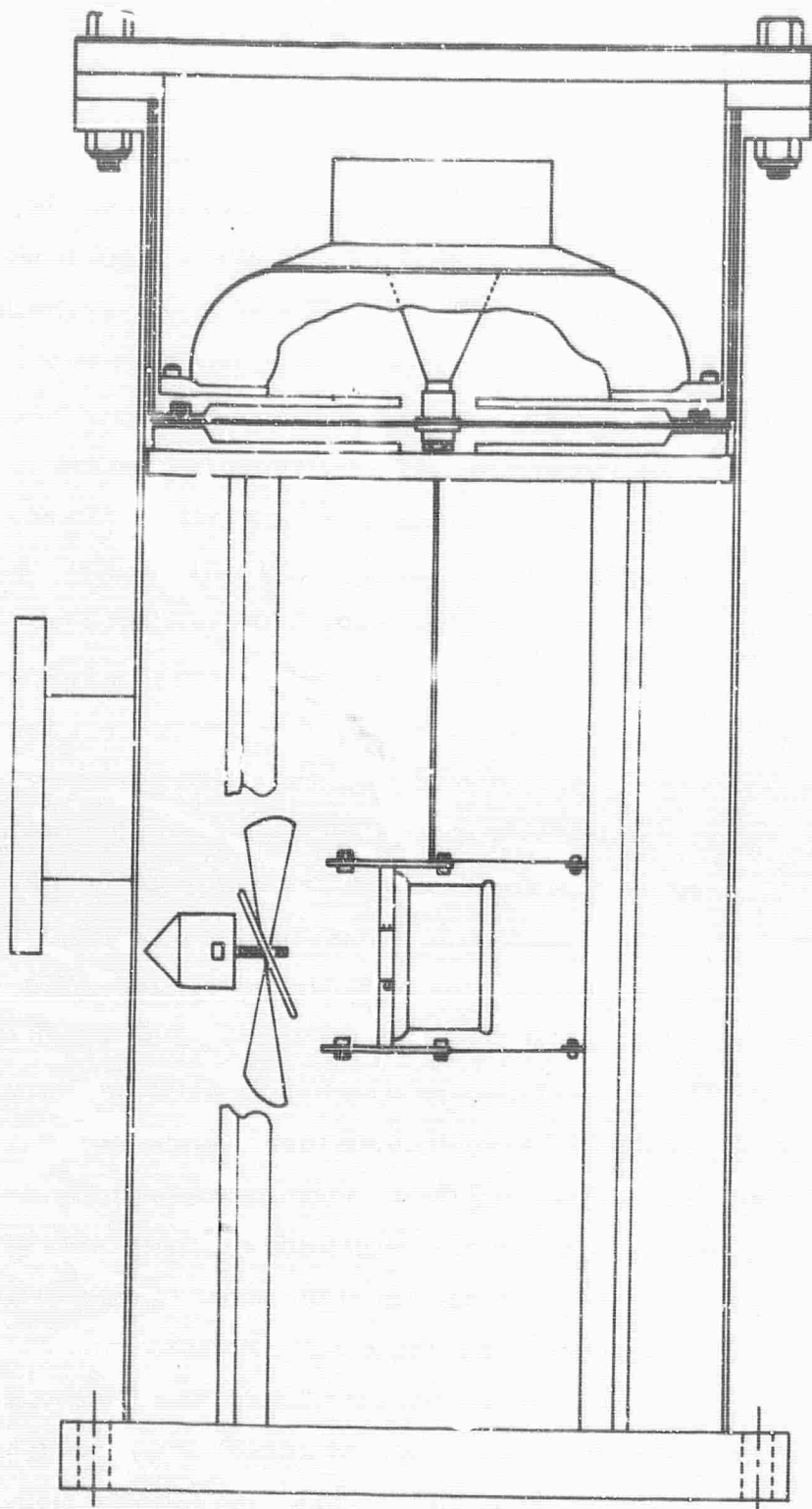


Fig. 3 Pressure-Mechanical Injector with Vapor Barrier Installed in the Injector Housing. A high vacuum is attained in the powder containing chamber while only a rough vacuum exists in the speaker chamber. Transverse motion is transmitted through a sealed stainless steel diaphragm.

of view of the instrumentation and installation expense, it was decided at this time to mount the shock tube horizontally and conduct further tests with the injection system already mentioned. Figure 4 is a view of the shock tube mounted in the horizontal position with the pressure-mechanical powder injector mounted at the end of the tube. For comparison purposes, shown in Fig. 5, is a view of the vertical shock tube, where the high pressure powder injection system is shown in use.

Further tests with the pressure-mechanical injector system in the horizontal position revealed that the action of the fan in the injection chamber tended to speed up the process of agglomeration of the initially small size particles, and to cause them, through the motion of the gas within the chamber, to strike on the walls of the injection tank and thereby stick to it. In addition, it was found that the driven section of the shock tube, which was glass in all tests so far conducted, attracted the highly desiccated injected powders by electrostatic charge and tended to collect the bulk of the injected powder within a few feet of the injection chamber. In order to overcome these latter difficulties, an all-metal shock tube was constructed, and the fan and sieve assembly was removed from the injector tank. In its place, the injection device shown schematically in Fig. 6 was installed. With this (presently used) powder injection technique, a predetermined amount of the selected material is loaded into a one-inch transfer chamber while in the dry-box. The transfer chamber is sealed off, removed from the dry-box and then attached to the air solenoid-actuated valve on the top of the injection tank. When the shock tube is sealed off from the atmosphere, the injection tank is evacuated through the large air solenoid-actuated

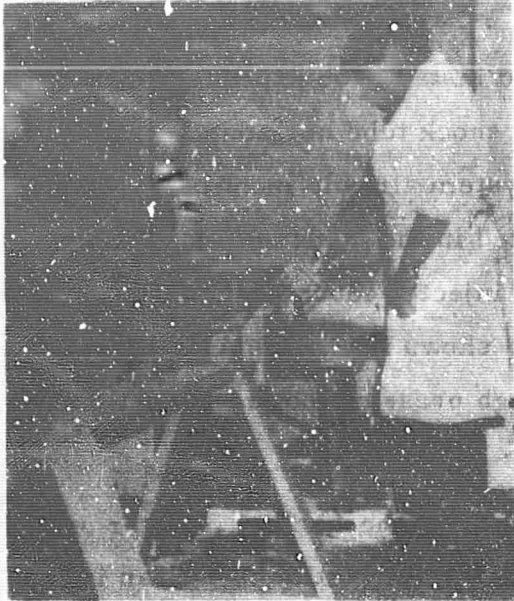


Fig. 4 Pressure-Mechanical
Injector

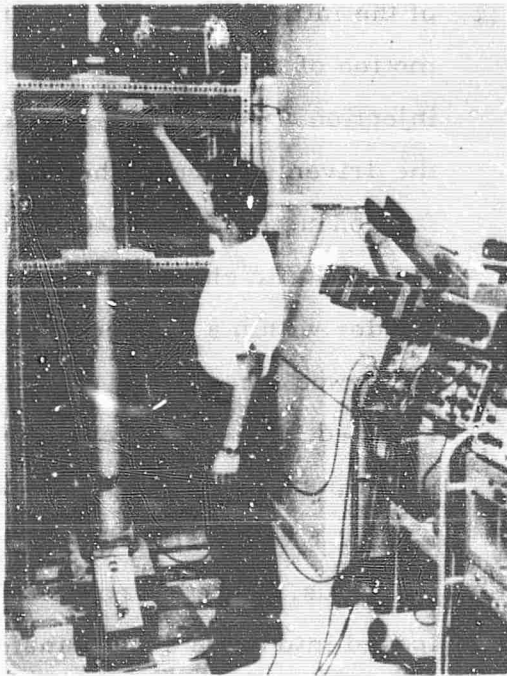


Fig. 5 Vertical Shock Tube

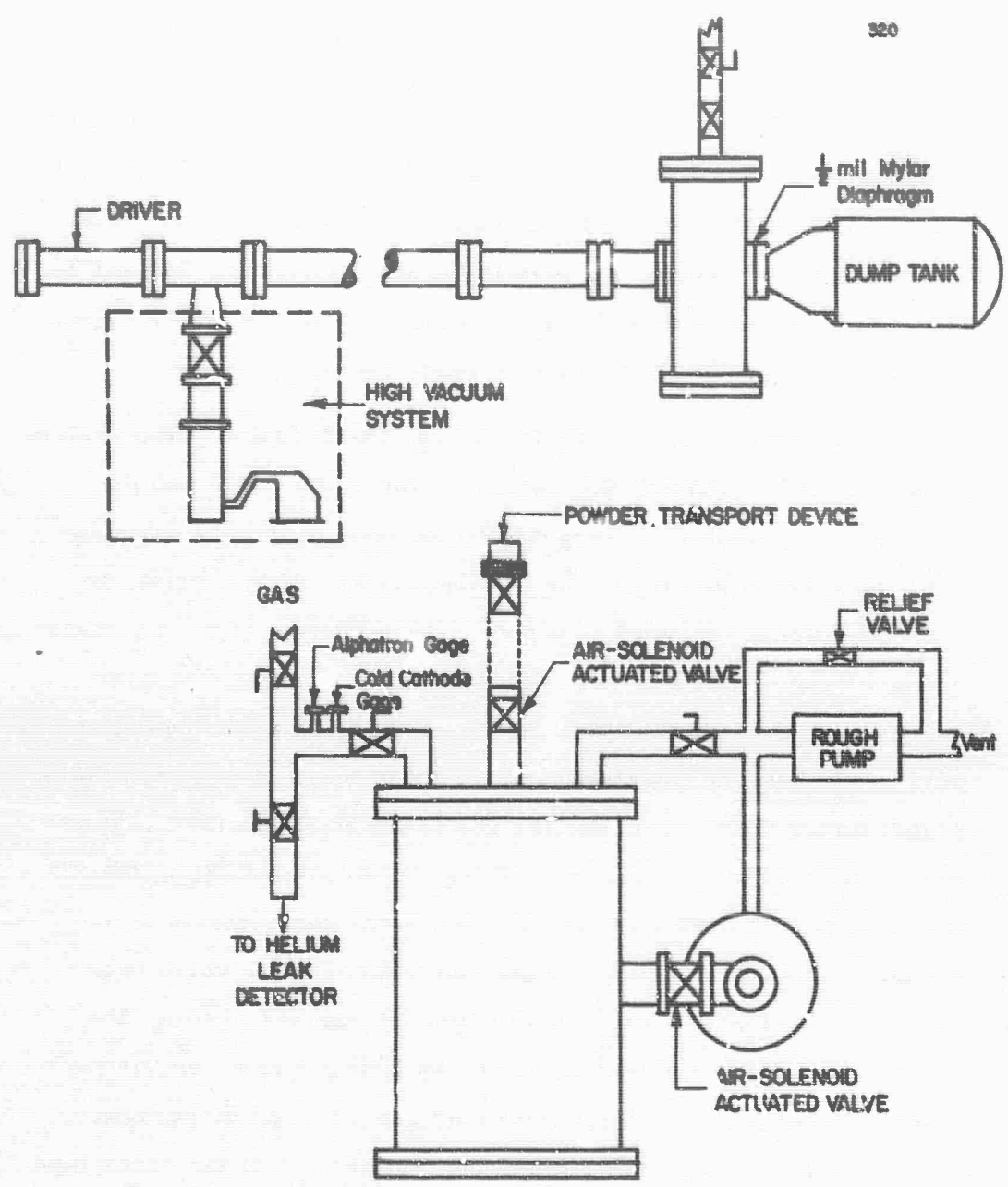


Fig. 6 Schematic of the Shock Tube and Powder Injection Chamber

valve connecting it to the driven section of the shock tube. When the ultimate vacuum is achieved (approximately 10^{-4} mm Hg), the valve between the shock tube and the injection tank is closed and the desired test gas pressure is loaded in the injection tank. The diffusion pump continues to maintain the ultimate vacuum on the driven section of the tube until the tube is ready for firing.

When the tube is ready for firing, the diffusion pump system is closed off from the driven section of the shock tube, and the powder transport device is opened to the body of the air solenoid-actuated valve on the top of the injection tank. At this point, an automatic firing sequence is begun. The initiation of the automatic firing sequence starts a multiple cam-switch timer. The first operation of this timer is to inject the powder from the powder transport device into the injection tank. This process of injection thoroughly mixes the powder with the gas in the injection tank. At a preset interval of time following this, the valve connecting the injection tank to the shock tube is opened and the contents of the injection tank are injected into the shock tube. This valve is then immediately closed. After another preset interval of time, the shock tube driver is actuated, either by firing it as a combustion driver, or by pressurizing it with cold gas until the diaphragm bursts. The injection system and the test section of the shock tube are shown in Figs. 7 and 8.

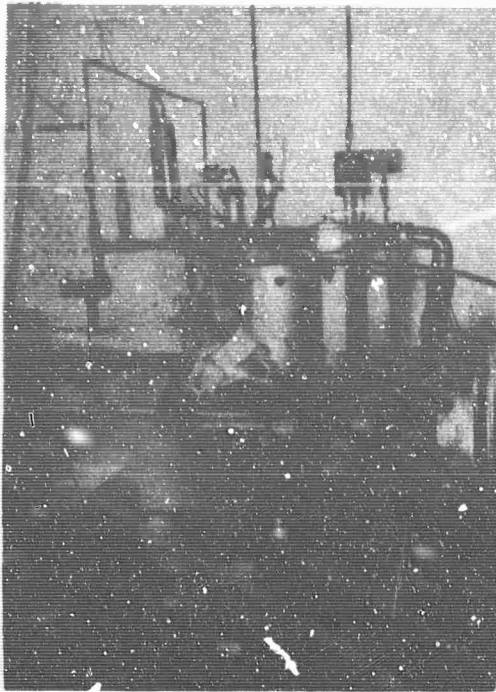


Fig. 7 View of the Powder Injection Chamber.



Fig. 8 View of the Shock Tube Test Section and Powder Injection Chamber.

4. POWDER DISTRIBUTION DETERMINED FROM WEIGHT MEASUREMENTS

The first measurements to determine the axial and radial distribution of powder within the driven section of the shock tube were made using light-scattering techniques.³ While these measurements showed that it was possible, under certain injection techniques, to obtain a relatively uniform scattering of light throughout the volume of the shock tube, it was not possible to deduce quantitatively the mass distribution of injected powder throughout the volume of the shock tube. In addition, these initial light-scattering measurements were made with the glass shock tube, wherein the electrostatic charging problems as mentioned before, were serious.

A more positive series of weight determinations was made when the all-metal shock tube was put into operation. With this system, a strip of aluminum foil approximately two inches wide was laid the full length of the shock tube. After the shock tube was prepared as if for a normal firing, the powder was injected and allowed to settle onto the aluminum foil. After several hours had been allowed for the powder to settle, the shock tube was slowly let up to atmospheric pressure with dry nitrogen, and the aluminum foil was carefully removed from the shock tube. At specified intervals, the strip was cut and three inch long sections of the foil were removed from the strip containing the powder which had settled out onto it. Each aluminum foil sample was immediately sandwiched between two glass cover slides and weighed on a semi-microbalance. After each aluminum foil glass sandwich had been weighed, the top cover slide was removed and the sandwich was re-weighed after

brushing off the accumulated powder. The net difference in weight for these two weighings was that due to the powder itself. Since these weight measurements were made in the room atmosphere, the dry powders removed from the shock tube on the aluminum foil tended to adsorb water vapor from the air, very rapidly, thereby producing an uncertainty in the weighings. In addition, the highly desiccated powders tended to stick by electrostatic attraction to the glass cover slides so that it was never possible to remove all of the powder from the glass surfaces adjacent to the aluminum foil. In order to overcome this problem, the entire semi-microbalance was mounted in the dry-box and all weighings were performed in the dry-box. The balance was installed on a massive marble slab within the dry-box to isolate vibrations of the dry-box assembly from the delicate semi-microbalance.

A new set of weight measurements was made in the shock tube by placing brass slides similar in size to the previous glass cover slides, in the shock tube at specified intervals. After a normal injection sequence the slides were removed from the shock tube and placed in covered Petri dishes. These Petri dishes were then placed in the dry-box and the brass slides were allowed to out-gas for a number of hours. It was found at this time that the semi-microbalance (in the dry-box) was weighing erratically. Even weighing a brass disk of known mass, the readings were not only erratic, but drifted continuously as though the brass was either gaining or losing weight. The problem was eventually traced to the presence of electrostatic charge within the mechanism of the semi-microbalance itself. This excess charge was brought about, presumably, by the very dry atmosphere within the dry-box. This

problem was cured by installing several 500-microcurie Polonium strips at various points within the balance itself. The radioactive Polonium strips ionized the gas within the dry-box and allowed the static charge responsible for the weight scatter to leak off. At the same time, the inner surface of the glass window of the dry-box was coated with an anti-static material. When these steps were taken, the operation of the balance was returned to normal.

Shown in Fig. 9 are the weight distributions obtained from two separate powder injection weighing tests. These tests were made with an injection sequence for which all valve operations were manually controlled. Based on the weight distribution measurements to be discussed later in the text (Section 5) it is felt that the scatter shown in Fig. 9 is attributable to the irreproducibility of a manually-injected operation. For both of the distributions shown in Fig. 9 it is seen that it was necessary to be approximately 150 cm from the location of the injector in order to avoid the substantial rise in weight distribution associated with the injection process. For both of the tests shown in Fig. 9, 7 grams of teflon 120 FEP powder was loaded in the transfer chamber. The final pressure in the shock tube after injection was 1 cm Hg. The ordinate on this graph corresponds to the weight per sample (brass slide) placed in the shock tube. The brass slides had dimensions of approximately 2 " x 3".

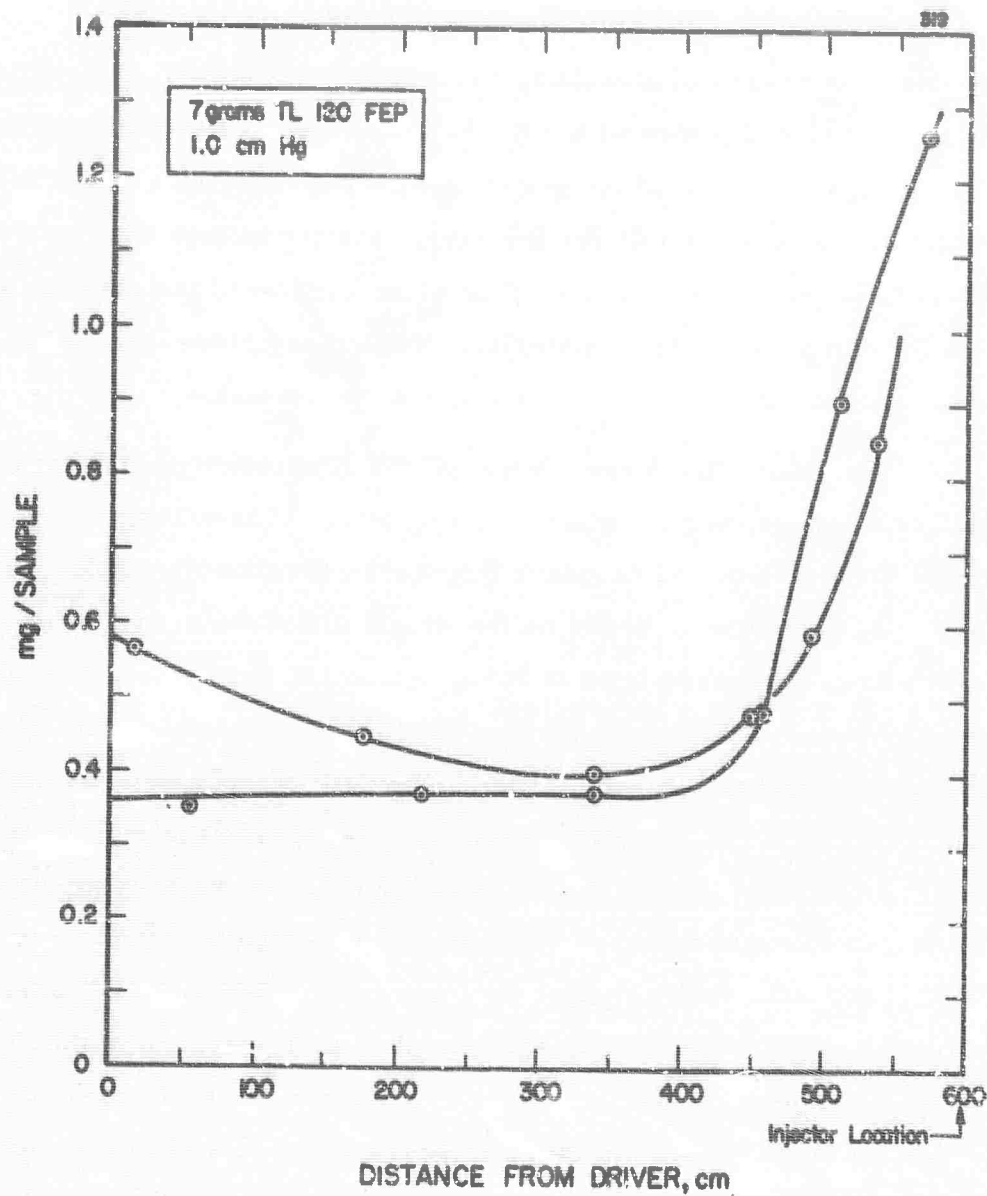


Fig. 9 Teflon powder weight distribution along the driven section of the shock tube, obtained by direct weight measurement. The sample holders were 2" x 3" brass slides.

5. TEFLON POWDER DISTRIBUTION DEDUCED FROM CF_2 ABSORPTION

The distribution of injected powder along the length of the driven section of the shock tube may be measured at one observation station as the driven gas/powder dispersion is swept by the observation station during a normal shock firing. If, under the conditions of the shock tube test, the teflon powder decomposes rapidly to form a stable gas phase compound, then a measurement of absorption by this gas phase compound will give a measure of the original powder distribution along the length of the tube. The valid use of this technique requires certain restrictions on the test conditions:

- a. The powder must vaporize in a time short compared to the shock tube testing time.
- b. A resulting equilibrium gas phase mixture must be formed in a time short compared to the shock tube testing time.

Calculations performed in Reference 4 have shown that the burnup times of micron and submicron-size teflon particles are short compared to experimental testing times in a conventional shock tube facility. In Fig. 10 are shown the results of these calculations for one micron diameter teflon particles in air. Also shown in Fig. 10 are measured and computed⁶ experimental testing times for the facility used, namely, one having driven section dimensions of 3 inch diameter by 15 feet length to the observation station. It is seen that the particle burnup time is approximately an order of magnitude smaller than the available experimental testing time and, therefore, criterion (a) above is satisfied.

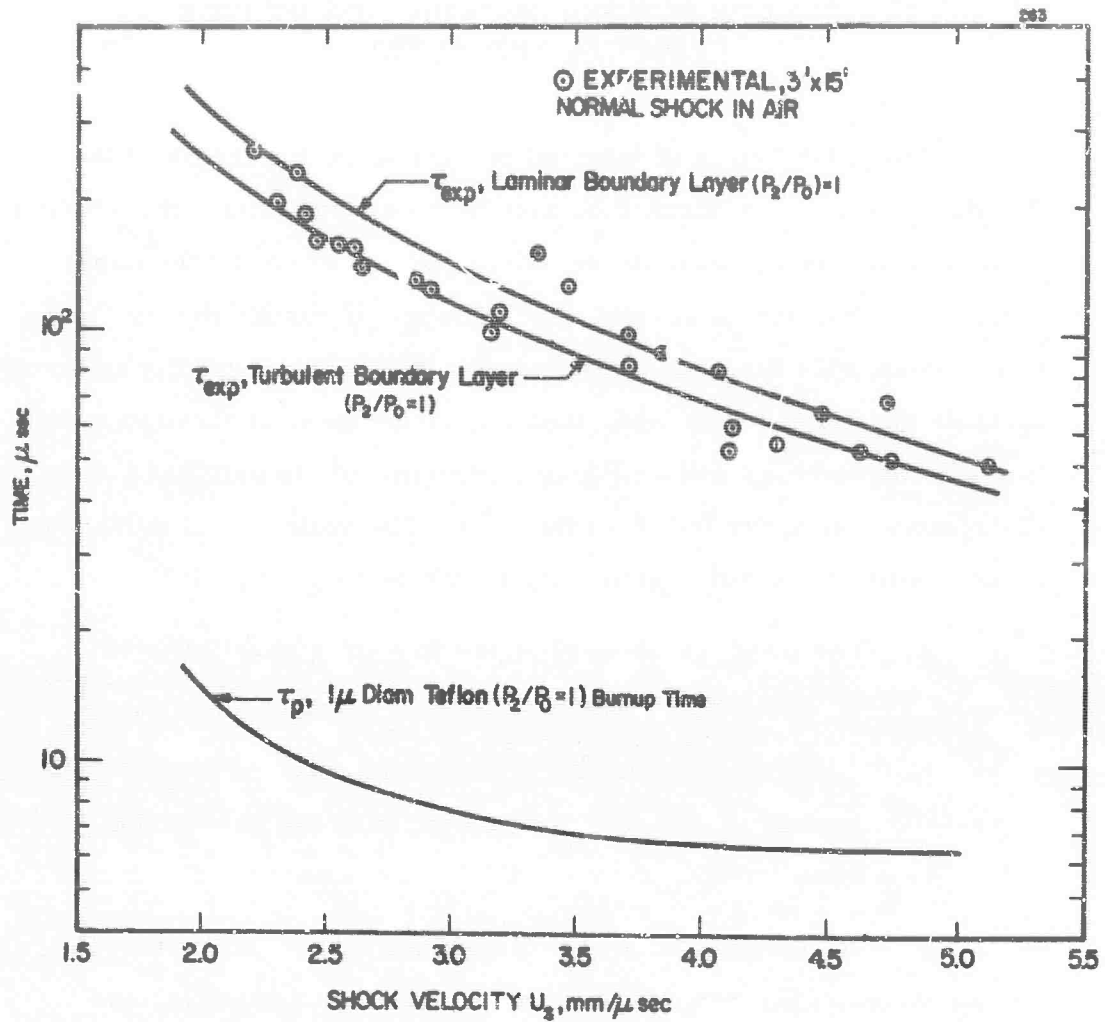


Fig. 10 Shock tube testing time and teflon powder burnup time versus shock velocity.

In References 7 and 8, Modica studied the dissociation of C_2F_4 to CF_2 and the dissociation of CF_2 to $CF + F$ in an argon diluent, and deduced reaction rate coefficients for these two reactions. This data was used in Reference 9 to show that over the temperature interval of 1750 to 3000°K, and at a total molar concentration of approximately 10^{-5} moles per cm^3 of argon, C_2F_4 dissociates rapidly to CF_2 and the resulting CF_2 concentration diminishes relatively slowly during the course of the experimental testing time in the shock tube. Based on this information, a measurement of CF_2 absorption behind a shock wave in a mixture of teflon and argon would provide a measure of the distribution of teflon powder along the length of the driven section of a shock tube and, therefore, criterion (b) above is satisfied.

We have used the CF_2 absorption technique to determine the uniformity of teflon powder injected into the driven section of our shock tube. Shown in Fig. 11 is a view of the test section of the shock tube used in these experiments, and the monochromator used for making the absorption measurements. This monochromator was a Bausch and Lomb Model 5 grating instrument (see Acknowledgment) with a deuterium light source and a 1F28 photomultiplier detector. After an initial series of measurements were made in which the various injection parameters were varied in order to obtain an optimum axial distribution of injected powder, the records shown in Fig. 12 were obtained. The records shown are from four successive tests in which all parameters were maintained constant in order to deduce reproducibility of injection. The initial driven argon pressure for these tests was maintained at 1 cm Hg, and the injected teflon powder was maintained at approximately 4 mole

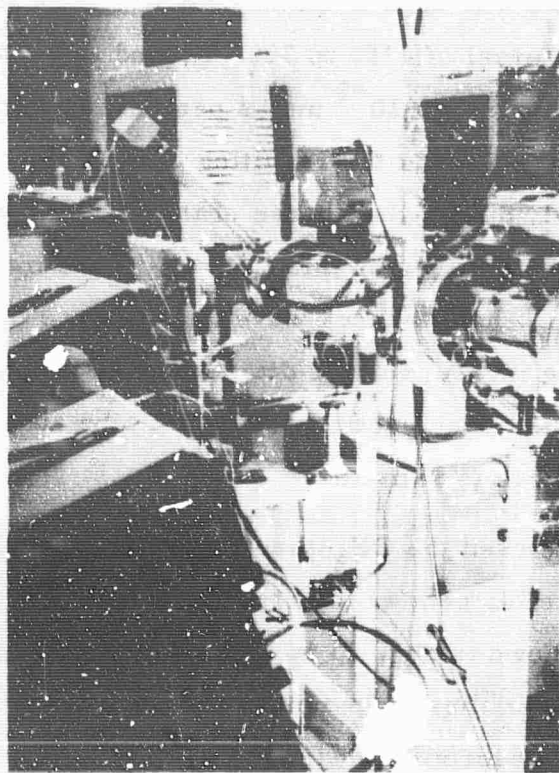


Fig. 11 View of the shock tube test section showing the grating monochromator (vertical).

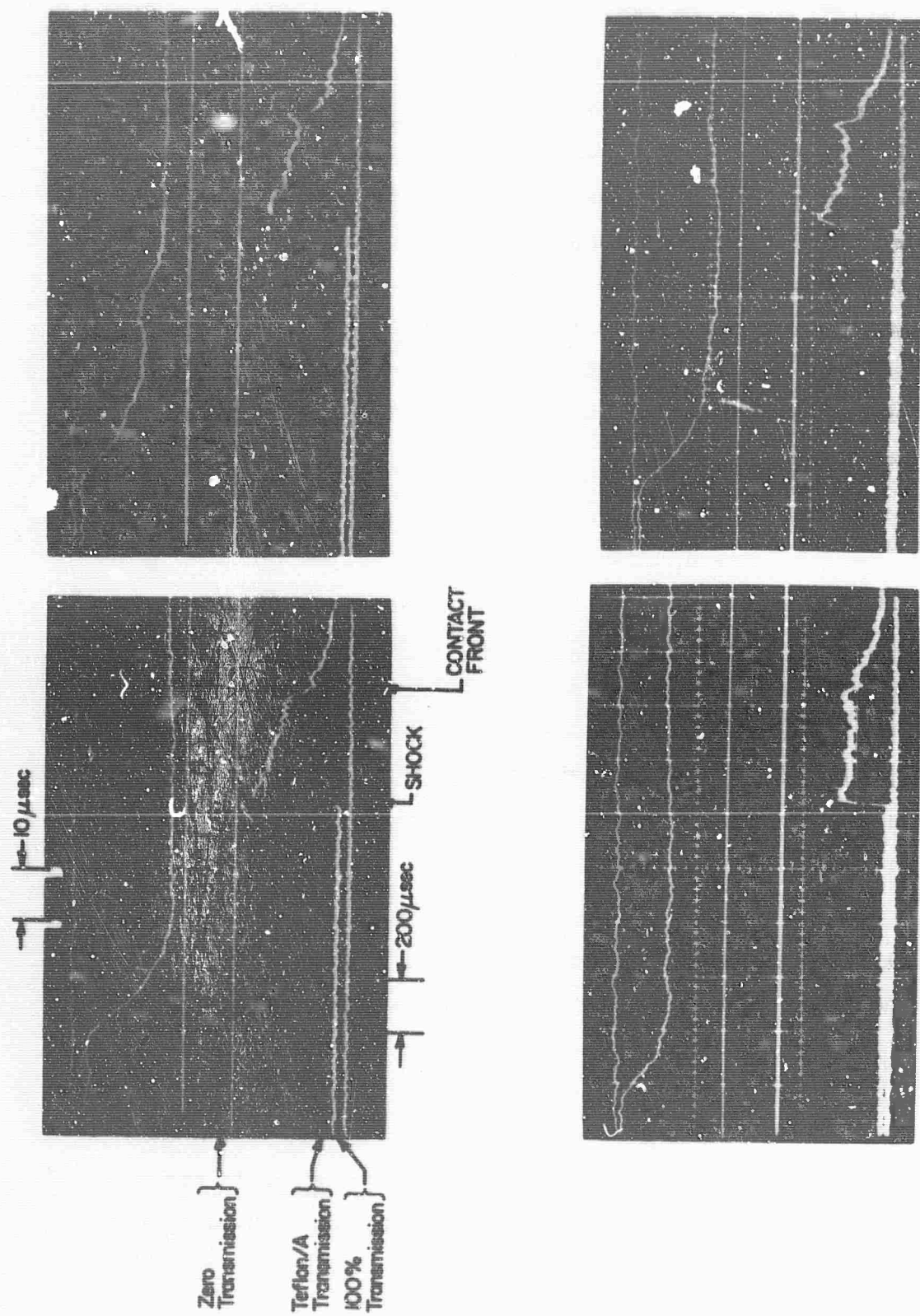


Fig. 12 Teflon powder distribution along the driven section of the shock tube as determined by CF₂ absorption at 2536Å. The gain and sweep rates are the same for all four test records shown. All injection operations were manually performed for these tests.

percent. In all four cases shown in Fig. 12, the powder injection operation was manually controlled. The top and bottom traces in each record are of the same event at different sweep rates. In each case, zero and one-hundred percent transmission lines were put onto the film shortly before the teflon powder/argon mixture was injected. The transmission through the teflon/argon mixture prior to shock arrival does not coincide with the one-hundred percent transmission line due to a scattering loss of light from the detector by the teflon powder.

While the general features of all four records in Fig. 13 are the same, it is seen that there is not exact reproducibility. Furthermore, the axial distribution of powder deduced from these records is not as constant as desired. In order to arrive at a more reproducible injection process, the automatic injection system referred to in Section 3 was installed. With this system, the timing sequence for each of the steps in the injection process was controlled by an electric timer, and all valve openings and closings were controlled by air actuators.

With the automatic equipment installed, a series of injection tests similar to that shown in Fig. 12, were conducted. The results of these tests are shown in Fig. 13. The test conditions for the records shown in Fig. 13 were maintained constant at an initial argon pressure of 4 cm Hg and a teflon powder concentration of approximately 1 mole percent. It is seen from Fig. 13 that the teflon powder concentration is virtually constant along the entire driven length of the shock tube. From the results of the tests shown by the records of Fig. 13, it is seen that it is possible to suitably

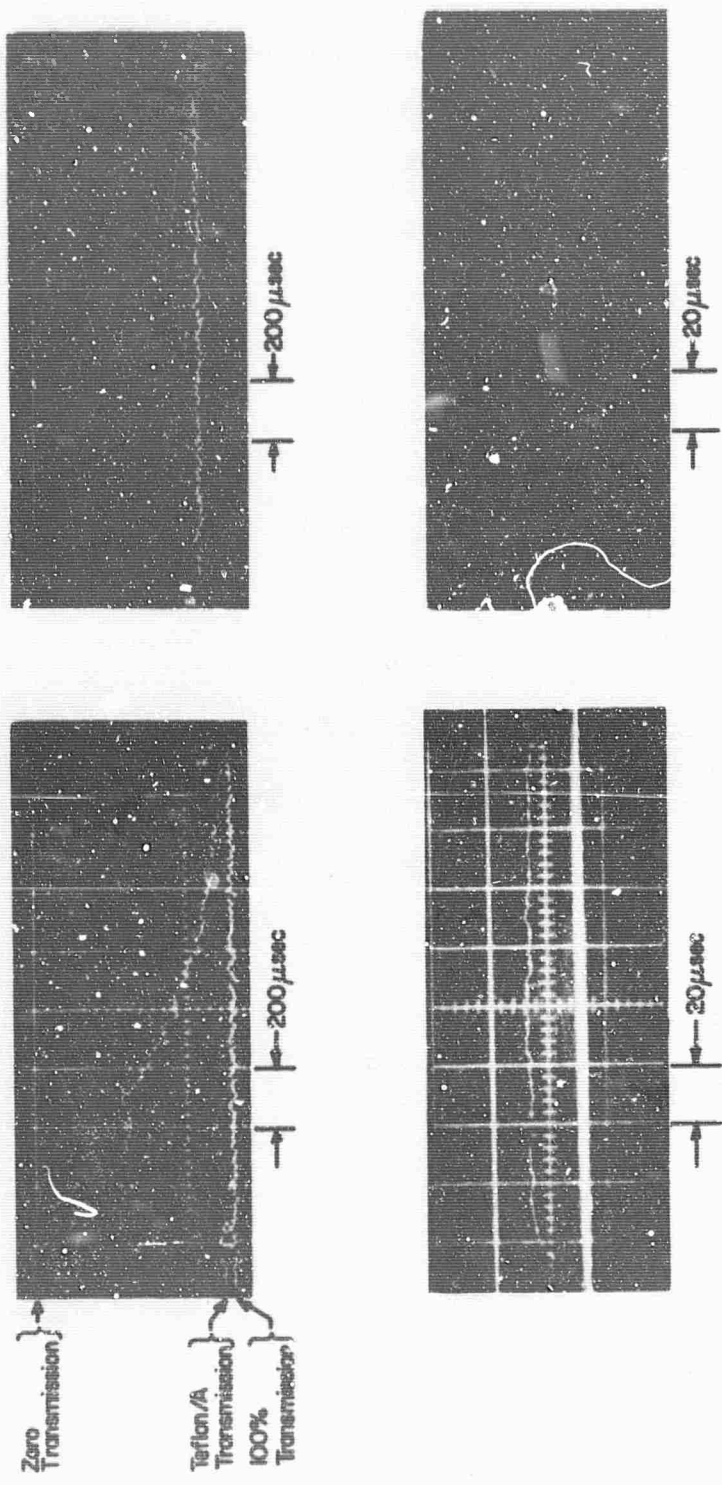


Fig. 13 Teflon powder distribution along the driven section of the shock tube as determined by CF_2 absorption at 2536\AA . The gain settings are the same for all four test records shown. All injection operations were automatically controlled for these tests.

adjust all of the operating parameters associated with the injection of teflon powder into the driven section of the shock tube such that a uniform, reproducible injection is obtained.

6. CONCLUSIONS

We have attempted to discuss some of the more important problems that arose during the process of developing the preser powder injection shock tube facility. While in hindsight, the solution to each of these problems seems relatively simple, each problem, nevertheless, presented its own peculiar challenge. While no claim is made that we have encountered and successfully solved all of the problems associated with the handling of finely divided submicron-size particles, what has been accomplished is to establish a procedure for a step-by-step operation of this facility, which produces the successful distribution tests shown in Fig. 13. Each new material handled in the future will most certainly present new problems, but with the limited experience that we have to date, the indication is that few materials are more difficult to handle than teflon. The results of future studies which require the modification of existing equipment, will be reported in subsequent reports.

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