T- 81425

N 69 19000

NASA CR 100186

PROPULSION DIVISION

SYSTEM ANALYSIS OF GELLED SPACE-STORABLE PROPELLANTS

CONTRACT NAS7-473, SA-I

Prepared for

Office of Advanced Research and Technology
National Aeronautics and Space Administration
Washington, D.C.

Summary Report 1038-02S July 1968

CASE FILE COPY

AEROJET
GENERAL TIRE
GENERAL

AEROJET-GENERAL CORPORATION
SACRAMENTO, CALIFORNIA

SYSTEM ANALYSIS OF GELLED SPACE-STORABLE PROPELLANTS

CONTRACT NAS7-473, SA-1

Prepared for

Office of Advanced Research and Technology
National Aeronautics and Space Administration
Washington, D.C.

Summary Report 1038-02S July 1968

AEROJET-GENERAL CORPORATION 43234T
A SUBSIDIARY OF THE GENERAL TIRE & RUBBER COMPANY

FOREWORD

Contract NAS 7-473, SA-1, "System Analysis of Gelled Space-Storable Propellants," is being performed by the Aerojet-General Corporation at Sacramento, California. This summary report describes the accomplishments for the second year of the contract, from May 1967 through April 1968.

The second year's effort was performed by the Research and Technology Department, Dr. P. L. Nichols, Jr., Manager. The Aerojet Program Manager and Project Engineer was Mr. R. H. Globus. The oxygen difluoride gel formulation and testing studies were performed under the direction of Mr. Globus with the assistance of Messers P. D. Beadle, R. L. Beegle and J. A. Cabeal. Dr. J. M. Adams designed the light extinction equipment required for measuring the particle size of chlorine trifluoride particles and reduced the data obtained. Mr. W. V. Timlin performed the analytical study on the effect of gelation on the in-space storability of cryogenic propellants. Mr. J. J. Bost assisted in the design of the flow viscometer for measuring the flow properties of gelled oxygen difluoride. Dr. S. D. Rosenberg and Dr. E. M. Vander Wall acted as consultants during this program.

The NASA Project Manager for Contract NAS 7-473, SA-1, is Mr. J. Suddreth, NASA Headquarters, OART; the NASA Technical Manager is Mr. D. L. Young of the Jet Propulsion Laboratory.

Report 1038-02S

ABSTRACT

A rapid condensation method for the preparation of fine particles of chlorine trifluoride was developed. These fine particles were used to gel liquid nitrogen and liquid oxygen difluoride.

Ten pounds of liquid oxygen difluoride gelled with solid chlorine trifluoride was prepared. The structure index and flow properties of these gels were measured. These gels exhibited extensive shear thinning with increasing shear rate. Within the limits of the tests performed, the OF_2/ClF_3 gels appear to be chemically and mechanically stable.

It was found that liquid nitrogen gelled with solid chlorine trifluoride possesses static and dynamic flow properties which closely approximate those of the OF_2/ClF_3 gels. Thus, N_2/ClF_3 gels may be used as suitable simulants for OF_2/ClF_3 gels.

TABLE OF CONTENTS

TABLE OF CONTENTS (Cont.)

TABLE LIST

TABLE LIST (Cont.)

APPENDIX I

FIGURE LIST

FIGURE LIST (Cont.)

APPENDIX

APPENDIX (Cont.)

SECTION I

INTRODUCTION

1. OBJECTIVE

The objective of this study was to gel oxygen difluoride (OF_2) using fine particles of a fully fluorinated oxidizer such as chlorine trifluoride (ClF_3) or chlorine pentafluoride (ClF_5). After the accomplishment of this primary objective, the secondary objectives were to develop the techniques required for the measurement of the structure and the flow properties of cryogenic gels, and to measure these properties of gelled OF_2.

2. ADVANTAGES OF GELATION

Gelled propellants offer the following improvements in rocket system performance: (1) positional stability in a zero-g environment, and (2) reduced sloshing. The first two advantages are of considerable importance when propellant systems are being considered for deep space missions. The disadvantages of gelled propellants are: (1) a reduction in performance; and (2) an impairment in restart capability if an inert, high-melting particulate material is used as the gelling agent. However, by the proper selection of gelling agents, performance losses can be reduced to a point that they are no longer significant and impairment of restart capability can be eliminated.

a. Positional Stability and Reduced Sloshing

The structure a gel possesses when at rest gives the material the properties of a coherent semi-solid. Consequently, until a shear force is applied, the material will remain in its original position. This property of positional stability which is imparted to a liquid by gelation offers

I, 2, Advantages of Gelation (cont.)

an important advantage to liquid propellants being considered for deep-space applications. It eliminates the need for a positive expulsion device, with its complexity and, in the case of fluorine-containing oxidizers, serious material problems, to ensure that the propellant will be delivered to the engine.

The reduction of sloshing caused by gelling a propellant was demonstrated by Aerojet under Contract NAS 7-473 (Reference 1). It was experimentally demonstrated that: (1) the resonant frequency for gels occurred at higher values than for ungelled liquid; and (2) the motion decayed in two cycles or less in gelled systems compared to 30 to 40 cycles for ungelled liquids.

b. Particulate Gelants

The reactivity of OF_2 prohibits the use of organic gelling agents. Consequently, particulate gelling agents prepared from materials that are not reactive in OF_2 are the only type of agent that can be considered for use with OF_2. A particle gels a liquid because of the surface properties of the particle. The ability to gel a liquid arises when the particles are small enough so that there is an attraction between the individual particles. Apparently, any particle, if it is small enough, possesses this attractive force which causes the particles to form interconnecting chains or networks that form micelles which trap the liquid. A gel-like material is the result. The property that makes this type of gelling agent uniquely distinctive is the fact that gel formation is independent of the chemical properties of the liquid. In other words, if the particles are small enough so that these chains or networks form, the particles will gel any liquid in which they are insoluble and nonreactive. Consequently the central problem of this program was the development of a technique to prepare fine particles, which would gel liquids, from the group of materials selected as potential gelling agents.

I, 2, Advantages of Gelation (cont.)

Two classes of materials are nonreactive in OF_2 and could serve as gelling agents; i.e., the inorganic fluorides and the fluorinated interhalogens. The disadvantages incurred by the use of inorganic fluorides are: (1) they possess no propellant value and consequently significantly degrade propellant performance; and (2) they are in many cases nonvolatile and would remain as a residue after propellant evaporation from the propellant lines and injector manifold during coast sequences. These residues are expected to impair restart capability.

The fluorinated interhalogens do not possess these advantages. Being excellent oxidizers in their own right, performance losses caused by gelation can be reduced to a point that they are not longer significant. Theoretical performance calculations were made to confirm this point. The specific impulse versus mixture ratio of OF_2, 0.9 OF_2 + 0.1 ClF_5 and 0.9 OF_2 + 0.1 ClF_3 has been calculated using diborane and methane as the fuels. The results of the calculations are presented in Figures 1 and 2. The calculations indicate that the theoretical performance degradation is 4 sec if ClF_5 is the gelling agent and 6 sec if ClF_3 is the gelling agent and diborane is the fuel. In the case of methane, the theoretical performance degradation is 3 sec when ClF_5 is the gelling agent or 5 sec when ClF_3 is the gelling agent. As the gelling agent concentration decreases, this difference become less.

The fluorinated interhalogens possess high vapor pressures at ambient temperature (25°C). Consequently, after shutdown, they will rapidly volatilize along with the oxygen difluoride remaining in the propellant lines and oxidizer manifold and will not impair restart capability.

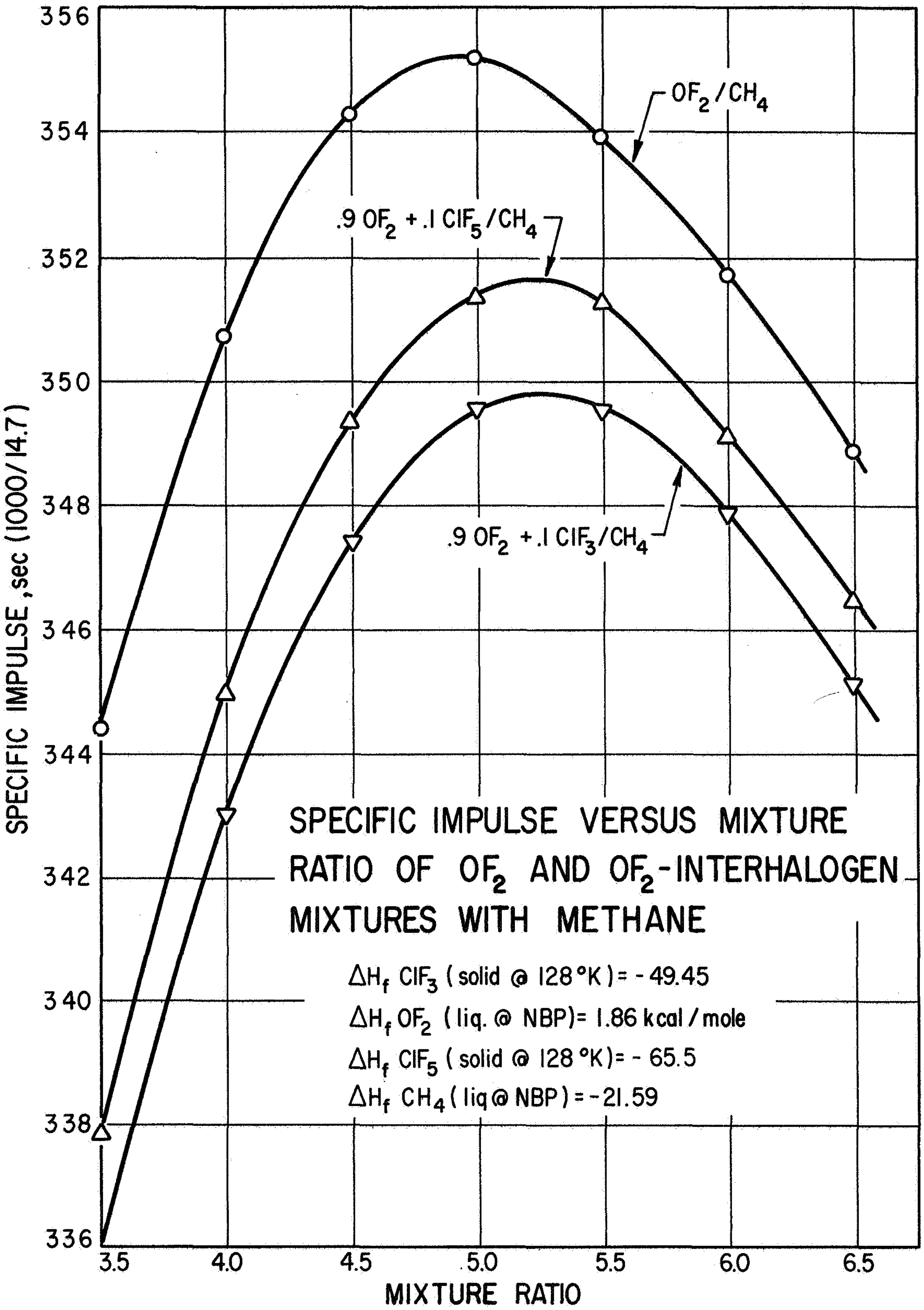


Figure 1. Specific Impulse versus Mixture Ratio of OF_2 and OF_2-Interhalogen Mixtures with Methane

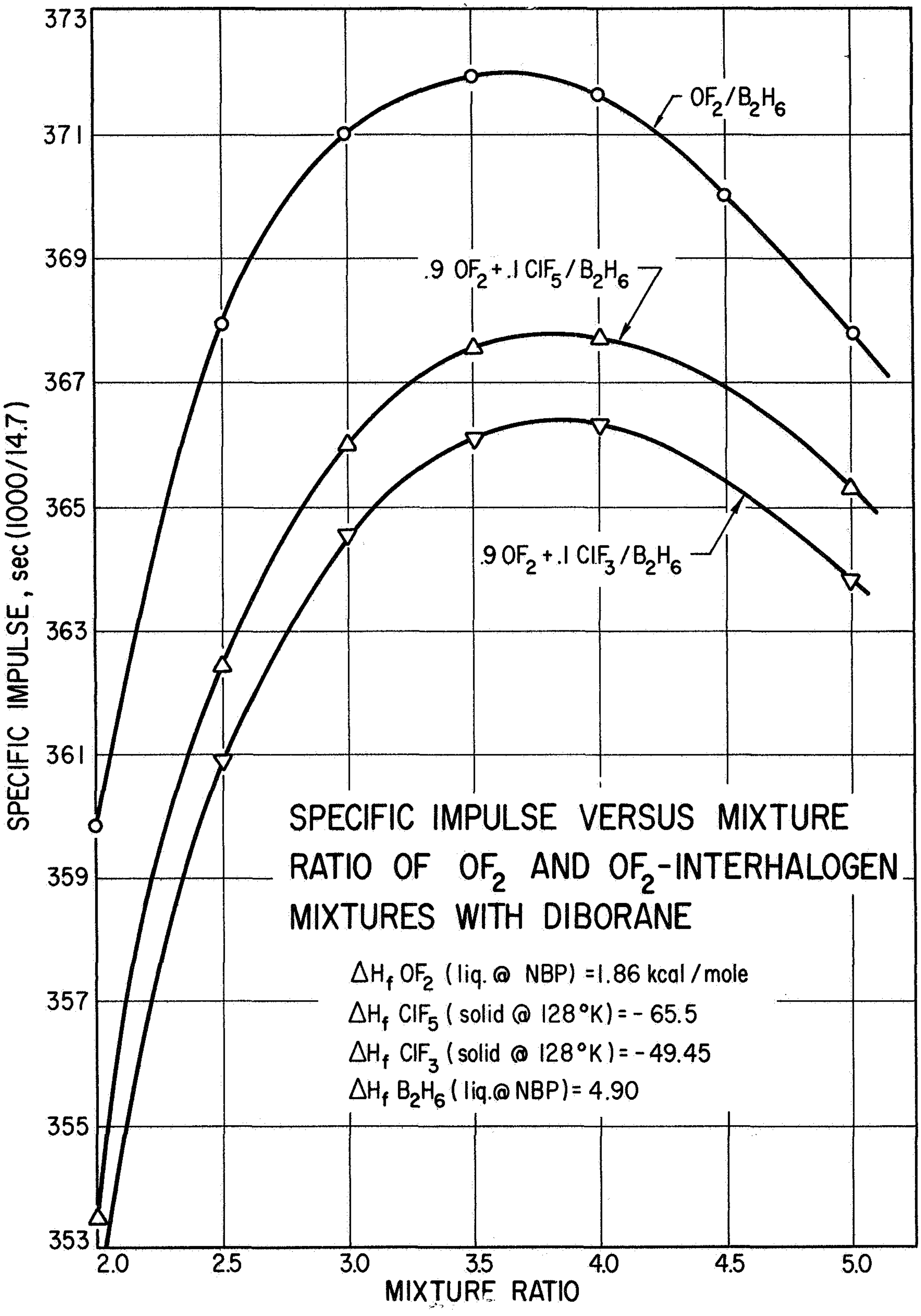


Figure 2. Specific Impulse versus Mixture Ratio of OF_2 and OF_2-Interhalogen Mixtures with Diborane

SECTION II

SUMMARY

The technical effort on this program was divided into four tasks. The work performed and the results and their significance are summarized below, as are recommendations for future work.

Liquid oxygen difluoride was gelled with fine particles of solid chlorine trifluoride. Three batches of gelled oxidizer, totaling 10 lb, were prepared.

The structure index of gelled oxygen difluoride was measured at several gelant concentrations. An excellent gel was obtained at a chlorine trifluoride concentration of 5.5 wt% and a structure index of 1400 dynes/cm^2. Experience with other gelled materials indicates that gels with this degree of structure are stable. As is typical of particulate gels, it was found that the degree of structure of $OF_{2(l)}/ClF_{3(s)}$ gels changed rapidly with changing gelant concentration.

The flow properties of gelled oxygen difluoride were measured in a novel flow viscometer. A gel containing 5.0 wt% chlorine trifluoride concentration exhibited an apparent viscosity ranging from 0.09 to 13.9 poise as the pressure drop ranged from 4.63 to 1.67 psig, respectively. The rapid decrease in apparent viscosity with increasing shear rate indicates that under flow conditions the propellant properties will be essentially the same as the neat liquid. This means that it will be possible to transfer the gelled propellant in conventionally designed pumps and lines.

The fine chlorine trifluoride particles were prepared by diluting the oxidizer vapor with helium and rapidly injecting the gaseous mixture through a small orifice positioned beneath the surface of liquid nitrogen. Oxygen difluoride gels were prepared by removing the liquid nitrogen by evaporation and adding liquid oxygen difluoride to the fine chlorine trifluoride particles.

II, Summary (cont.)

Liquid nitrogen was gelled with fine particles of chlorine trifluoride. The structure index of gelled nitrogen was measured; the rate of change of the structure index of gelled nitrogen was similar to that of gelled oxygen difluoride at comparable gelant concentrations. The flow properties of gelled nitrogen were measured; the rate of change in apparent viscosity with changing shear rate was similar to that of gelled oxygen difluoride at comparable gelant concentrations.

A measurement of the particle size of the solid chlorine trifluoride using a spectral technique indicated that, over the range of experimental parameters investigated, no measurable change in particle size distribution occurred during or after the preparation of solid chlorine trifluoride in liquid nitrogen. The sum total of these results indicate that liquid nitrogen gelled with fine chlorine trifluoride particles provides an excellent simulant for liquid oxygen difluoride gelled similarly.

The attempt to develop similar cryogenic gels by the substitution of chlorine pentafluoride for chlorine trifluoride did not succeed. The most likely cause of failure is the suspected solubility of solid chlorine pentafluoride in liquid nitrogen.

Based on the results obtained on this program, it is recommended that: (1) the properties of gelled oxygen difluoride be determined in detail; (2) the gelation of diborane, using the techniques developed on this program, be undertaken; and (3) a companion program for the gelation of FLOX and methane be initiated and the resulting gels characterized in detail.

SECTION III

TECHNICAL DISCUSSION

This study was a continuation of Contract NAS 7-473, System Analysis of Space-Storable Propellants, which consisted of five tasks: Task I--Preliminary Investigations; Task II--preliminary Analysis; Task III--Component Design Analysis; Task IV--System Design Analysis; and Task V--Documentation. This continuation was divided into four technical tasks: Task VI--Comprehensive Review; Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel; Task VIII--Gelation of Oxygen Difluoride with Fluorinated Oxidizers; and Task IX--Measurement of Engineering Properties of Gelled Oxygen Difluoride. The work performed and the results and their significance are discussed in this section.

1. TASK VI--COMPREHENSIVE REVIEW

During the original program, Aerojet-General conducted a literature search on gelled propellant technology. Approximately 400 technical reports were surveyed to determine those most applicable to the areas of interest on the present program. Of these reports, more than half were considered directly useful to this program and were abstracted. The abstract was presented in the first quarterly report prepared for Contract NAS 7-473, SA-1, "System Analysis of Gelled Space-Storable Propellants," Report 1038-01-Q1, 1 August 1966 (Ref. 1

This abstract was reviewed to select those reports pertinent to this study. This review considered the following specific areas:

(1) Potential gelling agents prepared by the in situ condensation of gases or volatile liquids in the propellant.

(2) Handling procedures used with oxygen difluoride.

(3) Engineering measurements made or attempted on cryogenic gels and other selected particulate gels.

In addition, the Aerojet-General technical library was searched for reports dated after May 1966 under the following headings: gelling of cryogenic propellants, gelling of cryogenic and storable oxidizers, Technidyne, Thiokol Chemical Corporation, Reaction Motors Division, and NASA Lewis Research Center, Advanced Rocket Technology Branch. The results of this review are presented below.

a. Preparation of Gelling Agents

The purpose of this research is to develop the techniques required to gel OF_2 with the fine particles of fluorinated oxidizers. These fine particles are to be prepared by freezing oxidizers such as chlorine trifluoride or chlorine pentafluoride. The nature of the proposed gelling agents requires that they be frozen in situ in the liquid OF_2. Consequently, the literature review was restricted to those programs in which attempts were made to prepare gelling agents by the in situ condensation of a gas or volatile liquid, and to the techniques available for dispersing or deagglomerating the particles after they have been prepared.

The feasibility of in situ gelation of liquid hydrogen using frozen particles of other fuels was investigated by Technidyne (Reference 2). The initial series of experiments were conducted using liquid N_2 as a simulant for liquid hydrogen. In the first experiment, n-pentane was introduced into liquid N_2 by bubbling in gaseous nitrogen saturated with n-pentane vapor. Fine crystals of pentane formed and showed a thickening effect, but condensation in the feed tube caused clogging. The experiment was repeated using propane. Apparently, it was believed that propane did not solidify, perhaps due to super-cooling. Nucleation sites for the crystallization of the propane were provided by adding small amounts of pyrogenic silica or acetylene black to the liquid N_2 prior to introduction of the propane. The propane coagulated in the experiment with the silica, but in the experiment with acetylene black, a gel formed. Attempts were made to crystallize methane in liquid N_2, but it was found that the methane was soluble in the liquid N_2. Fine crystals or particles of ammonia were prepared in liquid N_2. Particle size appeared to be (no actual measurement reported) less than 0.1μ; however, no gel was formed.

Attempts were made to gel liquid hydrogen with methane (References 2 and 3). The methane was diluted with helium and introduced by bubbling it into the liquid hydrogen. Unfortunately, condensation in the tube formed a plug. In a second experiment, it appeared that submicron particles were obtained; however, no gel was obtained. The type of experiment just described has recently been done at Los Alamos (Reference 4). Suspensions of frozen methane in liquid hydrogen were obtained. However, as the suspension was agitated, the particles agglomerated and settled.

Reaction Motors Division, Thiokol Chemical Corporation, attempted to gel OF_2 with fine particles of arsenic pentafluride (Reference 5). The arsenic pentafluoride was condensed from the gas phase in liquid OF_2. No gel formed, probably because the particles were too large.

All the agencies cited above concluded that the condensation of vapors to form fine particles was a promising approach to the preparation of particulate gelling agents. However, in no case was extensive experimental work conducted. The results of this portion of the review indicate that there was little prior art regarding the <u>in situ</u> condensation of vapors as a technique for preparing micron-size particles. However, it is apparent that the device used to inject the vapor to be condensed into the cryogenic liquid must be heated in order to prevent clogging of the gas injection tube.

b. Handling Procedures Used with Oxygen Difluoride

On a laboratory scale, oxygen difluoride is commonly transferred as a gas in a vacuum system and liquified at the point where the experimental work is planned (References 5, 6, 7, 8, 9, and 10). In order to reduce toxicity problems and in some cases explosive hazards, the quantities handled are usually quite small, 2 gm or less. Disposal of the OF_2 is usually handled either by venting through a hood if quantities are small, or by burning if larger quantities are involved.

While the use of a vacuum system for tranferring reactive materials is common, it is accompanied by difficulties. Quantities are strictly limited and the experimental work is frequently interrupted by leaks. Therefore, in this research it was planned to dilute the interhalogens with helium and to transfer with a driving pressure; OF_2 would be transferred by its own pressure.

As the quantities of OF_2 commonly used in experimental work, 1 and 10 gm, are completely inadequate for the evaluation of gel properties, it was decided to handle OF_2 remotely. The approach permitted the use of reasonably large quantities of OF_2, up to 1 liter, with safety. In addition, it was decided that initial work would be done with liquid N_2 so that experimental procedures could be checked out before work with OF_2 was attempted.

c. Engineering Measurements on Selected Particulate Gels

(1) Measurement of Static Properties

Two commonly measured static properties of a gelled system are yield stress and mechanical stability. Yield stress is a measurement of the degree of structure possessed by a gel when at rest. Mechanical stability measurements are used to determine whether the solid suspended in a gelled propellant settles during storage and/or if an exudate forms. In this phase of the propellant development program, only the degree of structure possessed by the gel is of interest.

The measurement of yield stress is the standard measurement used to determine the degree of structure of a gelled propellant. Technidyne calculated the yield stress of liquid hydrogen gelled with a particulate gelling agent by measuring the maximum torque required to start the rotation of the star rotor of a Rotoviscometer (Reference 2). They also measured the yield stress of these gels with cone penetrometers (Reference 14). Aerojet-General attempted to measure the yield stress of gelled liquid oxygen with a rising sphere rheometer. It was found that a steady-state force could not be obtained (Reference 12). The failure of the rising sphere rheometer to measure the yield stress of liquid oxygen gelled with a particulate gelling agent is probably due to the fact that a shear rate is applied to the gel during the measurement.

The results of the work reviewed indicated that either a Rotoviscometer or a cone penetrometer are feasible means of measuring the degree of structure possessed by a cryogenic gel. However, the reactive and toxic properties of OF_2 are such that experimental problems would be encountered.

(2) Measurement of Flow Properties

The measurement of the flow properties of propellants gelled with particulate gelling agents has been investigated by several agencies. Reaction Motors Division, Thiokol Chemical Corporation, attempted to measure the flow properties of OF_2 gelled with various particulate gelling agents with a capillary viscometer (References 5, 6, 7, 8 and 9). It was found that their instrument was not suitable. The gels separated under pressure and/or cored during the measurements and no useful results were obtained. Technidyne measured the flow properties of liquid hydrogen gelled with a particulate gelling agent with a Rotoviscometer (References 2, 3, 9 and 11). The results obtained were reproducible and showed that the gels shear-thinned. Aerojet-General measured the apparent viscosity of liquid oxygen gels at various shear rates with a Brookfield Viscometer (Reference 12) and good reproducibility was obtained. Aerojet-General has also studied the flow of water gelled with a particulate gelling agent from a small tank (Reference 13). It was found that a baffle across the outlet significantly reduced coring and hang-up. It was also concluded that the configuration of the tank outlet end closures would significantly affect the expulsion behavior of the particulate gel.

The results of this review clearly show that rotational viscometers have been the principal type of instruments which have been used to date to measure the flow properties of cryogenic liquids gelled with particulate gelling agents. However, the research by Union Carbide on the rheological properties of fluid hydrogen slush (Reference 15) and the recently published comments from the Cryogenics Division of NBS indicate that it is possible to measure the rheological properties of slushes in flow systems. Because of the encouraging results obtained by Union Carbide the NBS and because rotational viscometers do not provide information that is directly relatable to the behavior of the gel under flow conditions, it was planned to develop a technique to measure the flow properties of gelled OF_2 with a flow viscometer. This technique would provide quantitative data regarding flow properties and qualitative information regarding techniques required to expel the gelled OF_2 from the tanks.

d. In-Space Storability

In the course of this program, an analytical study was completed on the effect of gelation and the effect of the gelling agent selected on the storability of a cryogenic propellant in a space environment. The propellant considered was OF_2. The following paragraphs present a discussion of the assumptions made and the results of the heat transfer calculations performed.

The OF_2 would be stored in space at essentially zero acceleration and would be subjected to solar thermal radiation. A storage tank configuration has not been specified; for the sake of comparison, the analyses were done for 1.0 sq ft of tank surface oriented normal to the solar radiation. An optimistic tank surface condition was considered; i.e., a solar absorptivity of 0.1 and an emissivity of 1.0 were used.

The results of the comparison conducted are that neat OF_2 boils off most readily and that OF_2 gelled with an inert agent boils off least readily. Gelled OF_2 gelled with either ClF_3 or ClF_5 falls between these extremes, with OF_2 gelled by means of ClF_3 having a lower boiloff rate than OF_2 gelled with ClF_5. The conclusions drawn from this analytical study are: (1) if boiloff is a consideration in an unshielded tank, gelled OF_2 may offer an advantage over neat OF_2; and (2) if it is desired to use an oxidizer as a gelling agent, ClF_3 is better than ClF_5 from the point of view of boiloff.

In order to arrive at this comparison, two relatively simple thermal models were used, one for neat OF_2 and another for gelled OF_2. Both models considered a 1.0 ft^2 tank wall area with a solar absorptivity of 0.1 and an emissivity of 1.0 oriented normal to the solar radiation. For the neat model, it was determined that the OF_2 wetting the wall would experience nucleate boiling and hence the wall temperature could be approximated by the OF_2 saturation temperature [230°R at one atm (assumed tank pressure)]. Knowing this wall temperature, the heat radiated to space could be calculated and, hence, the net heat into the OF_2 (0.76×10^{-4} Btu/in.^{2}-sec) was known, i.e., absorbed solar radiation ($0.1 \times 8.52 \times 10^{-4}$ Btu/in.^{2}-sec) less heat reradiated to space (0.092×10^{-4} Btu/in.^{2}-sec). It was assumed that the OF_2 in the tank was saturated and, therefore, this net heat evaporated OF_2. With a heat of vaporization of 82 Btu/lb, this corresponds to 11.5 lb/day for 1.0 ft^2 of wetted surface area.

III, I, Task VI--Comprehensive Review (cont.)

The thermal model for the gelled OF_2 was slightly more complicated. It is shown in Figure 3. The basic assumption in setting up the model is that, as the OF_2 is evaporated at the tank surface, a gap is formed between the evaporating surface of the gelled OF_2 and the tank wall, and that this gap is filled with the gelling agent matrix plus OF_2 vapor. The gap grows as OF_2 evaporates; the larger the gap, the more thermal insulation is provided for the remaining gelled OF_2. Three equations are used to define gap width (and correspondingly OF_2 evaporated) as a function of time. Referring to Figure 3, they are:

$$Q_{COND} = \alpha Q_{SUN} - Q_{RERADIATION} \qquad \text{(Eq 1)}$$

$$Q_{COND} = h_{FG} \frac{dX}{d\Theta} \qquad \text{(Eq 2)}$$

$$Q_{COND} = k_G \frac{T_W - T_{SAT}}{X} \qquad \text{(Eq 3)}$$

where:

Q_{SUN} = solar radiation (8.54×10^{-4} Btu/in.^{2}-sec)

Q_{COND} = heat conduction across gap

$Q_{RERADIATION}$ = $\varepsilon \, \sigma \, T_W^4$

ε = tank wall emissivity (1.0)

α = tank wall solar absorptivity (0.1)

σ = Stefan-Boltzman constant

T_W = tank wall temperature (no gradient assumed in wall)

h_{FG} = OF_2 heat of vaporization, 82 Btu/lb

ρ = OF_2 density, 95 lb/ft^3

X = gap width

Θ = time

k_G = gap material conductivity (approximated by fluorine vapor conductivity, 0.003 Btu/ft-hr°R)

T_{SAT} = OF_2 saturation temperature

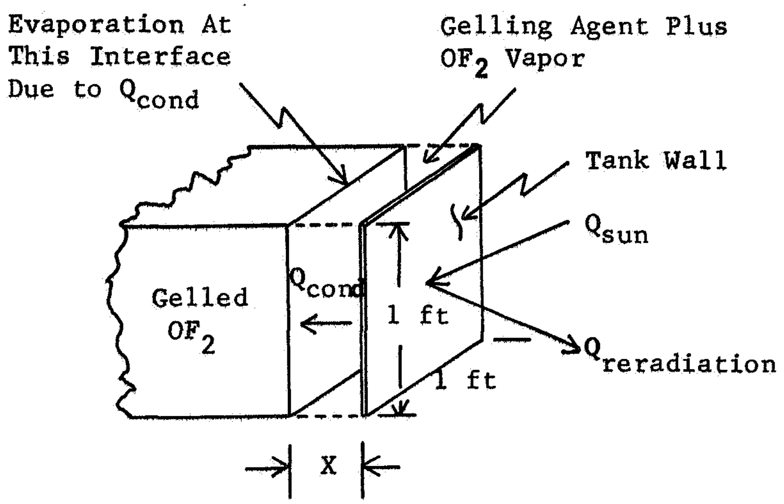


Figure 3. Thermal Model for Evaporation of Gelled OF_2

Using Equations 1 and 3, a relation is established between T_W and X. With this relationship, Equation 2 is then integrated to find the dependence of X upon Θ which defines evaporation with time as long as X is increasing. It is assumed that the gap width and tank wall temperature are increased until the melting point of the gelling agent is reached at the tank wall and then the gap widens while wall temperature remains constant. With the gap width constant, the evaporation rate is constant and is calculated from the net heat input to the tank wall, absorbed solar radiation minus reradiated heat. For OF_2 gelled with the inert agent, the gelling agent melting point is never reached and the gap width continues to grow providing more and more thermal protection for the OF_2. The ClF_5 gelling agent melts at -153°F and the ClF_3 melts at -105°F; therefore, the gap width is larger for the ClF_3 (0.3 in.) than for the ClF_5 (0.1 in.). Boiloff is, thereby, greater for the ClF_5 than for the ClF_3 gelling agent.

The model described above is idealized for the case of melting ClF_5 and ClF_3 gelling agents; however, it is belived to be conservative.

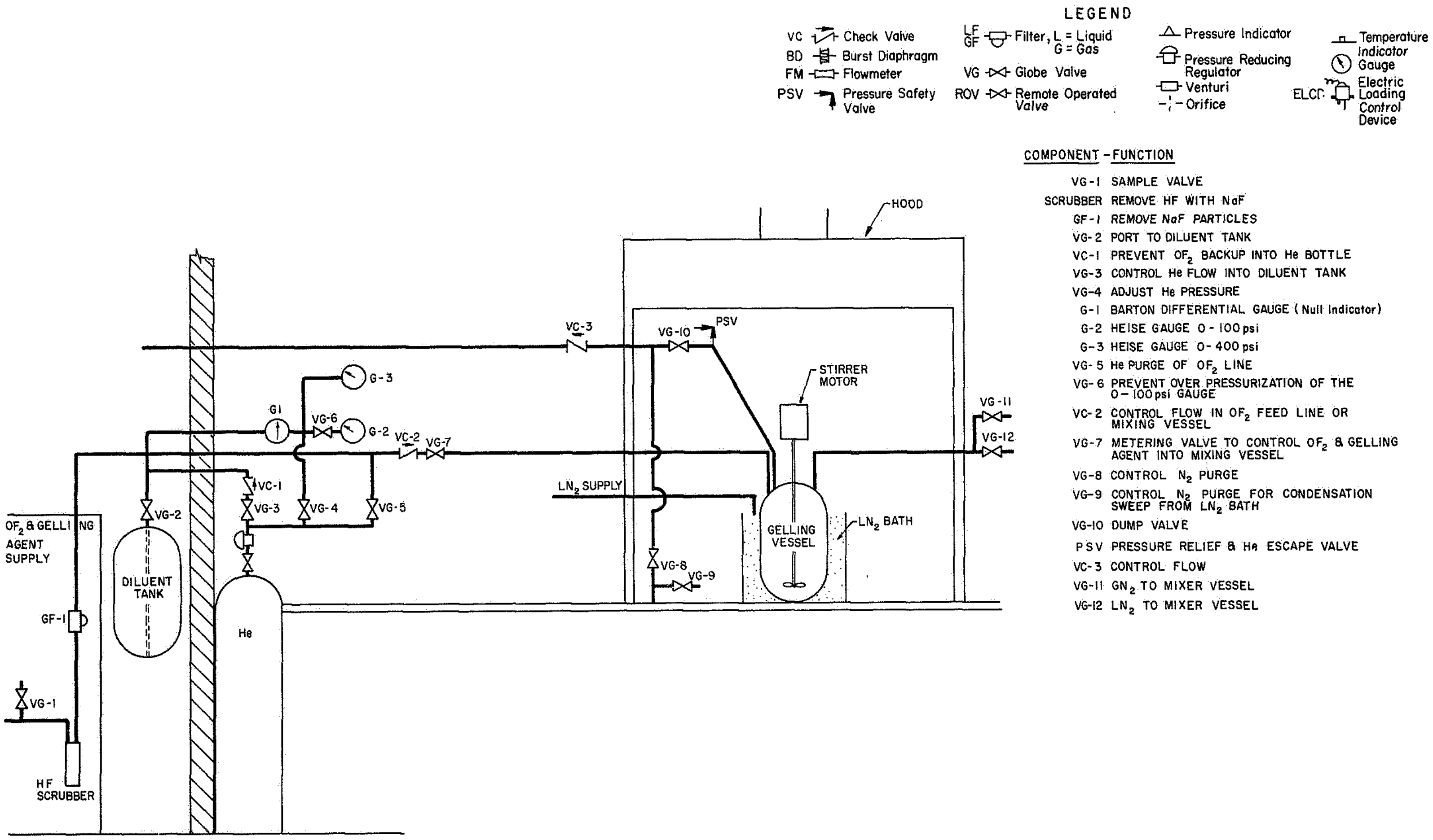
2. TASK VII--DEVELOPMENT OF TECHNIQUES TO GEL OXYGEN DIFLUORIDE AND TO MEASURE ENGINEERING PROPERTIES OF THE GEL

a. Design and Fabrication of Equipment

The design of the fluorine compound transferring equipment for preparing fine particles by freezing interhalogens was based on the following criteria: (1) the interhalogen was only to be handled as a gas until it was condensed as a fine particle; (2) the equipment must have the capability of handling liquid OF_2 which also would be handled as a gas until it was liquified; (3) the quantity of oxidizer on hand in the apparatus should not exceed the quantity required for a given experiment by more than 10%; (4) the equipment would have the capability of diluting the interhalogen with helium and be capable of varying the amount of diluent over a wide range; (5) the equipment should have the capability of measuring the pressure, flow rate and flow velocity at which the interhalogen was delivered to the vessel where it was condensed; and (6) the equipment must be flexible so that it could be modified easily and economically to meet changing requirements. Figure 4 presents a schematic of the equipment designed and fabricated to meet the above listed requirements.

All of the lines were fabricated from stainless-steel tubing. The gelling vessel and cap were of glass during the initial part of the program. During the latter part of the program, the glass cap was replaced with a cap fabricated from aluminum. This change was made because it was found that the glass cap was too fragile. The various reagent lines were attached to the cap by stainless-steel flex lines. All of the hardware was thoroughly cleaned with soap and water, rinsed with distilled water, pickled in a nitric acid-HF pickling solution, again rinsed with distilled water and dried. After drying, the hardware was flushed with methanol, then flushed with Freon 113 and again dried; it was then installed. During installation, special precautions were taken to keep the hardware clean. After installation, the system was passivated with ClF_3 vapors. After the initial ClF_3 passivation, the system was pressurized to working pressure with helium and exposed overnight to a mixture of helium and ClF_3. All valves in ClF_3 or OF_2 service were either 1/4-in. or 1/2-in. Control Component Valves Model No.'s MV6004T and MV6008T. Except for an occasional valve developing a small leak across the stem after long periods of service, no equipment problems were encountered.

The item of hardware which is used to add the gaseous gelling agent to the cryogenic liquid is called the injection tube. The tube contains a heater, the purpose of which is to ensure that the gelling agent does not freeze in the tube and clog it, a thermocouple for measuring gas temperature, and a nozzle. Figure 5 illustrates a typical injection tube. Early models had an asbestos



COMPONENT	FUNCTION
VG-1	SAMPLE VALVE
SCRUBBER	REMOVE HF WITH NaF
GF-1	REMOVE NaF PARTICLES
VG-2	PORT TO DILUENT TANK
VC-1	PREVENT OF_2 BACKUP INTO He BOTTLE
VG-3	CONTROL He FLOW INTO DILUENT TANK
VG-4	ADJUST He PRESSURE
G-1	BARTON DIFFERENTIAL GAUGE (Null Indicator)
G-2	HEISE GAUGE 0 - 100 psi
G-3	HEISE GAUGE 0 - 400 psi
VG-5	He PURGE OF OF_2 LINE
VG-6	PREVENT OVER PRESSURIZATION OF THE 0 - 100 psi GAUGE
VC-2	CONTROL FLOW IN OF_2 FEED LINE OR MIXING VESSEL
VG-7	METERING VALVE TO CONTROL OF_2 & GELLING AGENT INTO MIXING VESSEL
VG-8	CONTROL N_2 PURGE
VG-9	CONTROL N_2 PURGE FOR CONDENSATION SWEEP FROM LN_2 BATH
VG-10	DUMP VALVE
PSV	PRESSURE RELIEF & He ESCAPE VALVE
VC-3	CONTROL FLOW
VG-11	GN_2 TO MIXER VESSEL
VG-12	LN_2 TO MIXER VESSEL

Figure 4. Schematic - OF_2 Gelling Equipment

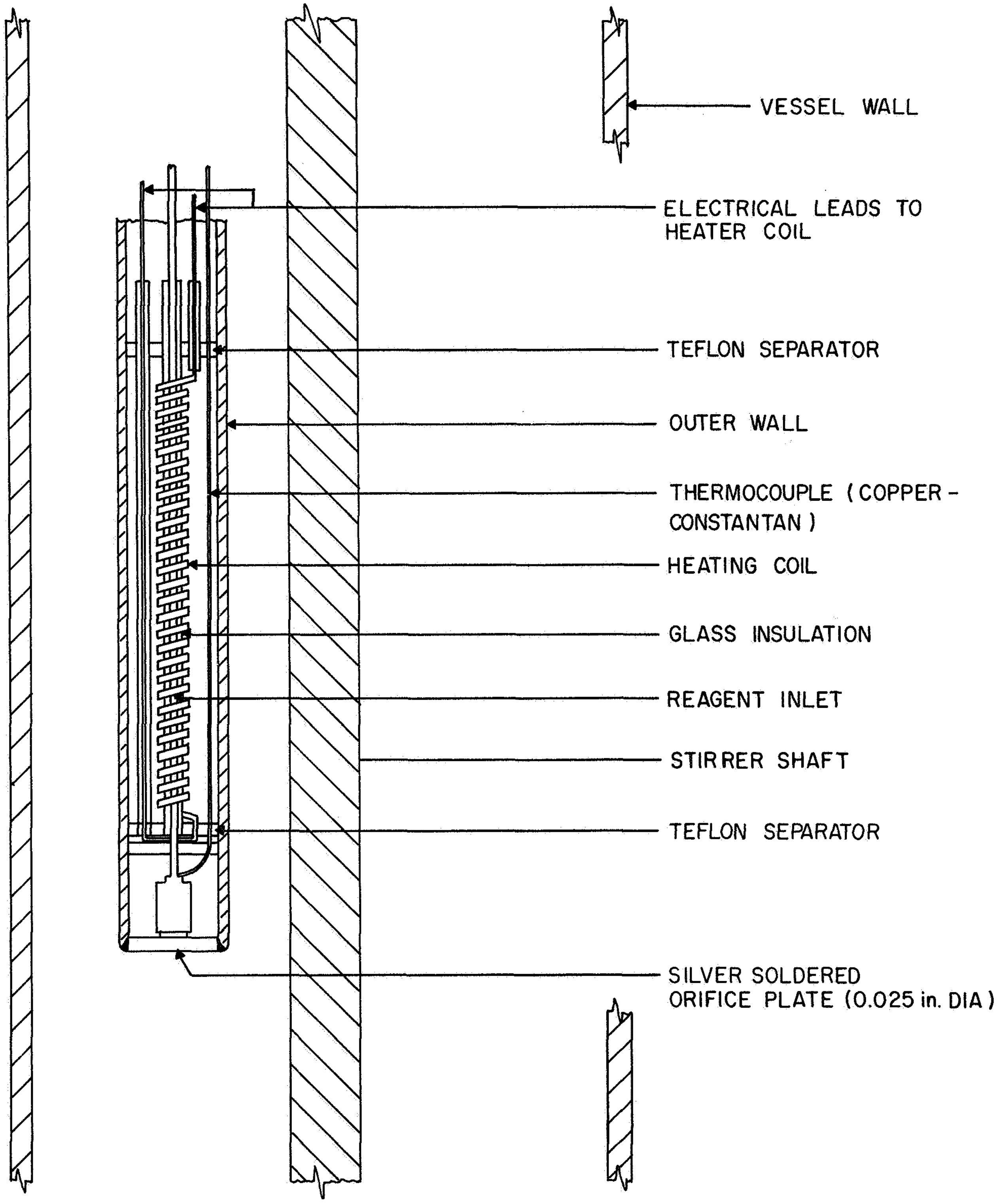


Figure 5. Injection Tube

liner and aluminum coil cover, and the orifice plate was attached by means of a copper cone, retainer and retainer cap. In later models, the asbestos liner and aluminum foil were eliminated. The orifice plate was silver soldered to the outer wall, eliminating the copper cone. This step greatly reduced the possibility of having the reagents leak into the heater section of the injection tube.

The outer portion of the injection tube external to the gelling vessel contained a helium inlet. This permitted the pressurization of the interior of the tube between the reagent tube and the outer wall, thus ensuring that there would be no leak into the heater section. Leakage into this section could not be permitted because the heater, insulation and potting compounds are not compatible with the gelling agent.

b. Development of Techniques to Prepare Fine Particles of Frozen Fluorinated Oxidizers

The fluorinated interhalogens were obvious choices as gelling agents, i.e., ClF_3 and ClF_5. The former was arbitrarily chosen for initial experimental work. To reduce the safety hazards, liquid N_2 was selected as a simulant for liquid OF_2.

(1) Preparation of Fine Particles of Chlorine Trifluoride

The first three experiments had two major purposes. First, to check-out the interhalogen delivery equipment, and second, to determine whether the injection tube design was satisfactory. In addition, the compatibility of optical equipment, which would be used later in the program with ClF_3, was determined. The fluorine compound delivery equipment was satisfactory and the final inlet tube design, described in the previous section, was developed during these experiments. The optical equipment was found to be compatible with ClF_3.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

The procedures used during the experiments to prepare fine particles of ClF_3 were essentially the same in all cases. The ClF_3 was diluted with helium and then injected into the liquid N_2 through a small orifice. The rate of injection was varied and the temperature of the injected gas mixture was monitored.

After the particles were made, their settling rate was measured so that particle size could be calculated. (Section III,2,c,(2) describes that portion of this program that was devoted to a precise study of the effort of experimental variables on particle size.) If the particles gelled the liquid N_2, the degree of gel structure was estimated by determining whether the gel, at various ClF_3 concentrations, would support a steel ball of known mass and diameter. (Section III,2,d,(1) describes the development of a more accurate method for assessing the degree of gel structure.) ClF_3 concentration was varied by adding or evaporating liquid N_2.

The results of the experimental work to prepare fine particles of ClF_3 are summarized in Table I. Specific features of each experiment and the conclusions drawn are discussed below.

Experiment 4

The inlet tube orifice was located 1/2 to 1-1/2 in. above the liquid N_2 level. A few large particles (14 microns) of ClF_3 formed, but most of the ClF_3 froze out on the vessel walls. It was concluded that the ClF_3-helium mixture must be injected below the liquid N_2 level.

Experiment 5

The inlet tube orifice was located approximately 6 in. below the liquid N_2 surface. Extremely fine particles were formed which apparently did not settle during an hour of observation. As the liquid N_2 was evaporated a gel formed; at a ClF_3 concentration of 17 wt% the gel would support a steel ball of 0.3596-in. dia. The ball weighed 4.047 gm. The calculated structure index is approximately 3000 $dynes/cm^2$.

TABLE I

THE PREPARATION OF FINE PARTICLES OF CHLORINE TRIFLUORIDE IN LIQUID NITROGEN

Exp. No.	Gelling Agent & Diluent	Dilution Vol of Gas / Vol of Gelling Agent	Orifice		Flow			Particles		Formed	Remarks
			Dia, in.	Location	Rate, $cm^3/sec^{(1)}$	Velocity, $cm/sec^{(2)}$	Reynolds No.	Made	Size, microns		
4	ClF_3 & He	39	0.025	1 1/2 in. above LN_2 level	120	2.8×10^4	8,000	Yes	15	No	No gel, concentration of ClF_3 in LN_2 not known because large quantity of ClF_3 froze out on gelling vessel walls.
5	ClF_3 & He	16.1	0.025	6 in. below LN_2 level	430	13.8×10^4	30,200	Yes	≤0.9	Yes	Gel formed; first indication at 6.1% ClF_3 by weight. At 17% ClF_3 by weight the gel would support a steel ball 0.4696 in. dia and weighing 4.047 gm. Structure Index ≈ 3000 dynes/cm^2. No visible settling of particles at any concentration. If the settling rate exactly equalled the LN_2 evaporation rate, the particle size would be 0.9 micron.
6	ClF_3 & He	---	---	---	---	---	---	---	---	No	Inlet tube clogged - No significant quantity of particles formed.
7	ClF_3 & He	---	---	---	---	---	---	---	---	No	Inlet tube clogged - No significant quantity of particles formed.
8	ClF_3 & He	35	0.025	6 in. below LN_2 level	340	10.9×10^4	24,500	Yes	0.9	Yes	Gel formed; first indication at 7.8% ClF_3 by weight. At 14.8% ClF_3 concentration gel would support a steel ball 0.3596-in. dia, 4.047 gm. Structure Index ≈ 3000 dynes/cm^2.
9	ClF_3 & He	36	0.025	6 in. below	669	21.6×10^4	48,300	Yes	≤0.7	Yes	Gel formed; first indication at 6.2% ClF_3 by weight. At 10.9% ClF_3 by weight the gel would support a steel ball 0.3596 in. dia and weighing 4.047 gm. Structure Index ≈ 2700 dynes/cm^2. No visible settling of the particles at any concentration. If the settling rate exactly equalled the LN_2 evaporation rate, the particle size would be 0.7 micron.
12	ClF_3 & He	35.4	0.025	6 in. below	370	12×10^4	26,800	Yes	0.8	Yes	Gel formed. At 11% ClF_3 by weight the gel would support a steel ball 0.321 in. dia and weighing 3.353 gm. Structure Index ≈ 2700 dynes/cm^2.

(1) cc at STP; T = 25°C
(2) At gelling vessel pressure.

LN_2 Analysis (by AGC) Experiment 4

Purity	99.96%
O_2, Ar	0.04%
PPM H_2O	14.5
PPM HC	<0.7

LN_2 Analysis (by vendor)

	Experiments 8 & 9	Experiment 5	Experiment 12
Purity	99.97% (including argon)	99.97% (includes argon)	99.97% (includes argon)
PPM HC	0.08	0.07	1.1
PPM Argon	470	470	470
PPM O_2	5	<5	5
H_2O	0.002 mg/liter	0.0009 mg/liter	0.0015 m/liter

Experiments 6 and 7

The inlet tube orifice was again located approximately 6 in. below the liquid N_2 surface. In both experiments the inlet tube clogged with frozen ClF_3 as soon as the ClF_3-helium mixture flow was started. It was concluded that clogging was caused by ClF_3 freezing during a drop in flow velocity in the inlet tube which occurred as the neat helium flow was cut off and the ClF_3-helium flow was started. Consequently, in Experiments 8 and 9 helium flow was maintained constant until the ClF_3-helium flow was established at the desired rate; then the neat helium flow was cut off.

Experiment 8

The inlet tube orifice was located approximately 6 in. below the liquid N_2 surface. Extremely fine particles were formed; however, the particles did slowly settle at the beginning of the experiment. No evidence of settling was observed after the concentration of the ClF_3 reached 7.8 wt%. A gel, with a structure index of approximately 3000 dynes/cm^2 formed at a ClF_3 concentration of 14.8 wt%.

Almost all of the liquid nitrogen was pumped off and fresh liquid nitrogen added. A second gel identical in appearance and properties to the first was formed. The fact that the particles did not loose their gelling ability after most of the liquid N_2 had been removed is of considerable importance. It means that the particles can be prepared in liquid N_2 and most of the liquid N_2 removed and liquid OF_2 added and gelled. This implies that particles can be prepared in advance, stored at the NBP temperature of nitrogen, and used at a later time. This ability should greatly reduce safety problems and simplify the eventual large-scale production of the particles.

Experiment 9

The inlet tube orifice was located approximately 6 in. below the liquid N_2 level. The flow rate of the ClF_3-helium mixture was increased above that of previous experiments. The particles formed appeared smaller than in the previous experiments. Visual observation indicated that a gel occurred at lower ClF_3 concentrations.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

Experiment 12

The purpose of this experiment was to demonstrate that the production of fine particles of ClF_3 which would gel liquid N_2 had been reduced to a routine laboratory operation. The inlet tube orifice was located approximately 6 in. below the liquid N_2 surface. Extremely fine particles were formed; however, the particles did slowly settle at the beginning of the experiment. No evidence of settling was observed after the concentration of ClF_3 reached 7.9 wt%. A gel with a Structure Index of approximately 3000 dynes/cm^2 formed at a ClF_3 concentration of 11%.

Figure 6 shows a suspension of ClF_3 particles in liquid N_2 shortly after it had been prepared. The liquid layer above the suspension is not an exudate. It is the liquid N_2 in the Dewar which surrounds the gelling vessel.

Figure 7 shows a small portion of the liquid nitrogen gelled with ClF_3 particles clinging to a spatula. It clearly shows that the gel does not flow at 1g unless sheared.

The series of experiments just described demonstrated that (1) particles of ClF_3 that would gel liquid N_2 could be prepared, (2) it is essential in the preparation of fine particles that the mixture of ClF_3 and helium be injected below the level of the liquid N_2, otherwise large particles formed and most of the gelling agent freezes out on the vessel walls, (3) the preparation of the particulate gelling agent has been reduced to a routine laboratory operation, and (4) the particles did not lose their gelling ability if the liquid N_2 was removed and more liquid N_2 added while maintaining the particles at the NBP of liquid N_2. The apparent particle size, on the basis of observation of the settling rate, was less than 1 micron.

During the course of this program, many liquid N_2 gels were prepared using ClF_3 particles as the gelant. In no case did the particles fail to gel liquid N_2 when the described procedure was used. Injection velocity

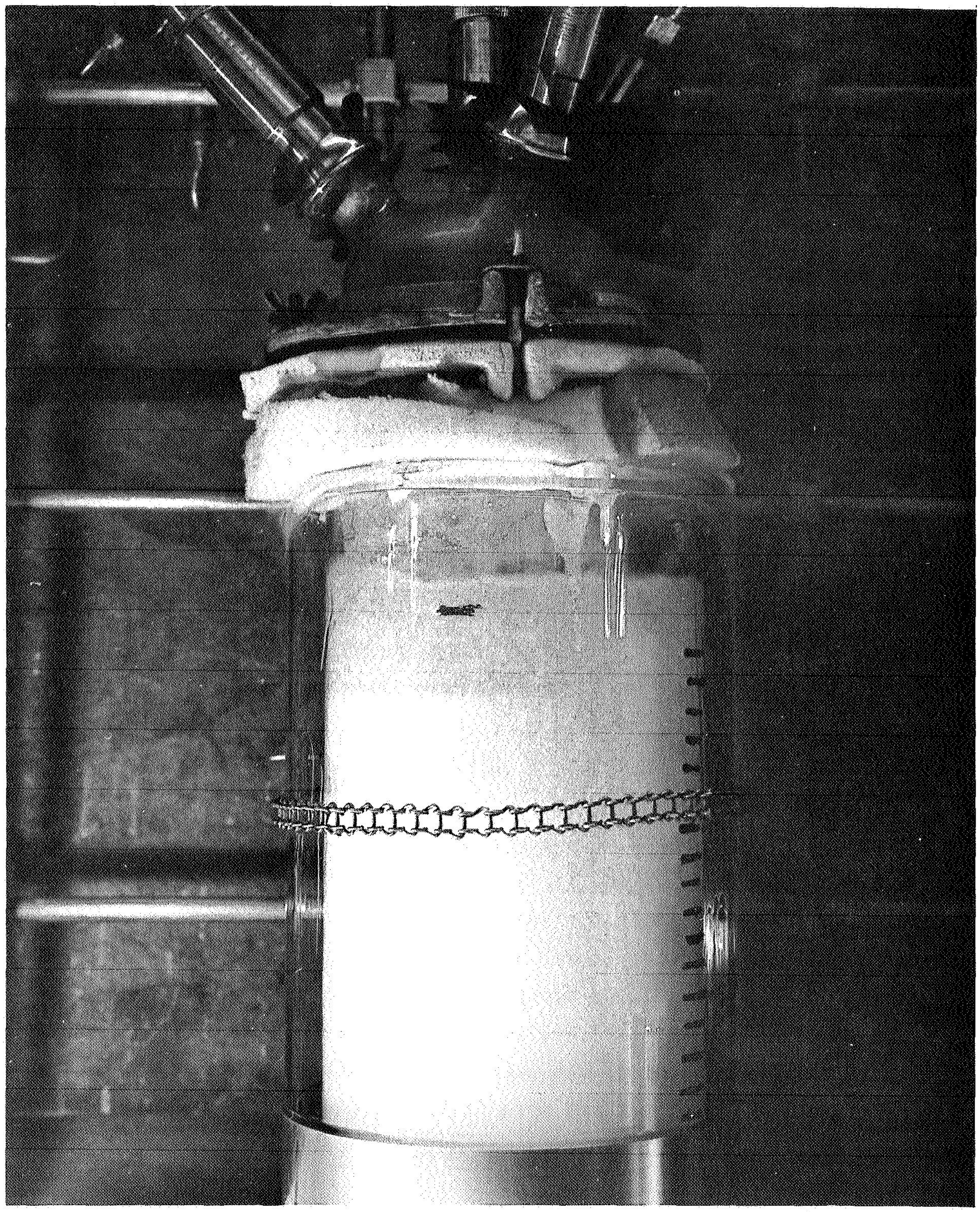


Figure 6. Suspension of ClF_3 Particles in Liquid Nitrogen

Figure 7. Liquid Nitrogen Gelled with ClF_3 Particles

ranged from 340 to 1900 cm^3(STP)/sec and dilution of the ClF_3 vapor ranged from 16 to 60 volumes of helium to 1 volume of ClF_3 vapor. The gels were formulated (1) by preparing the particles of ClF_3 in a large volume of liquid N_2 and then evaporating a portion of the liquid N_2 to obtain a gel of the desired thickness and (2) by preparing the quantity of particles required to give a gel of the desired consistency in the volume of liquid N_2 that was needed. Particles prepared from dilute mixtures of ClF_3 and helium appeared to form stiffer gels at a given gelling agent concentration. However, no quantitative data regarding this phenomenon were obtained, and the particle size measurements discussed in Section III,2,c,(2) did not confirm this observation.

(2) Preparation of Fine Particles of Chlorine Pentafluoride

A series of experiments were conducted to determine whether a particulate gelling agent could be prepared from ClF_5. The procedure used during the experiments to prepare fine particles of ClF_5 were essentially the same as the procedure used with ClF_3. The ClF_5 was diluted with helium and then injected into the liquid N_2 through a small orifice. The rate of injection was varied and the temperature of the injected gas monitored.

The results of the experimental work to prepare fine particles of ClF_5 are summarized in Table II. Specific features of each experiment and the conclusions drawn are discussed below.

Experiment 10

The inlet tube orifice was located approximately 6 in. below the liquid nitrogen surface. As the ClF_5-helium mixture was injected into the liquid nitrogen, particles of ClF_5 formed. On the basis of visual observation of the settling rate, the particle size was calculated to be 18 microns. No gel formed as the liquid N_2 was pumped off.

TABLE II

THE PREPARATION OF FINE PARTICLES OF CHLORINE PENTAFLUORIDE IN LIQUID NITROGEN

Experimental Parameters

Exp. No.	Gelling Agent & Diluent	Dilution Vol of Gas / Vol of Gelling Agent	Orifice Dia, in.	Orifice Location	Flow Rate, $cm^3/sec^{(1)}$	Flow Velocity, $cm/sec^{(2)}$	Flow Reynolds No.	Particles Made	Particles Size Microns	Gel Formed
10	ClF_5 & He	46	0.025	6 in. below LN_2 level	749	24.2×10^4	56,800	Yes	18	No
11	ClF_5 & He	91	0.025	6 in. below LN_2 level	583	18×10^4	33,400	Yes	23	No

It was concluded that possibly because of the substantially lower melting point of the ClF_5 (-103°C) compared to ClF_3 (-76°C), freezing of the particles was occurring at a substantially lower rate and the particles had time to grow.

Experiment 11

To reduce the quantity of ClF_5 present during the formation of the particles, thereby encouraging the formation of smaller particles, the dilution of the ClF_5 with helium was increased to the maximum capacity of the equipment and the injection rate was decreased.

As the ClF_5 helium mixture was injected into the liquid nitrogen, particles of ClF_5 formed. On the basis of visual observation of the settling rate, the particle size was calculated to be 23 microns. No gel formed as the liquid N_2 was pumped off.

The failure of the condensation technique for preparing fine particles of ClF_5 which would gel liquid N_2 has two possible causes. The first and most likely cause is that ClF_5 which melts at -103°C, has a slight solubility in liquid N_2. If a slight solubility exists, particle growth and a loss of gelling ability would occur as particle growth at the expense of the smaller particles is normal in liquid-solid systems where a slight solubility exists.

The second possible cause is that the particles of ClF_5 were forming at a slower rate because of the lower melting point of ClF_5. Consequently, the particles had an opportunity to grow to a relatively large size before growth was stopped. As the results of the analytical study of the effect of gelation on in-space storability had shown that ClF_3 gels possessed superior storage properties (see Section III,1,d), and the performance calculations indicated only a slight improvement in performance with ClF_5 compared to ClF_3 (see Section I,2,b), it was decided to discontinue work with ClF_5.

c. Particle Size Measurements

It was recognized that the ability to measure the size of the particles formed would be a valuable aid in directing the course of this program. It could provide a means for determining if changing an experimental variable was improving the quality of the particles prepared (e.g., the variable change resulted in the preparation of a smaller particle). Consequently, early in the program, a study was initiated to evaluate the various methods available for measuring particle size. After the evaluation was completed, the most applicable method was developed.

(1) Evaluation and Selection of Technique

The first technique considered was a light extinction procedure for measuring the settling rate of the particles and subsequently calculating particle size from Stokes law. The second technique considered was a modified BET procedure for measuring the surface area of the particles with the subsequent calculation of their size.

The light extinction procedure is a technique for determining the settling rate and homogenity of a polydispersion of solid particles in a liquid. It depends on the interaction of the dispersed particles with a monochromatic beam of light traversing the medium. Once the settling rate has been determined, particle size can be calculated from Stokes law.

The light passes to the gelling vessel where it is rendered parallel and passed into the liquid medium by means of an optical train. The transmitted light beams are received by receptor optics and passed to an optical sampler; the beam is continuously compared to a reference beam. This comparison is necessary to preclude any bias resulting from changes in lamp intensity or phototube sensitivity which can occur during the relatively long duration of an experiment.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

The electrical signals from the phototubes would be monitored on a standard oscillograph or visicorder. Because of the long duration of an experiment, the oscillograph would be operated using short pulses separated by relatively long quiescent periods. An automatic timer would be used for this mode of operation.

The second technique considered was modification of the BET procedures. It has been suggested that the adsorption isotherms exhibited by various gases at temperatures not far removed from their condensation point occurs because van der Waal forces are predominant under these conditions. Also, the same phenomenon permits multimolecular adsorption layers to be formed at successively higher adsorbate pressures. Consequently, if the quantity of gas adsorbed is measured at several pressures and the results plotted and extrapolated to zero pressure, it is possible to estimate the quantity of gas required to form a monomolecular layer on the particle. With this information it is then possible to calculate the surface area of a unit mass of particles.

The light extinction procedure was the method selected for measuring the particle size of the particles prepared. Appendix I, Light Extinction Measurements of Settling in Particulate Containing Liquids, presents a complete discussion of theory-principles of operation, apparatus, results, conclusions, and references. In addition, the particle size distributions of ClF_3 particles measured are presented for particles greater than 1 micron in Table I and less than 1 micron in Table II of Appendix I. The results obtained and their significance are discussed later in this report.

Appendix II presents the results of an analytical study of the feasibility of using the modified BET procedure which was briefly discussed above. It was concluded that the procedure was not applicable because, under the experimental conditions anticipated, the particles would not adsorb and desorb sufficient quantities of gas to permit meaningful measurements.

The possibility of measuring the settling rates of the particles, and subsequent calculation of particle size from Stokes law by measuring the change in intensity of a beam of radiation passing through the sample was also considered. The source of radiation could be one of several radioactive materials. However, this approach did not appear to offer any advantages over the light extinction procedure and would require more complex readout equipment.

(2) Experimental Measurements

A group of 11 experiments was conducted with the light extinction equipment to determine the manner in which injection velocity and the dilution ratio of helium to ClF_3 affect the size of the solid ClF_3 particles prepared by the condensation technique. The volume ratio of ClF_3 to helium ranged from 1:20.5 to 1:74.4; the injection velocity ranged from 168 to 1500 cm^3(STP)/sec. The final concentration of solid ClF_3 in the liquid N_2 varied from 0.08 to 1.84 gm/1. The results of these experiments are discussed in the following paragraphs.

Experiment 16

The purpose of this experiment was to check out the equipment under actual operating conditions. The injection velocity of the ClF_3-helium mixture was 320 cm^3 (STP)/sec and the quantity of ClF_3 added was 0.34 gm. The data obtained were not considered satisfactory because the optics had become misaligned during the assembly of the equipment. To prevent a recurrence of this problem, the prism mounts were made more rigid.

Experiments 17 through 26

In all cases the injection tube orifice was located approximately 6 in. below the liquid N_2 level. Appendix I presents a complete discussion of the theory, appratus, and results. Table I of Appendix I presents the results of the particle size distributions for particles of a diameter greater than 1 micron and Table II presents the results of the polydispersion properties of particles less than 1 micron in diameter as observed at the upper station. The experimental parameters which described the conditions under which the ClF_3 particles were prepared are presented in Table III.

Estimation of the accuracy of the results obtained from the particle size measurements presents certain difficulties. For example, if the particles form loose aggregates because of the attraction forces between the individual particles, the light extinction apparatus would see the aggregate as a single large particle. Consequently, the calculated particle size would be high and the calculated surface area would be low. In addition, if aggregate formation does occur, calibration of the device with other fine particles whose properties can be measured by other techniques would be fruitless because the degree of aggregation occurring among the ClF_3 particles could not be reproduced with a particle prepared from a different material.

The data obtained strongly indicates that aggregation is occurring. This is shown by the fact that the lower station measured a substantially smaller number of particles than did the upper station in every experiment except Experiment 24. This difference in the number of particles observed at the two stations supports the theory that aggregation is occurring in that the probability of the individual particles forming aggregates increases as the particles settled and the number of particles per unit volume increases. Table IV presents this information. In addition, the particles must show a strong tendency to form aggregates because the formation of interconnecting chains or networks of particles which trap the liquid to form a gel-like material is a requirement of a particulate gelling agent; it has been demonstrated that these particles form gels.

TABLE III

EXPERIMENTAL PARAMETERS
PARTICLE SIZE MEASUREMENTS

Exp. No.	Interhalogen & Diluent	Dilution Vol of Gas/ Vol of Gelling Agent	Injection Flow Rate, $\frac{cm^3 (STP)}{sec}$/	Injection Flow Velocity, cm/sec	Reynolds No.	ClF_3 Quantity Injected, gm
17	ClF_3 & He	37.2	723	23×10^4	50,900	1.11
18	ClF_3 & He	37.2	542	18×10^4	38,700	1.11
19	ClF_3 & He	37.2	1480	48×10^4	106,000	2.23
20	ClF_3 & He	37.2	170	5.5×10^4	12,200	1.11
21	ClF_3 & He	74.4	323	10×10^4	23,000	0.78
22	ClF_3 & He	74.4	1440	46×10^4	102,000	1.34
23	ClF_3 & He	74.4	168	5.4×10^4	12,000	0.54
24	ClF_3 & He	20.5	1500	48×10^4	106,600	1.82
25	ClF_3 & He	20.5	678	22×10^4	48,900	1.05
26	ClF_3 & He	20.5	≈ 170	$\approx 5 \times 10^4$	$\approx 12,000$	*

---

*Injection tube clogged almost immediately.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

TABLE IV

NUMBER OF PARTICLES OF ClF_3 OBSERVED
PARTICLE SIZE MEASUREMENTS

Experiment No.	Number of Particles x 10^{-3} per cm^3 Upper Stage	Lower Stage
17	15	8.6**
	19	6.9***
18	28	16
19	23	15
20	12*	-
21	20	9.4
22	67	17
23	56	7.0
24	6.4	10
25	31	7.3
26	32	12

---

*Note: The results from Experiment 20 should be discarded because the optical system was severely degraded by ClF_3 attack.

**First measurement of particle size.

***Sample restirred and particle size remeasured.

The mass of ClF_3 used in the experiments can be determined by two independent methods. First, the pressure drop observed for the ClF_3 holding tank during the transfer of the ClF_3-helium mixture from the tank to the apparatus can be used to calculate the mass of ClF_3 introduced. Second, the quantity of ClF_3 present in the apparatus can be calculated from the density, size and number of ClF_3 particles. A comparison of the values obtained by the two methods is presented in Table V and provides a means of estimating the reliability of the particle size measurements. The agreement between the two sets of data is quite good, the greatest difference being on the order of 40%.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

TABLE V

QUANTITY OF CHLORINE TRIFLUORIDE BASED ON EXPERIMENTAL DATA AND CALCULATED FROM PARTICLE SIZE MEASUREMENTS

	Experimental	Calculated From Particle Size Measurements		
		Upper Stage	Lower Stage	Mean
Exp. No.	$\frac{\text{gm ClF}_3}{\text{Liter LN}_2}$	$\frac{\text{gm ClF}_3}{\text{Liter LN}_2}$	$\frac{\text{gm ClF}_3}{\text{Liter LN}_2}$	$\frac{\text{gm ClF}_3}{\text{Liter LN}_2}$
17	0.968	0.977	1.76	1.37
		0.752	1.24	0.996
18	0.915	0.536	1.40	0.968
19	1.84	0.723	2.31	1.52
20	0.980	0.062*	---	---
21	0.660	0.710	0.860	0.785
22	1.102	0.592	2.21	1.40
23	0.486	0.418	0.334	0.376
24	1.528	0.624	1.56	1.09
25	0.933	1.13	1.56	1.34
26	-**	0.081	0.087	0.084

---

*Note: The results from Experiment 20 should be discarded because the optical system was severely degraded by ClF_3 attack.

**Injection tube clogged almost immediately.

Experiment 17 was designed to measure the particle size of the ClF_3 particles prepared under the experimental conditions, determine the reproducibility of the apparatus and to determine whether particle growth was occurring. To achieve these objectives, the particles were prepared and particle size measured over a period of one hour. Upon completion of this measurement the sample was restirred and the particle size again measured. The results of this experiment are tabulated in Table VI.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

TABLE VI

PARTICLE SIZE MEASUREMENTS, SUMMARY OF RESULTS OBTAINED IN EXPERIMENT 17

	Upper Station		Lower Station	
	Run 1	Run 2	Run 1	Run 2
Quantity ClF_3 gm/liter LN_2 (a)	0.977	0.752	1.76	1.24
Number of Particles				
$<1\mu/cm^3$	3.2×10^{10}	6.4×10^{10}	---	---
$>1\mu/cm^3$	1.5×10^6	1.9×10^6	---	---
$>1\mu/cm^3$	---	---	8.3×10^5	7.0×10^5
Total Surface Area cm^2/gm	3.60×10^3	5.5×10^3	1.6×10^3	0.7×10^3

(a) The quantity of ClF_3 injected into the liquid N_2 was 0.968 gm, quantity calculated from the measured pressure drop in the ClF_3 holding tank.

While the results of this experiment have not been rigorously analyzed, deviation from the mean of any calculated results based on the measurements taken from one station is not more than 33%. Consequently, it can be conservatively concluded that the measurements are probably reproducible to within ± 50%. In addition, it is concluded that, over a 2-1/2 hour period, there is no evidence of ClF_3 particle growth at the NBP of liquid N_2. This lack of particle growth indicates that the gel promises stability when stored.

The maximum resolution of the light extinction measurement apparatus is approximately 0.5 micron. However, the total quantity of particles less than 1 micron in diameter can be estimated from the difference between the transmittance of the dispersion at the termination of the run and the transmission occurring through neat liquid N_2 which was measured prior to the run.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

If varying experimental conditions had any measurable effect on particle size, there should be a significant change in the ratio (number of particles of ClF_3 less than 1 micron/number of particles of ClF_3 greater than 1 micron). This change in ratio should be greatest at the upper station where the particle density was lowest per unit volume thereby reducing the effect of particle aggregation. Consequently, the necessary calculations were done for all upper station measurements to estimate the number of particles present less than 1 micron in diameter. The calculation technique used is fully discussed in Appendix I. Using the result obtained above, the ratio (number of particles of ClF_3 less than 1 micron/number of particles of ClF_3 greater than 1 micron) was calculated. This ratio was then plotted against the three variables investigated during the particle size measuring work: (1) composition of injected gas; (2) injection rate; and (3) concentration of solid ClF_3 in the suspension. These plots are presented in Figures 8, 9, and 10, respectively. These plots show that, over the range of conditions studied, these variables have no measurable effect on the size of the particles prepared.

This lack of measurable effect on particle size when the composition of the injected gas and injection rate are varied provides some insight into the mechanism of particle formation. If the particles of ClF_3 were formed entirely within the bubbles of helium as they pass through the liquid N_2, the size of the particles should increase with increasing ClF_3 concentration and be independent of injection rate because bubble size is primarily determined by the surface tension of the liquid N_2 and, thus, would be independent of the rate of injection. Consequently, as the concentration of ClF_3 increased within the bubble, the size of the particle should increase.

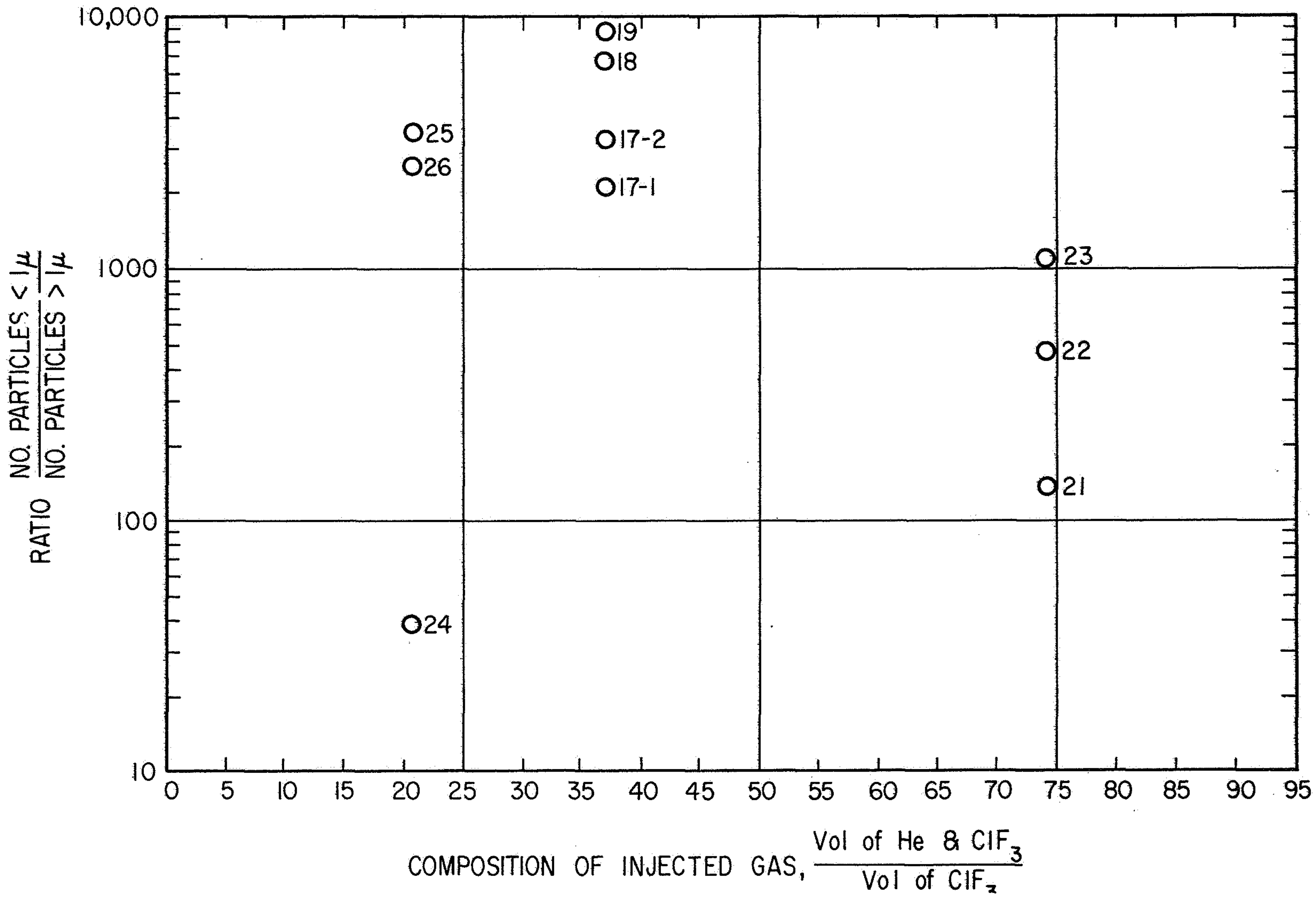


Figure 8. Ratio of $\frac{\text{No. of ClF}_3\text{ Particles} < 1\mu}{\text{No. of ClF}_3\text{ Particles} > 1\mu}$ vs Composition of Injected Gas

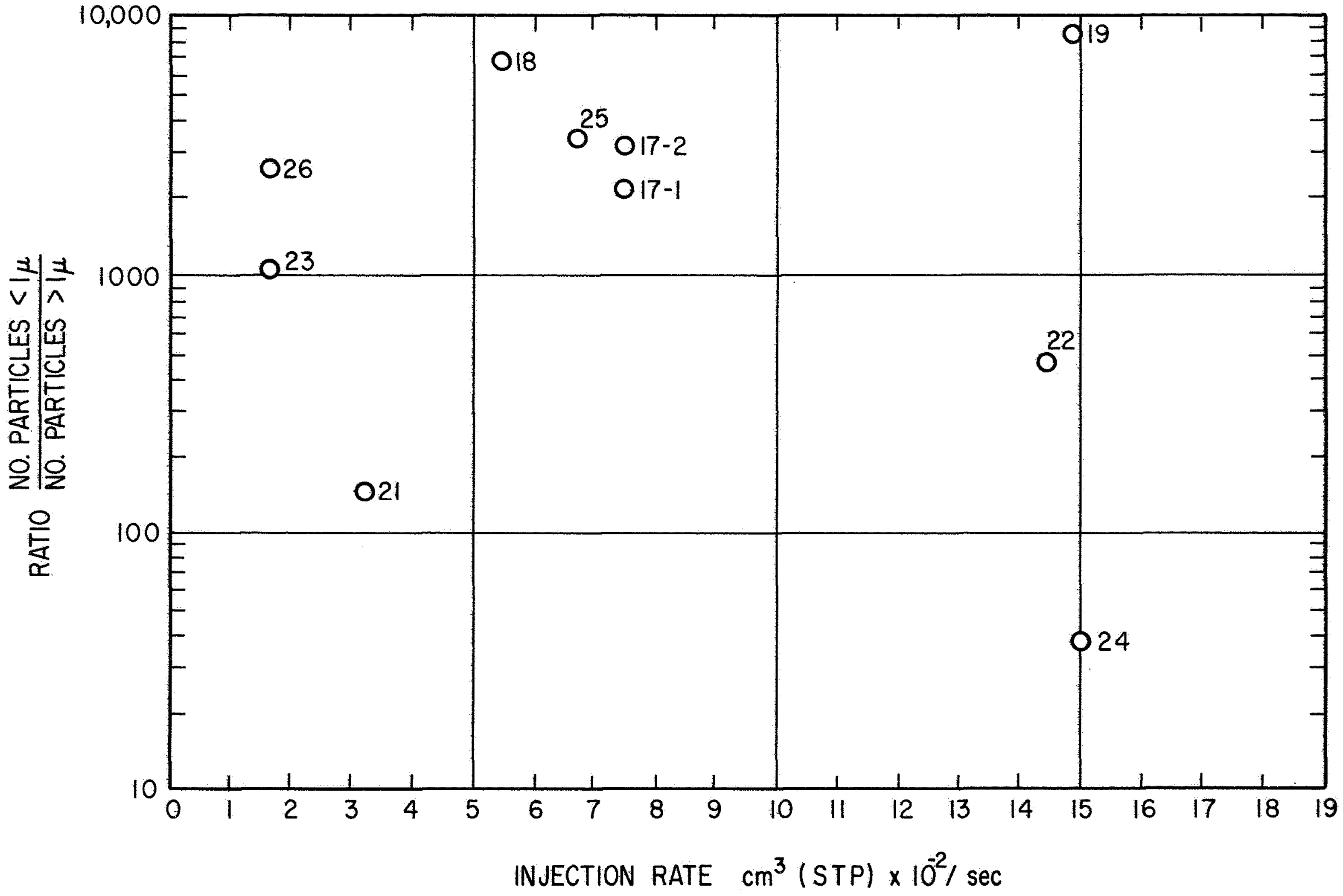


Figure 9. Ratio of $\frac{\text{No. of } ClF_3 \text{ Particles} < 1\mu}{\text{No. of } ClF_3 \text{ Particles} > 1\mu}$ vs Injection Rate

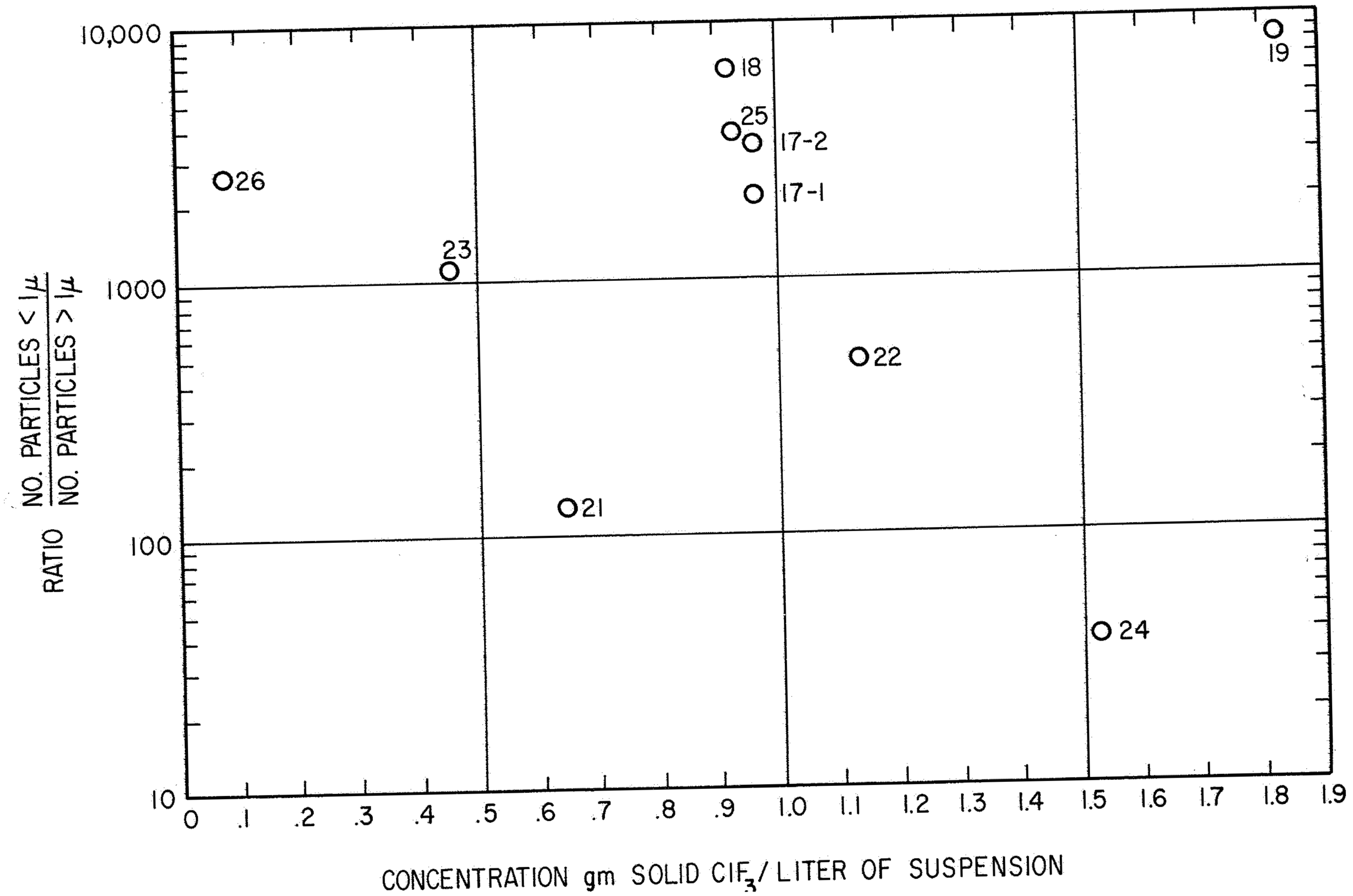


Figure 10. Ratio of $\frac{\text{No. of ClF}_3\text{ Particles} < 1\mu}{\text{No. of ClF}_3\text{ Particles} > 1\mu}$ vs Concentration of ClF$_3$ in the Suspension

However, if the particles were formed entirely within the liquid N_2, particle size should increase with increasing injection rates if concentration is held constant because increasing injection rate would increase the concentration of ClF_3 in the liquid N_2 if particle formation occurred external to the helium bubble. Figure 11 illustrates that the particle size of the ClF_3 particles prepared is independent of both injection rate and composition of injected gas over the range of conditions investigated. This lack of measurable effect on particle size when the composition of the injected gas and injection rate are varied indicates that particle formation is probably occurring both within the helium bubble and in the liquid N_2.

The following conclusions are drawn from the results of the particle size measurements: (1) the particles measured are, in all probability, aggregates of much smaller particles; (2) no measurable particle growth was observed over a 2-1/2 hour period at the NBP of liquid N_2; and (3) over the range of conditions investigated, no measurable effect was observed on the size of the particles prepared, indicates particle formation occurs within the helium bubbles <u>and</u> in the liquid N_2.

d. Development of a Technique for Measuring the Engineering Properties of Cryogenic Gels

(1) Static Properties

The two commonly measured static properties of a gelled system are yield stress and mechanical stability. Yield stress is a measurement of the degree of structure possessed by a gel when at rest. Mechanical stability measurements are used to determine whether the solid suspended in a gelled propellant settles during storage and/or if an exudate forms. At this stage in the propellant development, only the degree of structure possessed by the gel is of interest.

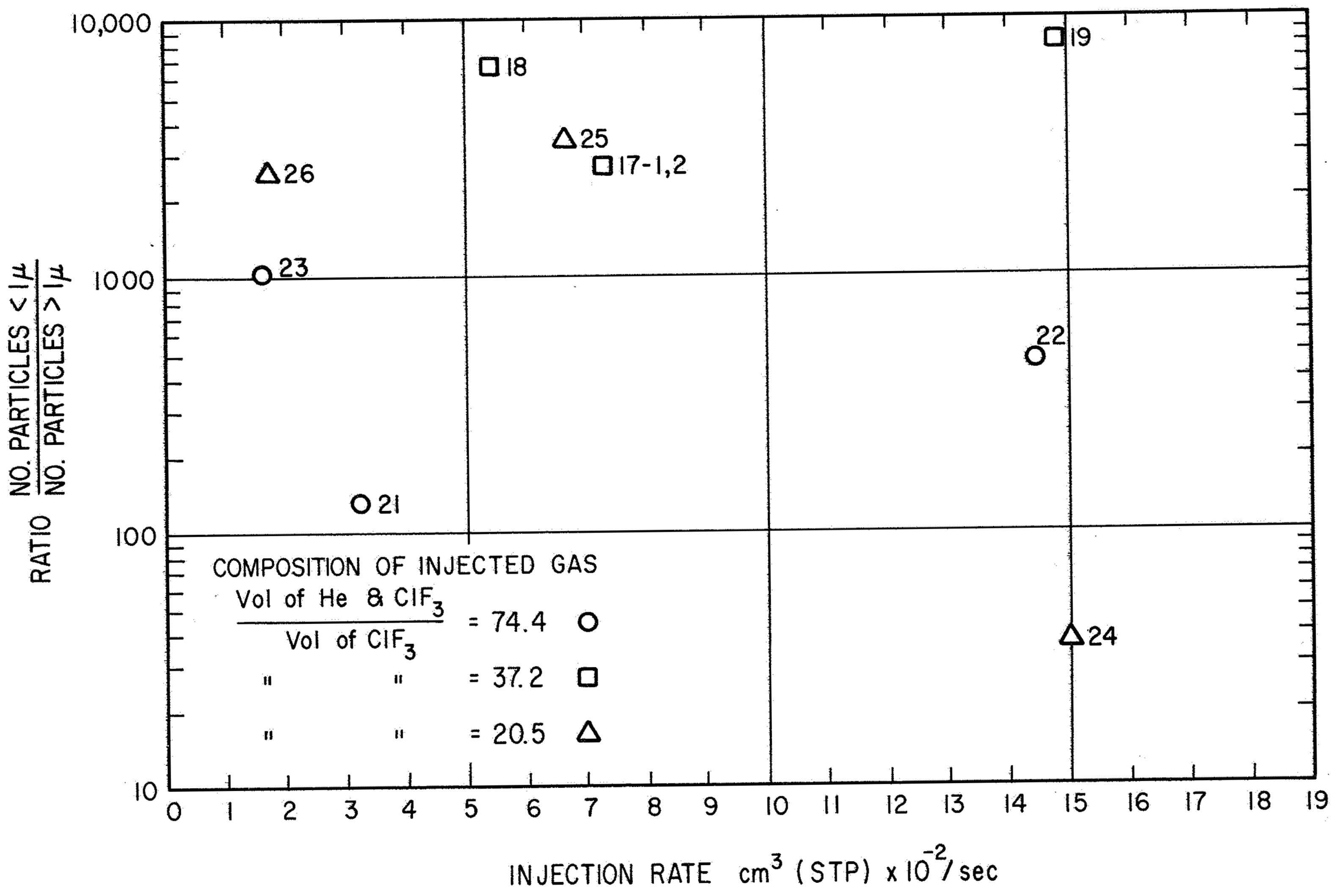


Figure 11. Ratio of $\frac{\text{No. of ClF}_3\text{ Particles} < 1\mu}{\text{No. of ClF}_3\text{ Particles} > 1\mu}$ vs Injection Rate

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

The discussion of the measurement of static properties of gels presented in Section III,1,c,(1), had shown that only Rotoviscometers and cone penetrometers had been used successfully to assess the degree of structure possessed by a cryogenic gel. In addition, the reasons for the failure of the Rising Sphere Rheometer to measure and assess the structure of particulate cryogenic gels was discussed. The most likely cause was attributed to the fact that the measurement of structure with a Rising Sphere Rheometer includes a shear rate component. Consequently, no meaningful result is obtained because cryogenic gels shear thin with extreme rapidity.

Initially a steel ball was slowly lowered into the gel, if the ball was supported by the gel, the degree of structure (called structure index, reported in units of dynes/cm^2) was calculated from the known mass and projected surface area of the steel ball. If the ball was not supported on the surface of the gel, additional nitrogen was allowed to evaporate and the degree of structure again measured. This was repeated until the gel possessed enough structure to support the steel ball. The results obtained with this technique are presented under the heading remarks in Table I.

As the work proceeded, a more precise method of assessing structure was developed. A series of glass spheres of constant diameter containing varying amounts of mercury to provide a range of weights were fabricated. All the spheres were heavy enough to sink beneath the surface of the neat liquid but some of them were light enough to be supported by the surface of the gelled liquid. This technique for measuring the degree of structure was particularly suited for work with extremely reactive cryogenic propellants such as OF_2, because manipulation of the propellant is held to a minimum. The maximum weight which can be supported by the surface of the gel and the projected area of the sphere are used to calculate the maximum force per unit area that can be supported by the surface. The value obtained in this manner is referred to as the structure index. It must be noted that this technique does not provide an absolute measurement of gel structure. However, the values obtained are an excellent measure of gel structure and show changes in the quantity of gel structure with changing gelling agent concentration.

In Experiment 15, the structure index of gelled liquid N_2 at various gelling agent (ClF_3) concentrations was measured. The experimental parameters and results of this experiment are presented in Table VII. Figure 12 shows the change in structure index with changing gelling agent concentration. The rapid change in structure observed with small changes in concentration is typical of particulate gels.

It was concluded from the results of the experiment that a workable technique for assessing the degree of structure of cryogenic particulate gels had been developed. This was the technique used for assessing the degree of structure of the OF_2 gels which were prepared.

While the procedure described above was satisfactory, it presented serious manipulation problems when used with reactive, air sensitive, toxic propellants. Consequently, work was conducted to determine whether it was possible to measure or estimate the structure index by use of an alternative procedure. The results of the experimental work are reported below (Experiment 40).

It is theoretically possible to estimate the structure index of a gel by extrapolating the results of a series of flow measurements to a zero flow condition. This extrapolation can be accomplished by plotting the logarithm of shear stress against fluidity (the reciprocal of viscosity). Therefore, a series of Freon-113 gels (gelling agent Cab-O-Sil H-5) were prepared and the structure index and flow properties measured. The technique for flowing the gels and the flow viscometer used was essentially identical to that used with the cryogenic gels. See Section III,2,d,(2) for a discussion of the flow viscometer. The results of the flow measurements conducted with these gels are presented in Table VIII. The results obtained are also presented as a

TABLE VII

EXPERIMENT 15--PREPARATION OF FINE PARTICLES OF CHLORINE TRIFLUORIDE

Interhalogen and Diluent	ClF_3 and He
Dilution	
$\frac{\text{vol of gas}}{\text{vol of gelling agent}}$	36.9
Injection Flow	
Rate, (cm^3/sec, STP)	550
Velocity, (cm/sec)	18 x 10^4
Reynolds No.	39,800

STRUCTURE INDEX OF GELLED LIQUID NITROGEN AT VARIOUS ClF_3 CONCENTRATIONS

wt % ClF_3	vol % ClF_3	Structure Index, ($dynes/cm^2$), mean
7.8	2.6	<500
10.5	3.6	700
11.9	4.2	1200
15.5	4.6	1700

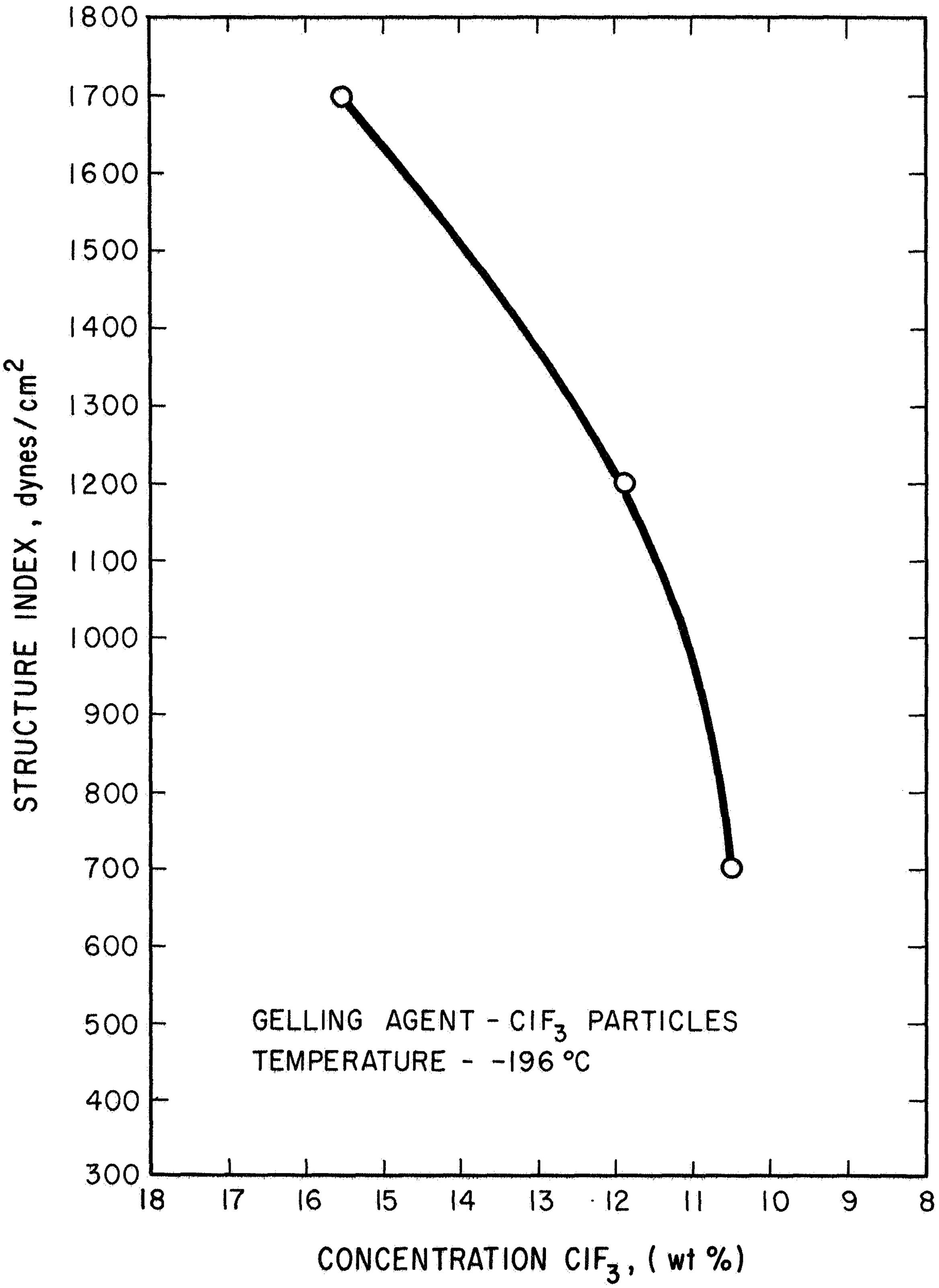


Figure 12. Structure Index of Gelled LN_2 at Various Gelling Agent Concentrations

TABLE VIII

EXPERIMENT 40--FLOW CHARACTERISTICS OF LIQUID FREON-113 GELLED WITH CAB-O-SIL H-5

Run No.	Gelling Agent Conc.,		Structure Index,	Δ Pressure,		Flow Parameters		Reynolds No.	Shear Rate	Shear Stress	Viscosity,	Fluidity,
	vol%	wt%	dynes/cm^2	dynes/cm^2 x 10^{-4}	psi	Rate, cm^3/sec	Velocity, cm/sec		$\frac{8V}{D}$, sec^{-1}	$\frac{D\Delta P}{4L}$, dynes/cm^2	poise	1/poise
1	1.6	2.7	700	26.2	3.8	1.59	8.8	14	146	68	0.47	2.15
2				4.62	6.7	4.47	24.7	67	411	114	0.28	3.60
3				6.76	9.8	7.37	40.7	133	677	158	0.23	4.29
4	1.7	2.8	1900	2.55	3.7	0.81	4.5	4	74	67	0.90	1.11
5				4.55	6.6	2.04	11.3	13	188	119	0.63	1.58
6				6.48	9.4	3.99	22.0	38	367	162	0.44	2.26
7				8.48	12.3	6.12	33.8	71	562	203	0.36	2.78
8	1.8	2.9	4000	2.55	3.7	0.36	2.0	1	33	69	2.1	0.48
9				7.52	10.9	1.37	7.6	4	126	199	1.6	0.63
10				9.17	13.3	1.64	9.0	4	150	241	1.6	0.63

plot of $\frac{D\Delta P}{4L}$ versus $\frac{8V}{D}$ in Figure 13; this mode of presentation is referred to as the "Characteristic Flow Curve." The results are also presented in a plot of $\frac{D\Delta P}{4L}$ versus fluidity in Figure 14. Theoretically, an extrapolation of the results of this plot to zero fluidity should permit an estimation of the structure index of the gel to be made. The results of this extrapolation and the measured structure index are presented in Table IX. Inspection of Table IX clearly shows that the extrapolation does not yield useful information.

In addition, the pressure required to initiate the flow of gels of various thickness was measured. The results of this measurement are presented in Table X.

These test results demonstrate that the properties of a particulate gel under static conditions are entirely different from the gel properties when the gel is being flowed. Consequently, any measurement of structure, if it is to provide reliable data, must not impose a shear rate upon the gel and the technique used in this program is most likely to measure the degree of structure possessed by a particulate gel reliably. A completely satisfactory technique awaits development.

The results of the measurement of pressure required to initiate flow in particulate gels of varying structure show that small applications of pressure will start flow.

(2) Flow Properties

The review of the measurement of flow properties, reported in Section III,1,c,(2), had shown that the measurement of the flow properties of cryogenic propellants gelled with particulate agents had been accomplished with

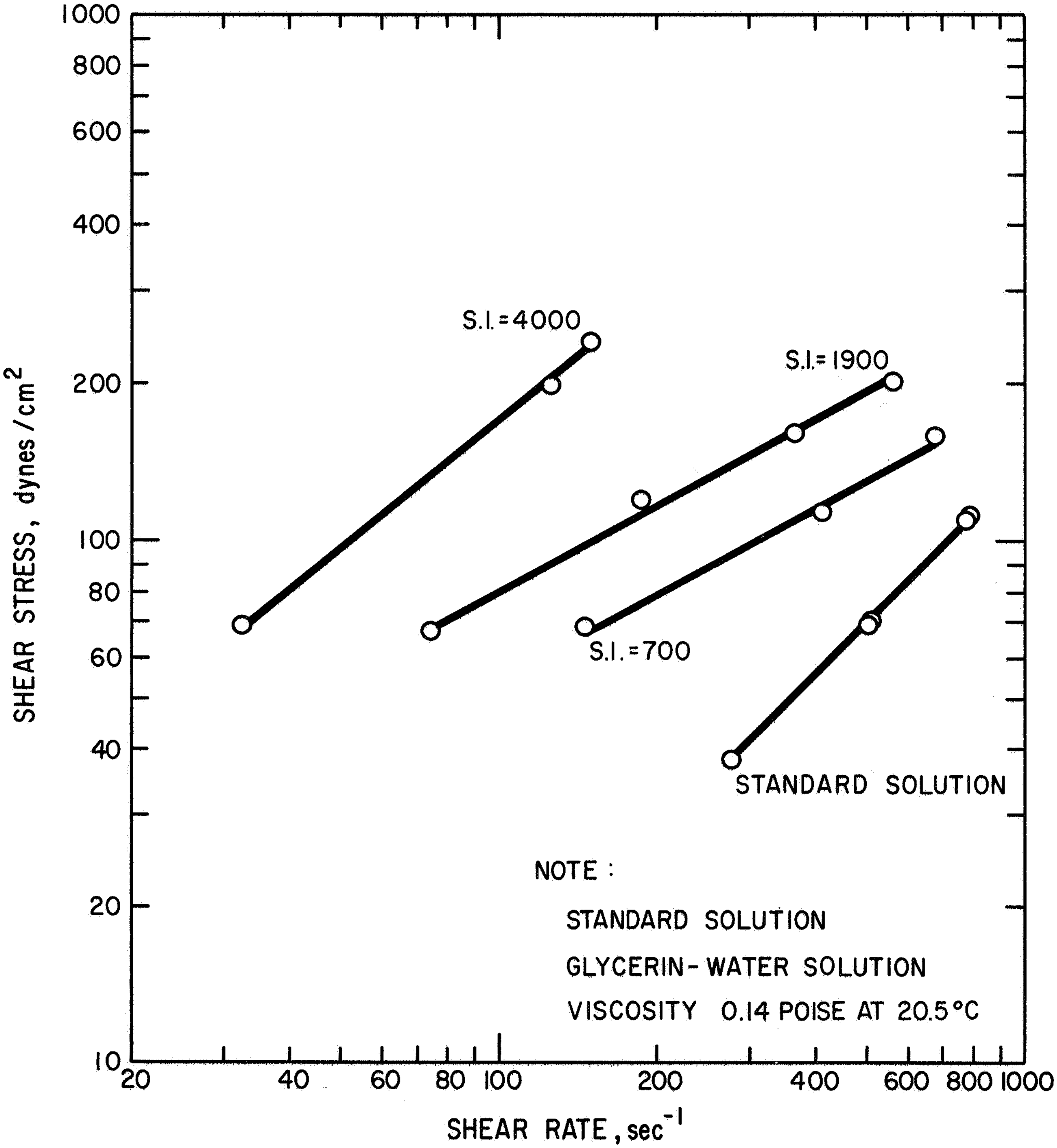


Figure 13. Characteristic Flow Curves of Freon-113 Gelled with Cab-O-Sil H-5

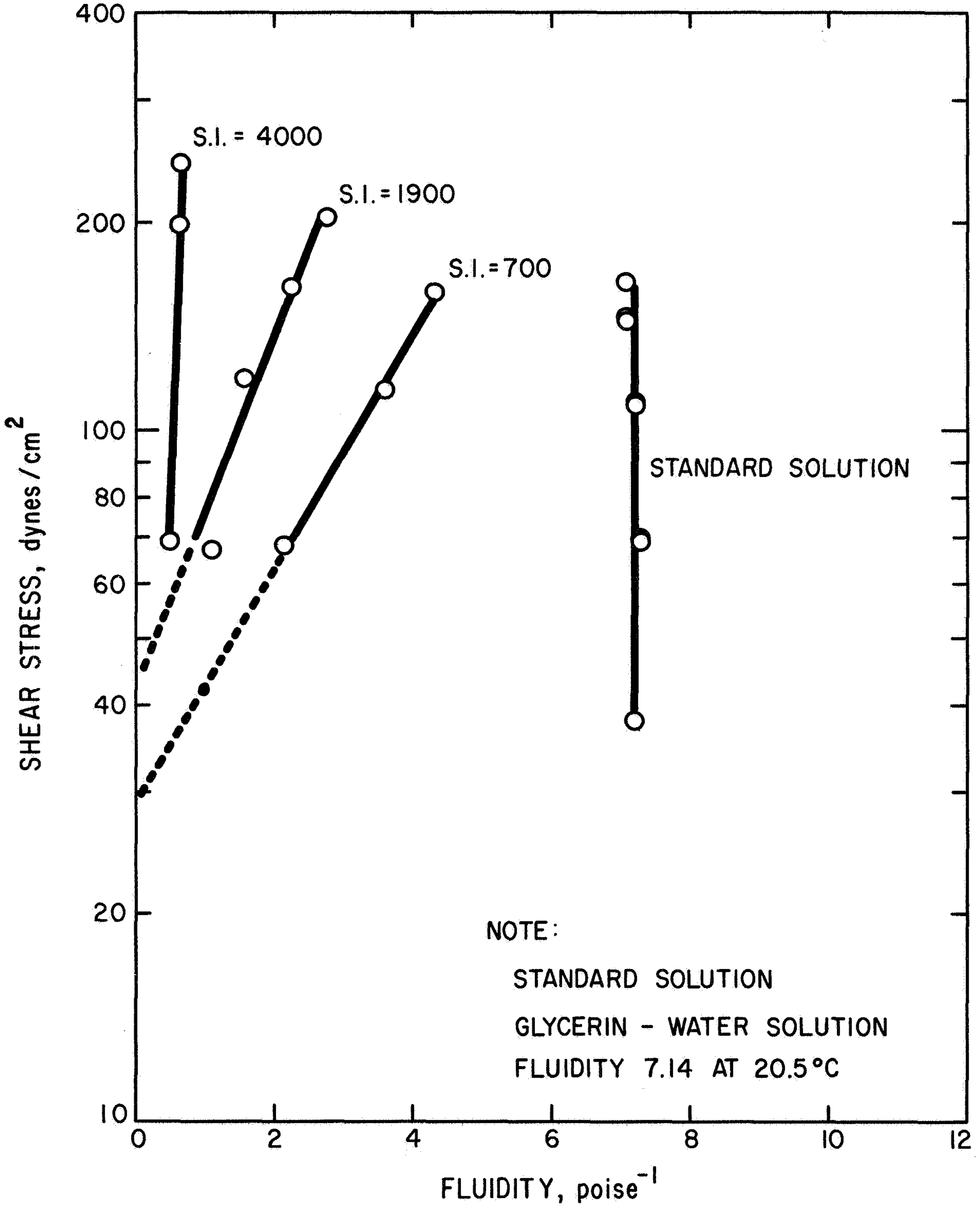


Figure 14. Fluidity of Selected Freon-113 Gel at Various Shear Stresses - Gelling Agent Cab-O-Sil H-5

TABLE IX

COMPARISON OF MEASURED STRUCTURE INDEX AND STRUCTURE INDEX ESTIMATED FROM FLOW PROPERTIES*

Measured Structure Index, dynes/cm^2	Estimated Structure Index, dynes/cm^2
700	29
1900	42
4000	1

---

*Freon-113 gelled with Cab-O-Sil H-5

TABLE X

PRESSURE REQUIRED TO START FLOW OF FREON-113 GELLED WITH CAB-O-SIL H-5

Structure Index, dynes/cm^2	Pressure Required to Start Flow, psi
700	0.01
1900	0.86
4000	2.64

rotational viscometers. Attempts to measure flow properties with flow viscometers had not been successful. However, in spite of the difficulties reported with the use of flow viscometers with cryogenic gels, it was decided to develop a flow viscometer to measure the properties of gelled OF_2. The reasons for this decision were: (1) flow-type viscometers are the only instrument that produces information directly related to the behavior of the gel under flow conditions; (2) techniques have been developed which permit the utilization of the information provided by a flow viscometer in preliminary system design work; and (3) during the course of measurements with a flow viscometer, information can be obtained regarding the coring and the hangup behavior of the gel.

Two parameters are measured during an experiment with a flow viscometer, i.e., the ΔP across the tubing and the flow rate. These two parameters are plotted in the form of $\frac{D\Delta P}{4L}$ versus $\frac{8V}{D}$, where D is the diameter of the tubing, L is the effective length of the tubing, and V is the velocity at which the gel is transferred through the tubing. This velocity is calculated from the measured flow rate. The term $\frac{D\Delta P}{4L}$ is commonly referred to as the shear stress, and the term $\frac{8V}{D}$ is referred to as the shear rate. The ratio of shear stress divided by the shear rate is the viscosity of the fluid at that particular shear rate.

The plot of the $\frac{D\Delta P}{4L}$ versus $\frac{8V}{D}$ produces a curve which is referred to as the "characteristic flow curve" for the fluid being tested. An example of such a curve for Alumizine, a suspension of Aluminum in gelled hydrazine is given in Figure 15. The principal curve in Figure 15 represents the laminar flow region; the branches from the laminar flow curve occur as the mode of flow becomes turbulent. This curve demonstrates that a greater pressure drop is incurred during the turbulent mode of flow than during the laminar mode for the transfer of material at equivalent flow rates.

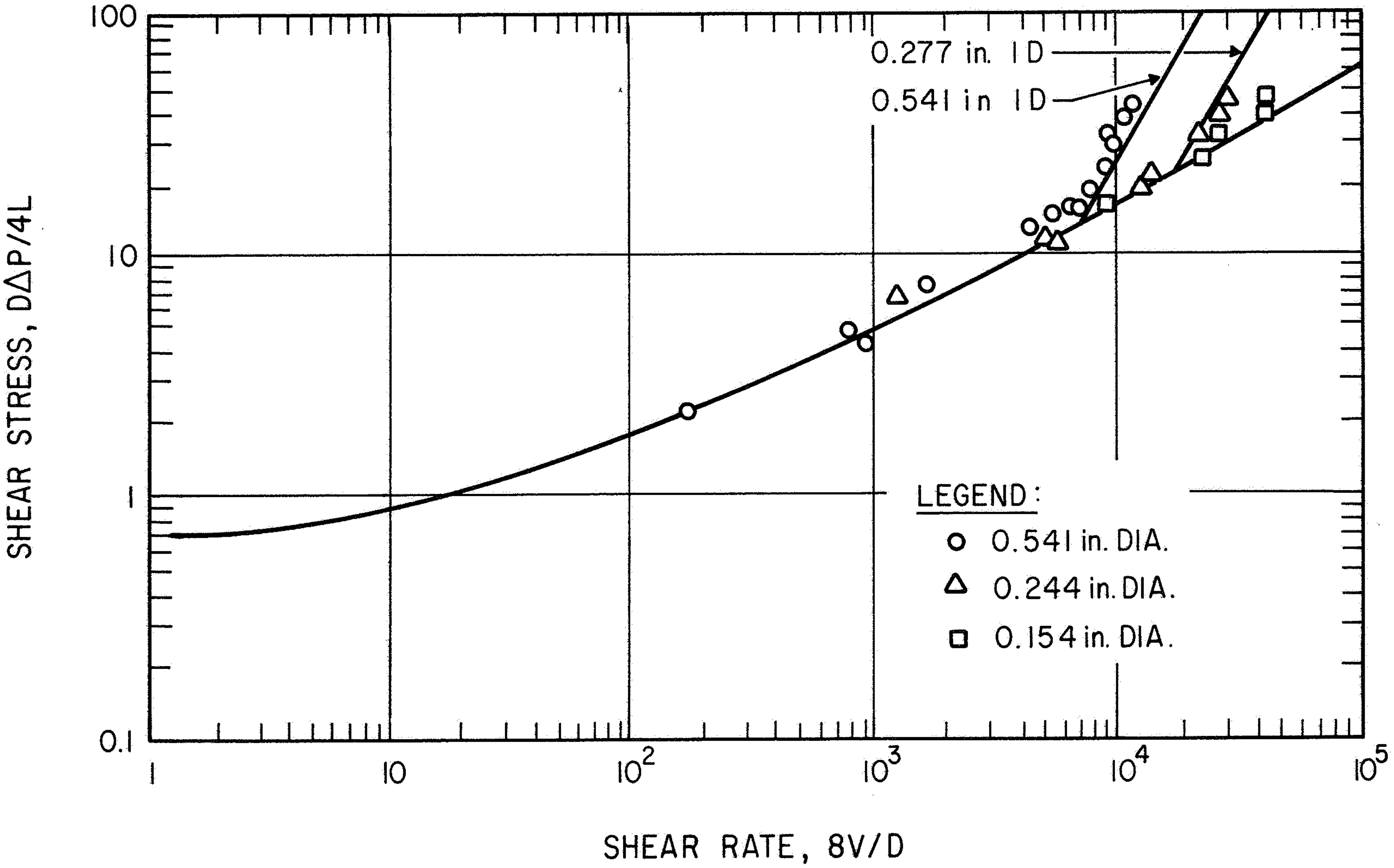


Figure 15. Characteristic Flow Curve of Alumizine

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

This method of data presentation, the characteristic flow curve, is useful in that it provides data necessary for the design of prototype hardware systems using the liquid or gel of interest. For example, in the region of laminar flow, if the flow rate and the diameter and length of the pipe are known, the pressure drop which will occur can be determined from the curve. Or, by selection of a pressure drop value for a length of pipe and flow rate, the required diameter of pipe can be calculated. Further, because the curve provides the viscosity value of the liquid over a range of shear rates, the Reynolds number can be calculated for the liquid at various flow conditions. Experimentally, the turbulent mode of flow can be determined by increasing the flow rate until the data display a sharp branching from the smooth laminar curve. If the flow measurements are repeated with another size of tubing and the flow rate is again increased until the turbulent mode is reached, the point at which the transition from laminar to turbulent flow occurs can be calculated from the data for any size piping and flow rate. The turbulent flow properties as represented by the branches from the laminar flow curve can be used for system design in the same manner as the data available in laminar region.

Before designing the flow viscometer, the research by Union Carbide on the rehological properties of fluid hydrogen slush (Reference 15) and the recently published comments from the Cryogenics Division of NBS were reviewed with regard to flowability through capillaries and transfer lines. The Union Carbide experiments, which are reported as qualitative, were based on the flow rate through 0.236-in. ID tubing. The results showed that the viscosity of 30 to 50% slush is approximately the same as that of liquid hydrogen. The NBS researchers have reported transferring slush hydrogen through 75 ft of 3/4-in. pipe using pressures of 14 to 15 psi successfully. The above information supplements the findings at Aerojet regarding flow and viscosity of gels prepared with the particulate type of gelling agents (e.g., carbon black and colloidal silica). The experience has been that, when the particulate gel is sheared by pressurization through a line, the rate of flow approximates that of neat liquid alone.

In the experiments conducted to this point in the program with gels of ClF_3 particles in liquid nitrogen, the behavior in handling indicated that, under shearing conditions, the flow rate and apparent viscosity will approximate that of the ungelled liquid (either liquid N_2 or liquid OF_2). In the case of liquid OF_2 thermostated by using liquid N_2 or liquid O_2, the viscosities are 0.010 poise (-196°C) and 0.007 poise (-183°C), respectively (Reference 16). This OF_2 fluidity is approximated by water (at room temperature) having a viscosity of 0.009 poise. Consequently, the design criteria for a macro-scale flow measurement device was based on handling a fluid such as water to yield precise data on volumes transferred over accurately measured times by the low pressures of 1 to 10 psig. The pressures available are limited by the experimental apparatus and safety considerations in the laboratory. Ideally, a 6 to 9 feet straight section of 1/4-in. stainless-steel tubing would serve for obtaining accurate flow measurements. However, this is not practical because of the experimental setup and the required temperatures. As a consequence, it was planned to use 6 to 8 feet of tubing in the form of a coiled section that will fit inside the Pyrex test vessel and be immersed in the gel.

The approach using a coiled section of tubing immersed in the test gel will eliminate temperature changes in the gel flowing through the tubing as long as the tubing remains beneath the surface level of the gel being tested. Temperature-conditioned helium will be used to provide the pressurization. Because flow in bends and coils encounters an additional resistance over that in straight sections, the equivalent length of the cylindrical coiled section will be calculated using the conventional relationships (Reference 17). The equivalent length used with a coiled section yields flow data that agree with flow in a corresponding length of straight section.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

Because flow in bends and coils encounters an additional resistance over that in straight sections, it was necessary to calculate the equivalent length of the coils. In other words, the length of straight tubing that would offer a resistance to flow equal to the resistance observed in the coil. This additional resistance caused by the bends in the coil changes with the flow velocity, i.e., changing shear rate.

The equivalent length at various shear rates of the flow coils fabricated during the course of this program was determined in the following manner. A solution of glycerol and water, viscosity 0.140 poise at 21°C, was flowed at various pressures through the coil and the volume of flow as a function of time measured at each pressure. The effective length of the coil was then calculated from the relationship $\frac{D\Delta P}{4L}$, dynes/cm^2, shear stress divided by $\frac{8V}{D}$, sec^{-1} shear rate which is equal to viscosity. The unit for viscosity (η) is poise, D is tube diameter (cm), ΔP is pressure drop (dynes/cm^2), L is equivalent tube length (cm), and V is flow velocity (cm/sec). Solving this relationship for equivalent length, L the expression is

$$L = \frac{D^2 \Delta P}{32 \eta V}$$

The calculated equivalent lengths were then plotted versus shear rate. It was then possible to determine the equivalent length of the coil for any flow rate, e.g., shear rate. The shear rate, $\frac{8V}{D}$, sec^{-1} could be calculated from the measured data obtained in a flow experiment (flow volume and flow time; tubing diameter being already known). Using the value for the shear rate, the plot could be entered and the equivalent length of the coil could be determined. Then the shear stress applied to the gel could be calculated from the expression $\frac{D\Delta P}{4L}$, dynes/cm^2. Knowing both the shear stress and the shear rate, the apparent viscosity of the gel under these particular flow conditions would then be calculated as viscosity is the shear stress divided by the shear rate.

The results obtained during the calibration of one of the flow coils used during this program is presented in Table XI and the plot of equivalent length versus shear rate derived from the calibration results is shown in Figure 16.

The final viscometer developed for measurement of the flow properties of liquids gelled with solid ClF_3 consisted of 3/8-in. dia copper tubing coiled to fit inside the Pyrex gelling vessel. The dia of the coil is approximately 4 in. and the total length of tubing including the straight inlet and outlet portions is 70.7 in. The coil is immersed in the gelled liquid at the beginning of the flow measurement. The inlet section is in the form of an inverted-J with the entrance near the bottom of the Pyrex vessel. This positioning of the entrance allows utilization of a major portion of the gel for the measurement and is intended to minimize any coring which might occur. Approximately 1 in. above the entrance a small vertical tube is installed to permit injection of helium into the copper tubing. The helium is cooled by passing through the 1/8 in. tubing coil above the copper coil which is also immersed in the gel. The injection of helium prevents the liquid from flowing through the tubing and eliminates the need for a valve in the flow system.

The inverted-J section of tubing is followed by the coiled section and the coiled section is followed by the straight outlet section. Figure 17 shows the flow tube and helium coil. The outlet section extends through the bottom of the Pyrex vessel by means of a ball-socket joint (glass/stainless steel). Figure 18 shows the assembled apparatus including the injection tube. The exit of the tubing is immersed in a bath of liquid N_2. After passage through the copper tubing, the gel is discharged into the liquid N_2 bath. The effluent will raise the level of the liquid N_2 bath immediately external to the Pyrex gelling vessel, and the volume of the effluent is determined quantitatively by measuring this change in level. Figure 19 shows the assembled apparatus. The box-like structure surrounding it is a moisture shield.

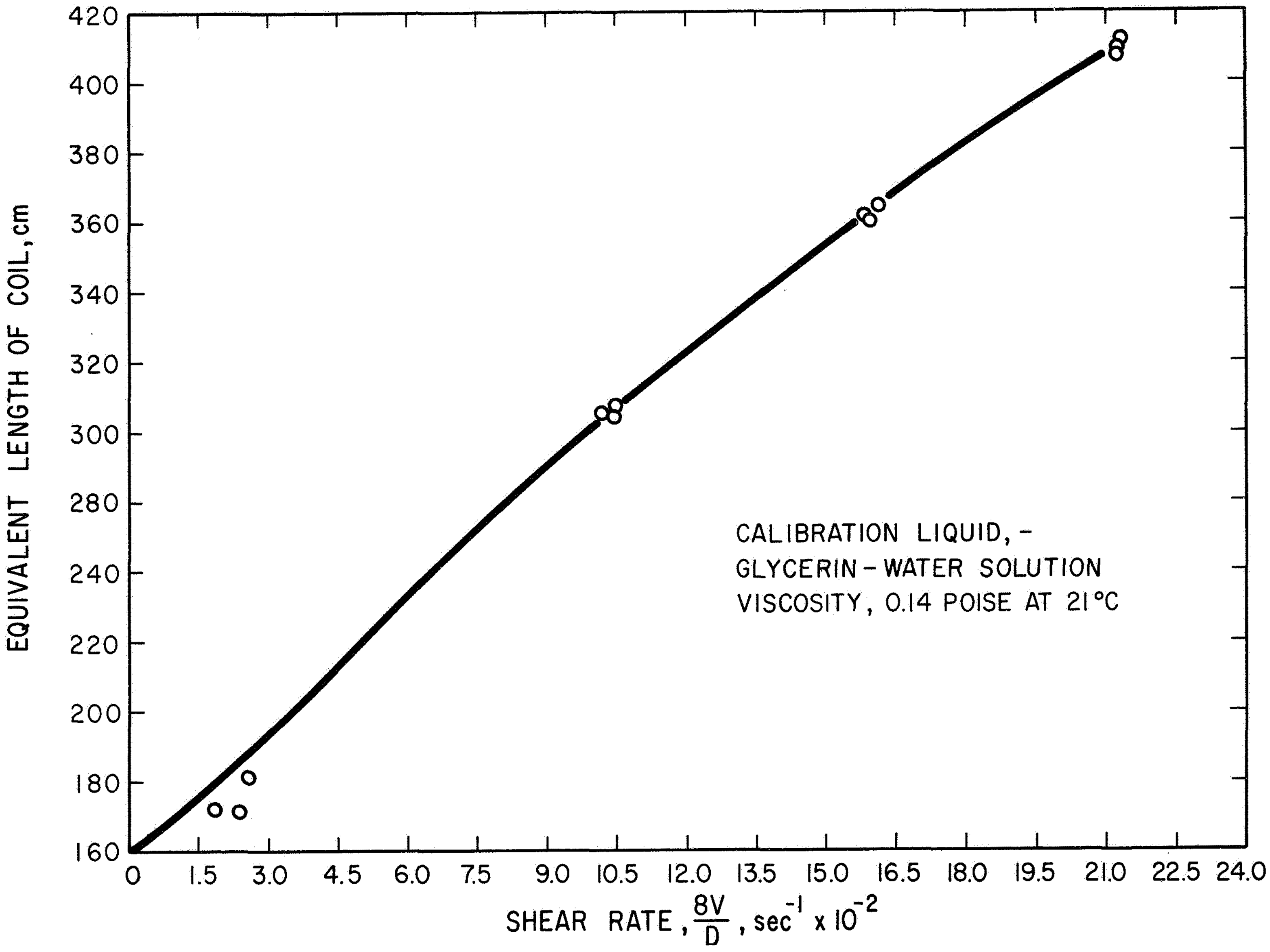


Figure 16. Equivalent Coil Length versus Shear Rate

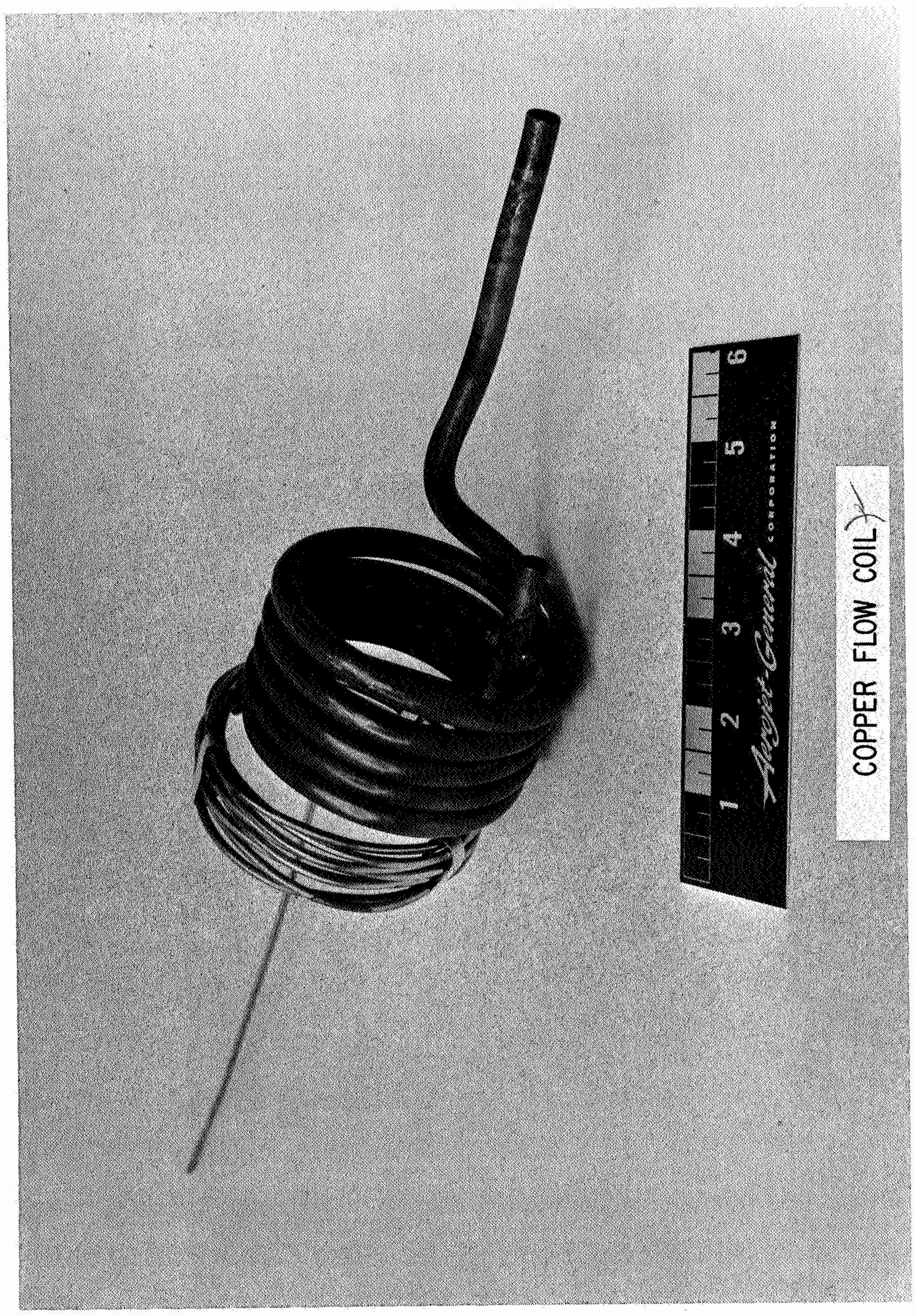


Figure 17. Copper Flow Coil

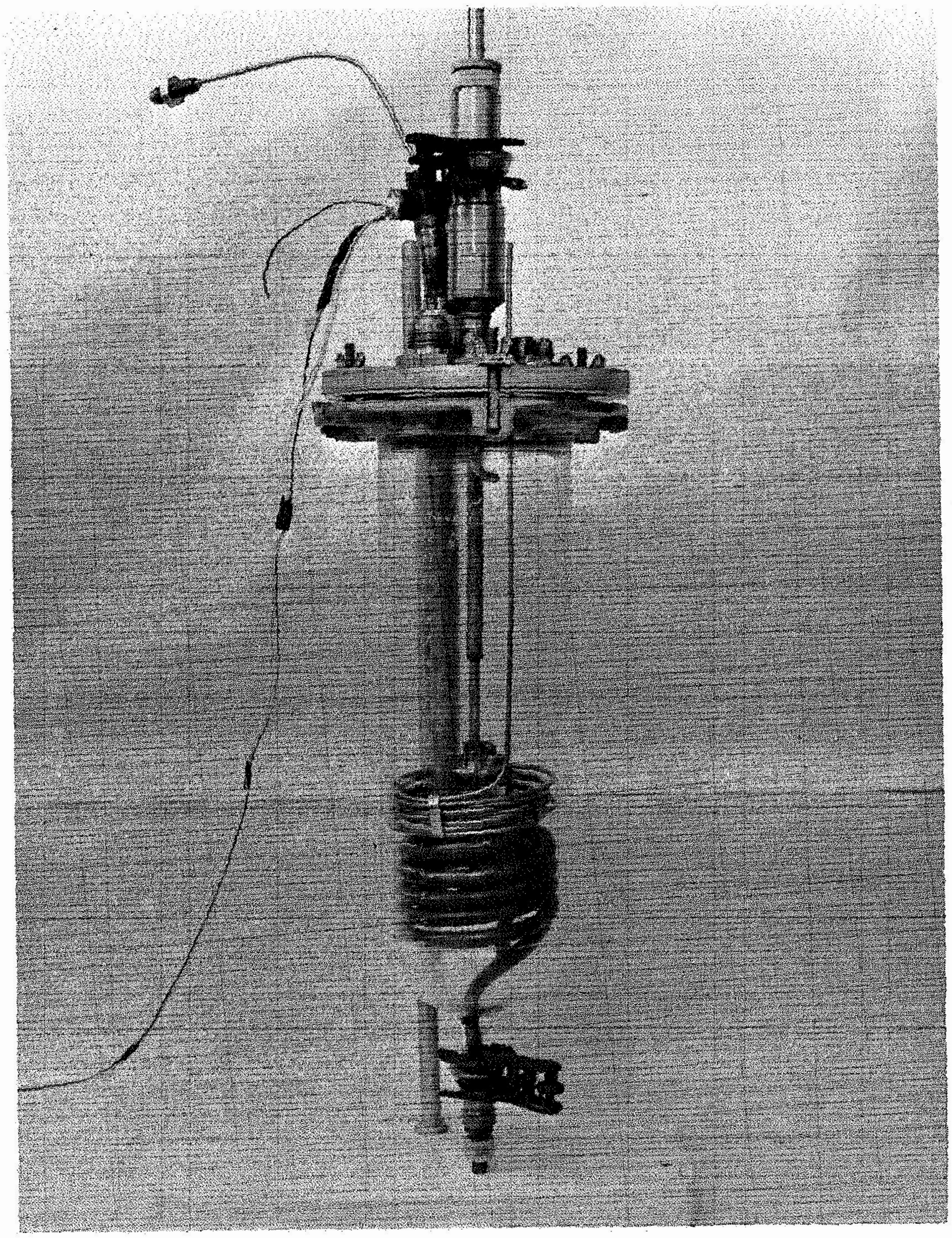


Figure 18. Cryogenic Gel Flow Viscometer

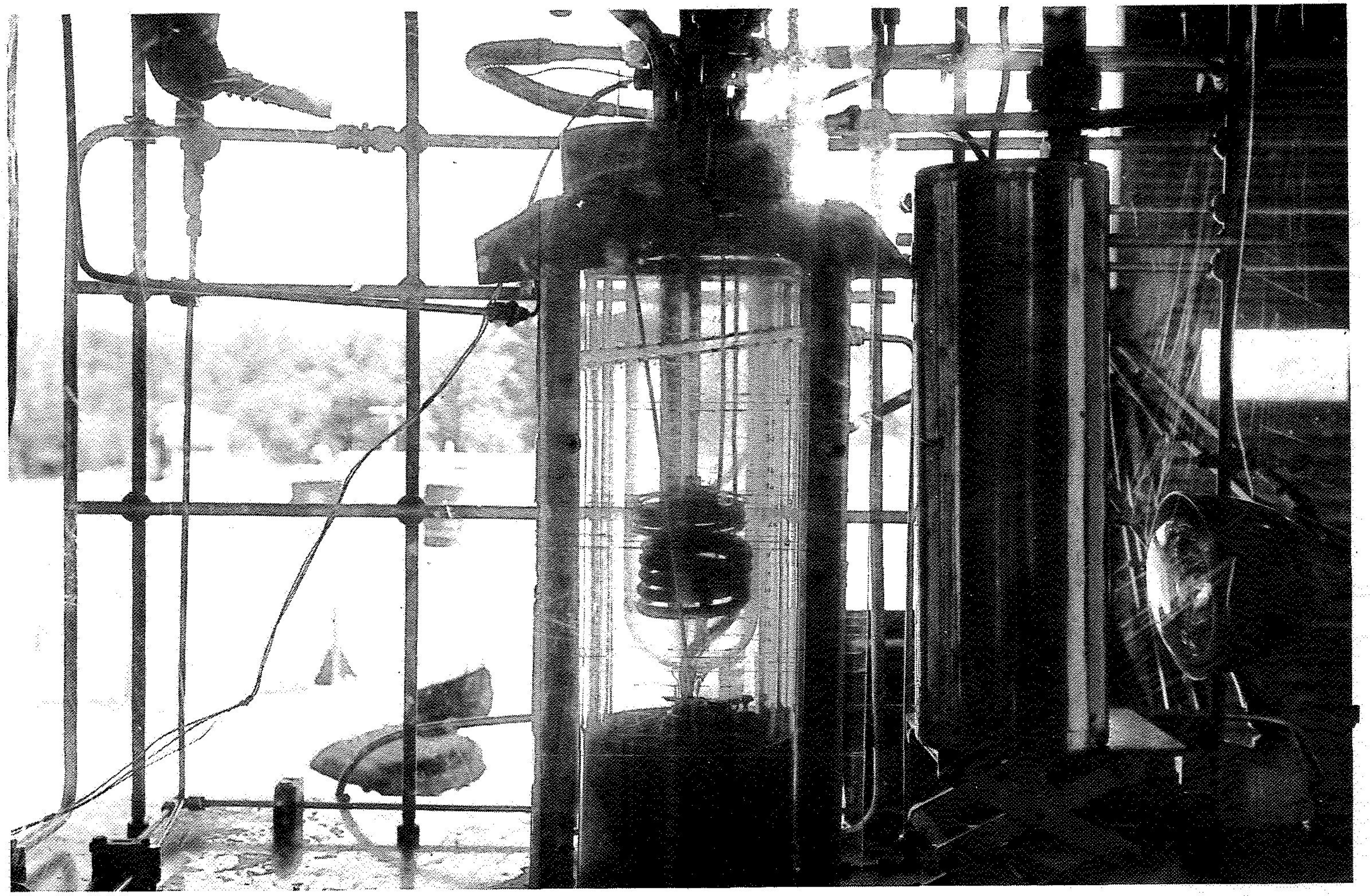


Figure 19. Installed Cryogenic Flow Viscometer

TABLE XI

CALIBRATION OF FLOW COIL NUMBER TWO

Run	Δ Pressure, psi	Δ Pressure, $\frac{\text{Dynes}}{\text{cm}^2}$ x 10^{-4}	Flow Time, sec	Flow Rate, $\frac{\text{cm}^3}{\text{sec}}$	Flow Velocity, $\frac{\text{cm}}{\text{sec}}$	Shear Rate $\frac{8V}{D}$, sec^{-1}	Effective Coil Length cm	Effective Coil Length in.	Remarks
1	0.56	3.86	58.5	12.4	26.0	262	201.9	79.5	Actual coil length 179.5 cm, 70.7 in.
2	0.41	2.83	87.8	9.6	20.2	207	190.3	74.9	
3	3.25	22.43	34.5	47.6	99.6	1021	350.9	120.4	Coil Diameter, OD 0.95 cm, 0.35 in.
4	3.33	22.93	32.7	48.6	101.8	1044	306.0	120.6	ID, 0.78 cm, 0.307 in.
5	3.31	22.85	34.6	48.6	101.7	1043	305.1	120.1	
6	6.11	42.12	22.3	75.0	156.9	1609	304.7	143.6	Calibration liquid Glycerol-water
7	5.95	41.02	23.9	74.0	154.9	1588	359.8	141.6	Solution Viscosity 0.140 poise
8	5.93	40.88	23.6	73.7	154.2	1581	360.2	141.8	at 21°C
9	9.02	62.21	15.4	99.0	207.2	2125	407.9	160.6	
10	9.17	63.21	15.5	99.5	208.3	2136	412.2	162.3	
11	9.14	63.01	16.2	99.4	208.0	2133	411.6	162.0	

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

The measurement of the flow properties was accomplished in the following manner. During preparation of the gel, helium at liquid N_2 temperatures, was injected through the small tube into the copper tubing to prevent gel from entering the flow-coil. After the gel was prepared, the helium flow was stopped, and the Pyrex gelling vessel was pressurized to a preselected value, flow began, and the volume of flow was recorded as a function of time. Flow was stopped at will by releasing the pressure on the gelling vessel and injecting helium into the copper tubing. The results of the flow experiments follows.

(a) Measurement of the Flow Properties of Gelled Liquid N_2 - Gelling Agent ClF_3 Particles

In all experiments the injection tube orifice was located approximately 6 in. below the liquid N_2 surface and the dilution of the ClF_3 with helium ranged from 30 to 46 volumes of helium to 1 volume of ClF_3 vapor. The injection rate varied from 784 to 1835 cc (STP)/sec. In Experiments 27, 28, and 29, after the ClF_3 particles were prepared in liquid N_2, a portion of the liquid N_2 was allowed to evaporate to form a gel of the desired consistency. In the later experiments, gels of the desired consistency were prepared directly without removal of excess liquid N_2 after injection was completed. The fact that the gels can be prepared directly at the desired concentration without an intermediate evaporation step is significant because one step is eliminated from the preparative procedure. The specific details of each experiment are discussed below. The structure index of the gels was not measured in this series of experiments. The values given were estimated from the measurements made earlier in the program. See Figure 12 for the structure index of gelled liquid N_2 at various ClF_3 gelling agent concentrations.

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

Experiment 37

A liquid N_2 gel containing 2.85 volume percent (8.9 wt%) ClF_3 particles was prepared. Based on previous experimental work, this gel would have a structure index of approximately 400 dynes/cm^2. The flow rate remained constant during the course of the measurement. During flow, no appreciable hang up of the gel on the vessel walls was observed. The quantity of gel flowed was 1127 cc in 12.4 sec at a Δp of 1.75 psi. The results of this experiment are presented in Table XII.

Experiment 38

A liquid N_2 gel containing 4.75 volume percent (13.45 wt%) ClF_3 particles was prepared. On the basis of previous experimental work, this gel would have a structure index of approximately 1400 dynes/cm^2. The flow rate remained constant during the course of the measurement. During flow, no appreciable hang up of the gel on the vessel walls was observed. The quantity of gel flowed was 1127cc in 43.4 sec at a Δp of 1.25 psi. The results of this experiment are presented in Table XIII.

Experiment 39

A liquid N_2 gel containing 5.28 volume percent (14.8 wt%) ClF_3 was prepared. On the basis of previous experimental work, this gel would have a structure index of approximately 1650 dynes/cm^2. During flow, only slight hang up of the gel on the vessel wall was observed. The sequence of flow was as follows:

(1) Initial flow was 334 cc in 11.5 sec at a Δp of 1.57 psi.

(2) Flow was then stopped and the system held at rest for 5 minutes.

(3) The gel was pressurized to 3.05 psi and 320 cc of gel flowed in 2.1 sec.

(4) Flow was then stopped and the system held at rest for 5 minutes.

(5) The gel was pressurized to 0.75 psi and 69 cc for gel flowed in 78.9 sec.

(6) The pressure was increased to 1.59 psi and 174 cc of gel flowed in 13.4 sec. The total volume of gel flowed was 897 cc.

TABLE XII

EXPERIMENT 37--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

Concentration of Gelling Agent 2.85 vol percent

Δ Pressure, dynes/cm^2 x 10^{-4}	Δ Pressure, psi	Flow Parameters Rate, cm^3/sec	Flow Parameters Velocity, cm/sec	Reynolds No.	Shear Rate $\frac{8V}{D}$, sec^{-1}	Shear Stress $\frac{D\Delta P}{4L}$ dynes/cm^2	Viscosity, poise
12.06	1.75	90.9	190.2	4000	1949	59	0.03
Ungelled liquid N_2							0.0016

TABLE XIII

EXPERIMENT 38--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

Concentration of Gelling Agent 4.75 vol percent

Δ Pressure, dynes/cm^2 x 10^{-4}	Δ Pressure, psi	Flow Parameters Rate, cm^3/sec	Flow Parameters Velocity, cm/sec	Reynolds No.	Shear Rate $\frac{8V}{D}$, sec^{-1}	Shear Stress $\frac{D\Delta P}{4L}$ dynes/cm^2	Viscosity, poise
8.62	1.25	26.0	54.3	280	557	68.9	0.124
Ungelled liquid N_2							0.0016

TABLE XIV

EXPERIMENT 39--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

Concentration of Gelling Agent 5.28 vol percent

Run	Δ Pressure, dynes/cm^2 x 10^{-4}	Δ Pressure, psi	Flow Parameters Rate, cm^3/sec	Flow Parameters Velocity, cm/sec	Reynolds No.	Shear Rate $\frac{8V}{D}$, sec^{-1}	Shear Stress $\frac{D\Delta P}{4L}$ dynes/cm^2	Viscosity, poise
1	10.82	1.57	29	60.7	315	623	83.8	0.135
2	20.97	3.04	152	318	9700	3261	80.2	0.023
3	5.20	0.75	0.87	1.82	0.3	18.7	74.1	3.97
4	11.09	1.59	13.0	27.2	53	279	100	0.36
Ungelled liquid N_2								0.0016

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

The results of this experiment are presented in Table XIV and the results obtained are also presented as a plot of $\frac{D\Delta P}{4L}$ versus $\frac{8V}{D}$ in Figure 20; this mode of presentation is referred to as the "Characteristic Flow Curve."

Experiment 28

A liquid N_2 gel containing 2.24 volume percent ClF_3 (6.7 wt%), structure index less than 500 dynes/cm^2, particles was prepared. Flow data were obtained and the results are presented in Table XV. As the level-monitoring device did not function properly, the time of flow can only be considered approximate. Therefore, the viscosity values are correct only to within an order of magnitude.

Experiments 29 and 30

In both experiments an excellent gel was obtained. However, no flow occurred even when the gel was pressurized to 9 psig. It was concluded that the gel was not flowing because a crust of ClF_3 had formed, clogging the viscometer. The most likely cause of crust formation would be evaporation of a portion of liquid N_2 in the gel which was in the flow line. The helium purge, used to prevent siphoning and to provide on-off capability of the flow system evidently carried the gaseous N_2 out of the line into the external bath and, thereby caused additional liquid N_2 evaporation from the gel near the entrance to the flow tube. To prevent this from recurring, the next series of tests were designed so that the helium purge always flowed into the gelling vessel. Helium flow in this direction would keep the tube clear of the gel until the flow test started and, thereby, eliminate the possibility of clogging the flow tube with a crust of ClF_3.

Experiment 31

A liquid N_2 gel containing 2.0 volume percent ClF_3 (6.0 wt%), structure index less than 500 dynes/cm^2, particles was prepared. Flow data were obtained and the results are presented in Table XVI. The level-monitoring device did not function, and the volume of material which flowed through the tube was estimated visually and is known approximately. Therefore, the viscosity values calculated are correct only to within an order of magnitude.

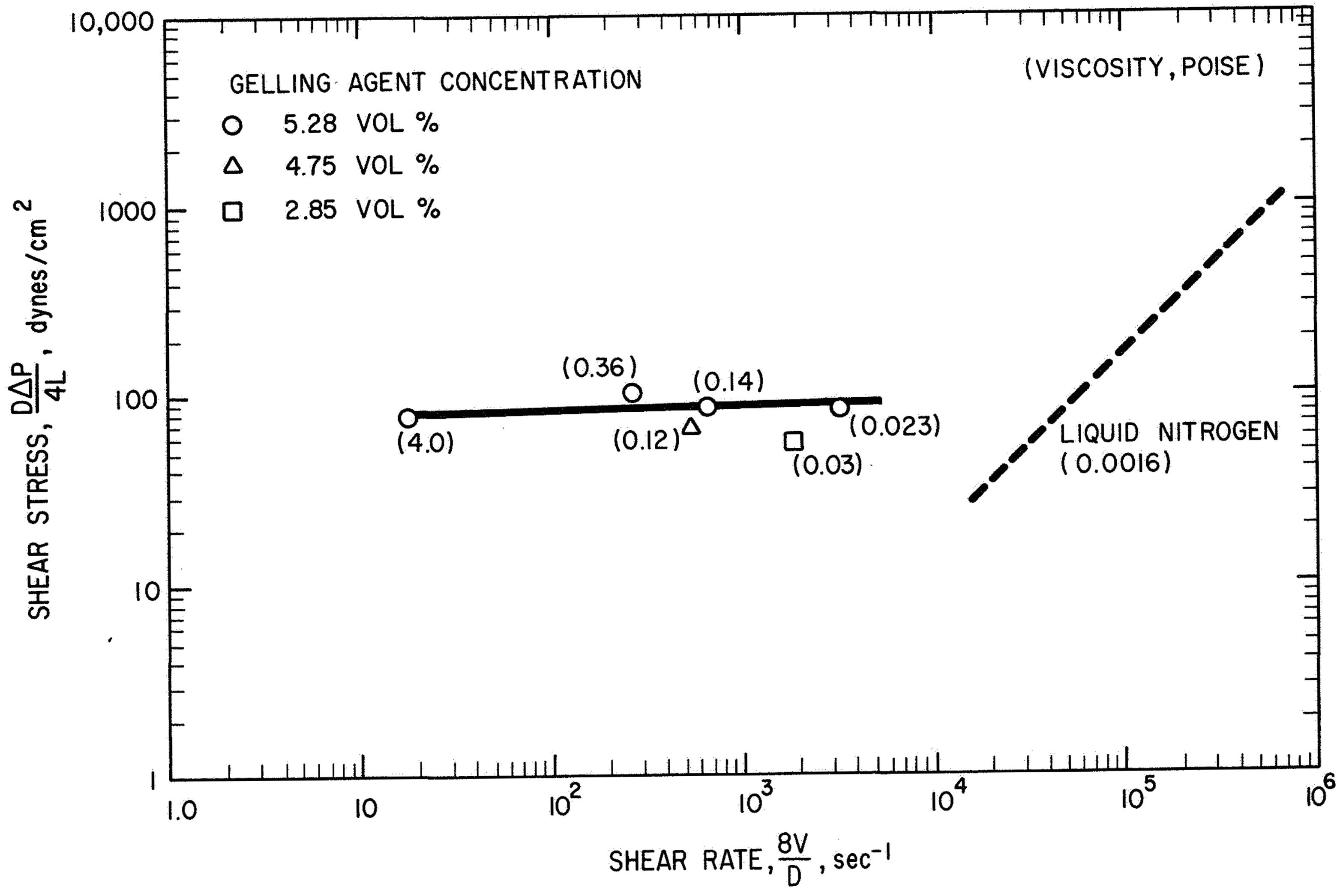


Figure 20. Characteristic Flow Curve of Liquid Nitrogen Gelled with Chlorine Trifluoride

TABLE XV

EXPERIMENT 28--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

Concentration of Gelling Agent 2.24 Vol%, 6.7 wt%

Δ Pressure,		Flow Parameters					
dynes/cm^2 x 10^{-4}	psi	Rate, cm^3/sec	Velocity, cm/sec	Reynolds No.	Shear Rate $\frac{8V}{D}$, sec^{-1}	Shear Stress $\frac{D\Delta P}{4L}$, dynes/cm^2	Viscosity, poise
22.7	3.2	6.1	33.4	130	544	58.1	0.10
14.6	2.0	4.5	24.6	106	408	38.2	0.09
Ungelled liquid nitrogen				---	---	---	0.0016

TABLE XVI

EXPERIMENT 31--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

Concentration of Gelling Agent 2.0 Vol%, 6.0 wt%

Δ Pressure,		Flow Parameters					
dynes/cm^2 x 10^{-4}	psi	Rate cm^3/sec	Velocity, cm/sec	Reynolds No.	Shear Rate $\frac{8V}{D}$, sec^{-1}	Shear Stress $\frac{D\Delta P}{4L}$ dynes/cm^2	Viscosity, poise
34.5	4.8	22.2	121	1570	2000	68.0	0.03
38.7	5.4	34.6	189	3680	3130	64.6	0.02
Ungelled liquid nitrogen				---	---	---	0.0016

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

After the experiment was completed, the thermopile used to monitor the liquid level was inspected. There was evidence of considerable corrosion on the exposed ends of the thermocouples. Therefore, the failure of the level-monitoring device was attributed to corrosion of the individual thermocouples in the thermopile. This corrosion occurred while the ClF_3 was being evaporated from the system after each experiment was completed. Consequently, the thermopile was removed and fiducial marks were placed on the external bath so that the volume of material which flowed during the experiment could be measured visually.

Experiment 32

A liquid N_2 gel containing 2.1 volume percent ClF_3 (6.2 wt%, structure index less than 500 dynes/cm^2) particles was prepared. Flow data were obtained and the results are presented in Table XVII. The results obtained are also presented as a plot of $\frac{D\Delta P}{4L}$ versus $\frac{8V}{D}$ in Figure 21; this mode of presentation is referred to as the "Characteristic Flow Curve."

Experiment 33

A liquid N_2 gel containing 3.1 volume percent ClF_3 (9.5 wt%, structure index less than 500 dynes/cm^2) particles was prepared. Flow of the gel was initiated at 2.2 psi; however, the flow rate gradually decreased and reached zero. The pressure was increased to 5.4 psi and flow began again but the flow rate gradually decreased and reached zero. An attempt to initiate flow again by pressurizing the system to 9.3 psi was unsuccessful. No usable data were obtained because the flow rate was not constant. The equipment was modified after the experiment so that small flow volumes could be measured.

Experiment 34

A liquid N_2 gel containing 3.0 volume percent ClF_3 (9.4 wt%, structure index less than 500 dynes/cm^2) particles was prepared. Flow data was obtained at 3.0 psi and is presented in Table XVIII. After the initial measurements, the flow rate gradually decreased and reached zero. After flow ceased, the pressure was increased to 7.75 psi but flow did not restart. One possible cause of the gradually decreasing flow rate is the boiling of the liquid N_2 as it flowed through that portion of the coil which was above the surface of the gel. Consequently, a new coil was fabricated that did not have any portion above the level of the gel.

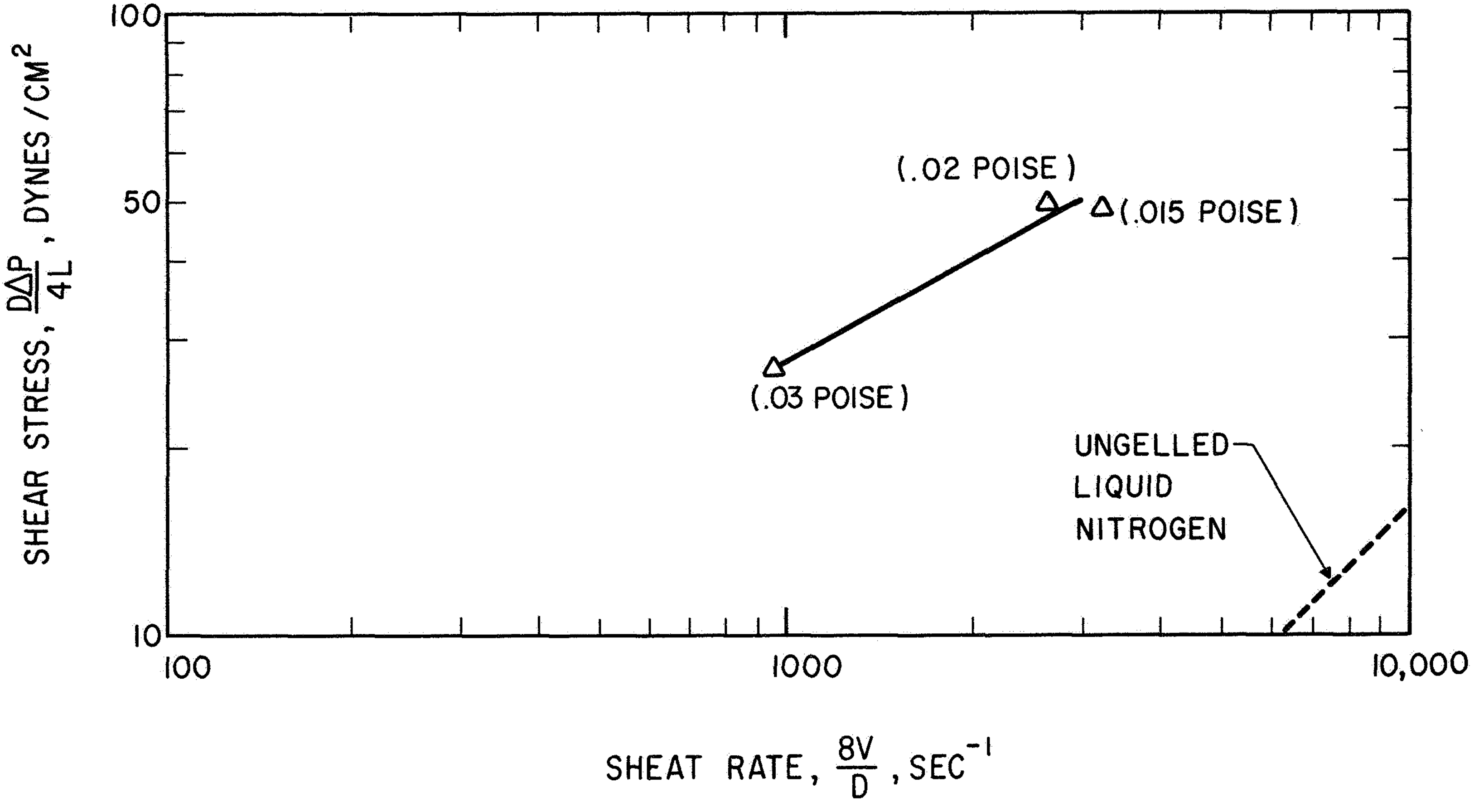


Figure 21. Preliminary Characteristic Flow Curve of Liquid Nitrogen Gelled with 2.1 Volume Percent Chlorine Trifluoride

TABLE XVII

EXPERIMENT 32--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

Concentration of Gelling Agent 2.1 Vol%, 6.2 wt%

Δ Pressure,		Flow Parameters					
dynes/cm^2 x 10^{-4}	psi	Rate, cm^3/sec	Velocity, cm/sec	Reynolds No.	Shear Rate $\frac{8V}{D}$, sec^{-1}	Shear Stress $\frac{D\Delta P}{4L}$ dynes/cm^2	Viscosity, poise
11.7	1.6	10.7	58.5	760	970	27.7	0.03
27.5	3.8	28.9	158	3080	2620	49.3	0.02
28.8	4.1	34.8	191	4960	3167	47.8	0.015
Ungelled liquid N_2							0.0016

TABLE XVIII

EXPERIMENT 34--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

Concentration of Gelling Agent 3.0 Vol Percent

Δ Pressure,		Flow Parameters					
dynes/cm^2 x 10^{-4}	psi	Rate, cm^3/sec	Velocity, cm/sec	Reynolds No.	Shear Rate $\frac{8V}{D}$, sec^{-1}	Shear Stress $\frac{D\Delta P}{4L}$ dynes/cm^2	Viscosity, poise
21.37	3.1	2.94	16.1	447	266	57.2	0.21
20.68	3.0	2.18	11.9	440	197	55.8	0.28
20.68	3.0	2.31	12.7	442	210	56.5	0.27
Ungelled liquid N_2							0.0016

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

Experiment 35

A liquid N_2 gel containing 2.0 volume percent ClF_3 (8.5 wt%, structure index less than 500 dynes/cm^2) particles was prepared. Flow data were initially obtained at 2.8 psi; however, the flow rate again gradually decreased and reached zero; flow could not be restarted at 7.5 psi. It was, however, possible to flow the gel back into the gelling vessel by pulling a slight vacuum on the gelling vessel. The gel (at a new concentration because of the neat liquid N_2 present in the outer jacket) was again flowed. This procedure was repeated several times and the results are presented in Table XIX. After the gel had been flowed back and forth two times, no further decrease in flow rates during each flow experiment was observed.

Another possible cause for the decreasing flow rate is the presence of lumps in the gel which were clogging the flow tube. Consequently, in Experiment 36, the stirrer was repositioned so that the gel could be sheared to break up any lumps that may form just before the gel entered the flow tube.

TABLE XIX

EXPERIMENT 35--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

			Δ Pressure,		Flow Parameters			Shear Rate	Shear Stress	
Run	vol%	wt%	Dynes/cm^2 x 10^{-4}	psi	Rate, cm^3/sec	Velocity, cm/sec	Reynolds Number	$\frac{8V}{D}$, sec^{-1}	$\frac{D\Delta P}{4L}$	Viscosity, poise
1	2.8	8.5	19.30	2.8	7.3	39.9	126	661	81.5	0.12
2	2.1	6.6	24.82	3.6	27.3	149	1409	2474	75	0.03
3	1.3	4.0	28.96	4.2	41.5	228	4686	3770	71	0.02
4	.8	2.3	16.55	2.4	27	149	2842	2465	50	0.02
ungelled liquid N_2										0.0016

III, 2, Task VII--Development of Techniques to Gel Oxygen Difluoride and to Measure Engineering Properties of the Gel (cont.)

Experiment 36

A liquid N_2 gel containing 3.3 volume percent ClF_3 (9.6 wt%, structure index approximately 500 dynes/cm^2) particles was prepared. Flow was initiated at 3.5 psi; however, the flow rate gradually decreased and reached zero. The observed gradual decrease in flow rate is presented in Table XX.

TABLE XX

EXPERIMENT 36--FLOW CHARACTERISTICS OF GELLED LIQUID NITROGEN - GELLING AGENT ClF_3

Run	Length of Time Gel had been Flowing, sec	Δ Pressure, dynes/cm^2 x 10^{-4}	Δ Pressure, psi	Flow Rate, cm^3/sec
1	6	24.13	3.5	22.7
	11.8	24.13	3.5	15
	21	24.13	3.5	7.0
	75.5	24.13	3.5	1.1
	Flow Stopped			
2	4.3	34.47	5.0	16.8
	8.8	34.47	5.0	12.2
	21	34.47	5.0	6.5
	38.7	34.47	5.0	4.5
	52	34.47	5.0	5.3
	Flow Stopped			

Because the most likely cause of the decreasing flow rate and eventual clogging of the flow tube at a constant pressure was small lumps in the gel, two additional steps were taken to eliminate this problem. First, the stirrer blade was replaced with two high shear turbine blades and second, a new flow tube was fabricated, calibrated, and installed which had an ID of 0.78 cm instead of 0.48 cm as originally used.

(b) Significance of the Measurement of the Flow Properties of Gelled Liquid Nitrogen - Gelling Agent ClF_3 Particles

The series of experiments just discussed with liquid N_2 gelled with ClF_3 had demonstrated that thin particulate cryogenic gels could be flowed without appreciable difficulty and that thick gels (structure index between 1400 and 1650 dynes/cm^2) could be flowed without difficulty if special care is taken to ensure that the gel is uniform. In addition, the ability to stop flow and then restart it after a rest period both at pressures higher and lower than the previous flow pressure was demonstrated. The extremely rapid shear thinning that was predicted for this type of gel was demonstrated; i.e., the viscosity decreases very rapidly with increasing shear rate (see Figure 20).

The results of the flow work just described indicate that to successfully employ this gelled propellant in a flight system special precautions will have to be taken to ensure that the gel is smooth, i.e., free from lumps. This means that it will be necessary to develop a quality control procedure for measuring the degree of smoothness possessed by the gel.

The extremely rapid shear thinning exhibited by the gel, i.e., rapid decrease in viscosity with increasing shear rate, strongly indicates that valves, propellant lines, and injectors can be designed as if the gelled propellant was a neat liquid.

3. TASK VIII--GELATION OF OXYGEN DIFLUORIDE WITH FLUORINATED OXIDIZERS

For the gelation of liquid OF_2, the particles of ClF_3 were prepared in liquid N_2. This approach was taken for safety reasons. The method used for the preparation of the fine particles of ClF_3 requires that the ClF_3-helium mixture be introduced into the cryogenic liquid at a rate of 375 cc (STP)/sec or higher. It was believed that the addition of ClF_3-helium mixtures at this rate into liquid OF_2 would create a potentially hazardous condition. Consequently, the approach taken was to prepare the ClF_3 particles in liquid N_2, pump or sparge off the liquid N_2 and then add the OF_2 to the gelling vessel.

The OF_2 was added by two procedures which appeared to be equally satisfactory. In one, OF_2 vapor was added to and condensed in the gelling vessel. Essentially complete removal of the LN_2 was assured by leaving the vent valve open during the initial portion of the OF_2 condensation. The heat of condensation of the OF_2 provided the energy required to vaporize the residual liquid N_2. The other procedure for gelation of OF_2 with ClF_3 particles was to add liquid OF_2 directly to the vessel in which the particles of ClF_3 were prepared in the liquid N_2 after most of the liquid N_2 had been removed. A helium sparge was then used to remove the residual N_2 which had a vapor pressure of 1 atmosphere while the vapor pressure of liquid OF_2 is approximately one Torr.

Gelled OF_2 was first prepared in Experiment 13. The ClF_3 particles were prepared in liquid N_2. The liquid N_2 was pumped off, and gaseous OF_2 was condensed to the liquid phase in the gelling vessel. Table XXI presents the experimental parameters.

Gelled OF_2 was again prepared in Experiment 14. The procedure was similar to that used in Experiment 13 except that the vent valve was left open for a longer period of time during the OF_2 condensation and the mixture was vigorously stirred until all of the liquid N_2 had vaporized. Table XXII presents the experimental parameters and Figure 22 is a photograph of the gelled OF_2.

The structure index of the gel was measured at three gelling agent concentrations at the NBP of liquid N_2 (see Section III,4,a for a discussion of the results). The gel was then gradually warmed to the NBP of liquid OF_2 with vigorous stirring. An explosion occurred which terminated the experiment, just as the gel reached the NBP of liquid OF_2.

TABLE XXI

EXPERIMENT 13--THE PREPARATION OF GELLED LIQUID OXYGEN DIFLUORIDE WITH CHLORINE TRIFLUORIDE PARTICLES

Preparation of Gelling Agent	
Gelling Agent and Diluent	ClF_3 and He
Dilution	
$\frac{\text{vol of gas}}{\text{vol of gelling agent}}$	35.8
Orifice	
Diameter (in.)	0.025
Location	6 in. below LN_2 level
Flow	
Rate (cm^3(STP)/sec)	364
Velocity (cm/sec)	11.7×10^4
Reynolds No.	26,000
Size of Particles Prepared (micron)	0.6(1)
Gelled Liquid N_2 Properties	
Structure Index (dynes/cm^2)	2750 at 14.7% ClF_3
Gelled OF_2 Properties	
Structure Index (dynes/cm^2)	2500 at -196C
Gelled OF_2 Composition	
ClF_3(wt%)	6
LN_2 (wt%)	3
OF_2 (wt%)	91
Quantity of Gel Prepared, g	1209 (2.66 lb)

---

(1) Particle size calculated from a visual estimate of the particle settling rate.

TABLE XXII

EXPERIMENT 14--PREPARATION OF GELLED OF_2 - GELLING AGENT ClF_3 PARTICLES

Preparation of Gelling Agent	
Gelling Agent and Diluent	ClF_3 and He
Dilution	
$\frac{\text{vol of gas}}{\text{vol of gelling agent}}$	35.8
Orifice	
Diameter, in.	0.025
Location	6 in. below LN_2 level
Flow	
Rate, cm^3(STP)/sec	375
Velocity, cm/sec	12.1×10^4
Reynolds No.	26,900
Size of Particles Prepared (micron)	0.9(1)
Gelled OF_2 Properties	
Gel Composition	Structure Index at -196°C
93.3% OF_2 6.7% ClF_3	1900 dynes/cm^2
94.4% OF_2 5.6% ClF_3	1450 dynes/cm^2
94.9% OF_2 5.1% ClF_3	950 dynes/cm^2
Quantity of Gel Prepared, g	1440 (3.17 lb)

---

(1) Particle size calculated from a visual estimate of the particle settling rates.

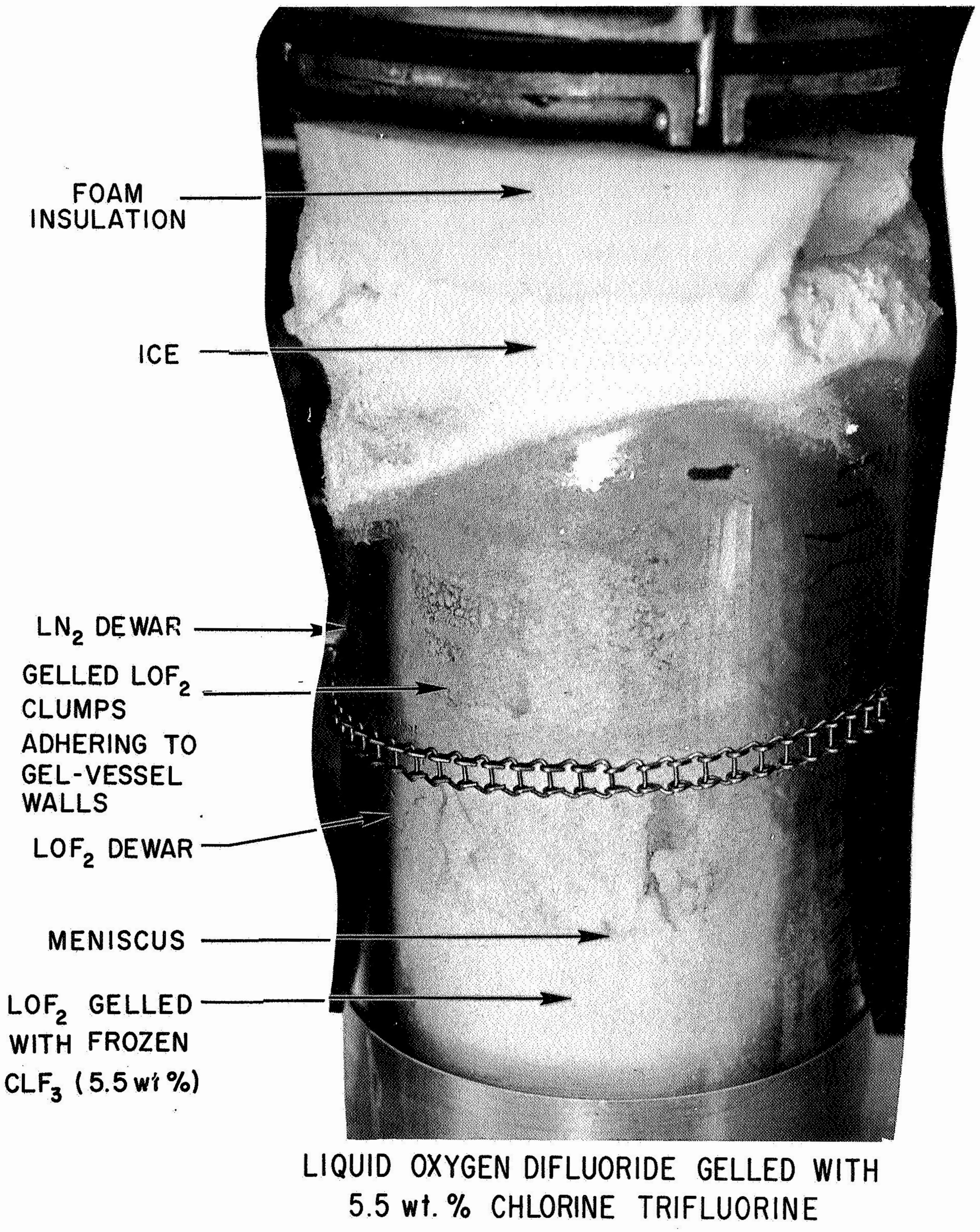


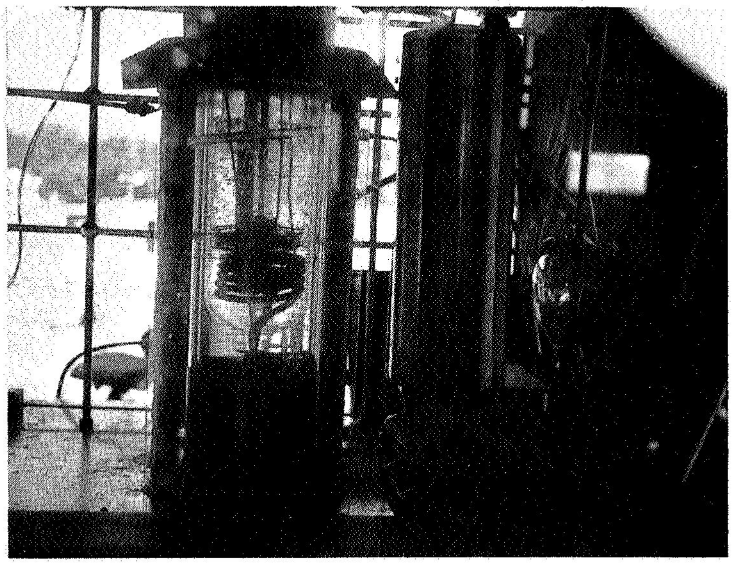
Figure 22. Liquid Oxygen Difluoride Gelled with 5.5 wt% Chlorine Trifluoride

Five possible causes of the explosion have been hypothesized: (1) ignition of the asbestos liner of the injection tube; (2) ignition of the aluminum foil liner of the inlet tube; (3) ignition of the Teflon tape on the injection tube; (4) ignition of the propane gas which could have been back pressured into the system from the burn pit; and (5) ignition of some of the silicone grease used on the Teflon O-ring between the gelling vessel and the vessel cap. Experimental work has shown that it was possible for the propane from the burn pit to be back pressured into the system. There was some visual evidence that indicated that the fire which caused the explosion was combustion of the asbestos. However, this evidence was not conclusive.

The following steps were taken to eliminate the possibility of a recurrence of this event: (1) a new injection tube was designed and fabricated which does not have an asbestos liner, an aluminum foil liner, or Teflon tape that can be exposed to OF_2 or the ClF_3; (2) the orifice has been silver soldered to the injection tube to eliminate the possibility of leaks into the interior of the injection tube; (3) the burn pit was eliminated, and (4) the use of the silicone grease on the Teflon O-ring was discontinued.

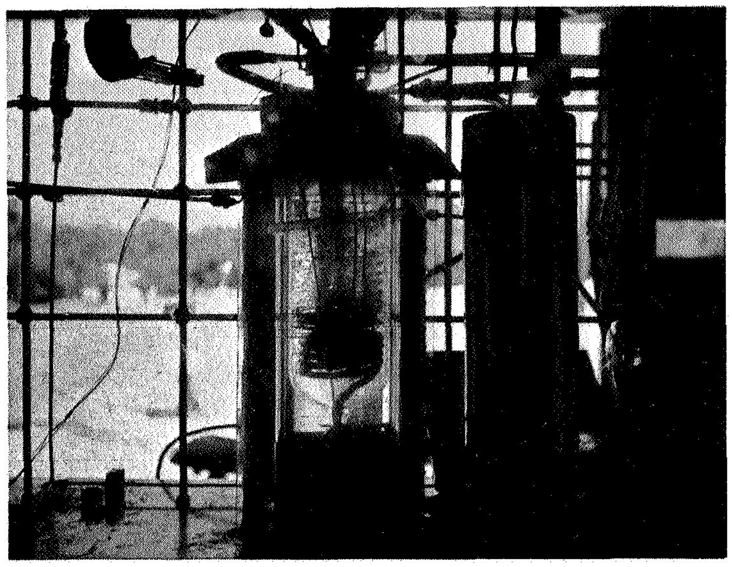
Gelled OF_2 was prepared in Experiment 42. The procedure was similar to that used in the previous experiments except that the OF_2 was liquified before it was added to the gelling vessel. After the addition was completed, the mixture was vigorously stirred and sparged with helium until all of the liquid N_2 was vaporized off. Figures 23-1 through 23-4 illustrate the preparation of the gelled OF_2. Table XXIII presents the experimental parameters. The OF_2 gel prepared in this experiment was used to measure flow properties (see Section III,4,b, for discussion of the results).

The results of the experimental work just described demonstrated that OF_2 can be gelled with fine particles of ClF_3. The procedure used was straightforward and substantial quantities of gelled OF_2 were prepared. In Experiment 13,



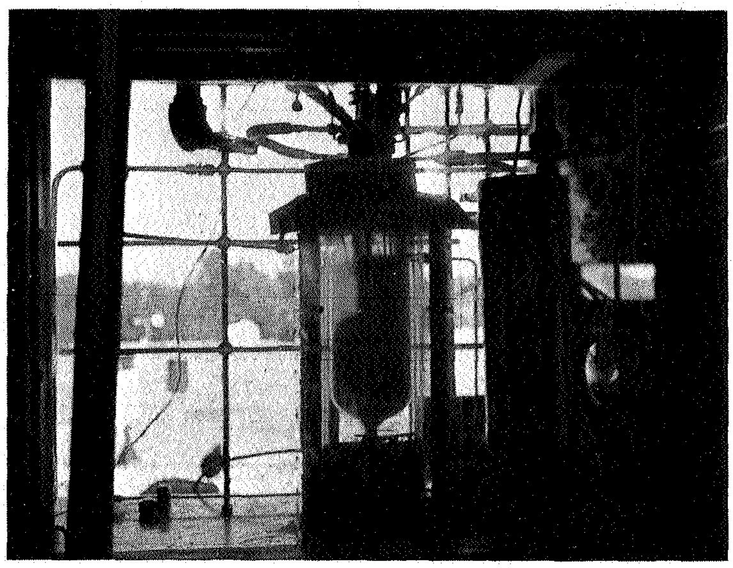
(Gelling vessel with flow viscometer installed is filled with liquid nitrogen)

Figure 23-1. Preparation of Gelled Oxygen Difluoride



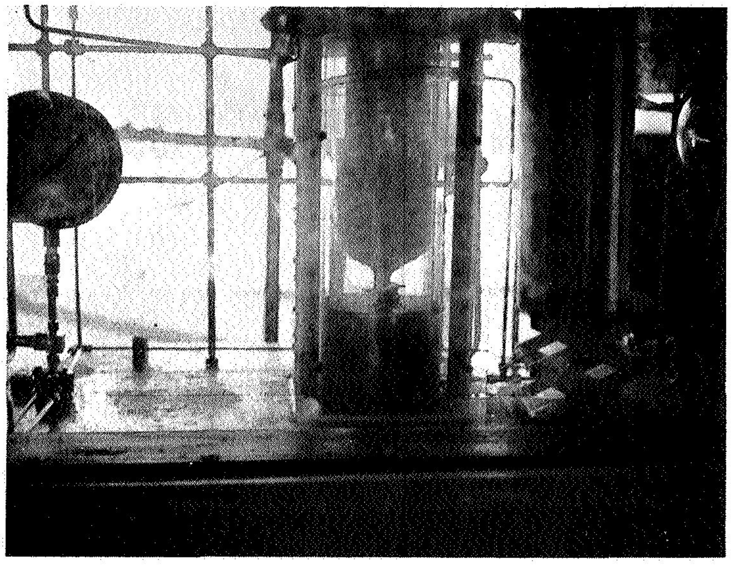
(Stirrer is on and neat helium is flowing through injection tube at programmed rate)

Figure 23-2. Preparation of Gelled Oxygen Difluoride



(Helium/chlorine trifluoride mixture is flowing through injection tube at programmed rate. The opaque area is ClF_3 particles in liquid N_2.)

Figure 23-3. Preparation of Gelled Oxygen Difluoride



(Liquid nitrogen is completely removed and liquid oxygen difluoride is added. Uniform, stable OF_2 /ClF_3 gel is formed by vigorous stirring

Gel composition:

$OF_{2(l)}$	95.0 wt%
$ClF_{3(s)}$	5.0 wt%
Amount	1893 g (4.17 lb)
Structure Index	950 dynes/cm^2)

Figure 23-4. Preparation of Gelled Oxygen Difluoride

TABLE XXIII

EXPERIMENT 42--PREPARATION OF GELLED OF_2 - GELLING AGENT ClF_3 PARTICLES

Preparation of Gelling Agent	
Gelling Agent and Diluent	ClF_3 and He
Dilution	
$\frac{\text{vol of gas}}{\text{vol of gelling agent}}$	60
Orifices	
Diameter, in.	0.025
Location	6 in. below LN_2 level
Flow	
Rate, cm^3(STP)/sec	1480
Velocity, (cm/sec)	47.7×10^4
Reynolds Number	106,000
Gelled OF_2 Properties	
Gel Composition	
95.0% OF_2	
5.0% ClF_3	
Estimated Structure Index, dynes/cm^2)	950
Quantity of Gel Prepared, g	1893 (4.17 lb)

Note: Gel used for flow property measurements.

2.66 lb of gel were prepared, 3.17 lb in Experiment 14 and 4.17 lb in Experiment 42. Thus, a total of 10 lb of gelled OF_2 was prepared in the course of the program. It is believed that the procedure for preparing the gelling agent and subsequent preparation of the gelled OF_2 can be scaled up to produce the gelled propellants in quantity. The successful gelation of OF_2 with fine particles of ClF_3 established that the primary objective of this program has been accomplished.

4. TASK IX--MEASUREMENT OF THE ENGINEERING PROPERTIES OF GELLED OXYGEN DIFLUORIDE

As was previously discussed in the Introduction to this report, a particle gels a liquid because of the properties of the particle. The ability to gel a liquid arises when the particle is small enough so that there is an attraction between the individual particles. Apparently any particle, if it is small enough, possesses this attractive force which causes the particles to form interconnecting chains or networks that form micelles which trap the liquid. A gel-like material is the result. The property that makes particulate gelling agents uniquely distinctive is that gel formation is essentially independent of the chemical properties of the liquid. In other words, if the particles are small enough, and if there are enough particles per unit volume so that chains or networks form, the particles will gel any liquid in which they are insoluble and nonreactive.

A correllary that arises from this theory of the cause of particulate gel formation is that the rate of change of gel properties of any series of gelled liquids with changing gelling agent concentration, if gelled with a common particulate gelling agent, will be primarily determined by the gelling agent rather than the liquid. This means that the rate of change of a gel property expressed as a function of the volume concentration of the particulate gelling agent should be essentially similar from liquid to liquid if a common gelling agent is used. The significance of this expected similarity in the behavior of gelled liquids, under both static and flow conditions is that initial development work for the gelation and utilization of energetic propellants can be conducted using an inert liquid simulant gelled with the particulate gelling agent selected. Consequently, only a few tests with the actual propellant are required to confirm the results obtained with the simulant. The savings in time and money that could be achieved, if this expected similarity in gel properties could be established, is readily apparent. The primary dependence of particulate gel formation on the particle rather than on the nature of the liquid is indicated by the fact that the ClF_3 particles gel both liquid OF_2 and liquid N_2.

This portion of the report presents the results of the measurements of the properties of liquid OF_2 gelled with ClF_3 particles. In addition, these results are compared to the results of the measurements of the properties of liquid N_2 gelled with ClF_3 particles. The comparative data is presented as a function of volume because particles gel a liquid when a sufficient number of particles are present in a unit volume of liquid to form the chains and networks which are required to produce the gel-like structure.

a. Static Properties

The development of the technique used for measuring the static properties of cryogenic particulate gels was described in Section III,2,d,(1). The specific method used for measuring the structure index of gelled OF_2 was discussed on Page 45 of that section.

The structure index of gelled OF_2 was measured at three gelling agent concentrations in Experiment 14. The results of these measurements are presented in Table XXIV and in Figure 24.

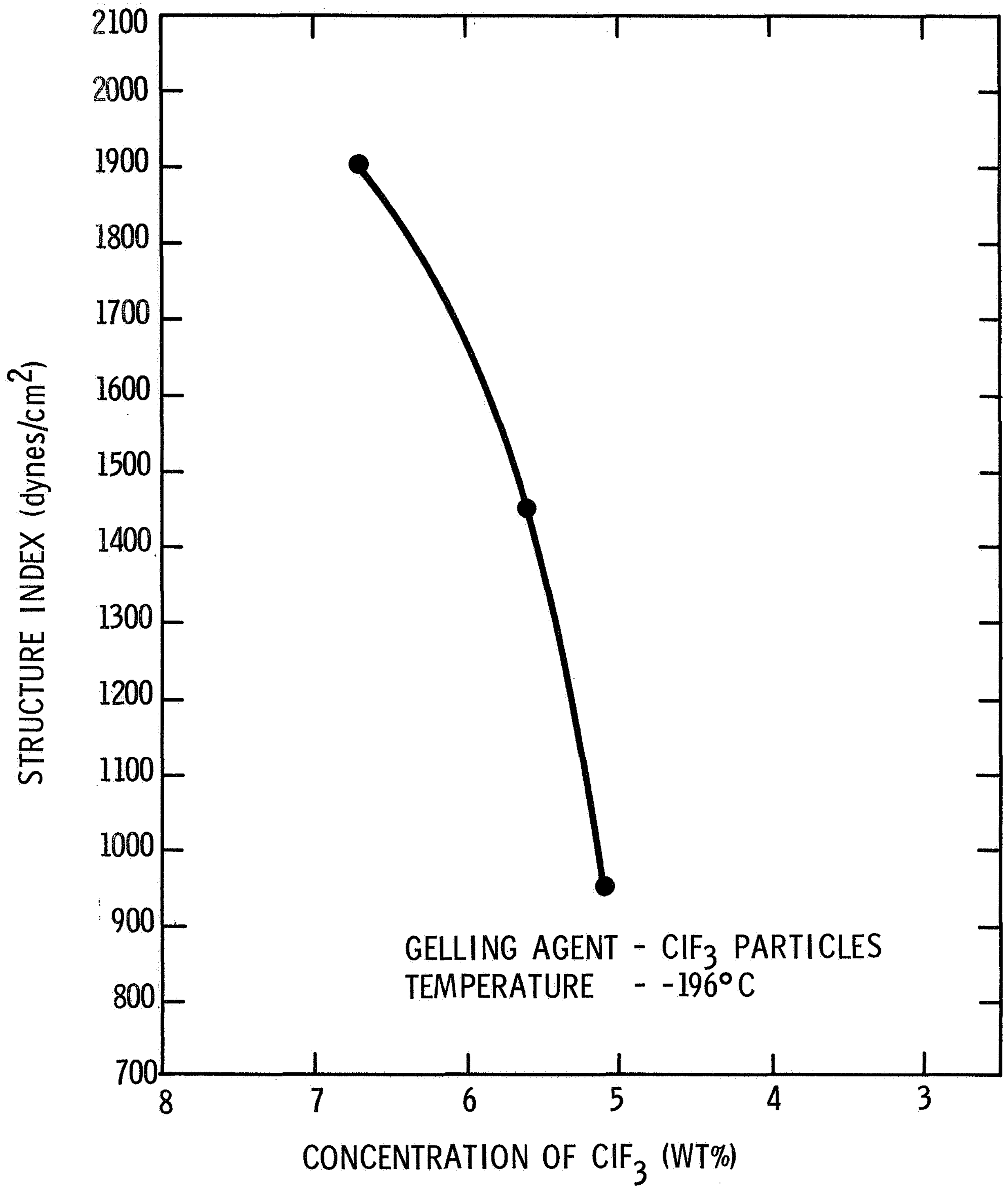


Figure 24. Structure Index of Gelled OF_2 at Various Gelling Agent Concentrations

III, 4, Task IX--Measurement of the Engineering Properties of
Gelled Oxygen Difluoride (cont.)

The structure index of gelled liquid OF_2 and gelled liquid N_2 is presented in Figure 25. The data are presented as a function of the concentration of ClF_3 in vol%. As theory predicts, the incremental volume of ClF_3 particles required to achieve a given increase in the degree of structure is comparable.

TABLE XXIV

STRUCTURE INDEX OF GELLED OF_2
AT VARIOUS GELLING AGENT CONCENTRATIONS

Gelling Agent ClF_3

Gel Composition, wt%	Structure Index at -196°C, dynes/cm^2
93.3% OF_2 6.7% ClF_3	1900
94.4% OF_2 5.6% ClF_3	1450
94.9% OF_2 5.1% ClF_3	950

b. Flow Properties

The development of the technique used for measuring the flow properties of cryogenic particulate gels was described in Section III,2,d,(2). The specific apparatus used for measuring the flow properties of gelled OF_2 was discussed on Pages 50 through 65 of that section. The preparation of the gelled OF_2 used for the flow measurement was described in Section III,3.

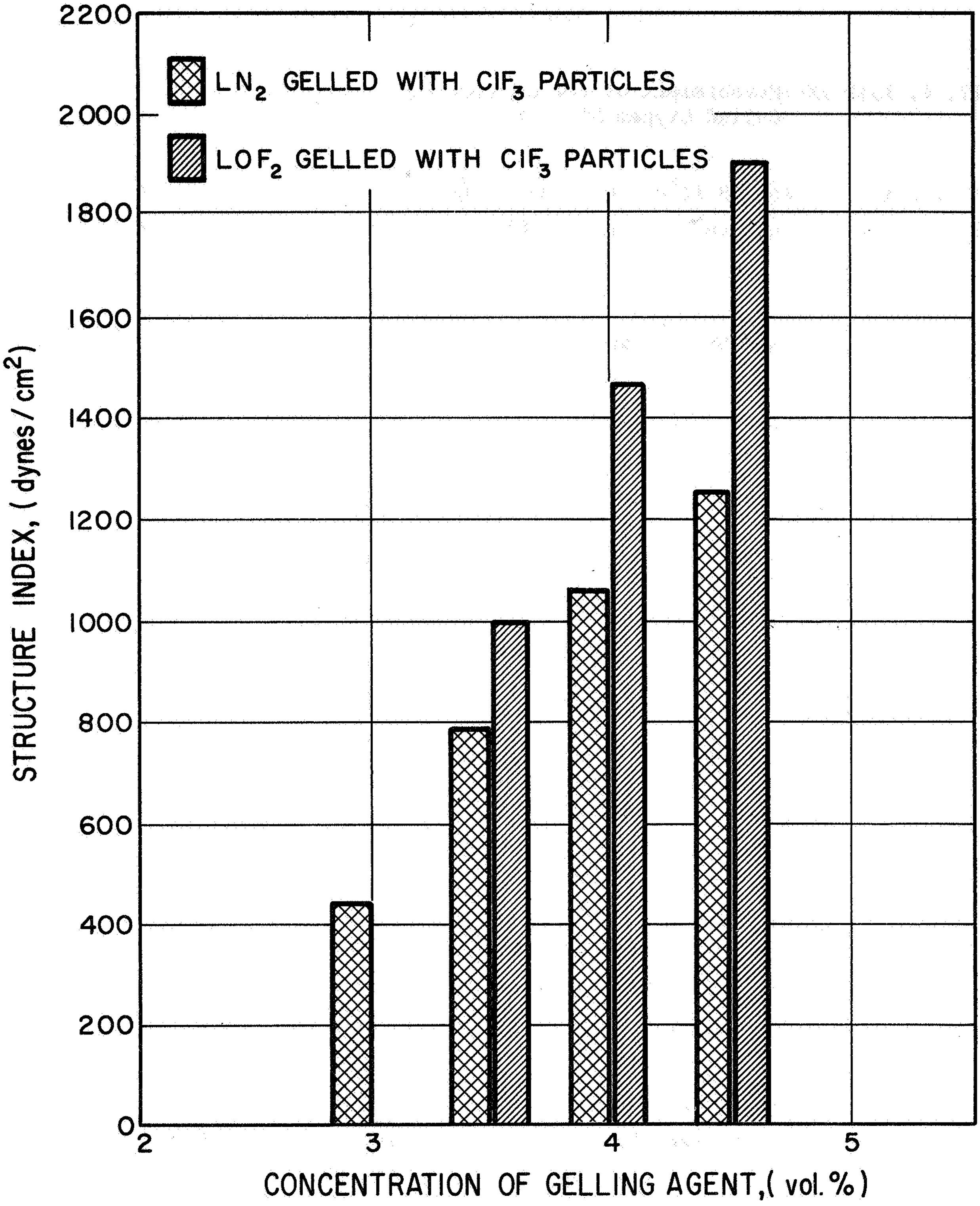


Figure 25. Structure Index of Gelled OF_2 and Liquid Nitrogen Gelled with Chlorine Trifluoride Particles

III, 4, Task IX--Measurement of the Engineering Properties of Gelled Oxygen Difluoride (cont.)

Figures 17, 18, and 19 illustrate the cryogenic gel viscometer. Figure 26 illustrates the test bay side of the OF_2 gellation equipment and the cryogenic flow viscometer. Figure 27 illustrates the laboratory side of the equipment.

A liquid OF_2 gel was prepared and its flow properties measured in Experiment 42. The gel contained 3.57 vol% (5.0 wt%) ClF_3. On the basis of previous experimental work, this gel had a structure index of approximately 950 dynes/cm^2. During flow, only slight hangup of the gel was observed. The sequence of flow was as follows:

(1) Initial flow was 246 cc in 44.3 sec at a ΔP of 2.94 psi.
(2) Flow was then stopped and the pressure changed.
(3) The gel was pressurized to 4.03 psi and 297 cc of gel flowed in 8.2 sec.
(4) Flow was then stopped and the pressure changed.
(5) The gel was pressurized to 4.63 psi and 147 cc of gel flowed in approximately 2 sec.
(6) Flow was then stopped and the pressure changed.
(7) The gel was pressurized to 1.67 psi and 68 cc of gel flowed in 182 sec. The total volume of flow was 758 cc.

The results of this experiment are presented in Table XXV and a plot of $\frac{D\Delta P}{4L}$ versus $\frac{8V}{D}$ in Figure 28. This mode of presentation is referred to as the Characteristic Flow Curve.

The Characteristic Flow Curves of gelled liquid OF_2 and gelled liquid N_2 are presented in Figure 29. Note that the sets of data produce parallel lines which means that the viscosities of the gelled liquids decrease at the same rate with increasing shear rates. This observation is in agreement with the theory that the properties of a particulate gel are determined by the gelling agent rather than by the liquid gelled.

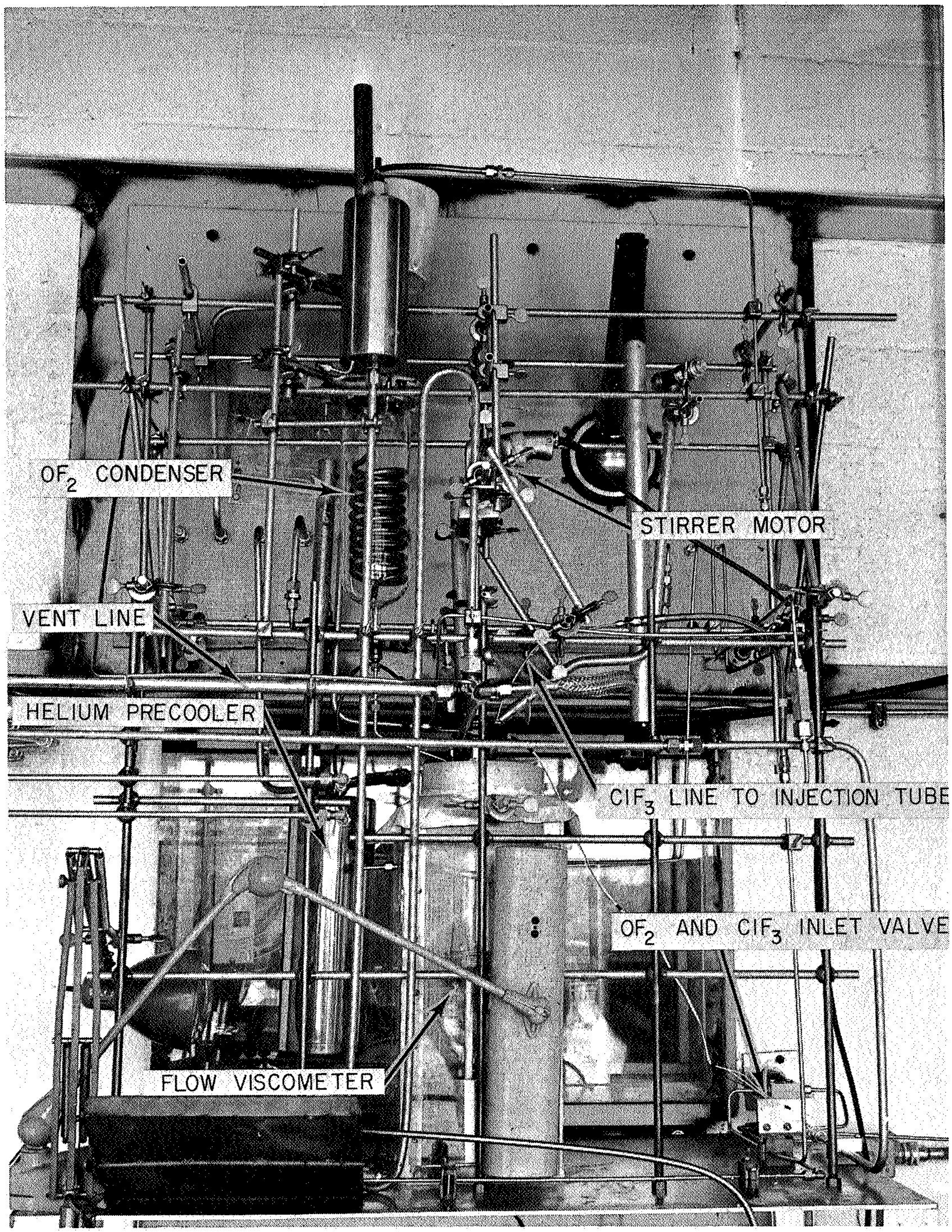


Figure 26. Facility for Gelation of Oxygen Difluoride and the Cryogenic Flow Viscometer (Test Bay Side)

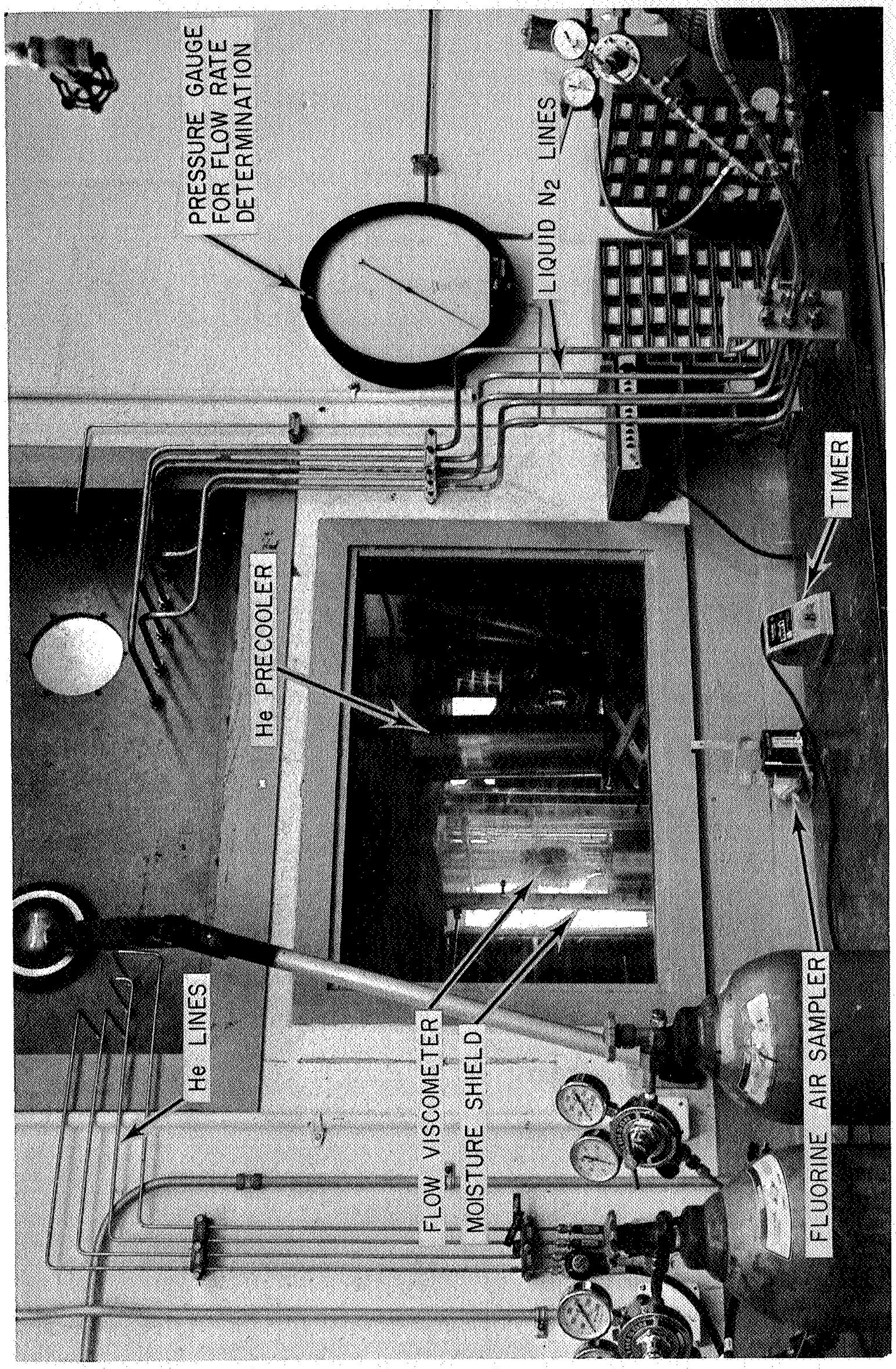


Figure 27. Facility for Gelation of Oxygen Fluoride and the Cryogenic Flow Viscometer (Laboratory Side)

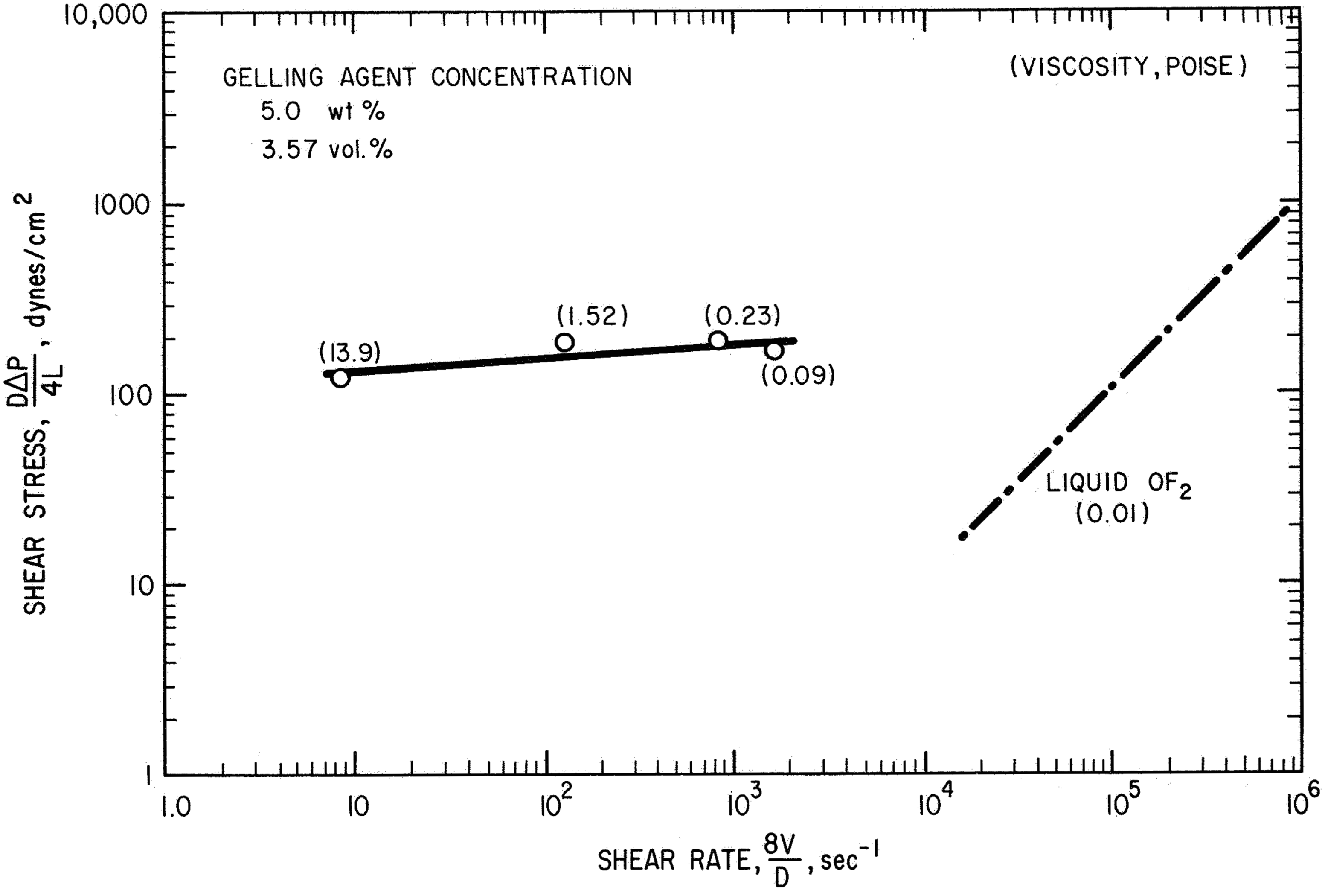


Figure 28. Characteristic Flow Curve of Liquid OF_2 Gelled with Chlorine Trifluoride

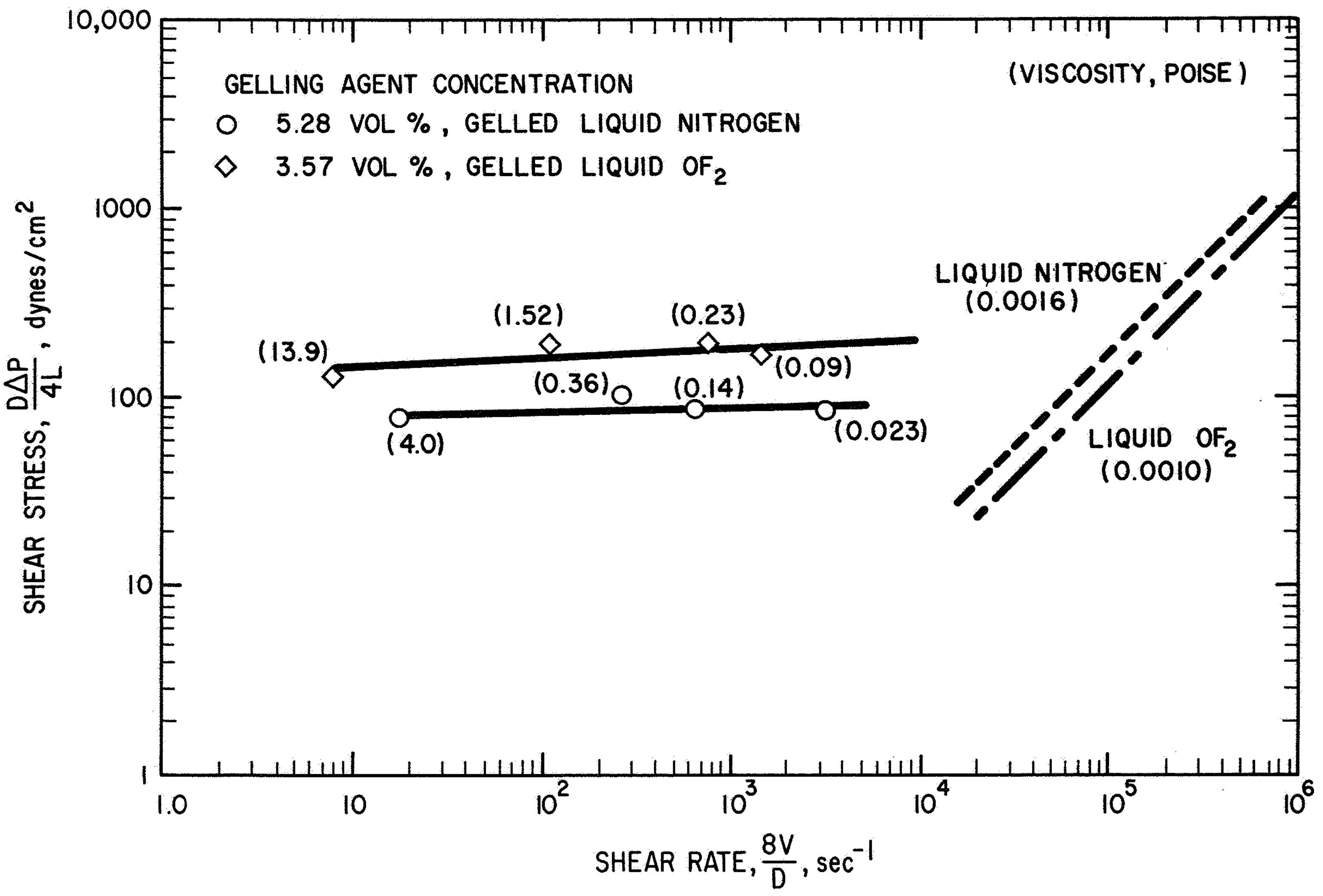


Figure 29. Characteristic Flow Curves of Liquid Nitrogen and Oxygen Difluoride Gelled with Chlorine Trifluoride

III, 4, Task IX--Measurement of the Engineering Properties of Gelled Oxygen Difluoride (cont.)

During the experimental work with gelled OF_2, the variation in gel structure with gelling agent concentration was measured and the gelled OF_2 was successfully flowed. The ability to stop flow and then restart it at higher and lower pressures was demonstrated. The extremely rapid shear thinning that was predicted for this type of gel was demonstrated, i.e, the viscosity decreases very rapidly with increasing shear rate (see Figure 28). In addition the similarity in behavior of two gelled liquids, gelled with a common gelling agent predicted by theory was conclusively demonstrated.

TABLE XXV

FLOW CHARACTERISTICS OF GELLED LIQUID OF_2 - GELLING AGENT ClF_3

Concentration of Gelling Agent 5.0%

Run No.	Δ Pressure, psi	Δ Pressure, dynes/cm^2	Flow Parameters Rate, cc/sec	Flow Parameters Velocity, cm/sec	Flow Parameters Reynolds No.	Shear Rate $\frac{8V}{D}$, sec^{-1}	Shear Stress $\frac{D\Delta P}{4L}$ dynes/cm^2	Viscosity $\frac{D\Delta P/8V}{4L/D}$, poise
1	2.94	2.027×10^5	5.55	12.24	11.0	128.9	195.5	1.52
2	4.03	2.770×10^5	36.24	79.90	482	841	191.3	0.227
3	4.63	3.192×10^5	74	163	2360	1716	162.6	0.09
4	1.67	1.151×10^5	0.376	0.83	0.08	8.74	121.5	13.9

Ungelled Liquid OF_2

SECTION IV

CONCLUSIONS AND RECOMMENDATIONS

1. CONCLUSIONS

The conclusions which are drawn from this research program are:

1. A simple and practical rapid condensation technique for the preparation of micron size particles of chlorine trifluoride has been developed and reduced to a routine operation.

2. Excellent gels of OF_2 have been prepared using micron size particles of chlorine trifluoride prepared by the rapid condensation technique.

3. If the particles are formed below the level of the liquid N_2 under the experimental conditions studied, varying the rate at which the gaseous ClF_3-helium mixture and/or the degree of dilution of ClF_3 in He does not have a measureable effect on the particle size.

4. This rapid condensation technique does not result in the preparation of micron particles of ClF_5 that will gel liquid N_2. The most likely cause of the failure of ClF_5 particles to gel liquid N_2 is that the particles are slightly soluble in liquid N_2. If a slight solubility exists, particle growth occurs, resulting in a loss of gelling ability.

5. A technique for measuring the degree of structure (structure index) of cryogenic particulate gels was developed and the structure index of gelled liquid N_2 and gelled liquid OF_2 at various gelling agent concentrations was measured. As theory predicts, the incremental volume of ClF_3 particles required to achieve a given increase in the degree of structure is comparable for the two liquids.

IV, 1, Conclusions (cont.)

6. A technique for measuring the flow properties of cryogenic gels was developed and the flow properties of gelled liquid OF_2 and gelled liquid N_2, each at one gelling agent concentration, were measured. As theory predicts, the viscosities of the gelled liquids decrease at the same rate with increasing shear rate. In addition, the predicted extremely rapid decrease in viscosity with increasing shear rate was observed.

7. An exploratory study to determine whether the degree of structure possessed by a particulate gel could be estimated by extrapolating the results of flow measurements to zero flow conditions demonstrated that this approach to assessing gel structure did not yield useful information. A completely satisfactory method for assessing the degree of structure of reactive, toxic gelled cryogenic propellants awaits development.

8. An analytical study of the effects of gelation on the in-space storability of a cryogenic propellant indicated that gelation improves the storability of the propellant.

9. Performance calculations have shown that the performance degradation incurred by the gelation of OF_2 with ClF_3 particles is within tolerable limits.

2. RECOMMENDATIONS

The recommendations which are made are as follows:

1. The properties of liquid OF_2 gelled with ClF_3 particles should be completely characterized. This characterization should include storage tests over the anticipated use temperature range and complete characterization of the flow properties of gelled oxygen difluoride.

IV, 2, Recommendations (cont.)

2. As there is no reason to believe that the rapid condensation technique would not be suitable for the preparation of fine particles of fuels which could serve as gelling agents for diborane, a program to gel diborane, using the techniques developed during this program should be initiated. After the diborane has been gelled, flow and storage properties should be determined.

3. Pertaining to the technology developed during this program, it is recommended that consideration be given to taking advantage of the benefits that would arise from the gelation of FLOX and methane. In addition, the advantage to be gained by the utilization of slush cryogenic propellants should be considered as gelation makes feasible the preparation of uniform solid-liquid mixtures. While the specific impulse is not benefited by using the solid phase of a propellant, the density-impulse and the directly related vehicle-payload capability is increased by the higher density of solid-liquid slushes.

SECTION V

PROGRAM PERSONNEL

The NASA Project Manager for this program was Mr. J. Suddreth, NASA Headquarters, OART; the NASA Technical Manager was Mr. D. L. Young of the Jet Propulsion Laboratory.

The Aerojet Program Manager and Project Engineer was Mr. R. H. Globus. The interhalogen particle preparation and gel formulation and testing studies were performed under the direction of Mr. Globus with the assistance of Messrs P. D. Beadle, R. L. Beegle, and J. A. Cabeal. Dr. J. M. Adams designed the light extinction equipment for measuring the particle size of ClF_3 particles and reduced the data obtained. Mr. W. V. Timlen performed the analytical study on the effect of gelation on the in-space storability of cryogenic propellants. Mr. J. J. Bost assisted in the design of the flow viscometer for measuring the flow properties of cryogenic gels.
Drs. S. D. Rosenberg and E. M. Vander Wall acted as consultants during this program.

REFERENCES

1. System Analysis of Gelled Space-Storable Propellants, Aerojet-General Corp., Report No. 1038-011R, Vol. I, Interim Report, May 1967, Contract NAS 7-473, p. 10.

2. Gelling of Liquid Hydrogen, Technidyne Incorporated, West Chester, Pennsylvania, Report RR 64-47, Report NASA CR-5405-5, Final Report, July 31, 1964. Contract No. NAS 3-2568.

3. Gelling of Liquid Hydrogen, Technidyne Incorporated, West Chester, Pennsylvania, Monthly Progress Report No. 11, September 24 through October 23, 1965, Contract No. NAS 3-4186.

4. Private communication, Dr. Fred J. Edeskuty, Los Alamos Scientific Laboratories to Mr. R. H. Globus, Aerojet-General Corp.

5. Gelling of Liquid Oxygen Difluoride, Reaction Motors Division, Thiokol Chemical Corporation, Denville, New Jersey, Report RMD 5039-4, November 18, 1963 to 17 November 1964, Contract No. NAS 3-4180.

6. Gelling of Cryogenic Oxidizer, Reaction Motors Division, Thiokol Chemical Corporation, Denville, New Jersey, Report RMD 5070-Q-2, 5 August 1965 to 4 November 1965, Contract No. NAS 3-6286.

7. Ibid, RMD 5070-ML-5, 4 November to 4 December 1965.

8. Ibid, RMD 5070-ML-6, 5 December to 4 January 1966.

9. Ibid, RMD 5070-Q-3, 5 November 1965 to 4 February 1966.

10. Ibid, RMD 5070-ML-7, 5 February 1966 to 4 March 1966.

11. Gelling of Liquid Hydrogen, Technidyne Incorporated, West Chester, Pennsylvania, Monthly Progress Report No. 12, October 24 through September 23, 1965, Contract No. NAS 3-4186.

12. Feasibility of a Tripropellant Feed System, Aerojet-General Corporation, Sacramento, California, Report AFRPL-TR-65-29, December 1963 through September 1964. Contract No. AF 04(611)-957. Confidential

13. System Analysis of Gelled Space-Storable Propellants, Aerojet-General Corporation, Sacramento, California, Report 1038-01-Q3, November 1966 through January 1967, Contract NAS 7-473.

14. Gelling of Liquid Hydrogen, Technidyne Incorporated, West Chester, Pennsylvania, Quarterly Progress Report No. 5, 24 June through 23 September 1965, Contract No. NAS 3-4186.

REFERENCES (Cont.)

15. Dwyer, R. F., et al., Laboratory Production of Fluid Hydrogen Slush, A. and E. C., Product Res. and Dev., Quarterly 3, w16 (1964).

16. Design Handbook for Oxygen Difluoride, Reaction Motors Division, Thiokol Chemical Corporation, Denville, New Jersey, Report RTD-TDR-63-1084, November 1963, Contract AF 04(611)-8400, Unclassified.

17. Flow of Fluids Through Valves, Fittings, and Pipe, Technical Paper No. 410, Crane Co., Chicago, (p. 2-12, 4.7).

APPENDIX I

LIGHT EXTINCTION MEASUREMENTS OF SETTLING

LIGHT EXTINCTION MEASUREMENTS OF SETTLING

ABSTRACT

By application of the Mie theory of light scattering by spherical particles and Stoke's law of settling, a measurement of the extinction of a monochromatic light beam over a known path length through a turbid medium allows a determination of the particle size distribution within the medium.

Apparatus was fabricated and used successfully for the determination of particle size distributions of interhalogen compounds suspended in liquid nitrogen. The optical probe assemblies were constructed of a size small enough to fit into a four inch vessel. Optical coupling was accomplished with flexible light guides between the light source, probes, and detectors. To eliminate errors due to changes in the light source intensity or detector sensitivity over the relatively long duration of a run, a continuous reference measurement was provided by optical sampling.

Light extinction measurements were made on particulate suspensions of $ClF_{3(s)}$ undergoing tranquil settling in liquid nitrogen. The results indicate that the apparent particle size distribution was more strongly dependent on particulate concentration in the liquid than on the characteristics of formation. Thus, it appears that agglomeration of the particles was occurring rapidly in the vessel and that these agglomerates, rather than individual particles, were being observed. Where low particulate concentrations were studied, the results indicate a specific surface of about 0.5 m^2/gm.

INTRODUCTION

During the recent development of techniques to form particulate gels in cryogenic liquid propellant systems,(1) it has become necessary to determine the size distributions of particles within a cryogenic liquid. A particularly useful method of determining characteristics of a polydispersion was suggested by Gumprecht and Sliepcevich,(2,3) and results from a knowledge of the interaction of the dispersed particles with a monochromatic beam of light traversing the medium. By application of the Mie theory of light scattering by spherical particles(4) and Stoke's law of settling, a measurement of the extinction of a monochromatic light beam over a known path length through the turbid medium allows a determination of the particle size distribution and concentration in the light path.

This quantitative optical method is of particular value for conditions where the medium is not readily accessible for direct analysis. Furthermore, because of the small container volume necessitated by the hazardous nature of the particulate phase in the present case (ClF_3), a compact optical system was required. The resulting apparatus and method, which allows continuous, precise measurement of the light extinction coefficient in a settling polydispersion, is described in the following paragraphs.

THEORY - PRINCIPLES OF OPERATION

Light Extinction by Spherical Particles

With the optical system shown in Figure 1, a monochromatic light beam of incident intensity I_o is passed through a polydispersion where it is attenuated from absorption and scattering by particles in the light path. The light beam strikes the acceptance lens L2 with intensity I, given by

$$I = I_o \exp\left(- t \int_o^\infty K(\gamma_a + \gamma_s) \tilde{N} \, d D\right) \qquad (1)$$

where $\tilde{N}dD$ is the number density of particles lying in the size range between D and D+dD, cm^{-3}.

D is the particle diameter

t is the extinction path length, cm. (Figure 1)

γ_a, γ_s are the optical absorption and scattering cross sections for the particle diameter D.

K is a correction factor to account for scattered light received by the acceptor lens, L2(2).

With the Mie theory applied to spherical particles, the cross sections γ_a and γ_s are uniquely determined from the refractive index, m, of the dispersed phase relative to the continuous phase, the wavelength of the light and the particle diameter D. The complete analysis of these cross sections, together with many simplified formulae, has been presented by Van de Hulst(5). A computer program for calculation of optical cross sections for polydispersions has been given by Erickson(6).

Application of Stoke's Law

On small spheres moving in a medium of viscosity μ, the drag force is given by

$$\vec{F}_D = -3\pi\mu D\vec{u} \qquad (2)$$

where u is the velocity of the particle relative to the surrounding medium.

The gravitational force acting on the same particle of density ρ_S in a liquid of density ρ_L is positive downward when expressed by

$$\vec{F}_G = \frac{\pi D^3}{6} (\rho_S - \rho_L) \vec{g}_L \qquad (3)$$

At terminal velocity, the net force on the particle in the medium is zero, and we have, by equating the sum of equations 2 and 3 to zero:

$$D = \sqrt{\frac{18\ \mu\ u}{(\rho_S - \rho_L)\ g_L}} \tag{4}$$

which relates the diameter of spherical particles to their terminal velocity.

If stirring of the suspension is stopped at time $\theta = 0$, equation 4 can be used to determine the time θ at which particles of diameter D disappear from the light path which is located a distance h under the liquid surface, i.e.

$$D = C\ \theta^{-1/2}, \tag{5}$$

where

$$C = \sqrt{\frac{18\ \mu\ h}{g_L\ (\rho_S - \rho_L)}}. \tag{6}$$

Equation 6 neglects the time required to accelerate the particles to their terminal velocity, assumes interparticulate forces are small (i.e., interparticulate distances are large) and is strictly applicable only when the settling time is long in relation to the time required for convection currents.within the liquid to cease. When the solids density is lower than that of the liquid, equation 5 is still applicable where h is now the distance from the bottom of the vessel to the optical path.

Transient Light Extinction Equation

Taking the logarithm of equation 1 and differentiating, we obtain

$$d \ln I = - t \tilde{N} \gamma_t K dD. \tag{7}$$

where* $\gamma_t = \gamma_a + \gamma_s$.

By differentiating equation 5, substituting and rearranging, we obtain from equation 7:

$$F = K \gamma_t \tilde{N} = \frac{2 \theta^{3/2}}{tC} \frac{d \ln I}{d\theta} = \frac{2 \theta^{3/2}}{tC} \cdot \frac{1}{I} \cdot \frac{dI}{d\theta} \tag{8}$$

Thus with an optical system such as that shown in Figure 1, located a depth h beneath the surface of a suspension, the quantity F can be determined at any given time from a measurement of I versus time θ. From equation 5, the diameter corresponding to F can be determined and used in turn to evaluate K and γ_t. The particle number density function $\tilde{N}$, is then calculated from the equation

$$\tilde{N} = \frac{F}{K \gamma_t} , \tag{9}$$

and a plot of $\tilde{N}$ versus D, the desired size distribution curve for the polydispersion, can be thus obtained.

---

* With most quasi-dielectric media at low temperatures, $\gamma_a << \gamma_s$ and γ_a can be neglected.

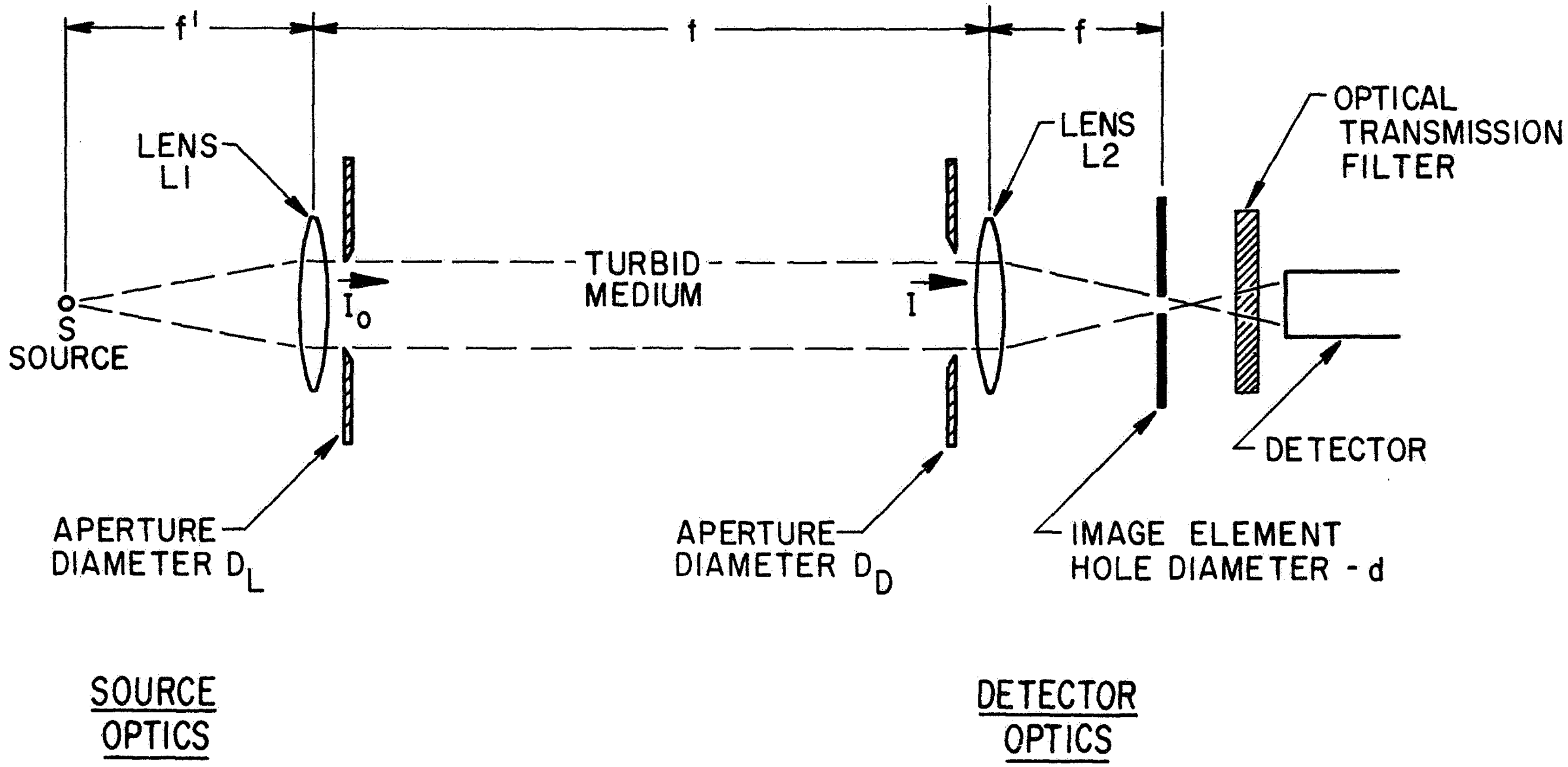


Figure 1. Optical Schematic for Light Extinction Measurement

APPARATUS

A photograph of the optical system used to perform the transient light extinction measurements is shown in Figure 2. The linkage of the various components in the system can be better understood from a schematic of the apparatus, presented in Figure 3. An intense source of light was supplied by a 200 watt mercury arc lamp* housed and supported within a 4 inch x 4 inch x 6 inch enclosure and air-cooled by natural convection. One wall of the enclosure, shown in Figure 4, served as a retainer for four one-sixteenth inch diameter fiber-optic light guides+ which were positioned so that their centerlines intersected at the center of the arc lamp.

Six flexible light guides to transmit light between the light sources, the optical probe assemblies and the detectors were arranged in a manner shown in Figure 3. To account for any change in characteristics of the detectors or the light source during an experiment, light guides were connected directly between the light source and the detectors.

Optical Sensing Probes

The light extinction optical system shown in Figure 5 was designed for total immersion and operation in a corrosive liquid environment. The size of the complete optical probe assembly was such that two complete assemblies, a stirring drive shaft and several sampling tubes could be placed within a vessel having an inside diameter of only four inches. A photograph of the two complete assemblies attached to the vessel lid is shown in Figure 6.

The optical sensing probes were fabricated entirely from stainless steel with the dimensions shown in Figure 5. The ratio of the light guide retainer aperture diameter d (image element, Figure 1) and the focal length f of the intermediate lens L2 on the detector probe was fixed by the requirement(7)

$$\arctan (d/2f) \leq 1.5^\circ \, , \qquad (10)$$

---

* George W. Gates & Co., Inc. HBO 200W/2 lamp and low ripple direct-current power supply P210DV.

+ American Optical Co. LGM 2-72.

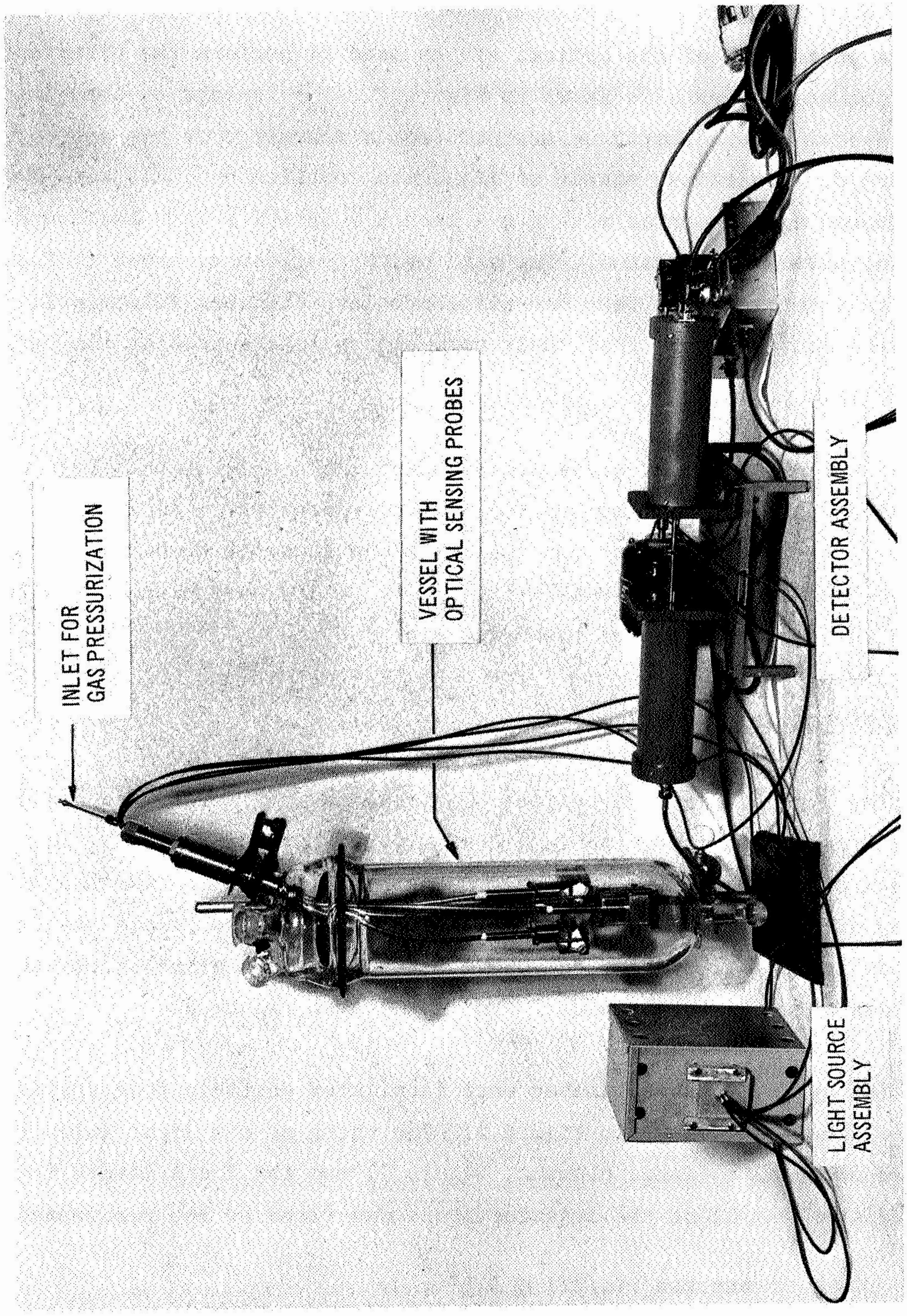


Figure 2. Light Extinction Apparatus

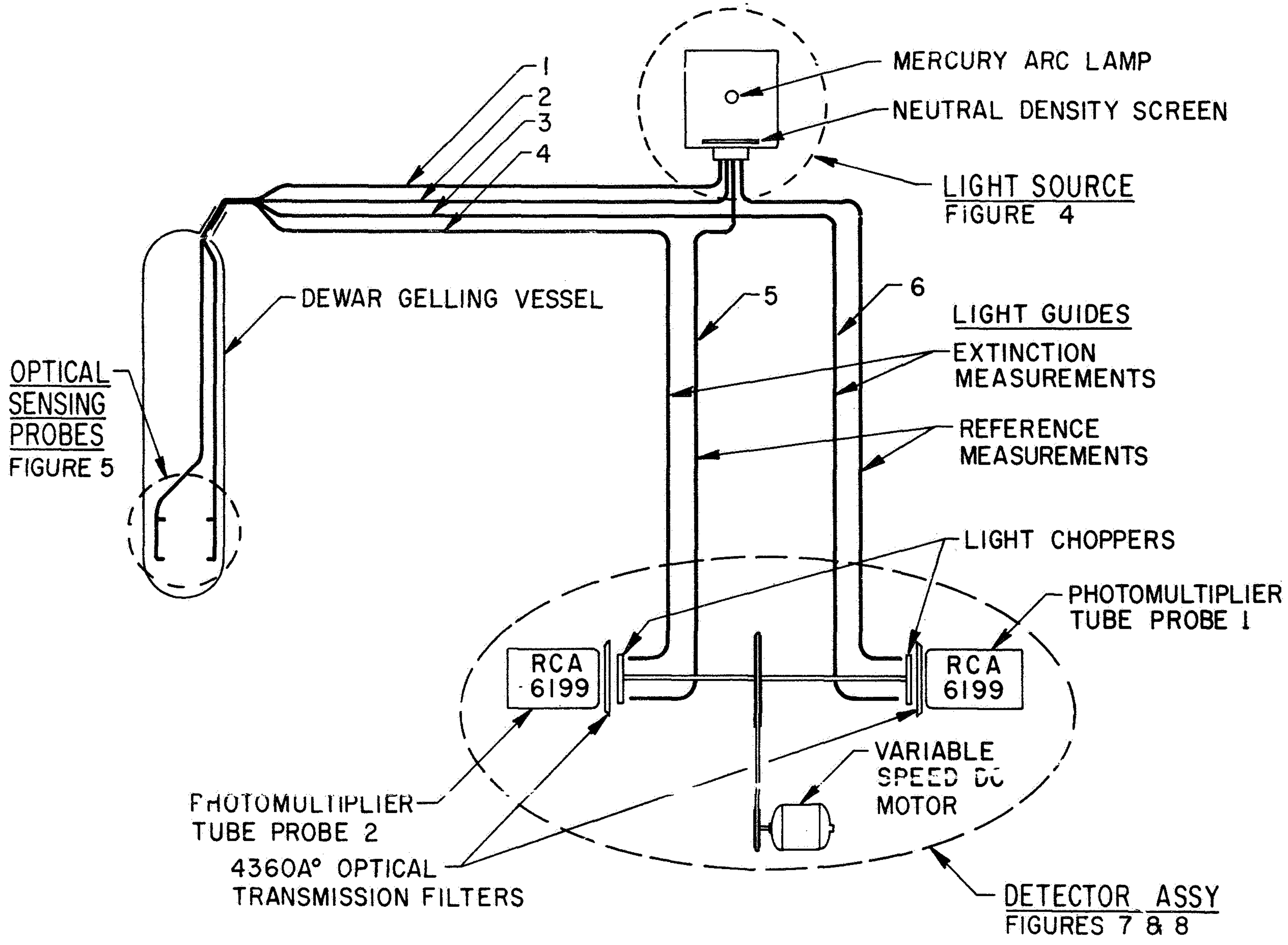


Figure 3. Schematic of Light Extinction Apparatus

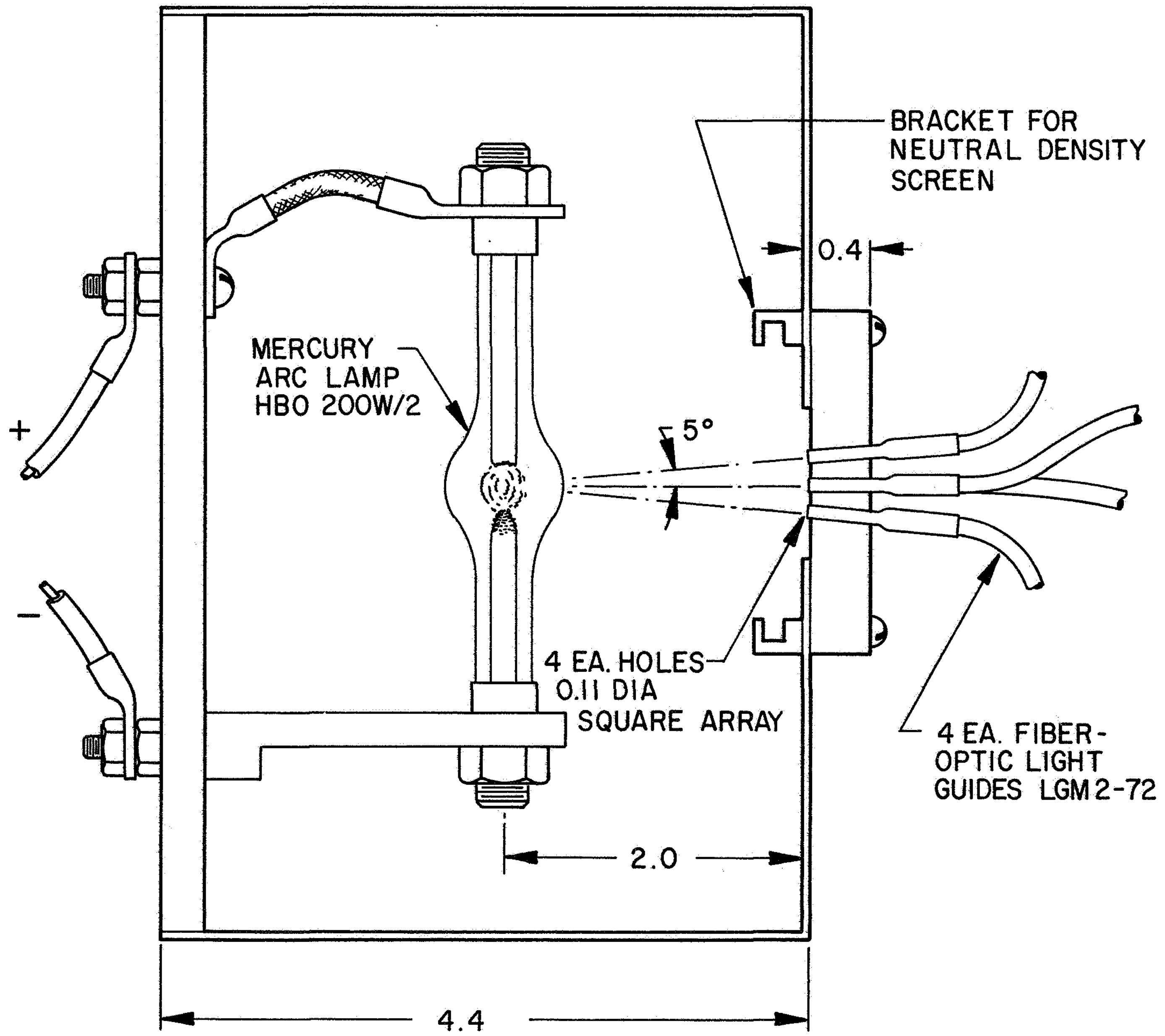


Figure 4. Light Source

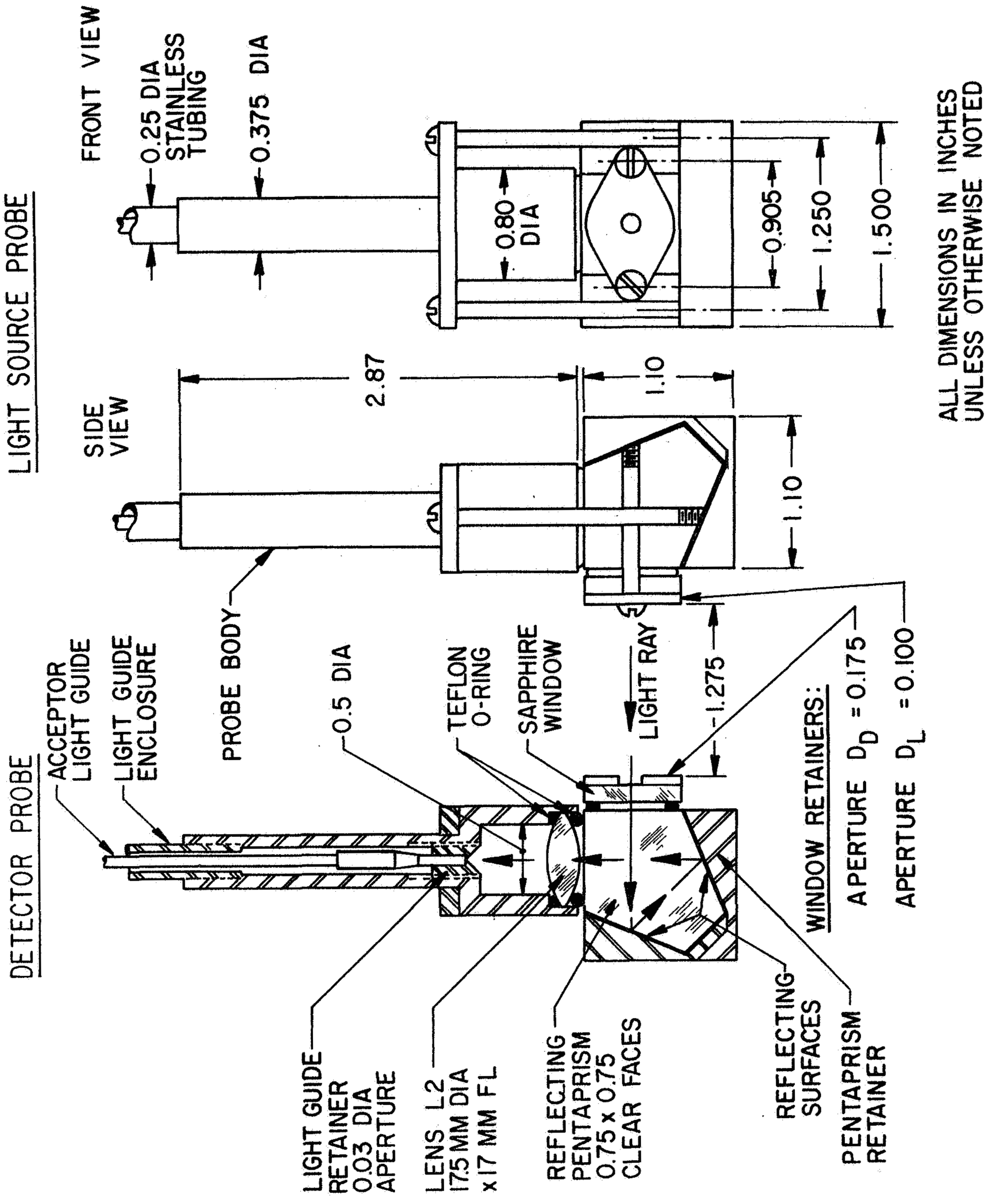


Figure 5. Optical Sensing Probes

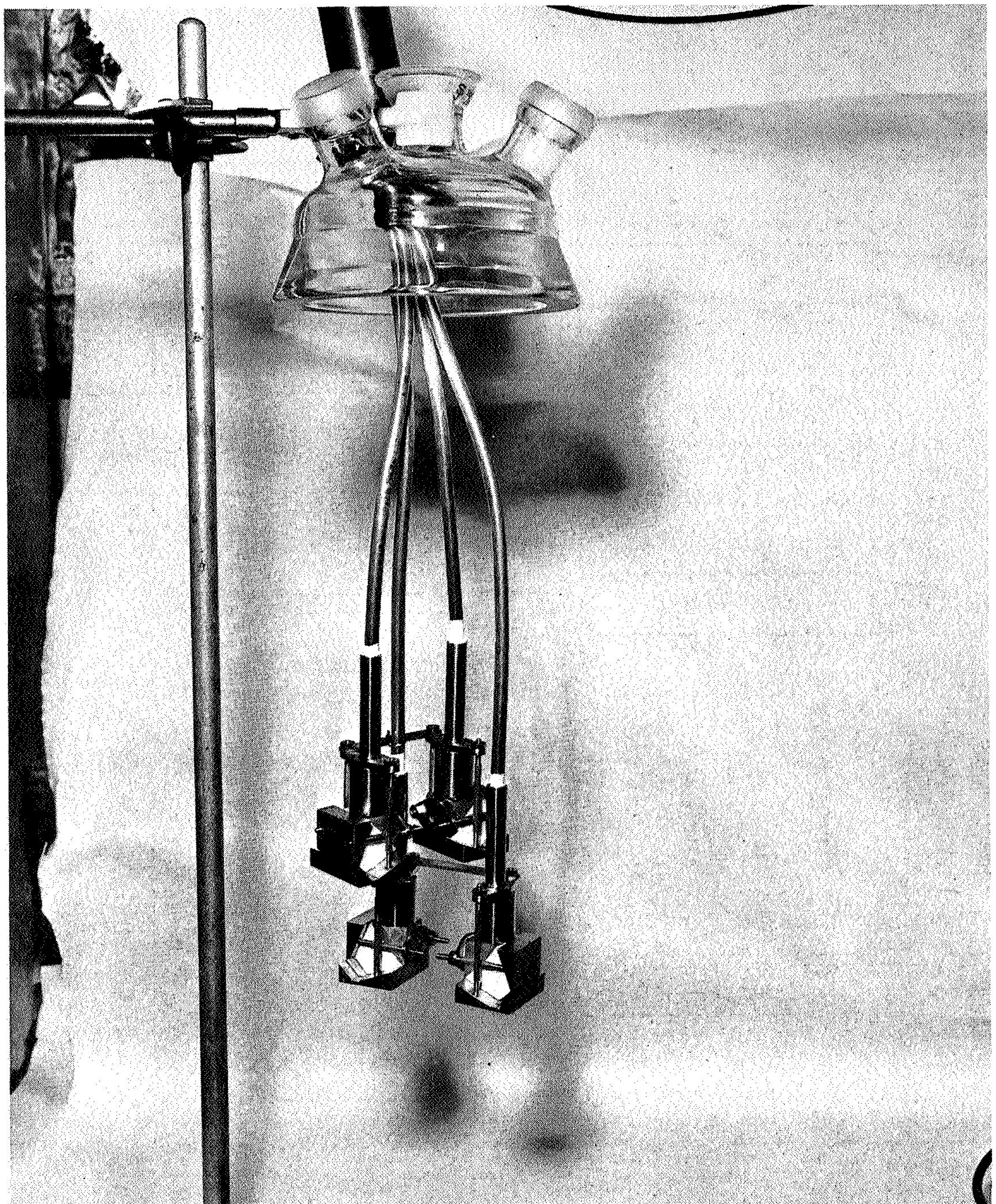


Figure 6. Optical Sensing Probes Attached to Vessel Lid

with the additional requirement on the diameter of the window retainer-apertures given by (7)

$$D_D \geq \frac{dt}{f} + D_L \tag{11}$$

where D_D is the aperture diameter of the detector probe and D_L is that of the light source probe (see Figure 1). The diameter D_L was chosen to obtain reasonable spacial resolution. A maximum value for D_D is dictated by the numerical aperture of the light guides. With the lens L1 placed at a distance from the end of the light guide equal to its focal length, a collimated light beam of diameter D_L was formed by the light source probe which passed through the dispersion to be intercepted by the detector probe and focused by lens L2 on the end of the acceptor light guide. Adjustment of the distance between the light guide ends and the lenses was accomplished with the threaded light guide retainer. Identical lenses were used in the four probes.

The use of the pentaprisms provided horizontal orientation of the light beam in a minimum of space while offering two additional advantages: 1) protection of the reflecting surface was afforded without the inherent multiple reflections of a standard second-surface reflector; 2) slight misalignment of the prisms and the probe centerline did not affect the optical path, i.e., the only rigid requirement for perfect alignment is that the vertical centerlines of the two probes be parallel.

Support for the optical sensing probes within the vessel and protection for the light guides was provided by heavy-wall stainless steel tubing which was threaded for direct connection to the probe body. Each of the four tubes was welded at the top end to a flange for attachment to the standard ball and socket fitting which in turn is connected to the vessel lid. At the flange, provision was made for sealing the light guide enclosure, thus enabling pressurization of each of the optical probes and light guide enclosures to approximately five psig. The helium gas pressurization was provided to prevent any leakage of liquid into the probe body.

Detector Assembly

Continuous monitoring of the light intensity from the optical sensing probes was accomplished with two multiplier phototubes, mounted as shown in Figures 7 and 8. The table-mounted hangers* served as supports for the phototube housings, the light-guide ends and the rotating shaft to drive both light choppers, as shown in the figures. Two light guides were optically coupled to each of the phototubes and were alternately sampled with the rotating light chopper, directly in front of each phototube face. As described previously, one of the light guides transmitted light from the light source directly to the phototube to serve as a reference, the other transmitted light from the optical sensing probe within the vessel (see Figure 3). The light chopper was fabricated by cutting a 30° slot along a radius of 0.5 inches in a commercial dial plate*, which was shaft-mounted and rotated with a direct-current variable-speed electric motor.

Between the light chopper and the phototube face, an optical interference filter[+] was placed to provide spectral discrimination at a wavelength of 4360A°, the brightest region in the mercury arc spectrum.

The phototubes each were electrically coupled to the standard ten-resistor bridge and connected to a well-regulated high-voltage power supply. The tubes were operated at a voltage from 850-1000 volts. Magnetic shields of mu-metal were provided for each of the tubes; these were electrically coupled to anode potential (- 1000 volts) and were insulated from the outer aluminum housing with a tube of polymethylmethacrylate. The output from each phototube was monitored with an operational amplifier[§], the input of which was coupled as a current-follower from cathode to positive ground.

---

* Precision Instrument Components, Inc.
+ Thin Film Products, 1/2 power bandwidth 100 Angstroems, blocked to 1 micron.
§ Analogue Devices, Model 107C.

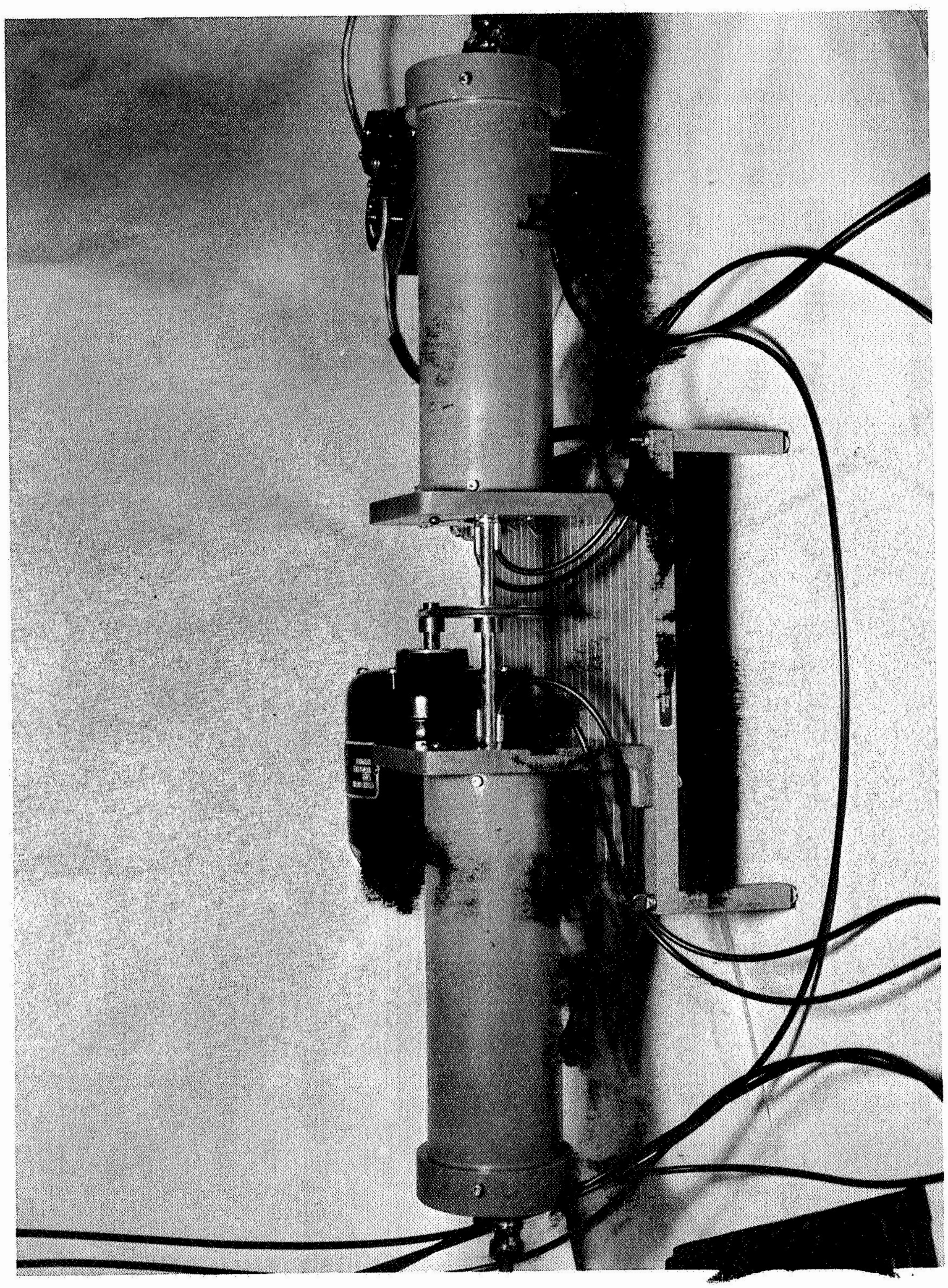


Figure 7. Detector Assembly

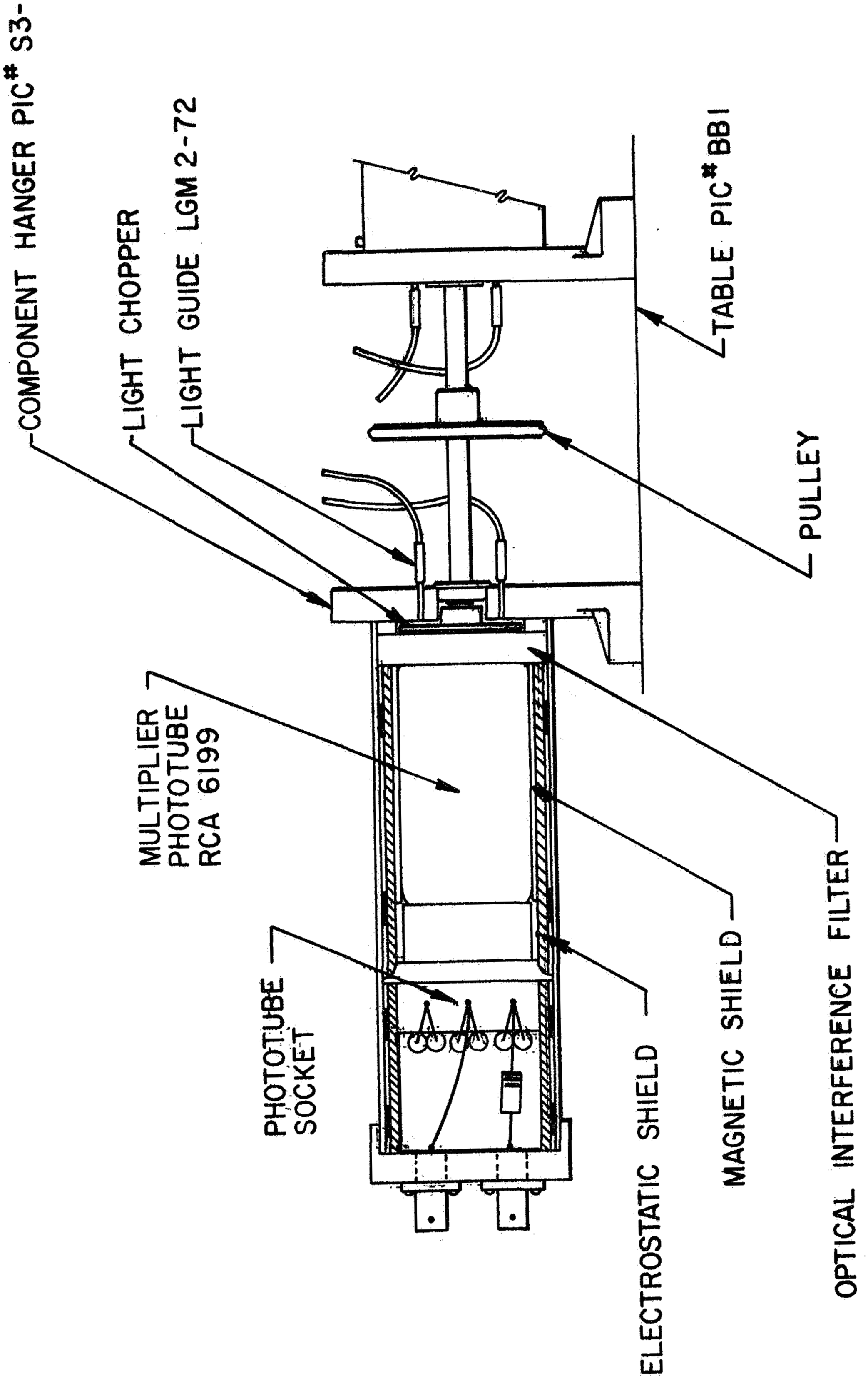


Figure 8. Sketch of Detector Assembly

Data Recording

Data from each of the phototubes was in the form of two square pulses per rotation of the light-chopper. One pulse represented the reference signal coming directly from the light source and the other was the light intensity transmitted through the polydispersion. A continuous display of these repetitive pulses was produced by using the output from the operational amplifiers to drive a galvanometer in a standard oscillograph recorder. One galvanometer was used for each phototube.

Because of the required duration of an experiment (two to four hours) and limited length of an oscillograph record (two hundred feet of paper), the oscillograph had to be operated intermittently. This was accomplished with a commercial cam-timer unit*, which actuated the oscillograph for 2.5 seconds every thirty seconds. At the start of an experiment, immediately after the stirring action was stopped, resolution of the heavier particles (see equation 5) was obtained by operating the oscillograph continuously for the first thirty or sixty seconds. Automatic operation then continued to record the data. A typical data record is shown in Figure 9.

Calibration

To obtain the best possible precision in the measurements it was necessary to adjust the intensity of the reference signal to closely equal the intensity from the sensing probes. This was accomplished by placing a glass neutral density screen** in front of the two reference light guides. This screen was mounted on an aluminum backing plate which was fastened in position in the light source housing (see Figure 4). The remaining two light guides, which transmitted light to the sensing probes, were unblocked by the screen. A screen of density - 2.0 was sufficient to obtain a rather good match of light intensities between the reference and probe signals. Some additional fine adjustment was possible by movement of the light guide ends in their support.

---

* Industrial Timer Corp., No. CM-2 with 30 second cam foundation unit No. A-12.

** Optics Technology Set 5A.

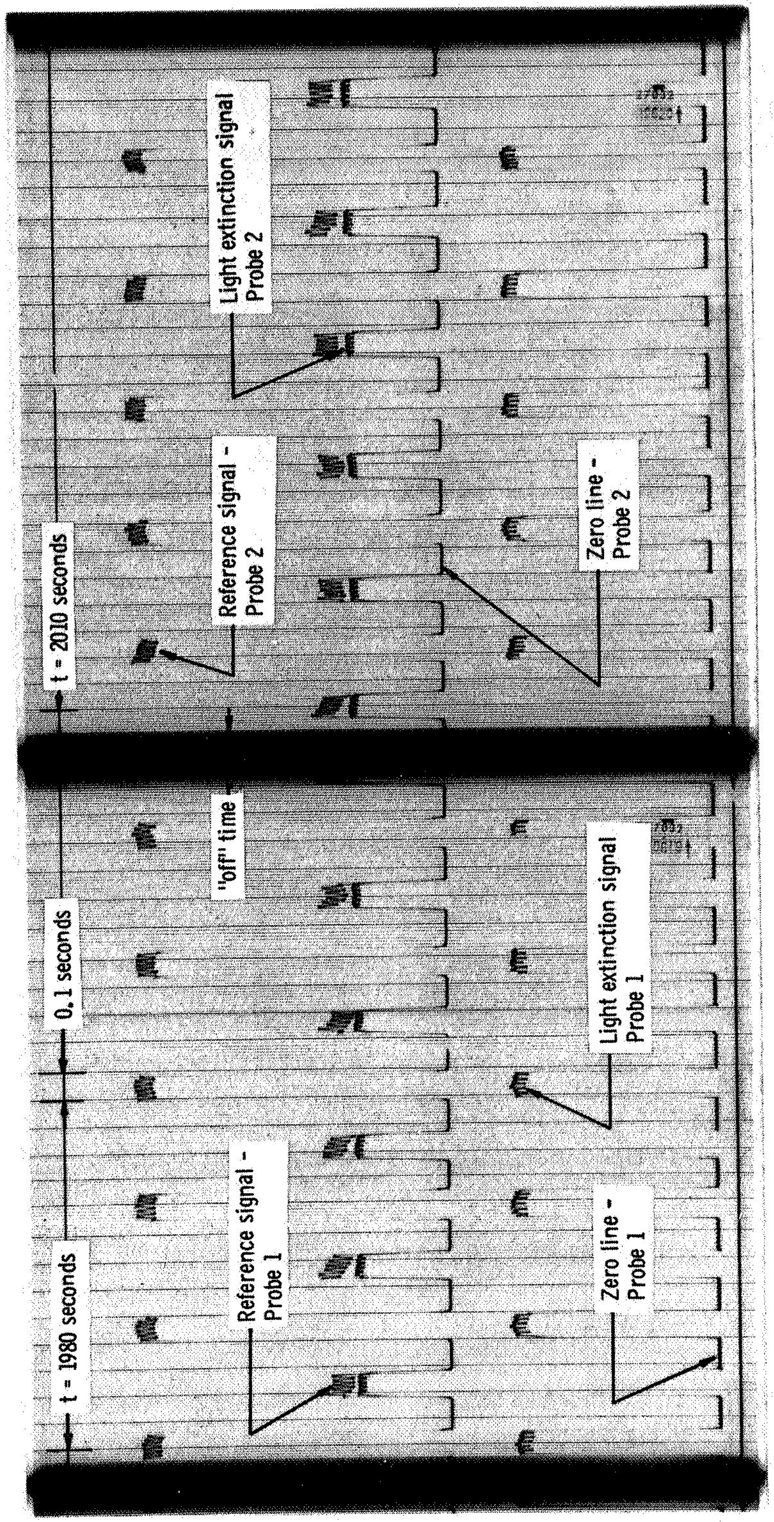


Figure 9. Example of Record Data

Prior to a run, non-linearities in the response of the phototubes, amplifiers and galvanometers were determined by interposing a calibrated, neutral-density stepping screen in the optical path of each of the sensing probes. By drawing the stepping screen through the optical path while operating the oscillograph, a graduated calibration through twenty-two changes in light intensity was obtained.

RESULTS

With the sensing probes submerged in a polydispersion of $ClF_{3(s)}$ particles suspended in liquid nitrogen, measurements of relative light intensity at each of the two stations were obtained over periods up to 7500 seconds. This period, corresponding (equation 5) to a particle diameter of about 0.5 micron passing by the upper station, appeared to be the limit of resolution of the instrument. That is, for times greater than 7500 seconds, noise and fluctuations in the signal were of the same magnitude as the time rate of change of light intensity, $dI/d\theta$ (v. equation 8). A typical size distribution measured with this device is shown in Figure 10.

Results obtained with the apparatus are presented in Appendix I in the form of tables. Table I presents the number density, specific volume and surface of the polydispersion over increments of 1 micron. Measurements taken with the upper station (denoted by the letter U in the experiment number) begin at one micron, the nominal lower limit of resolution. Measurements taken with the lower station (L in the experiment number) begin at two microns for the same reason. The upper limit on particle size was chosen according to the magnitude of the contribution from that increment, but in no case was less than twenty microns.

The contribution to the size distribution between D=o and the lower limit of particle diameter given in Table I was obtained from the spectral transmittance of the dispersion at the termination of the run. That is, from equation 1

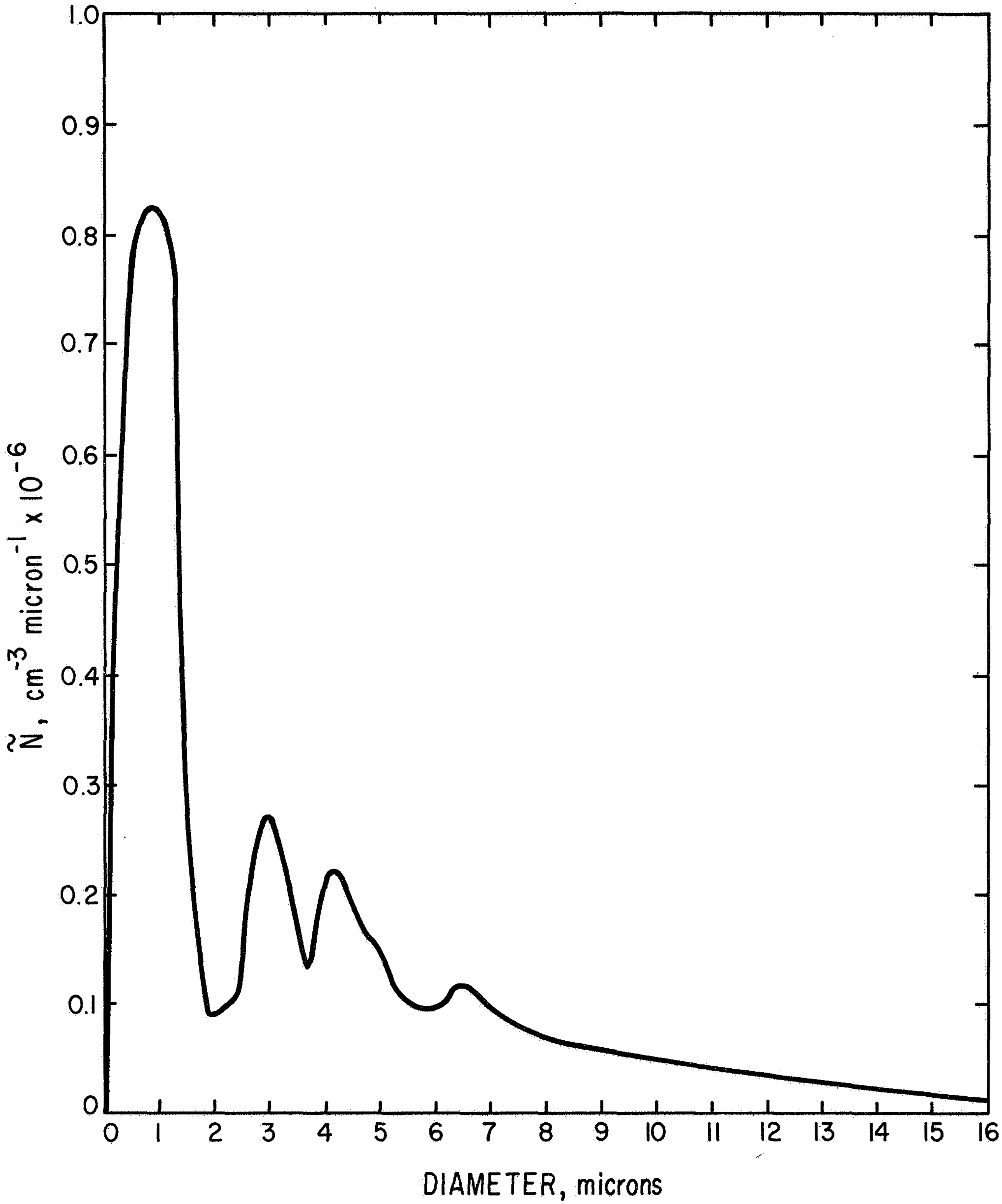


Figure 10. Measured Particle Size Distribution of $ClF_{3(5)}$ in Liquid Nitrogen

TABLE I

PARTICLE SIZE DISTRIBUTIONS
DIAMETER > 1 MICRON

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671205-1U (17, RUN NO. 1)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  2.510 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER     NUMBER       PARTICLE      SURFACE
     INCREMENT    DENSITY       VOLUME        AREA
      MICRONS     PER GM       CU CM/GM     SQ CM/GM
      1 -  2    0.4054E 09   0.4987E-03   0.2183E 02
      2 -  3    0.1498E 09   0.1582E-02   0.3472E 02
      3 -  4    0.1948E 09   0.4310E-02   0.7369E 02
      4 -  5    0.1923E 09   0.9060E-02   0.1208E 03
      5 -  6    0.1111E 09   0.9573E-02   0.1045E 03
      6 -  7    0.1104E 09   0.1597E-01   0.1468E 03
      7 -  8    0.8272E 08   0.1814E-01   0.1453E 03
      8 -  9    0.6746E 08   0.2166E-01   0.1528E 03
      9 - 10    0.4943E 08   0.2205E-01   0.1394E 03
     10 - 11    0.4295E 08   0.2604E-01   0.1487E 03
     11 - 12    0.3711E 08   0.2954E-01   0.1540E 03
     12 - 13    0.3503E 08   0.3582E-01   0.1718E 03
     13 - 14    0.2705E 08   0.3465E-01   0.1542E 03
     14 - 15    0.2168E 08   0.3456E-01   0.1430E 03
     15 - 16    0.1726E 08   0.3356E-01   0.1300E 03
     16 - 17    0.1286E 08   0.3014E-01   0.1097E 03
     17 - 18    0.9000E 07   0.2513E-01   0.8630E 02
     18 - 19    0.5924E 07   0.1953E-01   0.6345E 02
     19 - 20    0.3684E 07   0.1422E-01   0.4384E 02
     20 - 21    0.2385E 07   0.1071E-01   0.3138E 02

CHECK =   0.10000E 01

PARTICLE NUMBER DENSITY =   0.1541E 07 PER CU CM

SOLIDS CONCENTRATION =   0.9769E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.21767E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671205-1L  (17)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  8.850 CM

EXTINCTION PATH LENGTH =  2.965 CM

REFRACTIVE INDEX. SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER      NUMBER        PARTICLE      SURFACE
     INCREMENT     DENSITY        VOLUME         AREA
      MICRONS      PER GM       CU CM/GM      SQ CM/GM
       2 -  3    0.7372E 08   0.6542E-03   0.1515E 02
       3 -  4    0.6733E 08   0.1593E-02   0.2665E 02
       4 -  5    0.7098E 08   0.3324E-02   0.4442E 02
       5 -  6    0.2811E 08   0.2380E-02   0.2613E 02
       6 -  7    0.1860E 08   0.2647E-02   0.2448E 02
       7 -  8    0.1104E 08   0.2434E-02   0.1946E 02
       8 -  9    0.1053E 08   0.3409E-02   0.2398E 02
       9 - 10    0.1430E 08   0.6486E-02   0.4079E 02
      10 - 11    0.2187E 08   0.1344E-01   0.7640E 02
      11 - 12    0.2420E 08   0.1925E-01   0.1004E 03
      12 - 13    0.2448E 08   0.2509E-01   0.1203E 03
      13 - 14    0.2188E 08   0.2810E-01   0.1249E 03
      14 - 15    0.1711E 08   0.2720E-01   0.1126E 03
      15 - 16    0.1218E 08   0.2373E-01   0.9188E 02
      16 - 17    0.1249E 08   0.2944E-01   0.1069E 03
      17 - 18    0.1258E 08   0.3534E-01   0.1211E 03
      18 - 19    0.1392E 08   0.4636E-01   0.1501E 03
      19 - 20    0.1512E 08   0.5868E-01   0.1805E 03
      20 - 21    0.1083E 08   0.4852E-01   0.1423E 03
      21 - 22    0.3660E 07   0.1869E-01   0.5248E 02

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.8550E 06 PER CU CM

SOLIDS CONCENTRATION =  0.1764E-02 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.16014E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671205-2U (17, RUN NO. 2)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  2.510 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

        DIAMETER      NUMBER        PARTICLE      SURFACE
        INCREMENT     DENSITY       VOLUME         AREA
         MICRONS      PER GM        CU CM/GM      SQ CM/GM
          1 -  2     0.6700E 09    0.1175E-02    0.4501E 02
          2 -  3     0.5563E 09    0.3756E-02    0.9470E 02
          3 -  4     0.1206E 09    0.2585E-02    0.4469E 02
          4 -  5     0.1564E 09    0.7841E-02    0.1024E 03
          5 -  6     0.2138E 09    0.1870E-01    0.2032E 03
          6 -  7     0.2596E 09    0.3771E-01    0.3463E 03
          7 -  8     0.1699E 09    0.3701E-01    0.2970E 03
          8 -  9     0.1346E 09    0.4339E-01    0.3057E 03
          9 - 10     0.1089E 09    0.4860E-01    0.3073E 03
         10 - 11     0.8522E 08    0.5142E-01    0.2940E 03
         11 - 12     0.4899E 08    0.3842E-01    0.2013E 03
         12 - 13     0.2798E 08    0.2847E-01    0.1368E 03
         13 - 14     0.2092E 08    0.2685E-01    0.1194E 03
         14 - 15     0.1477E 08    0.2343E-01    0.9714E 02
         15 - 16     0.7994E 07    0.1540E-01    0.5985E 02
         16 - 17     0.2845E 07    0.6572E-02    0.2403E 02
         17 - 18     0.7422E 06    0.2045E-02    0.7053E 01
         18 - 19     0.2941E 06    0.9739E-03    0.3159E 01
         19 - 20     0.3438E 06    0.1334E-02    0.4106E 01
         20 - 21     0.2398E 06    0.1076E-02    0.3155E 01

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.1956E 07 PER CU CM

SOLIDS CONCENTRATION =  0.7522E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.26967E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671205-2L (17, RUN NO. 2)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  8.850 CM

EXTINCTION PATH LENGTH =  2.965 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

      DIAMETER       NUMBER         PARTICLE       SURFACE
      INCREMENT      DENSITY         VOLUME          AREA
       MICRONS       PER GM        CU CM/GM.      SQ CM/GM
        2 -  3     0.7891E 08     0.7763E-03     0.1732E 02
        3 -  4     0.1240E 09     0.2669E-02     0.4609E 02
        4 -  5     0.5372E 08     0.2470E-02     0.3321E 02
        5 -  6     0.2799E 08     0.2402E-02     0.2626E 02
        6 -  7     0.2178E 08     0.3144E-02     0.2893E 02
        7 -  8     0.1615E 08     0.3563E-02     0.2847E 02
        8 -  9     0.1663E 08     0.5381E-02     0.3786E 02
        9 - 10     0.1532E 08     0.6859E-02     0.4332E 02
       10 - 11     0.2371E 08     0.1464E-01     0.8312E 02
       11 - 12     0.2833E 08     0.2253E-01     0.1175E 03
       12 - 13     0.2766E 08     0.2835E-01     0.1359E 03
       13 - 14     0.2498E 08     0.3210E-01     0.1427E 03
       14 - 15     0.2111E 08     0.3366E-01     0.1393E 03
       15 - 16     0.1776E 08     0.3460E-01     0.1339E 03
       16 - 17     0.1643E 08     0.3867E-01     0.1405E 03
       17 - 18     0.1489E 08     0.4173E-01     0.1431E 03
       18 - 19     0.1229E 08     0.4069E-01     0.1320E 03
       19 - 20     0.1043E 08     0.4047E-01     0.1245E 03
       20 - 21     0.9331E 07     0.4206E-01     0.1231E 03

  CHECK =  0.10000E 01

  PARTICLE NUMBER DENSITY =  0.6962E 06 PER CU CM

  SOLIDS CONCENTRATION =  0.1240E-02 GMS SOLID/CU CM LIQUID

  TOTAL SURFACE AREA =  0.16775E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671207U (18)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  3.300 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER      NUMBER        PARTICLE      SURFACE
     INCREMENT     DENSITY        VOLUME         AREA
      MICRONS      PER GM        CU CM/GM      SQ CM/GM
       1 -  2     0.5137E 08    0.1553E-03    0.5054E 01
       2 -  3     0.1180E 10    0.1006E-01    0.2356E 03
       3 -  4     0.8344E 09    0.1966E-01    0.3293E 03
       4 -  5     0.1684E 10    0.8149E-01    0.1077E 04
       5 -  6     0.8296E 09    0.6952E-01    0.7662E 03
       6 -  7     0.3183E 09    0.4483E-01    0.4159E 03
       7 -  8     0.1380E 09    0.2986E-01    0.2402E 03
       8 -  9     0.1096E 09    0.3511E-01    0.2479E 03
       9 - 10     0.5612E 08    0.2460E-01    0.1565E 03
      10 - 11     0.1197E 08    0.6884E-02    0.4001E 02
      11 - 12     0.3269E 07    0.2667E-02    0.1379E 02
      12 - 13     0.9737E 07    0.1015E-01    0.4838E 02
      13 - 14     0.1756E 08    0.2269E-01    0.1007E 03
      14 - 15     0.1439E 08    0.2282E-01    0.9462E 02
      15 - 16     0.6606E 07    0.1265E-01    0.4926E 02
      16 - 17     0.1536E 07    0.3516E-02    0.1290E 02
      17 - 18     0.4000E 05    0.1044E-03    0.3667E 00
      18 - 19     0.0000E 00    0.0000E 00    0.0000E 00
      19 - 20     0.0000E 00    0.0000E 00    0.0000E 00

CHECK =   0.10000E 01

PARTICLE NUMBER DENSITY =  0.2821E 07 PER CU CM

SOLIDS CONCENTRATION =  0.5358E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.38347E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671207L  (18)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  9.500 CM

EXTINCTION PATH LENGTH =  2.965 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER       NUMBER        PARTICLE      SURFACE
     INCREMENT      DENSITY        VOLUME         AREA
      MICRONS       PER GM       CU CM/GM      SQ CM/GM
       2 -  3     0.2274E 09   0.2111E-02   0.4825E 02
       3 -  4     0.1634E 09   0.3607E-02   0.6175E 02
       4 -  5     0.1360E 09   0.6461E-02   0.8594E 02
       5 -  6     0.7830E 08   0.6768E-02   0.7381E 02
       6 -  7     0.7081E 08   0.1023E-01   0.9411E 02
       7 -  8     0.5421E 08   0.1196E-01   0.9559E 02
       8 -  9     0.6466E 08   0.2106E-01   0.1478E 03
       9 - 10     0.6846E 08   0.3080E-01   0.1942E 03
      10 - 11     0.8737E 08   0.5330E-01   0.3037E 03
      11 - 12     0.7371E 08   0.5823E-01   0.3044E 03
      12 - 13     0.4147E 08   0.4193E-01   0.2019E 03
      13 - 14     0.2445E 08   0.3136E-01   0.1395E 03
      14 - 15     0.2132E 08   0.3394E-01   0.1405E 03
      15 - 16     0.1475E 08   0.2863E-01   0.1109E 03
      16 - 17     0.1037E 08   0.2429E-01   0.8843E 02
      17 - 18     0.6369E 07   0.1772E-01   0.6092E 02
      18 - 19     0.3021E 07   0.9904E-02   0.3223E 02
      19 - 20     0.1023E 07   0.3909E-02   0.1209E 02
      20 - 21     0.1322E 06   0.5720E-03   0.1697E 01

CHECK =   0.10000E 01

PARTICLE NUMBER DENSITY =  0.1608E 07 PER CU CM

SOLIDS CONCENTRATION =  0.1402E-02 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.21981E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671219-1U  (19)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  3.275 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER      NUMBER       PARTICLE      SURFACE
     INCREMENT     DENSITY       VOLUME         AREA
      MICRONS      PER GM       CU CM/GM     SQ CM/GM
       1 -  2    0.4566E 09   0.1015E-02   0.3639E 02
       2 -  3    0.5591E 09   0.4398E-02   0.1056E 03
       3 -  4    0.2837E 09   0.6568E-02   0.1106E 03
       4 -  5    0.4385E 09   0.2133E-01   0.2814E 03
       5 -  6    0.4441E 09   0.3991E-01   0.4296E 03
       6 -  7    0.5023E 09   0.7004E-01   0.6520E 03
       7 -  8    0.2301E 09   0.5020E-01   0.4027E 03
       8 -  9    0.1557E 09   0.4964E-01   0.3510E 03
       9 - 10    0.6862E 08   0.2994E-01   0.1907E 03
      10 - 11    0.2699E 08   0.1614E-01   0.9259E 02
      11 - 12    0.1505E 08   0.1190E-01   0.6222E 02
      12 - 13    0.1347E 08   0.1381E-01   0.6623E 02
      13 - 14    0.1101E 08   0.1410E-01   0.6277E 02
      14 - 15    0.8038E 07   0.1278E-01   0.5295E 02
      15 - 16    0.5947E 07   0.1155E-01   0.4476E 02
      16 - 17    0.4460E 07   0.1045E-01   0.3805E 02
      17 - 18    0.3380E 07   0.9458E-02   0.3245E 02
      18 - 19    0.2600E 07   0.8595E-02   0.2789E 02
      19 - 20    0.2020E 07   0.7826E-02   0.2409E 02
      20 - 21    0.1579E 07   0.7106E-02   0.2081E 02

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.2337E 07 PER CU CM

SOLIDS CONCENTRATION =  0.7230E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.30852E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671219-1L (19)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  9.530 CM

EXTINCTION PATH LENGTH =  3.245 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER      NUMBER        PARTICLE       SURFACE
     INCREMENT     DENSITY       VOLUME          AREA
      MICRONS      PER GM        CU CM/GM       SQ CM/GM
      2 -  3      0.2231E 09    0.1876E-02    0.4416E 02
      3 -  4      0.7726E 08    0.1672E-02    0.2880E 02
      4 -  5      0.5508E 08    0.2622E-02    0.3485E 02
      5 -  6      0.2766E 08    0.2354E-02    0.2580E 02
      6 -  7      0.2016E 08    0.2913E-02    0.2680E 02
      7 -  8      0.1572E 08    0.3461E-02    0.2767E 02
      8 -  9      0.1602E 08    0.5182E-02    0.3647E 02
      9 - 10      0.1492E 08    0.6689E-02    0.4223E 02
     10 - 11      0.1300E 08    0.7929E-02    0.4519E 02
     11 - 12      0.1619E 08    0.1299E-01    0.6759E 02
     12 - 13      0.2756E 08    0.2852E-01    0.1363E 03
     13 - 14      0.3468E 08    0.4475E-01    0.1987E 03
     14 - 15      0.3597E 08    0.5759E-01    0.2380E 03
     15 - 16      0.3180E 08    0.6170E-01    0.2391E 03
     16 - 17      0.2148E 08    0.5030E-01    0.1831E 03
     17 - 18      0.1407E 08    0.3924E-01    0.1348E 03
     18 - 19      0.8325E 07    0.2742E-01    0.8911E 02
     19 - 20      0.5290E 07    0.2045E-01    0.6301E 02
     20 - 21      0.4236E 07    0.1910E-01    0.5590E 02

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.1531E 07 PER CU CM

SOLIDS CONCENTRATION =  0.2312E-02 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.17178E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671219-2U  (20)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  2.238 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

        DIAMETER       NUMBER        PARTICLE      SURFACE
        INCREMENT      DENSITY        VOLUME         AREA
         MICRONS       PER GM        CU CM/GM      SQ CM/GM
          1 - 2      0.8670E 10    0.1654E-01    0.6245E 03
          2 - 3      0.5805E 10    0.4539E-01    0.1092E 04
          3 - 4      0.1881E 10    0.4098E-01    0.7043E 03
          4 - 5      0.1688E 10    0.7983E-01    0.1063E 04
          5 - 6      0.7409E 09    0.6268E-01    0.6885E 03
          6 - 7      0.4932E 09    0.6988E-01    0.6470E 03
          7 - 8      0.1974E 09    0.4224E-01    0.3410E 03
          8 - 9      0.5714E 08    0.1750E-01    0.1254E 03
          9 - 10     0.8992E 07    0.3947E-02    0.2509E 02
         10 - 11     0.1011E 08    0.6204E-02    0.3528E 02
         11 - 12     0.8184E 07    0.6438E-02    0.3370E 02
         12 - 13     0.3955E 07    0.3977E-02    0.1919E 02
         13 - 14     0.9489E 06    0.1177E-02    0.5297E 01
         14 - 15     0.1552E 05    0.2262E-04    0.9649E-01
         15 - 16     0.0000E 00    0.0000E 00    0.0000E 00
         16 - 17     0.0000E 00    0.0000E 00    0.0000E 00
         17 - 18     0.0000E 00    0.0000E 00    0.0000E 00
         18 - 19     0.0000E 00    0.0000E 00    0.0000E 00
         19 - 20     0.0000E 00    0.0000E 00    0.0000E 00
         20 - 21     0.0000E 00    0.0000E 00    0.0000E 00

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.1219E 07 PER CU CM

SOLIDS CONCENTRATION =  0.6235E-04 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.54055E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671226-1U (21)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  3.505 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER      NUMBER       PARTICLE      SURFACE
     INCREMENT     DENSITY       VOLUME         AREA
      MICRONS      PER GM       CU CM/GM      SQ CM/GM
       1 -  2    0.9044E 09   0.1701E-02   0.6425E 02
       2 -  3    0.3216E 09   0.2613E-02   0.6192E 02
       3 -  4    0.2121E 09   0.4780E-02   0.8127E 02
       4 -  5    0.4082E 09   0.2054E-01   0.2681E 03
       5 -  6    0.3074E 09   0.2624E-01   0.2874E 03
       6 -  7    0.2173E 09   0.3102E-01   0.2865E 03
       7 -  8    0.1315E 09   0.2878E-01   0.2306E 03
       8 -  9    0.1024E 09   0.3282E-01   0.2316E 03
       9 - 10    0.6702E 08   0.2978E-01   0.1885E 03
      10 - 11    0.4793E 08   0.2891E-01   0.1653E 03
      11 - 12    0.3098E 08   0.2445E-01   0.1279E 03
      12 - 13    0.2277E 08   0.2323E-01   0.1115E 03
      13 - 14    0.1680E 08   0.2149E-01   0.9571E 02
      14 - 15    0.1180E 08   0.1878E-01   0.7778E 02
      15 - 16    0.9147E 07   0.1779E-01   0.6890E 02
      16 - 17    0.7841E 07   0.1843E-01   0.6703E 02
      17 - 18    0.6848E 07   0.1918E-01   0.6579E 02
      18 - 19    0.5427E 07   0.1794E-01   0.5823E 02
      19 - 20    0.4027E 07   0.1558E-01   0.4798E 02
      20 - 21    0.2823E 07   0.1268E-01   0.3716E 02

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.2014E 07 PER CU CM

SOLIDS CONCENTRATION =  0.7097E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.26240E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671226-1L (21)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  9.750 CM

EXTINCTION PATH LENGTH =  3.245 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

       DIAMETER        NUMBER          PARTICLE        SURFACE
       INCREMENT       DENSITY          VOLUME           AREA
        MICRONS        PER GM          CU CM/GM        SQ CM/GM
         2 -  3      0.3864E 09      0.3093E-02      0.7428E 02
         3 -  4      0.1496E 09      0.3577E-02      0.5966E 02
         4 -  5      0.1266E 09      0.5787E-02      0.7797E 02
         5 -  6      0.4224E 08      0.3593E-02      0.3939E 02
         6 -  7      0.3298E 08      0.4833E-02      0.4424E 02
         7 -  8      0.3627E 08      0.8022E-02      0.6405E 02
         8 -  9      0.4583E 08      0.1502E-01      0.1052E 03
         9 - 10      0.6168E 08      0.2755E-01      0.1741E 03
        10 - 11      0.4512E 08      0.2726E-01      0.1558E 03
        11 - 12      0.3292E 08      0.2608E-01      0.1362E 03
        12 - 13      0.2845E 08      0.2911E-01      0.1396E 03
        13 - 14      0.2439E 08      0.3132E-01      0.1393E 03
        14 - 15      0.2015E 08      0.3212E-01      0.1329E 03
        15 - 16      0.1646E 08      0.3203E-01      0.1240E 03
        16 - 17      0.1341E 08      0.3149E-01      0.1145E 03
        17 - 18      0.1086E 08      0.3043E-01      0.1043E 03
        18 - 19      0.8872E 07      0.2935E-01      0.9525E 02
        19 - 20      0.7349E 07      0.2848E-01      0.8767E 02
        20 - 21      0.6131E 07      0.2761E-01      0.8084E 02

CHECK =   0.10000E 01

PARTICLE NUMBER DENSITY =   0.9417E 06 PER CU CM

SOLIDS CONCENTRATION =   0.8596E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.19498E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671226-2U(22)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  3.043 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER       NUMBER        PARTICLE      SURFACE
     INCREMENT      DENSITY        VOLUME         AREA
      MICRONS       PER GM        CU CM/GM      SQ CM/GM
       1 -  2     0.5196E 10    0.9269E-02    0.3624E 03
       2 -  3     0.2023E 10    0.1649E-01    0.3912E 03
       3 -  4     0.6728E 09    0.1316E-01    0.2344E 03
       4 -  5     0.1399E 10    0.5941E-01    0.8224E 03
       5 -  6     0.5615E 09    0.4971E-01    0.5379E 03
       6 -  7     0.4941E 09    0.6963E-01    0.6459E 03
       7 -  8     0.2184E 09    0.4721E-01    0.3798E 03
       8 -  9     0.1169E 09    0.3720E-01    0.2632E 03
       9 - 10     0.6737E 08    0.2990E-01    0.1894E 03
      10 - 11     0.4253E 08    0.2553E-01    0.1462E 03
      11 - 12     0.2082E 08    0.1630E-01    0.8548E 02
      12 - 13     0.9562E 07    0.9674E-02    0.4657E 02
      13 - 14     0.4991E 07    0.6352E-02    0.2833E 02
      14 - 15     0.2460E 07    0.3886E-02    0.1613E 02
      15 - 16     0.1101E 07    0.2119E-02    0.8237E 01
      16 - 17     0.3741E 06    0.8624E-03    0.3156E 01
      17 - 18     0.3095E 05    0.8175E-04    0.2860E 00
      18 - 19     0.0000E 00    0.0000E 00    0.0000E 00
      19 - 20     0.0000E 00    0.0000E 00    0.0000E 00
      20 - 21     0.0000E 00    0.0000E 00    0.0000E 00

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.6414E 07 PER CU CM

SOLIDS CONCENTRATION =  0.5922E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.41614E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671226-2L(22)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  9.287 CM

EXTINCTION PATH LENGTH =  3.245 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER       NUMBER        PARTICLE      SURFACE
     INCREMENT      DENSITY        VOLUME         AREA
      MICRONS       PER GM       CU CM/GM     SQ CM/GM
       2 -  3     0.2548E 09   0.2075E-02   0.4932E 02
       3 -  4     0.9141E 08   0.1923E-02   0.3345E 02
       4 -  5     0.4246E 08   0.1956E-02   0.2629E 02
       5 -  6     0.1489E 08   0.1306E-02   0.1417E 02
       6 -  7     0.2060E 08   0.2992E-02   0.2747E 02
       7 -  8     0.1809E 08   0.3982E-02   0.3184E 02
       8 -  9     0.2726E 08   0.9012E-02   0.6296E 02
       9 - 10     0.4776E 08   0.2156E-01   0.1358E 03
      10 - 11     0.5700E 08   0.3475E-01   0.1980E 03
      11 - 12     0.5617E 08   0.4474E-01   0.2333E 03
      12 - 13     0.5942E 08   0.6082E-01   0.2917E 03
      13 - 14     0.2370E 08   0.3039E-01   0.1352E 03
      14 - 15     0.2064E 08   0.3289E-01   0.1361E 03
      15 - 16     0.1673E 08   0.3255E-01   0.1260E 03
      16 - 17     0.1361E 08   0.3195E-01   0.1162E 03
      17 - 18     0.9966E 07   0.2784E-01   0.9559E 02
      18 - 19     0.6912E 07   0.2281E-01   0.7408E 02
      19 - 20     0.4762E 07   0.1841E-01   0.5672E 02
      20 - 21     0.3295E 07   0.1480E-01   0.4339E 02

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.1740E 07 PER CU CM

SOLIDS CONCENTRATION =  0.2206E-02 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.18879E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671227U (23)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  1.875 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER       NUMBER        PARTICLE       SURFACE
     INCREMENT      DENSITY        VOLUME          AREA
      MICRONS       PER GM        CU CM/GM      SQ CM/GM
      1 -  2      0.8534E 10    0.1380E-01    0.5469E 03
      2 -  3      0.3144E 10    0.2359E-01    0.5756E 03
      3 -  4      0.7109E 09    0.1522E-01    0.2631E 03
      4 -  5      0.4027E 09    0.1874E-01    0.2510E 03
      5 -  6      0.1654E 09    0.1414E-01    0.1548E 03
      6 -  7      0.1289E 09    0.1836E-01    0.1697E 03
      7 -  8      0.6580E 08    0.1445E-01    0.1156E 03
      8 -  9      0.7059E 08    0.2282E-01    0.1606E 03
      9 - 10      0.5465E 08    0.2435E-01    0.1540E 03
     10 - 11      0.4315E 08    0.2609E-01    0.1491E 03
     11 - 12      0.3021E 08    0.2388E-01    0.1248E 03
     12 - 13      0.2313E 08    0.2361E-01    0.1133E 03
     13 - 14      0.1791E 08    0.2296E-01    0.1021E 03
     14 - 15      0.1363E 08    0.2170E-01    0.8985E 02
     15 - 16      0.1042E 08    0.2026E-01    0.7848E 02
     16 - 17      0.8079E 07    0.1895E-01    0.6896E 02
     17 - 18      0.7094E 07    0.1989E-01    0.6820E 02
     18 - 19      0.6003E 07    0.1985E-01    0.6443E 02
     19 - 20      0.4715E 07    0.1825E-01    0.5621E 02
     20 - 21      0.3521E 07    0.1583E-01    0.4638E 02

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.5625E 07 PER CU CM

SOLIDS CONCENTRATION =  0.4184E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.33537E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERMENT  671227L (23)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  7.928 CM

EXTINCTION PATH LENGTH =  3.245 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

      DIAMETER      NUMBER        PARTICLE      SURFACE
      INCREMENT     DENSITY        VOLUME         AREA
       MICRONS      PER GM       CU CM/GM      SQ CM/GM
        2 -  3    0.1013E 10   0.7745E-02   0.1879E 03
        3 -  4    0.2866E 09   0.6367E-02   0.1087E 03
        4 -  5    0.2352E 09   0.1113E-01   0.1482E 03
        5 -  6    0.1326E 09   0.1137E-01   0.1243E 03
        6 -  7    0.1230E 09   0.1774E-01   0.1633E 03
        7 -  8    0.6421E 08   0.1393E-01   0.1119E 03
        8 -  9    0.4404E 08   0.1411E-01   0.9963E 02
        9 - 10    0.2758E 08   0.1222E-01   0.7746E 02
       10 - 11    0.2618E 08   0.1599E-01   0.9111E 02
       11 - 12    0.2358E 08   0.1870E-01   0.9765E 02
       12 - 13    0.2167E 08   0.2220E-01   0.1064E 03
       13 - 14    0.2038E 08   0.2622E-01   0.1165E 03
       14 - 15    0.1896E 08   0.3029E-01   0.1252E 03
       15 - 16    0.1690E 08   0.3290E-01   0.1274E 03
       16 - 17    0.1421E 08   0.3336E-01   0.1213E 03
       17 - 18    0.1160E 08   0.3248E-01   0.1114E 03
       18 - 19    0.9366E 07   0.3098E-01   0.1005E 03
       19 - 20    0.7520E 07   0.2913E-01   0.8968E 02
       20 - 21    0.6625E 07   0.2987E-01   0.8743E 02

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.7032E 06 PER CU CM

SOLIDS CONCENTRATION =  0.3342E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.21967E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS
EXPERIMENT 671229U  (24)  RERUN-1

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  2.778 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

    DIAMETER      NUMBER        PARTICLE      SURFACE
    INCREMENT     DENSITY        VOLUME         AREA
     MICRONS      PER GM       CU CM/GM      SQ CM/GM
      1 -  2     0.2056E 09    0.1756E-03    0.8864E 01
      2 -  3     0.5842E 08    0.5051E-03    0.1178E 02
      3 -  4     0.2764E 08    0.6078E-03    0.1042E 02
      4 -  5     0.3305E 08    0.1566E-02    0.2085E 02
      5 -  6     0.4124E 08    0.3707E-02    0.3991E 02
      6 -  7     0.1510E 09    0.2235E-01    0.2040E 03
      7 -  8     0.1455E 09    0.3176E-01    0.2547E 03
      8 -  9     0.6136E 08    0.1879E-01    0.1347E 03
      9 - 10     0.1238E 08    0.5615E-02    0.3531E 02
     10 - 11     0.2926E 08    0.1810E-01    0.1026E 03
     11 - 12     0.4780E 08    0.3859E-01    0.2003E 03
     12 - 13     0.8110E 08    0.8214E-01    0.3953E 03
     13 - 14     0.7118E 07    0.8524E-02    0.3881E 02
     14 - 15     0.2382E 08    0.3913E-01    0.1603E 03
     15 - 16     0.3832E 08    0.7457E-01    0.2887E 03
     16 - 17     0.1937E 08    0.4469E-01    0.1635E 03
     17 - 18     0.2228E 07    0.5947E-02    0.2073E 02
     18 - 19     0.0000E 00    0.0000E 00    0.0000E 00
     19 - 20     0.0000E 00    0.0000E 00    0.0000E 00
     20 - 21     0.0000E 00    0.0000E 00    0.0000E 00

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.6143E 06 PER CU CM

SOLIDS CONCENTRATION =  0.6236E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.20913E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 671229L (24) RERUN-1

SOLID DENSITY =  0.8080 GMS/CU CM

LIQUID DENSITY =  2.5200 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  9.023 CM

EXTINCTION PATH LENGTH =  3.245 CM

REFRACTIVE INDEX. SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER      NUMBER       PARTICLE      SURFACE
     INCREMENT     DENSITY       VOLUME        AREA
      MICRONS      PER GM       CU CM/GM     SQ CM/GM
       2 -  3    0.5177E 10   0.3668E-01   0.9175E 03
       3 -  4    0.3809E 09   0.7860E-02   0.1376E 03
       4 -  5    0.1330E 09   0.6126E-02   0.8233E 02
       5 -  6    0.1496E 08   0.1043E-02   0.1225E 02
       6 -  7    0.1801E 06   0.3235E-04   0.2772E 00
       7 -  8    0.2815E 08   0.6513E-02   0.5125E 02
       8 -  9    0.5307E 08   0.1724E-01   0.1211E 03
       9 - 10    0.4852E 08   0.2170E-01   0.1371E 03
      10 - 11    0.6149E 08   0.3778E-01   0.2147E 03
      11 - 12    0.8141E 08   0.6528E-01   0.3396E 03
      12 - 13    0.1050E 09   0.1077E 00   0.5163E 03
      13 - 14    0.9078E 08   0.1166E 00   0.5185E 03
      14 - 15    0.7791E 08   0.1241E 00   0.5139E 03
      15 - 16    0.6136E 08   0.1192E 00   0.4618E 03
      16 - 17    0.4410E 08   0.1031E 00   0.3756E 03
      17 - 18    0.2907E 08   0.8128E-01   0.2789E 03
      18 - 19    0.2351E 08   0.7788E-01   0.2526E 03
      19 - 20    0.2218E 08   0.8615E-01   0.2649E 03
      20 - 21    0.2209E 08   0.9972E-01   0.2917E 03
      21 - 22    0.2327E 08   0.1213E 00   0.3384E 03

CHECK =  0.10000E 01

PARTICLE NUMBER DENSITY =  0.1015E 07 PER CU CM

SOLIDS CONCENTRATION =  0.1568E-03 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.58272E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS
EXPERIMENT 680102U  (25)  RERUN-1

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  2.000 CM

EXTINCTION PATH LENGTH =  3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER       NUMBER        PARTICLE      SURFACE
     INCREMENT      DENSITY        VOLUME         AREA
      MICRONS       PER GM        CU CM/GM     SQ CM/GM
       1 -  2     0.9715E 09    0.1474E-02    0.5917E 02
       2 -  3     0.4126E 09    0.3123E-02    0.7623E 02
       3 -  4     0.4136E 09    0.8574E-02    0.1506E 03
       4 -  5     0.1248E 09    0.5875E-02    0.7840E 02
       5 -  6     0.7553E 08    0.6477E-02    0.7084E 02
       6 -  7     0.8366E 08    0.1231E-01    0.1126E 03
       7 -  8     0.1019E 09    0.2268E-01    0.1807E 03
       8 -  9     0.1173E 09    0.3799E-01    0.2673E 03
       9 - 10     0.1088E 09    0.4886E-01    0.3082E 03
      10 - 11     0.1464E 09    0.8927E-01    0.5088E 03
      11 - 12     0.8522E 08    0.6684E-01    0.3503E 03
      12 - 13     0.4250E 08    0.4270E-01    0.2060E 03
      13 - 14     0.1319E 08    0.1670E-01    0.7465E 02
      14 - 15     0.7814E 07    0.1239E-01    0.5137E 02
      15 - 16     0.4587E 07    0.8872E-02    0.3442E 02
      16 - 17     0.2563E 07    0.5979E-02    0.2179E 02
      17 - 18     0.1344E 07    0.3738E-02    0.1285E 02
      18 - 19     0.6283E 06    0.2060E-02    0.6704E 01
      19 - 20     0.2133E 06    0.8134E-03    0.2517E 01
      20 - 21     0.1296E 05    0.5531E-04    0.1649E 00

CHECK =   0.10000E 01

PARTICLE NUMBER DENSITY =   0.3073E 07 PER CU CM

SOLIDS CONCENTRATION =   0.1132E-02 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =   0.25739E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS
EXPERIMENT 680102L  (25)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  8.165 CM

EXTINCTION PATH LENGTH =  3.245 CM

REFRACTIVE INDEX. SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER      NUMBER        PARTICLE      SURFACE
     INCREMENT     DENSITY        VOLUME         AREA
      MICRONS      PER GM       CU CM/GM      SQ CM/GM
       2 -  3     0.9295E 08   0.6916E-03   0.1689E 02
       3 -  4     0.3935E 08   0.8333E-03   0.1448E 02
       4 -  5     0.2611E 08   0.1270E-02   0.1676E 02
       5 -  6     0.1586E 08   0.1355E-02   0.1484E 02
       6 -  7     0.1174E 08   0.1689E-02   0.1556E 02
       7 -  8     0.9262E 07   0.2048E-02   0.1635E 02
       8 -  9     0.8702E 07   0.2828E-02   0.1987E 02
       9 - 10     0.1010E 08   0.4563E-02   0.2873E 02
      10 - 11     0.1997E 08   0.1234E-01   0.7003E 02
      11 - 12     0.2183E 08   0.1732E-01   0.9045E 02
      12 - 13     0.2539E 08   0.2622E-01   0.1253E 03
      13 - 14     0.4271E 08   0.5576E-01   0.2466E 03
      14 - 15     0.6641E 08   0.1065E 00   0.4401E 03
      15 - 16     0.5957E 08   0.1152E 00   0.4470E 03
      16 - 17     0.1912E 08   0.4367E-01   0.1603E 03
      17 - 18     0.8911E 05   0.2298E-03   0.8104E 00
      18 - 19     0.0000E 00   0.0000E 00   0.0000E 00
      19 - 20     0.3811E 04   0.1596E-04   0.4789E-01
      20 - 21     0.9211E 06   0.4171E-02   0.1219E 02

CHECK =   0.10000E 01

PARTICLE NUMBER DENSITY =  0.7345E 06 PER CU CM

SOLIDS CONCENTRATION =  0.1563E-02 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =  0.17365E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

EXPERIMENT 680105U  (26)  RERUN-1

SOLID DENSITY =   2.5200 GMS/CU CM

LIQUID DENSITY =   0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =   1.633 CM

EXTINCTION PATH LENGTH =   3.240 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

     DIAMETER        NUMBER         PARTICLE        SURFACE
     INCREMENT       DENSITY         VOLUME           AREA
      MICRONS        PER GM         CU CM/GM       SQ CM/GM
       1 -  2      0.3037E 11     0.4363E-01     0.1796E 04
       2 -  3      0.5973E 10     0.4239E-01     0.1053E 04
       3 -  4      0.9951E 09     0.2096E-01     0.3643E 03
       4 -  5      0.6959E 09     0.3297E-01     0.4389E 03
       5 -  6      0.3378E 09     0.2868E-01     0.3147E 03
       6 -  7      0.2174E 09     0.3095E-01     0.2861E 03
       7 -  8      0.1336E 09     0.2923E-01     0.2342E 03
       8 -  9      0.9298E 08     0.2965E-01     0.2096E 03
       9 - 10      0.5029E 08     0.2222E-01     0.1409E 03
      10 - 11      0.2939E 08     0.1765E-01     0.1011E 03
      11 - 12      0.1382E 08     0.1079E-01     0.5667E 02
      12 - 13      0.7222E 07     0.7353E-02     0.3532E 02
      13 - 14      0.5758E 07     0.7404E-02     0.3291E 02
      14 - 15      0.5371E 07     0.8584E-02     0.3549E 02
      15 - 16      0.5150E 07     0.1005E-01     0.3888E 02
      16 - 17      0.5041E 07     0.1185E-01     0.4309E 02
      17 - 18      0.4329E 07     0.1212E-01     0.4158E 02
      18 - 19      0.3440E 07     0.1137E-01     0.3691E 02
      19 - 20      0.2624E 07     0.1015E-01     0.3128E 02
      20 - 21      0.1945E 07     0.8748E-02     0.2562E 02

CHECK =   0.10000E 01

PARTICLE NUMBER DENSITY =   0.3162E 07 PER CU CM

SOLIDS CONCENTRATION =   0.8117E-04 GMS SOLID/CU CM LIQUID

TOTAL SURFACE AREA =   0.53177E 04 SQ CM/GM
```

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS
EXPERIMENT 680105L (26)

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

LIQUID LEVEL ABOVE STATION =  7.878 CM

EXTINCTION PATH LENGTH =  3.245 CM

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS

      DIAMETER       NUMBER          PARTICLE        SURFACE
      INCREMENT      DENSITY          VOLUME          AREA
       MICRONS       PER GM          CU CM/GM       SQ CM/GM
        2 -  3     0.9723E 10     0.5767E-01     0.1530E 04
        3 -  4     0.1510E 10     0.3520E-01     0.5923E 03
        4 -  5     0.1084E 10     0.5023E-01     0.6736E 03
        5 -  6     0.4055E 09     0.3421E-01     0.3761E 03
        6 -  7     0.2355E 09     0.3345E-01     0.3094E 03
        7 -  8     0.1309E 09     0.2856E-01     0.2291E 03
        8 -  9     0.8969E 08     0.2864E-01     0.2024E 03
        9 - 10     0.5172E 08     0.2291E-01     0.1452E 03
       10 - 11     0.3347E 08     0.2016E-01     0.1153E 03
       11 - 12     0.2022E 08     0.1594E-01     0.8341E 02
       12 - 13     0.1367E 08     0.1393E-01     0.6693E 02
       13 - 14     0.9589E 07     0.1227E-01     0.5464E 02
       14 - 15     0.6842E 07     0.1088E-01     0.4507E 02
       15 - 16     0.4842E 07     0.9395E-02     0.3641E 02
       16 - 17     0.3266E 07     0.7642E-02     0.2783E 02
       17 - 18     0.2124E 07     0.5927E-02     0.2035E 02
       18 - 19     0.1339E 07     0.4413E-02     0.1433E 02
       19 - 20     0.8087E 06     0.3120E-02     0.9619E 01
       20 - 21     0.4974E 06     0.2232E-02     0.6544E 01

  CHECK =   0.10000E 01

  PARTICLE NUMBER DENSITY =  0.1164E 07 PER CU CM

  SOLIDS CONCENTRATION =  0.8740E-04 GMS SOLID/CU CM LIQUID

  TOTAL SURFACE AREA =  0.45393E 04 SQ CM/GM
```

$$\int_{0}^{D_1} K \gamma_t \tilde{N} \, dD = \frac{1}{t} \ln \frac{I_o}{I(\theta_1)}$$

where $D_1 = C\theta_1^{-1/2}$, from equation 5.

With a properly chosen average cross section, $\overline{\gamma}_t$ we have

$$\overline{N} = \int_{0}^{D_1} \tilde{N} \, dD = \frac{1}{K\overline{\gamma}_t t} \ln \frac{I_o}{I(\theta_1)}$$

where $\overline{N}$ is the number density of particles (cm^{-3}) in the size range between 0 and D_1. These results are presented in Table II.

CONCLUSIONS

The results indicate that the apparent size distribution was more strongly dependent on particulate concentration in the liquid than on the characteristics of particle formation, at least in the range of variables studied. Thus, it appears that agglomeration of the particles was occurring rapidly in the vessel and that these agglomerates, rather than individual particles, were being observed. The tendency for this agglomeration to occur was drastically reduced under conditions of low concentration, where significant fractions of submicron particles were observed (v. Experiment No. 671219-2U (20)). This phenomenon of agglomeration was also apparent from the lower station measurements which in all cases indicated a larger mean particle diameter than did results from the upper station readings.

From the sequential measurements on the same dispersion which were interrupted by stirring (Experiment Nos. 671205-1, and -2), it was concluded that the measurements were reproducible to within the limits of error, and that no dissolution of the discontinuous phase was apparent.

A maximum specific surface area of about 5.0 m^2/gm was recorded for the lowest mass concentration studied.

TABLE II

POLYDISPERSION PROPERTIES
DIAMETER < 1 MICRON

```
LIGHT EXTINCTION MEASUREMENTS OF SETTLING IN PARTICULATE GELS

SOLID DENSITY =  2.5200 GMS/CU CM

LIQUID DENSITY =  0.8080 GMS/CU CM

VISCOSITY = 0.001400 GMS/CM SEC

REFRACTIVE INDEX, SOLID/LIQUID = 1.2120 + 0.000000 I

WAVELENGTH = 0.4360 MICRONS


      EXPERIMENT          PROPERTIES IN 0 - 1 MICRON REGION
        NUMBER            SOLIDS         NUMBER        SURFACE
                        CONCENTRATION    DENSITY         AREA
                          GM/CU CM       PER GM        SQ CM/GM
     671205-1U (17)      0.9770E-03    0.3348E 13    0.1468E 04
     671205-2U (17)      0.7522E-03    0.6613E 13    0.2900E 04
     671207U   (18)      0.5358E-03    0.2107E 14    0.9245E 04
     671219-1U (19)      0.7230E-03    0.1121E 14    0.4917E 04
     671219-2U (20)      0.6235E-04    0.4035E 14    0.1769E 05
     671226-1U (21)      0.7100E-03    0.3997E 12    0.1753E 03
     671226-2U (22)      0.5922E-03    0.2726E 13    0.1195E 04
     671227U   (23)      0.4184E-03    0.1069E 14    0.4690E 04
     671229U   (24)      0.6236E-03    0.1568E 11    0.6880E 01
     680102U   (25)      0.1132E-02    0.1161E 14    0.5093E 04
     680105U   (26)      0.8117E-04    0.1036E 15    0.4546E 05
```

REFERENCES

1. Globus, R. H., Adams, J. M., Beadle, P. D., and Bost, J. J., "System Analysis of Gelled Space-Storable Propellant" Fourth Qtrly Progress Report to Office Adv. Res. and Tech., NASA, Contract No. NAS 7-473 (10 August 1967).

2. Gumprecht, R. O., and Sliepcevich, D. M., "Scattering of Light by Large Spherical Particles" J. Phys. Chem. 57 (Jan, 1953) 90-95.

3. Gumprecht, R. O., and Sliepcevich, D. M., "Measurement of Particle Sizes in Polydispersed Systems by Means of Light Transmission Measurements Combined with Differential Settling" J. Phys. Chem. 57 (Jan, 1953) 95-97.

4. Mie, G., Ann. Physik 25 (1908) 377.

5. Van de Hulst, H. C., Light Scattering by Small Particles, Inley, New York (1957).

6. Erickson, W. C., "Light Scattering: A Technique for Studying Soot in Flames" PhD Thesis in Chemical Engineering, Massachusetts Institute of Technology (1962).

7. Gumprecht, R. O., "Particle Size Measurements by Light Scattering" PhD Thesis, University of Michigan (1952).

APPENDIX II

MEASUREMENT OF THE PARTICLE SIZE OF FULLY FLUORINATED OXIDIZERS BY A MODIFIED BET(1) PROCEDURE

MEASUREMENT OF THE PARTICLE SIZE OF FULLY-FLUORINATED OXIDIZERS BY A MODIFIED BET(1) PROCEDURE

I. INTRODUCTION

A study to determine the feasibility of using a modified BET procedure for measuring the surface area of particles of frozen interhalogens with subsequent calculation of their particle size has been completed. It is concluded the procedure is not applicable.

II. TECHNICAL DISCUSSION

A. BET THEORY

It has been suggested that the adsorption isotherm exhibited by various gases at temperatures not far removed from their condensation point occurs because van der Waal forces are predominant under these conditions. Also, the same phenomenon permits multimolecular adsorption layers to be formed at successively higher adsorbate pressures. Consequently, if the quantity of gas adsorbed is measured at several pressures and the results plotted and extrapolated to zero pressure, it is possible to estimate the quantity of gas required to form a monomolecular layer on the particle. With this information it is then possible to calculate the surface area of a unit mass of particles.

B. BET PROCEDURE

The powder is placed in a sample container and then evacuated, if possible at elevated temperatures, to drive off the adsorbed gases. Then the sample is cooled to the NBP of the gas which is to be adsorbed and the quantity of gas adsorbed at one or more partial pressures is measured. In

---

(1) S. Brunnauer, P. H. Emmet, E. Teller

II, B, Bet Procedure (cont.)

practice two basic types of measuring techniques are used. The first technique involves measurement of the pressure at various volumes of a fixed quantity of gas in an apparatus containing the sample of particulate matter. Based on the deviation of data obtained from ideal gas behavior, the quantity of gas adsorbed at zero pressure can be calculated. The second procedure measures changes in the concentration of a mixture of gases, usually helium and nitrogen, as one of the gases is being adsorbed or desorbed by the particles. The concentration of the adsorbed or desorbed portion of the gaseous mixture is calculated from measured changes in the thermal conductivity, and based on these data the amount of adsorbed gas can be calculated. In order to obtain maximum sensitivity, the volume of the particles closely approach the volume of the sample holder.

C. APPLICATION OF THE BET PROCEDURE TO THE MEASUREMENT OF FINE PARTICLES OF FROZEN INTERHALOGENS

During the initial phase of this study, fine particles of frozen interhalogens will be prepared by the rapid condensation and freezing of gaseous interhalogens in liquid nitrogen. Liquid nitrogen has been selected as a simulant for liquid OF_2 in order to reduce the hazards and toxicity problems associated with OF_2.

A test vessel of approximately 3-liters volume has been selected so that 1-liter samples of gelled material can be prepared. This quantity was selected in order to have sufficient quantities to make measurements of the engineering properties of the gel.

Because of the reactivity of the interhalogens and their low melting points, extensive manipulation of the particles or large temperature changes within the system present formidable experimental difficulties.

II, C, Application of the BET Procedure to the Measurement of Fine Particles of Frozen Interhalogens (cont.)

Consequently for any particle size measuring procedure to be practical, it must be possible to conduct the measurement in the test vessel at the NBP of liquid N_2.

1. BET Measurement of Desorbed N_2

After the particles are formed in the liquid N_2, it would be theoretically possible to pump off all of the liquid N_2 present except the monomolecular layer of N_2 on the particle. (See the section on BET Theory.) After the free liquid N_2 was removed, the sample could then be warmed until the particles melted and the residual quantity of N_2 remaining measured. While this is a single point measurement, rather than the usual 3 measurements, the surface area and relative particle size could be calculated and a fairly reliable value obtained. Consequently the feasibility of this approach has been investigated.

The following system properties were either assumed or calculated from the assumptions.

TABLE I

1.	Particle Size	10 microns
2.	Particle Shape	spheres
3.	Quantity of Particles	80.8 g
4.	Density of Particulate Material	2.5 g/cc
5.	Number of Particles Present	6.2×10^9
6.	Surface Area of Particles	1.9 m^2
7.	Quantity of N_2 Adsorbed at 100 mm Hg pressure, T 77.4°K(2)	0.45 cc N_2 STP

---

(2) Note: The quantity of N_2 adsorbed at 100 mm Hg pressure at 77.4°K by 1.9 m^2 of particles was calculated from Table 3-1 presented in Instructions - The Perkin-Elmer Shell Model 212D Sorptometer, March 1966, prepared by the Perkin-Elmer Corp., Norwalk, Conn.

Item 7 in Table I shows that the quantity of N_2 to be measured would be approximately 0.45 cc N_2 at STP. If this volume of N_2 is expanded to the 3-liters volume of the test vessel, it would exert a pressure of approximately 100 microns of Hg. It is expected that the vapor pressure of the melted interhalogen and the additional pressure generated by traces of SiF_4 formed by the reaction between the glass test vessel and the interhalogens and other noncondensible impurities would result in a total system pressure substantially higher than the pressure of N_2 caused by the desorption of the N_2 from the particles. Consequently a direct pressure measurement would not measure the quantity of N_2 present. A direct measurement of the quantity of N_2 present by mass spectrographic or other means was considered. However for this to be practical the entire system would have to be completely free of leaks. Because of the complexity of the experimental system and the reactive nature of the reagents it is doubtful if the equipment can be so leak free that an acceptable leak rate can be obtained.

Because of the above described difficulties it is concluded that the measurement of the quantity of desorbed N_2 from frozen particles of interhalogens can not be made with sufficient accuracy to provide useful information regarding surface area and/or particle size.

2. BET Measurement of Desorbed Gases other than Nitrogen

The feasibility of adsorbing a layer of a gas other than N_2 and measuring the quantity adsorbed has been investigated. It was felt that this approach would offer the following advantages. A gas that is not a common contaminant could be used, thereby reducing the requirement for a leak free system. The gases considered for use were neon, argon, krypton and xenon. Neon was rejected because its NBP is too low, i.e. system temperature is too far above the saturation temperature of neon.

II, C, Application of the BET Procedure to the Measurement of Fine Particles of Frozen Interhalogens (cont.)

The approach considered was to pump off the liquid N_2, add argon at a selected partial pressure and then bring up the system pressure to 1 atmosphere with helium. The same conditions were used as described in Table 1. The system would then be allowed to warm up until the interhalogen melted. A sample of system atmosphere would be taken, analyzed for argon content, and the result of this analysis used to calculate surface area and particle size.

It is expected that under the conditions just described that total system pressure would be approximately 1000 mm of Hg and the argon pressure would be 0.1 mm of Hg. A concentration of .01%. The sensitivity of the best available analytical procedure is ± .02%. Consequently no useful information could be obtained. The same results would be obtained if krypton or xenon were used.

III. CONCLUSION

For the reasons discussed in the section titled Technical Discussion, it is concluded that a modification of the BET procedure is not suitable for measuring the surface area of frozen interhalogen particles.

APPENDIX III

DISTRIBUTION LIST FOR SUMMARY REPORT
CONTRACT NAS 7-473

Copies	Recipient	Designee
4	Chief, Liq. Prop. Technology Program, RPL NASA Washington, D.C. 20546	(X)
1	Director, Launch Vehicles and Propulsion, SV NASA Washington, D.C. 20546	(X)
1	Director, Advanced Manned Missions, MT Washington, D.C. 20546	(X)
1	Director, Mission Analysis Division NASA Ames Research Center Moffett Field, California 94035	(X)
1	Air Force Rocket Propulsion Laboratory Research and Technology Division Air Force System Command Edwards, California 93523	
	Mr. G. Allen Beale	(X)
3	U.S. Naval Ordnance Test Station China Lake, California 93553	
	Mr. Duane Williams	(X)
	Mr. James Dake	(X)
	Mr. David Oliver	(X)
2	Jet Propulsion Laboratory 4800 Oak Grove Drive Pasadena, California 91103	
	Mr. Donald L. Young	(X)
1	NASA Pasadena Office 4800 Oak Grove Drive Pasadena, California 91103	
	Contracting Officer	(X)
1	Office of Technical Information and Patent Matters	(X)
25	Scientific and Technical Information Facility Attn: NASA Representative, Code CRT P.O. Box 5700 Bethesda, Maryland 20014	(X)
1	Lewis Research Center 21000 Brookpark Road Cleveland, Ohio 44135	
	Mr. Paul Herr	(X)

1	Marshall Space Flight Center Huntsville, Alabama 35812 Mr. Kieth Coates	(X)

NASA FIELD CENTERS

2	Ames Research Center Moffett Field, California 94035	Mission Analysis Div. Mr. Clarence A. Syvertson
2	Goddard Space Flight Center Greenbelt, Maryland 20771	Merland L. Moseson Code 620 Mr. D. Grant
2	Jet Propulsion Laboratory California Institute of Technology 4800 Oak Grove Drive Pasadena, California 91103	Henry Burlage, Jr. Propulsion Div., 38
2	Langley Research Center Langley Station Hampton, Virginia 23365	Dr. Floyd L. Thompson Director
2	Lewis Research Center 21000 Brookpark Road Cleveland, Ohio 44135	Dr. Abe Silverstein Director Mr. S. Cohen
2	Marshall Space Flight Center Huntsville, Alabama 35812	Hans G. Paul Code R-P+VED Werner Voss R-P and VE-PM
2	Manned Spacecraft Center Houston, Texas 77001	Dr. Robert R. Gilruth Director Mr. H. Pohl
2	Western Support Office 150 Pico Boulevard Santa Monica, California 90406	Robert W. Kamm Director
2	John F. Kennedy Space Center, NASA Cocoa Beach, Florida 32931	Dr. Kurt H. Debus
1	NASA Test Facility Propulsion Engineering Office White Sands, New Mexico	I.D. Smith Staff Chemist

GOVERNMENT INSTALLATIONS

1	Aeronautical Systems Division Air Force Systems Command Wright-Patterson Air Force Base Dayton, Ohio 45433	D. L. Schmidt Code ASRCNC-2
1	Air Force Missile Development Center Holloman Air Force Base New Mexico 88330	Maj. R. E. Bracken Code MDGRT
1	Air Force Missile Test Center Patrick Air Force Base, Florida	L. J. Ullian
1	Air Force Systems Division Air Force Unit Post Office Los Angeles 45, California 90045	Col. Clark Technical Data Center
2	Technical Library AFFTC (FTBPP-2) Edwards AFB, California 93523	Myrtle C. Jones
1	Arnold Engineering Development Center Arnold Air Force Station Tullahoma, Tennessee 37388	Dr. H. K. Doetsch
1	Bureau of Naval Weapons Department of the Navy Washington, D.C. 20546	J. Kay RTMS-41
1	Defense Documentation Center Headquarters Cameron Station, Building 5 5010 Duke Street Alexandria, Virginia 22314 Attn: TISIA	
1	Headquarters, U.S. Air Force Washington 25, D.C. 20546	Col. C. K. Stambaugh AFRST
1	Picatinny Arsenal Dover, New Jersey 07801	I. Forsten, Chief Liquid Propulsion Laboratory, SMUPA-DL
1	Air Force Rocket Propulsion Laboratory Research and Technology Division Air Force Systems Command Edwards, California 93523	RPRR/Mr. H. Main 1/Lt. D. Shantz

1 W. J. Wilcox
Oak Ridge Gaseous Diffusion Plant
(ORGDP) P. O. Box P
Oak Ridge, Tennessee 37830

1 U.S. Army Missile Command
Redstone Arsenal
Alabama 35809
Dr. Walter Wharton

1 U.S. Naval Ordnance Test Station
China Lake, California 93557
Code 4562
Chief, Missile Propulsion Div.

1 National Bureau of Standards
Boulder, Colorado
Cryogenic Division
Mr. Dudley Chelton

CPIA

1 Chemical Propulsion Information Agency
Applied Physics Laboratory
8621 Georgia Avenue
Silver Spring, Maryland 20910
P. Martin

INDUSTRY CONTRACTORS

1 Aerojet-General Corporation
P.O. Box 296
Azusa, California 91703
L. F. Kohrs

1 Aerojet-General Corporation
P.O. Box 1947
Technical Library, Bldg. 2015, Dept. 2410
Sacramento, California 95809
R. Stiff

1 Aeronutronic Division
Philco Corporation
Ford Road
Newport Beach, California 92663
Mr. N. Stern

1 Aerospace Corporation
2400 East El Segundo Boulevard
P.O. Box 95085
Los Angeles, California 90045
Mr. M. J. Russi
Mr. H. Greer
Propulsion Dept.

1 Arthur D. Little, Inc.
20 Acorn Park
Cambridge, Massachusetts 02140
E. Karl Bastress

1	Astropower Laboratory Douglas Aircraft Company 2121 Paularino Newport Beach, California 92663	Dr. George Moc Director, Research
1	Astrosystems International, Inc. 1275 Bloomfield Avenue Fairfield, New Jersey 07007	A. Mendenhall
1	Atlantic Research Corporation Edsall Road and Shirley Highway Alexandria, Virginia 22314	A. Scurlock
1	Beech Aircraft Corporation Boulder Division Box 631 Boulder, Colorado 80302	J. H. Rodgers
1	Bell Aerosystems Company P.O. Box 1 Buffalo, New York 14240	Mr. N. R. Roth J. Flanagan
1	Bendix Systems Division Bendix Corporation 3300 Plymouth Road Ann Arbor, Michigan 48105	John M. Brueger
1	Boeing Company P.O. Box 3707 Seattle, Washington 98124	J. D. Alexander
1	Missile Division Chrysler Corporation P.O. Box 2628 Detroit, Michigan 48231	John Gates
1	Wright Aeronautical Division Curtiss-Wright Corporation Wood-Ridge, New Jersey 07075	G. Kelley
1	Missile and Space Systems Division Douglas Aircraft Company, Inc. 3000 Ocean Park Boulevard Santa Monica, California 90406	R. W. Hallet Chief Engineer Advanced Space Tech. Mr. A. Pisciotta, Jr.
1	Aircraft Missiles Division Fairchild Hiller Corporation Hagerstown, Maryland 21740	J. S. Kerr

1	General Dynamics Convair Division 5001 Kearny Villa Road P.O. Box 1628 San Diego, California 92112	E. R. Peterson V.P., Research and Eng. Frank Dore
1	Missile and Space Systems Center General Electric Company Valley Forge Space Technology Center P.O. Box 8555 Philadelphia, Pennsylvania	F. Mezger
1	Advanced Engine and Technology Dept. General Electric Company Cincinnati, Ohio 45215	D. Suichu
1	Grumman Aircraft Engineering Corp. Bethpage, Long Island New York 11714	Joseph Gavin
1	Hughes Aircraft Co. Aerospace Group Centinela and Teale Streets Culver City, California	E. H. Meier V.P. and Div. Mgr., Research and Dev. Div.
1	Ling-Temco-Vought Corporation Astronautics P.O. Box 5907 Dallas, Texas 75222	Garland Whisenhunt
1	Lockheed Missiles and Space Co. Attn: Technical Information Center P.O. Box 504 Sunnyvale, California 94088	Y. C. Lee
1	Lockheed Propulsion Company P.O. Box 111 Redlands, California 92374	H. L. Thackwell J. E. Fitzgerald
1	The Marquardt Corporation 16555 Saticoy Street Van Nuys, California 91409	Warren P. Boardman, Jr. Mr. S. Minton
1	Baltimore Division Martin Marietta Corporation Baltimore, Maryland 21203	John Calathes (3214)
1	Denver Division Martin Marietta Corporation P.O. Box 179 Denver, Colorado 80201	J. D. Goodlette (A-241) A. J. Kullas

1	Orland Division Martin Marietta Corp. Box 5837 Orlanda, Florida	Mr. J. Ferm
1	McDonnell Aircraft Corporation P.O. Box 516 Municipal Airport St. Louis, Missouri 63166	R. A. Herzmark Mr. R. E. Martens
1	Space and Information Systems Division North American Aviation, Inc. 12214 Lakewood Boulevard Downey, California 90241	H. Storms
1	Rocketdyne (Library 586-306) North American Aviation, Inc. 6633 Canoga Avenue Canoga Park, California 91304	E. B. Monteath
1	Northrop Space Laboratories 3401 West Broadway Hawthorne, California 90250	Dr. William Howard
1	Reaction Motors Division Thiokol Chemical Corporation Denville, New Jersey 07832	Arthur Sherman Mr. Robert Gere
1	Republic Aviation Corporation Farmingdale, Long Island, New York	Dr. William O'Donnell
1	Space General Corporation 9200 East Flair Avenue El Monte, California 91734	C. E. Roth
1	Stanford Research Institute 333 Ravenswood Avenue Menlo Park, California 94025	Lionel Dickinson
1	TRW Systems One Space Park Redondo Beach, California 90278	Mr. D. Lee Mr. V. Moseley
1	Thiokol Chemical Corporation Huntsville Division Huntsville, Alabama 35807	John Goodloe

1	United Technology Center 587 Methilda Avenue P.O. Box 358 Sunnyvale, California 94088	B. Adelman
1	Florida Research and Development Pratt and Whitney Aircraft United Aircraft Corporation P.O. Box 2691 West Palm Beach, Florida 33402	R. J. Coar
1	Vickers Inc. Box 302 Troy, Michigan	
1	Sunstrand Aviation 2421 11th Street Rockford, Illinois 61101	Mr. R. W. Reynolds
1	Hamilton Standard Division United Aircraft Corp. Windsor Locks, Connecticut 06096	Mr. R. Hatch
1	Technical Library Air Research Mfg. Co. 9851 Sepulveda Boulevard Los Angeles, California 90009	Mr. C. S. Coe

AEROJET
GENERAL TIRE
GENERAL

AEROJET-GENERAL CORPORATION