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CONTINUOUS PARTICLE SAMPLER
STUDY PROGRAM

By
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Final Report, Contract Nonr-3819(00)(X)
Department of the Navy, Office of Naval Research,
monitored by Naval Research Laboratory

Meteorology Research, Inc.
2420 North Lake Avenue
Altadena, California

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ABSTRACT

Several versions of a continuous particle sampler had been developed and used on cloud physics programs to collect and permit analyzing of cloud droplets and ice crystals. The units yielded significant data but demonstrated various operational difficulties. The present project has been directed at studying the fundamental factors on which the technique is based, and at developing and testing engineering improvements to the device.

A method of absolute calibration to relate replica size to original droplet size was developed. Aspergillus niger spores, 3 microns in diameter, do not dissolve in the solvents used to soften the Formvar coating. These spores are mixed in the water from which the droplets are formed. Then counting the spores visible in the droplet replica permits estimating the mass of the original droplet. The correction factor (replica diameter/droplet diameter) is unity for small droplets of diameter thinner than the Formvar, and greater than unity for larger droplets. The correction factor appears to be a unique function of the final dry Formvar thickness, implying that the droplet final shape is defined after much of the solvent has evaporated. Theoretical studies involving a computer program were also conducted on droplet deformation as a function of water/Formvar solution/air interface surface tensions.

The various interrelated problems of flocculation, encapsulation and replication, blushing, blistering, crystal melting and droplet evaporation, solution blowoff, bubble formation, and film sticking are considered, and the design of an improved device (built and delivered to NRL) is discussed. The design factors are derived both from laboratory studies and field projects, including a set of NRL/MRI comparison flights.

Appendices give data on special studies and tests related to the program: surfactant tension and viscosity of the Formvar solutions, absorption and evaporation of the solvents, and wick design criteria.
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I. INTRODUCTION

This is the final report covering the research and study program which has been conducted by Meteorology Research, Inc. for the Department of the Navy, Office of Naval Research, under Contract Nonr-3819(00)(X).

The scope of this contract was a study of the problem areas connected with the design and operation of a continuous cloud particle collector or sampler which would adequately serve Navy research needs for such airborne instrumentation. The continuous collector has been employed by Meteorology Research, Inc. and the affiliated Atmospheric Research Group on various cloud physics projects since 1961. Its continued development has been supported by MRI, the National Science Foundation, the U.S. Army Electronics Research and Development Laboratory, and the Advanced Research Projects Agency, as well as by the present contract.

The continuous collector is a continuous version of the Formvar* replication scheme devised by Schaefer (1956). A Formvar-solvent solution is coated on a continuous film which moves past a slit in the airborne device. Particles impact on this soft viscous coating and are immediately completely encapsulated by the Formvar solution. The solvent quickly evaporates, leaving a hard permanent replica of the particle. Ice crystal replicas have the exact particle shape; water droplet replicas can be distorted, because surface tension effects flatten droplets which are thicker than the Formvar solution. The replicas are viewed by projecting the transparent film with a stop-motion microscope projector.

The feasibility of the principle upon which the design development was based was demonstrated by Todd (1951) and MacCready (1962). The subsequent work then has involved the construction and testing of a series of field breadboard systems which have culminated in the evolution of the present version of the field unit. The system, though simple in concept, is complicated in application by the wide range and antagonistic nature of the environment in which it must operate.

In order to eliminate, as far as possible, the trial and error approach and to obtain a calibration of the system, approximately one-third of the project effort has been devoted to investigation of the physics of the coating and replication phenomena. Another third of the effort was devoted to development and testing of the sampler in actual cloud flight operation.

The machine, although not completely perfected from an operational standpoint, has rapidly become indispensable as a source of permanent,

continuous, and definitive records of the distribution of water and ice particles in the fine structure of atmospheric clouds.

The analytical and empirical investigations of the physics of the replication phenomena have progressed successfully so that records of water and ice particles in the five to fifty micron diameter range may be considered definitive both quantitatively and qualitatively.
II. DISCUSSION OF THE PROBLEM

Studies in observational cloud physics are handicapped by the difficulty of obtaining accurate and frequent samplings of the distribution, size, and form of the particles which make up the fine structure of atmospheric clouds. The continuous cloud particle sampler which has been the subject of this development program has, therefore, been designed as a practical answer to fill the requirement for such a piece of airborne research instrumentation.

In the past there have been many attempts to create such an instrument but none appear to be very satisfactory when they are evaluated in terms of the actual need. The present system was, therefore selected on the basis of its fulfillment of some of the criteria which are now discussed.

Of basic importance is the necessity for this instrument to provide clear and accurate records of the size, form, and distribution of ice and water particles. In the investigation of large clouds this equipment must be capable of extended and uninterrupted operation in untended aircraft mountings. The records must be unambiguously time referenced. The form of the recorded data must lend itself to rapid evaluation. The size, weight, and power requirements of the instrument must be such as to be compatible with being mounted and operated on a light aircraft. It should be capable of functioning in a saturated atmosphere at pressure altitudes of up to thirty thousand feet and at temperatures ranging from 40°C down to -40°C. It must be able to withstand and operate reliably in the vibrating environment of an aircraft mounting while exposed to two hundred mile per hour slip stream in heavy icing, hail, or repeated applications of the violent conditions normally associated with strongly convective cells.
III. HISTORY OF DEVELOPMENT

In consideration of the foregoing requirements, it appeared that the most promising approach was to record plastic replicas of the particles on a clear 16 mm movie film base. Trials of this method, during the summer of 1960 on a project conducted by the Atmospheric Research Group for the National Science Foundation, gave substantial encouragement to this approach. During the next winter at the Yellowstone Project (Schafer, 1961) Koenig and MacCready tested and compared various particle sizing methods, and it was concluded that Schafer's Formvar technique offered such special virtues (primarily the permanent exact replica) that it should be adapted to continuous operation despite the obvious difficulties. A continuous particle sampler was built and used in the summer of 1961 on the MRI and ARG Flagstaff cumulus studies (see Todd, 1961). A duplicate of this developmental unit was procured by NRL. In the fall the MRI instrument was adapted to ground use by building a small wind tunnel around the collector slot to duplicate the airplane wind blast. This system was taken to France to the "Meeting for the Study and Comparison of Droplet Collectors at L'Observatoire du Puy de Dome", and a calibration of replica diameter vs. droplet diameter obtained by comparison with the droplet measuring devices (see MacCready, 1962).

Flight testing of the breadboard system during the summer of 1961 provided much useful research data and indicated the areas in which further development was required. An extension of the MRI and ARG support for Flagstaff studies indicated modifications to be incorporated, and further testing of a new system (as well as the original system) was carried out in 1962.

During the 1962 period of testing, a considerable improvement was evident and again significant research data were obtained. A new coating principle was evolved which gave more consistent results than those previously obtained. A de-icer was incorporated and operational problems were studied. In the fall of 1962 comparison flights were conducted between the new coating system which was mounted on a MRI Aero-Commander and the original system acquired by NRL which was mounted in the Navy S2F aircraft. The records which were obtained from the contractor-operated aircraft were edited at MRI and turned over to NRL for evaluation.

A further no-cost extension of the present ONR-NRL contract allowed the testing of critical components of the latest improved machine in connection with the MRI cloud physics studies at Flagstaff in the summer of 1963.

Each family of breadboard configurations has been flight tested and in turn each series of flight tests has suggested further modifications; however, in each series of flight tests which has been conducted in conjunction with a cloud physics research program, increased importance has been attached to
the data gathered by this instrument, and now it is considered to be relatively indispensable. The equipment in its present form must still be considered as an experimental prototype since it does not invariably replicate all the desired spectrum of particles to which it may be exposed on a routine basis on each flight.
IV. MECHANICAL DESIGN AND OPERATION

The system consists of a 1350-foot magazine from which the clear 16 mm film is drawn first over a coating device, then past a window where it is exposed to the air stream, along electrically heated drying tubes, and finally wound onto a take-up reel. The general mechanical setup can be seen in Fig. 1 through 5, especially in Fig. 1 which shows the layout of the latest version. As the film passes the window water particles are captured by the Formvar solution which quickly hardens, leaving a permanent plastic replica of the shape of the particles. The encapsulated water eventually diffuses through the plastic skin and evaporates.

A polyester base film (Mylar) has proven to be most satisfactory. This film is obtainable from duPont, Wilmington, Delaware, designated Cronar, P40A Leader, Polyester Photographic Film Base. It is regular 16 mm silent movie film perforated along both edges. It has the highest strength of the regularly available film bases and appears to be entirely compatible with the Formvar coating and the chlorinated hydrocarbon solvents. The following pertinent mechanical properties were obtained from duPont.

- Thickness of film: 0.004 inches
- Tensile strength: 26,000 P.S.I.
- Load at 10% elongation: 13,000 P.S.I.
- Elongation at break: 140%
- Operating temperature range: -40°C to +125°C
- Ignition temperature: 525°C
- Thermal expansion linear: 36 x 10^-6 °C^-1
- Refractive index: 1.639

No difficulties have been encountered due to low operating temperatures but deformation of the perforations may occur at the drive sprocket under high starting loads if the film has been softened by excessive drying temperatures. A thermostat has been installed on the final drying tube and an operating temperature of about 50°C has given satisfactory results.

The coating refers to the Formvar coating which is applied to the film. The purpose of the coating is to provide a continuous fluid bed of uniform depth and controlled composition and viscosity in which to capture and encapsulate the cloud particles.

The first continuous cloud particle collector used a method in which the film was precoated and dried in the laboratory. Just before exposure to the air stream the hardened coating was softened by immersion in a bath of solvent. The coating in the laboratory was accomplished with a broad pen. Formvar solution was carried into the pen by siphon feed from the supply bottle. The pen then applied the solution through a slit which was one-fourth
of an inch wide. The siphon head pressure, the speed of the film, and the viscosity of the solution controlled the thickness and to a small extent the width of the coating.

Coating speeds of up to eighteen inches per second were reliably achieved. It was found that if a solution of at least 6 per cent Formvar was used the coating would not orange-peel or wrinkle during the drying process. In general the thicker solution produced smoother and more uniform coatings. It was found that with a reliable transport system it was possible to coat up to eight 1300-foot spools of film a day.

A considerable effort was also expended in attempting to develop a continuous applicator using a wick. The results were not encouraging and further work on this method was discontinued. Details of the wick method are presented in Appendix E. In general the wick or pad always left a streaked surface, large feed pressures were required in order to produce acceptable coating rates, and the method generally involved numerous operational difficulties.

The roll-on coating wheel proved to be quite reliable operationally, and while it has never attained the uniform and consistent standards of the laboratory controlled pen coatings it has routinely produced acceptable coatings during intermittent operation and while mounted remotely on an aircraft. The roll-on method is superior to the pre-coated and dipped system in that it does not require two passes through two separate coating and drying devices and the concomitant insertion of another set of variables. Further, the concentration of the solution of the coating is uniform in depth when presented to the air stream rather than sharply graded towards the base. In the case of the pre-coat and dipped method, the film is unnecessarily and undesirably cooled by the evaporation of large surpluses of solvent.

In the coating wheel method the moving film is pressed against the top edge of a thin, large diameter wheel causing it to rotate. The bottom portion of the wheel lies in a reservoir of the coating solution, and as the wheel rotates a stripe of this solution is transferred to the film at the point of contact. In practice the edge of this wheel, which is about one-eighth of an inch wide, is machined to form a trench. The edges of the trench or lands provide enough contact area with the film to ensure that the compressive yield strength of the film is not exceeded when sufficient contact pressure is applied to ensure reliable starting and rotation of the coating wheel. If the adhesive forces at the Formvar film and Formvar wheel interfaces are relatively unaffected by temperature, then it is reasonable to assume that the thickness of the coatings deposited from this trench will also be unaffected by temperature and will only be a function of the geometry of the trench. These same forces also cause a heavier coating to be laid down at the intersection of the film and the normal surfaces of the wheel. In order to minimize this deposition of excess Formvar
at the edges of the stripe, an adjustable scraper was fitted. This scraper consists of thin stainless steel blades which are in contact with the perimeter and sides of the wheel and serve to wipe off any excess solution which adheres to these surfaces. These shim stock blades are mounted on a block of teflon which may be adjusted in order to vary the wheel clearance.

In order to facilitate the calibration of data during analysis it is important that a strip at least 200μ wide and centered on the stagnation point of the sample be provided with as uniform a coating as possible. In practice the thickness of this coating varies by perhaps ±10 per cent after exposure to the air stream.

It is possible that a multiple roller system similar to those employed in large printing machines might further improve the uniformity and reduce the heavy edge coatings, however of the many configurations which have been tried the roll-on method which has been described proved to be by far the most practical.

A discussion of the coater system must also include some consideration of the requirements of the coating material. The fluid must be stable in the supply tank in a flight environment for periods of several hours. Water must be relatively insoluble in the coating solution or the replicas will be neither sharply defined nor predictably reproducible. The relative surface energies of the solution and the water particles must be such as to encourage a high collection efficiency and rapid encapsulation of impacted droplets and ice crystals. Hardening of the coating must be sufficiently rapid to be compatible with a practical airborne instrument. A brief evaluation indicated that these criteria would be most probably satisfied by a solution of chloroform or ethylene dichloride. The physical properties of these solutions were therefore investigated. Appendices A, B, C, and D detail the pertinent properties of these materials.

While it is recognized that the solvent is evaporating from the coating from the moment it leaves the coating wheel, the design is such as to minimize a change in concentration of the coating solution prior to its exposure at the window. It appears from flight tests that a concentration of between 3.5 per cent and 4 per cent in the coater tank is about optimum. When the concentration is less than 3.5 per cent the air stream displaces the coating at relatively low air speeds. When the concentration is greater than 4 per cent the collection efficiency appears to decrease, and with large increases in concentration the replication phenomenon becomes intermittent and unreliable. 3-1/2 per cent chloroform solutions have afforded good replication at indicated air speeds of 170 mph (which was the structural limitation of the aircraft in which some of these tests were carried out).

If the coating is to arrive at the sampling window with a concentration essentially unchanged from that in the coating tank, and if the concentration
of the solution in the coating tank is to remain unchanged throughout a flight of perhaps several hours, then it is obvious that either no premature evaporation of solvent can be allowed to take place or solvent must be replaced as rapidly as it is lost.

The vapor pressure of the two solvents is very high (see Appendix A) and any flow of air over the surface of their solutions causes evaporation in sufficient quantities to maintain the partial pressures in equilibrium. Since the coated film is exposed to total head pressure at the window and it has not been found practical to place a seal between the window and the coater tank, then it follows that in order to prevent a flow of unsaturated air over the solution the tank and film magazine portion of the system must be pressurized or sealed so that their atmospheres are in equilibrium with the stagnation pressure. As an example it may be cited that a slight leak in the magazine of a well sealed system caused a loss of up to 5cc per minute of chloroform from a surface area of approximately 30 square centimeters agitated in a flight environment and under a pitot static differential corresponding to 120 mph.

The window through which the coated film is exposed to the air stream is a normal slot in a thin walled cylinder. The film is drawn up a channel inside this cylinder which bends it laterally so that the portion which is exposed to the air stream conforms to the inside surface of this cylinder without actually touching it. In this way the sample is obtained at the stagnation point of a simple surface of which the aerodynamic characteristics are well known. Collection efficiencies can then be computed by standard procedures. In order to prevent ice from blocking the window or modifying the flow about it a section of about 5 cm on either side of the slot is electrically heated (see Figs. 1 through 4, especially Fig. 4). The heater ribbon is designed to develop about 67 watts at 28 volts which was estimated to be sufficient to afford a small safety factor under the worst possible conditions at speeds of up to 200 mph. Operational experience suggests that the worst conditions are rarely encountered and that when not required it may be advantageous to operate the de-icer at less than maximum power to minimize possible melting of ice crystals.

After exposure at the window the coated film and impacted droplets are passed through the drying tubes to the take-up spool. The drying tubes are fabricated from aluminum extrusion with an inside diameter slightly larger than 16 mm. The outer surfaces are hard anodized to a depth of .002 inch and wound with a suitable nickel-chrome heating ribbon. The two heaters are connected in series with a thermostatically controlled switch which is physically located in good thermal contact with the larger of the two heaters.

The purpose of the heated tubes or film guides is to facilitate the drying of the coating. This drying or evaporation of the solvent from the plastic encapsulant must be accomplished in such a way as to minimize alteration
of the replica and completely enough to allow spooling without adhesion. Impacted water droplets are captured and contained beneath the Formvar surface very rapidly, within milliseconds, and then seem to be well protected from quick evaporation which would decrease their sizes. It is possible that ice crystals take somewhat longer to encapsulate. During the period when crystals are being filmed over by the Formvar, water vapor must not condense on their surfaces causing growth. After the filming over of the crystal, too much heating of the film base without sufficient ventilation to ensure evaporation of the solvent from the coating will cause the ice crystal to melt before the coating has become permanent and thus cause the resultant replica to appear to be that of a droplet. Sometimes such replicas appear to be slightly deformed. The evaporation of the solvent cools the coating, and if some heat is not added water vapor is condensed on the surface in the form of many small droplets of about a micron diameter. This surface phenomenon is commonly known as blushing and if severe can obscure the density variations due to actual replicas. The conditions which encourage blushing may also cause spurious crystal growth. In all cases the cure for blushing is to keep the surface of the film above the dewpoint of the air with which it is in contact. The most severe blushing usually occurs within the first few inches of the exposure window where the film and coating are exposed to the air stream and evaporative cooling is at its peak. It has been found that the heat supplied by the window de-icer is generally sufficient to maintain blushing at this point within acceptable limits. In general, for the replication of ice crystals it is necessary to maintain the 16 mm film and coating at a temperature just below 0°C until the replica is permanent. After the crystal has been covered by a film of Formvar, rapid evaporation of the solvent should be encouraged by good ventilation and the addition of just enough heat to raise and retain the temperature of the coating just below 0°C.

Very little trouble has been experienced in replicating water droplets in the size range five to fifty microns on a more or less routine basis. If the solution is of less than the recommended concentration, groups of droplets will tend to coalesce to form clumps or strings (flocculation) or occasionally one big apparent droplet (coalescence).

In the equipment which is the subject of this report, an attempt has been made to meet the drying criteria for ice crystals without the aid of sensors and servo controls. The ice crystal must first be filmed over or encapsulated by the highly mobile Formvar solution and then without allowing any melting of the crystal enough solvent must be evaporated from the surrounding coating to make permanent and solid the Formvar replica. Obviously until this latter point is reached the coating temperature must not be allowed to rise above 0°C. In the coating tank the film and its coating are approximately in equilibrium with the outside air temperature and surrounded by a stagnant solvent-saturated atmosphere. Ventilation of the drying circuit is brought about by the pitot static differential which prevails between the window and the interior of
the case which is vented to static pressure through the cover which protects the electric drive motor. As the coated film reaches the window rapid evaporation is initiated by the fresh air flow and the film surface is therefore cooled. At the same time this flow of air is heated by the de-icer which must be operated in icing conditions. Ideally the flow and the heating are adjusted so that the film surface is maintained at a temperature high enough to prevent severe blushing, but below 0°C. The heated window insert is connected by good thermal conductors to the support fairing in order to dissipate excess heat from the window. In this way the rate of evaporation is reduced and a little delay is allowed for the filming-over of the ice crystals.

The film now passes up the first drying tube. Based upon the heat of vaporization of chloroform (59 cal/gram), a 3.5 per cent solution and an initial coating thickness of .009 inch, the heat required to replace that lost by complete evaporation of the solvent would be equivalent to about 18 watts. The window de-icing heater has an output of approximately 60 watts and under equilibrium conditions with no icing the equivalent of 20 watts could be transferred to the air in the drying tube. It is estimated that even the initial rate of evaporation causes cooling equivalent to only 5 watts and therefore it is obvious that some net heating of the air and the film is likely to occur. At air speeds between 100 and 200 mph it is estimated that the rise in air temperature may amount to more than 5°C but is probably less than 10°C. In marginal conditions this rise may be enough to melt ice crystals and therefore a small thermostat has been mounted in the drier air flow just down stream from the window assembly. The switch element has been placed in series with the window heater and is shunted by a resistance of about 6 ohms. The thermostat switch has been set to open at just above the freezing point and in so doing approximately halve the power input to the window heater.

The heating elements of the first and second heating tubes are connected electrically in series with one another and a thermostatic switch mounted on the exit end of the second tube. The element of the first tube is wound so as to concentrate the heating at the down stream or upper end. The first heater develops about 37 watts and the second 42 watts. The thermostat is set so that in operation the Formvar film just hardens at the take-up reel. Since the rate of hardening is dependent upon the rate of evaporation, which in turn is dependent to a large extent on the partial pressure of evaporant in the adjacent air, then ventilation of the drying tubes is of paramount importance. Since ventilation depends upon the pitot static differential and no auxiliary blower is supplied, a pitot pressure must be simulated at the window for laboratory adjustments.

A 24-volt electric motor drives the film transport system through a gear train and adjustable overload clutch assembly. The motor is irreversible, regulated, and of adequate power rating. No difficulties or failures have occurred with this type of motor in several hundred hours of operation. Cooling is encouraged by venting the instrument case through slots on the inside of
the motor cover. All electrical connections are brought out to a barrier terminal strip and from these to a flange-mounted seventeen-pin AN receptacle.

The configuration used on the USAERDL-ARPA Flagstaff program in the summer of 1963 is shown in Fig. 5. It was built into the nose of the twin-engine Apache aircraft. A continuous projector was installed in the cockpit so the film could be examined immediately (considering the 70 seconds of film transport time from passing the slot to reaching the projector). The projector worked well, but was not used often because the flight tests which were usually initiated were not ones which benefited particularly from immediate assessment. The airborne projector method should prove valuable in engineering (in optimizing the heating of future units), and in cloud seeding operations (in searching out areas where ice crystals do or do not exist).
V. REVIEW OF OPERATIONAL DIFFICULTIES

An informal interim report of activities carried out under this contract was presented to ONR/NRL (Todd and Hindman, 196Z) during the fall of 1962 in which certain operational problem areas were outlined. These are now reiterated and discussed in the light of later developments.

1. Ice crystal growth on the film after collection
2. Droplet flocculation on the film after collection
3. Blushing (condensation of water vapor on the film as the solvent evaporates during replication)
4. Film sticking in the sampler
5. Collecting window icing
6. Insufficient softening of emulsion at low temperatures
7. Emulsion blown off at high aircraft speeds

1. Ice Crystal Growth

Ice crystals which were collected in the dip tank system were subject to considerable crystal growth due to moisture provided during cooling of the large amount of chloroform on the film. Some reduction in growth was effected by substituting ethylene dichloride for the chloroform since the rate of evaporation is lower. Heating of the solvent tank and removal of excess solvent by wiping the back of the film also caused some reduction of growth. The coater system has produced excellent ice crystal replicas with little or no growth due to the small amount of solvent used and subsequent reduction in chilling and water condensation on the film. This phenomenon appears to be well within acceptable limits for the present heated coating system.

2. Flocculation

Flocculation is the term used to describe the adhesion of several droplets to one another beneath the Formvar film. If these droplets join to form larger droplets, they are then said to have coalesced. This problem though rarely severe has been most frequently observed when thin, low percentage solutions are employed. It seems to occur when the droplet diameters are greater than the thickness of the coating. In the case for droplets which are encapsulated but larger in diameter than the thickness of the film the surface at the Formvar air interface is displaced. If two such droplets are adjacent it can be shown
that the resultant forces of the displaced encapsulant tend to
force them together. All the mechanisms of flocculation and coalescence as
they concern cloud particles in the Formvar solution are not completely
understood; however as a matter of operational fact it is a phenomenon which
is rarely encountered when the suggested 4 per cent solution is used in the
present coater system. Considerable flocculation can occur apparently
without coalescence, and so the droplet size and concentration information is
not lost by flocculation.

3. Blushing

Blushing is caused by condensation of atmospheric water on the surface
of the coating in the form of droplets which have a diameter of about a micron.
The condensation occurs when the solvent cools the film below the local dew-
point by evaporation. Considerable blushing was characteristic of the dip
tank system because of the large excesses of solvent. Some alleviation of
this condition was effected by heating the solvent tank. The coater system
has been much less subject to blushing and, in practice, blushing has been
easily controlled by adjusting the power input to either the window heater or
the drying system.

4. Sticking

Sticking of the film transport system due to the bonding of moving parts
by hardened Formvar was a frequent cause of failure in the early dipper and
coater systems. The sticking usually occurred either in the coating tank or
at the lower edge of the window insert. The problem has been almost entirely
eliminated by preventing the hardening of the Formvar in these areas. This
has been accomplished by sealing the coating tank and supply reel to the entry
of all except total head pressure air at the window and thus ensuring that all
moving parts on these assemblies are constantly surrounded by a stagnant
atmosphere which is saturated with solvent vapor. An adjustment for the
spring load on the friction drive for the coater wheel has also been provided.
During the last series of flight tests no failures occurred due to sticking of
the coater wheel or the film.

5. Icing

Window icing has been countered by the installation of a de-icer heating
element to the outer surface of the window insert. The thin resistive ribbon
may be seen as a helical winding on the hard anodized surface of the assembly
shown in Fig. 4. It was estimated that a power input of about 60 watts would
be enough to de-ice this area with a small safety margin at 200 mph in air at
-15°C with a freezing water content of 5 gm per cubic meter. Such severe
conditions have not been encountered on any of the MRI flight tests and, with
such heating, no failures have been attributable to icing over of the window.
6. **Low Temperatures**

The coater system has been operated at temperatures down to -20°C without any apparent difficulty. The dip tank system worked down to -20°C when used in conjunction with a heater which raised the temperature of the solvent in the tank to about +5°C at a -20°C ambient.

7. **High Aircraft Speeds**

The coater system replicated quite reliably at speeds up to 170 mph indicated air speed, which was the maximum structurally allowable in the type aircraft which was used for these tests. Prior to the scaling off of the coater tank and the film supply magazine it was felt that high air speeds caused excessive air flow through the tank and over the freshly coated film. This flow, coupled with window heat, caused the concentration of the solution to increase rapidly as a function of flight time and the coating on the film to be skimmed over by a hardened film prior to exposure at the window. The result of the skinning over was a loss of replication except where the air stream ruptured the skin and exposed the fluid beneath. It is invariably observed that the thickness of the coating is irregular after exposure to the air stream. These irregularities are an increasing function of air speed. A large proportion of cloud particles are found in the five to fifteen micron range. In order to obtain accurate dimensional data from replicas of the larger particles, it is necessary to know the coating thickness. Therefore in the interests of obtaining a uniform coating it is preferable to sample at lower rather than higher flight speeds. It might also be mentioned that collection efficiency for the smaller particles is an increasing function of air speed.

8. **Blisters and Bubbles**

When too much heat is applied to the coated film in an attempt to accelerate the drying process the surface of the coating jells, further evaporation underneath this skin causes a blister to form and a spurious replica appears. In general these are easily recognized since the very irregular sizing and large volumes distinguish them from normal cloud particles. Blisters have never been a serious problem and have been treated as a symptom of incorrect drying conditions.

There is another type of false replica which has been observed on some flight records but not on records made on the ground or in the air with the window masked. These are small (large compared to droplets), irregular bubbles occurring more or less frequently but distinguishable from cloud particles in that when they occur they appear indiscriminately on given series of records without reference to cloud entry and cloud exit. While not completely explained it is thought that these are caused by air and water
contamination of the coating solution in the tank. This contamination may have resulted in flight from air leaks in the tank and supply magazine which induced a flow of air from the window. Consequent evaporation of the solvent in the tank produces cooling which causes condensation and the subsequent encapsulation of false droplets in the coating solution. In cloud the same flow carries additional contaminants into the tank, particularly when the film transport is stopped and not carrying rain drop and ice crystal fragments on up into the drying chamber. In general those false replicas are not frequently observed and stand out by virtue of their irregularity and presence both in and out of cloud. There are mechanical contrivances for trapping spurious water at a point before it can give the effects described above, whether the water is from impacted droplets or condensing from the vapor form.

The use of a narrow (in the film movement direction) window slot rather than a wide one appears to decrease spurious effects of bubbling and other deleterious effects associated with high air speeds. The slot width and film speed (often 5 to 20 cm/sec) are adjusted to keep the number of droplets collected, in clouds with the maximum liquid water contents, down to where flocculation or overlapping of droplets is statistically insignificant.
VI. SAFETY CONSIDERATIONS

There are no unusual hazards connected with the operation of this equipment but two important factors must be mentioned. First, although the drive motor is of good construction and well shielded, neither it nor the rest of the electrical system may be considered spark-proof, and therefore in order to eliminate the possibility of what could be quite a violent explosion it is recommended that a non-inflammable solvent such as chloroform be used. Secondly, since the chlorinated hydrocarbons and in particular chloroform are toxic and their effect is to some extent cumulative, the equipment should be mounted and vented in such a way that personnel are not exposed to sensible concentrations of vapor.
VII. JOINT NRL/MRI COMPARISON FLIGHT TESTS

Joint flight tests conducted in California were designed to study the comparative results of sampling clouds using various sampler configurations and aircraft. The aircraft used in the studies were a Navy S2UF operated by the Naval Research Laboratories and an Aero-Commander equipped with superchargers operated by Meteorology Research, Inc.

The Naval Research Laboratories retained all of their samples, obtained with their sampler mounted on their airplane. Meteorology Research, Inc. edited the samples obtained by the MRI aircraft and sent them immediately to Naval Research Laboratory. These samples are to be considered a portion of this report. Due to the fact that MRI had only cursory views of the samples there can be no detailed analysis of the results by them as an integral part of the report. Much of the value of the comparison flights was gained through the intimate cooperation of the two organizations during the field tests.

One of the major problems for study in the comparison tests was determining the cause of the pseudo droplets noted primarily by NRL on their sampler. Tests were conducted in which the Aero-Commander was flown at speeds between 100 and 250 miles per hour indicated air speeds. From these tests it appeared that the pseudo droplets were a product of higher air speeds. The sampling window of the sampler was then covered with tape and tested. There were then no pseudo droplets. This seemed to establish that the pseudo droplets were indeed the result of the changing air speeds. However, it is possible that changing angles of attack could affect the creation of these pseudo droplets by permitting the air stream to blow more readily up or down the main vertical tube after entering the window.

A number of flights were made through clouds at various temperatures and altitudes with the two aircraft flying tight formation. The comparison of these films is being handled by NRL.

Only the Aero-Commander had heating apparatus on its sampler. Therefore, the tests of the effects of heating could not be considered comparative. However, it was shown that the blushing was mainly controlled by the heat applied at the window. Further control of blushing was obtained by heating the first two feet of film as it was drying. After that the heating served mainly to hasten the drying of the solvent.

The result of the comparison tests may be summarized as follows: The technique of flying close formation through the clouds worked out well as long as there was good communication between the two aircraft. There should be one person on one of the aircraft who acts as the flight director for both aircraft. The occurrence of pseudo droplets was a function of air speed, not coating or type of aircraft used.
These flights were conducted from March 15, 1963 through March 26, 1963. The operations took place out of Ontario Airport, Ontario, California. Most of the flights were within a 10-mile radius of the airport. One was conducted above the San Francisco Bay area.
XIII. DROPLET DISTORTION

A. The Basic Problem

When an ice crystal impacts on the softened Formvar film, a thin skin of Formvar solution covers over the ice crystal, completely encapsulating the ice crystal in the Formvar solution. The solvent evaporates from the Formvar, leaving the Formvar in its hard, solid form. Subsequently the ice crystal will sublimate or melt and evaporate; in either case, the water comprising it will disappear, leaving a perfect, hollow, three-dimensional replica of the crystal in the Formvar.

If the impacting particle is a liquid droplet, the situation is much more complex because the droplet will distort. The Formvar will still encapsulate the droplet (probably in a millisecond for small droplets), the solvent will evaporate and leave a Formvar replica, the droplet will eventually disappear by evaporating through the thin Formvar skin. However, the exact shape the droplet assumes will depend on the original spherical droplet diameter, the thickness of the Formvar coating, and the surface tensions of the Formvar solution to air and to water. As the solvent evaporates, the Formvar coating thickness changes, and the surface tensions change as the Formvar solution gets denser (and the solution concentration will get denser more quickly in the thin skin over the droplet than in the main film). These varying factors will keep readjusting the droplet shape until the Formvar solution viscosity increases to the point where no further shape change will take place (the Formvar gets so viscous it is effectively solid).

To give some numbers to this, consider that in a typical case of coating with a 4 per cent Formvar-chloroform solution the initial solution thickness may be on the order of 150μ, while the final dry thickness is about 7 or 8μ. A droplet of 6μ diameter will always be completely under the flat surface, will remain always spherical, and the replica will be spherical. A droplet of 100μ diameter will initially be spherical in the 150μ thick film, but will distort as the solvent evaporation decreases the film thickness below 100μ. The distortion will get greater until the solution locally gets rather stiff.

In measurement one measures the resulting replica diameter, in order to infer from that the original spherical droplet diameter. It would obviously be virtually impossible to derive an exact mathematical solution for finding the relation between replica diameter and original droplet diameter. The course followed here is rather to make calculations based on a physical picture which is simple enough to permit calculation, and then to use the calculations to describe trends from empirical calibrations.
In the foregoing it has been assumed that the droplet and solvent do not merge with each other. As noted in Appendix A, actually the solvent may dissolve in the water to the extent of about one per cent of the water mass. Thus an indeterminacy in true original droplet mass can arise, but the amount is negligible compared to other errors associated with the measurement and correction of droplet diameters.

B. Computational Analysis

The calculation of the distortion of the droplet is made by assuming a very simple picture: the droplet, originally spherical with radius \( r \), falls into a Formvar solution of thickness \( h \), and is squashed to an elliptical cross section with semi-major axis \( a \) and semi-minor axis \( b \); the Formvar skin or cap which coats over the top of the droplet has radius \( c \). These relationships are shown in Fig. 1. In the actual case the droplet is flattened but not in such an ideal oblate spheroid shape, and the Formvar solution and Formvar cap skin have varying strengths as the solvent evaporates. Nevertheless, for simplicity the computations are based on the picture illustrated in Fig. 1, and it is assumed the computations will give the correct form of the relationships but that empirical data will be required to give quantitative points.

When the spherical droplet, with surface area \( S_S \) is distorted into the oblate spheroid shape, its area will be increased to \( S_E \) and energy

\[ \gamma_F/W (S_E - S_S) \]

will have to be added. \( \gamma_F/W \) refers to the surface tension of the Formvar solution with respect to water. The energy available to cause this distortion is derived from raising the Formvar skin which flows over the top of the droplet. The film which originally had area \( A_C \) is distorted into a cap which has area \( S_T \), and so the energy is

\[ \gamma_F/A (S_T - A_C) \]

\( \gamma_F/A \) refers to the surface tension of the Formvar solution with respect to air.

Equating the energies,

\[ \gamma_F/W (S_E - S_S) = \gamma_F/A (S_T - A_C) \]

or

\[ \frac{\gamma_F/A}{\gamma_F/W} = \frac{S_E - S_S}{S_T - A_C} \].
The ratio $\gamma_F/A/\gamma_F/W$ is given for various concentrations of Formvar-chloroform and Formvar-ethylene dichloride in Appendix B. The problem then is to find $(S_E - S_S)/(S_T - A_e)$ as a function of the desired $r$ and the measurable $h$ and $a$. Once this is done, a nomograph can be drawn up on which, for a given $h$ and a given Formvar solution concentration, $r$ vs. $a$ will be shown, i.e., original droplet size can be obtained from replica radius.

These computations were made, utilizing a computer to assist in the rather complex calculations required. Fig. 7 is the resulting nomograph. It must be reemphasized that the nomograph cannot be deemed quantitatively informative, due to the oversimplification of the picture on which it is based.

One result of the nomograph is to show that the correction factor $a/r$ does depend on the surface tension ratios $\gamma_F/A/\gamma_F/W$ but that for normal values of $\gamma_F/A/\gamma_F/W$ the dependence is not very strong. The interpretation of the nomograph also assists in understanding the results which will be described in the next section.

C. Absolute Calibration

The method chosen for a rough absolute calibration was an adaptation of the one employed by Farlow and French (1956). It provides for the simple and rapid determination of micro-drop volumes by the inclusion of uniform size solid particles in the water drops.

The principle involves the use of a suspension, in water, of solid particles of uniform size. If the suspension is vigorously stirred, any small droplet will contain the same concentration of particles as the suspension from which it originated. The calibration of droplet size with replica size was effected by first aspirating droplets from the stock suspension onto the coated film. The concentration of particles per unit volume in the stock solution was determined by means of a hemacytometer. The diameter of many replicas was measured and a count made of the number of solid particles each contained. The number of solid particles in replicas of the same diameter was averaged and the original volume of such a droplet thus computed. The density of the particles is greater than that of water but both are practically insoluble in chloroform Formvar solutions. The original volume of the droplet was taken to include that occupied by the particles.

In order to calibrate droplets in the range fifty down to ten microns in diameter it seemed most convenient to obtain a uniform particle in the region of one to two microns diameter which could be easily seen with standard optics but would yet be small enough to calibrate the smaller volumes. It was found that polystyrene latex balls were dissolved by the
chloroform solvent during the encapsulation. Some samples of alumina optical fining powder were sized by repeated time selective settling methods but their irregular shape and non-uniform size caused a wide statistical spread in apparent distribution due to the difficulty of resolving individual particles. Some fluorescent particles were also unsatisfactory for the same basic reason.

A survey of available natural particles led to the selection of the spores of the common mold, aspergillus niger. The particles were quite uniform and had a very convenient diameter of about three microns. A small quantity were grown. The spores were shaken from the mold and placed in suspension in distilled water. The stock solution was carefully drawn from this initial solution. The stock solution was vigorously stirred and maintained in the bath of an ultrasonic cleaner. Some of the stock solution was aspirated at the window of the coater system equipment. The pitot static differential at the window was equivalent to an air speed of about seventy miles per hour. Because of the small quantity of the stock solution only two test runs were possible, one run on a solution of 3-1/2 per cent Formvar and one on 6 per cent Formvar. The thickness of the dried coating of Formvar from these two runs was arrived at by averaging many readings of a micrometer screw gauge and also by deducing the thickness by gravimetric procedures. Both methods gave results which were in good agreement. A total of 320 counts were made for calibration of the stock solution and somewhat over a hundred for replica droplet volume. The small number of replicas counted was due to the poor yield of usable sizes due to the crudeness of the aspiration equipment and the small volume of calibration suspension produced. The probable error for computations of volume from single points on the presented curves must be considered to be slightly over 20 per cent based upon the mean deviations of the experimental data, however the apparently good fit of the curves indicate that this might be pessimistic. The results are presented on Fig. 8. The scope of this experimental calibration was to demonstrate the feasibility of this method. It does appear that a well thought out and rigorously executed development of this approach might yield quite precise results.

D. Interpretation of the Absolute Calibration

Consideration of Fig. 8 shows an important point, namely, that the curve for the 6 per cent solution and the curve for the 3.5 per cent solution are about the same when plotted in terms of \( r/h_f \) or \( a/h_f \) where \( h_f \) is the final dry thickness of the film. This plot has been made as Fig. 9. As would be predicted from the model discussed previously, for \( a/h_f < 0.5 \), the droplet is entirely below the surface and is undistorted, \( a/r = 1 \). For larger droplets there is distortion...
and $a/r > 1$. Both the 6 per cent curve (corresponding to $h_f = 23 \mu$) and the 3.5 per cent curve (corresponding to $h_f = 10 \mu$) are the same, to within the accuracy of the measurements on which they are based. One would hypothesize that, as a reasonable first approximation, the same relation holds for other Formvar solution concentrations.

The curves for various concentrations will be a unique function of $a/h_f$ only if the original wet solution thickness is of no importance. For the 6 per cent solution used here the original wet solution thickness is on the order of 400$\mu$, while for the 3.5 per cent solution it is on the order of 360$\mu$. The curves of Fig. 8 would not coincide if plotted vs. $a/h_0$, where $h_0$ is the starting film thickness. Apparently the final shape of the droplets is determined by droplet size and the Formvar solution details after the solution has evaporated until it is considerably more concentrated than 6 per cent. Therefore the initial concentration is of no concern (except for other problems such as film coating, complete encapsulation, flocculation, blushing, etc.).

When the calibration factors shown on Fig. 9 are compared with the nomograph of Fig. 7, derived from the simplified theoretical treatment, another factor becomes evident. For thick films, where $a/h_f$ is not much larger than one, the final distortion is in agreement with theory for rather low $\gamma_F/A/\gamma_F/W$ ratios -- ratios which imply a very thick solution from which almost all the solvent must have evaporated. This will of course come about because until the concentration is thick, the film thickness will exceed the spherical droplet diameter. For thin films (or large droplets) where $a/h_f$ is large, say 5 or 10, the final distortion is in agreement with theory for higher $\gamma_F/A/\gamma_F/W$ ratios -- ratios which imply a rather thinner solution. It is presumed in this second case that the Formvar cap gets stiff from evaporation, rather quickly; thus the shape and hence the calibration ratio $a/r$ is determined by the stiff cap while the main solvent film is still not very thick.

The mean curve of Fig. 9 can be considered as the basic calibration for chloroform solutions of any starting concentration. In a practical case, $h_f$ can be measured from the film and then Fig. 9 used to find $r$ for the values of $a$ encountered. In actual use, $h_f$ will be found to be rather constant, and so from Fig. 9 a family of curves resembling those of Fig. 8 can be derived. From this plot, $r$ can be read off directly from $a$ on the appropriate concentration line.

The absolute calibration has been done only for chloroform solutions. For ethylene dichloride, one calibration is available from MacCready (1962). For the droplets studied, $a = 15 \mu$, and $r = 10 \mu$, $a/r = 1.5$. The film thickness was not known accurately. It was estimated at 10$\mu$. If it had been 6$\mu$, this point would fit perfectly on Fig. 9. The ethylene
dichloride would be expected to have a somewhat different calibration than the chloroform, because of the differing surface tension ratios, but as implied by the theoretical calculations the effect should not be large.
IX. CONCLUSIONS AND RECOMMENDATIONS

The unique value of the continuous Formvar collection principle has been established over the last three years of development. In its most efficient form, it collects solid and liquid particles in a manner which permits relatively easy analysis. Some collection scheme such as this seems to be the only satisfactory approach to measuring small ice crystals on cloud physics research programs; the shape of the crystals is desired, as well as size (which must be defined with respect to shape) and concentration. For droplet measurements, the Formvar technique is good but some other optical method is a possibility. The optical method, light scattering or light shadow, measures the droplets in the air, avoiding some collection efficiency problems as well as avoiding droplet distortion and breakup on a surface; also in some cases the data acquisition is electronic and so further data reduction is unnecessary. The continuous collector is simpler than optical methods, and more convenient to carry in small vehicles. In summary, the measurement of ice crystals apparently requires the continuous collector, and the same device is effective for droplet measurements although competitive systems are a possibility.

The most basic problem with the continuous sampler is that, although in final form the device is relatively simple, it is based on many complex factors and interrelationships (nonlinear viscosity and surface tension variations in coating and encapsulations, various contradictory requirements for rapid evaporation of solvent without blushing or crystal melting, etc.). From the development studies it is apparent that all of these practical problems are solvable, because all have been solved at one time or another. The difficult matter is to find a compromise which solves them all simultaneously over a broad range of atmospheric situations with adjustments which are not impractically critical. For the optimum collection of droplets one configuration is best, while for the optimum collection of crystals, another collection is best. Collection of droplets and crystals simultaneously has been performed very effectively, and in the opinion of the authors the present type of apparatus is capable of doing this reliably over the desired meteorological range with a single instrument provided many more systematic field development tests are conducted.

Experience with the present units of course has created ideas of how future designs can be improved. One suggestion to make the design requirements less critical is to give the film two separate parallel coatings, one of a type optimized for droplet collection, the other optimized for crystal collection. It would appear that the technique of vapor softening of precoated film warrants further consideration. It should be more quantitative and controllable than the normal coating wheel or dipper tank methods, and may offer enough advantages to justify the additional development effort required. The sampler probe and slot may conveniently be altered to a configuration in
which the film moves around an idler wheel and the slot lets droplets impact during about 30° of the idler rotation. The film would not have to be curled, as it is in the present tubular configuration, and the collection efficiency could be slightly improved. One could not then employ the standard cylinder collection efficiency calculations, but new ones are derivable.

Any system, whether substantially new or merely an adaptation of the present one, requires a series of systematic tests at various air speeds and ambient temperatures with various solution concentrations, with various amounts of drying heat, for collecting both crystals and droplets. A final version, if made to cover a wide meteorological range, will have controllable heaters (initially controlled manually, later automatically). The number of interrelated variables means final development must be by empirical testing. The tests to date have shown that operation of the collector becomes more difficult as the speeds get above about 150 knots because the sort of Formvar film which collects and replicates best is not so effective in minimizing blowoff. For higher speeds it thus seems that the lower speed unit should be used with a diffuser system which locally slows the air flow impinging on the collector. The diffuser is aerodynamically simple, to design, but it will complicate the calculation of collection efficiency.

Counting and sizing methods have not been treated in this report because they are secondary to the problem of obtaining good replicas. The simple counting techniques associated with projecting the film by a stop-motion 16 mm projector and counting by hand have worked well. A fair qualitative measure of particle numbers, sizes, and types can be found surprisingly quickly by this method. In fact, even movie projection at 16 frames per second conveys significant information. Quantitative measurements take only a few minutes per frame, a time which is so short that the development of automatic techniques to cut down the time is hardly justified now.

The absolute calibration technique described in this report shows that one of the primary questions pertaining to this collector method has been solved in principle. More detailed calibrations using the same method should be made, including tests at various temperatures and Formvar concentrations. It would be satisfying to make some basic microscopic studies of the true physical shape of the distorting droplets, but it appears such studies may not be needed for practical application of the system.

In summary, the continuous particle sampler in its present form produces unique and useful data unobtainable by any other method. More engineering development is required before the device can be easily applied to a broad variety of meteorological situations.
X. ACKNOWLEDGMENTS

Many people and organizations have participated actively in the development of the continuous droplet sampler. Mr. Clement J. Todd of MRI and ARG is the originator of the basic instrument and has been the key person in its continued development. Mr. J. A. K. Lake, Mr. Ronald Sloane, Mr. Edward Hindman, and Mr. Harold Hutchinson, all of MRI, contributed significantly to the present contract. Mr. Lake served as Project Director during the early phases of the work; Mr. Sloane handled the computer program associated with the theoretical calculations and did much of the laboratory work on materials; Mr. Hindman aided in the laboratory investigations, especially the evaporation-absorption work; Mr. Hutchinson provided the viscosity and wick-coating data. Mr. Williamson, co-author of this report, completely handled the absolute calibration procedure and the design of the latest mechanical unit.

The sponsorship of NRL is gratefully acknowledged. Mr. Robert Ruskin of NRL has been particularly helpful in the cooperative phases of the work. The program has been materially aided by concurrent sponsorship of cloud physics studies using and developing this type of sampler: support to MRI from the U. S. Army Electronics Research and Development Laboratory (with the support of the Advanced Research Projects Agency), and support to ARG by the Atmospheric Sciences Section of the National Science Foundation.
REFERENCES


Fig. 4. CLOSEUP OF WINDOW AND HEATER
Fig. 5. SAMPLER INSTALLATION IN APACHE, 1963

TOP RIGHT - DROP SAMPLER ON APACHE

TOP LEFT - SPECIAL PROJECTOR SYSTEM IN APACHE SHOWING CONTROL BOX AND MAGAZINE INSTALLATION

LOWER LEFT - APACHE NOSE INSTALLATION ON FRONT BULKHEAD WITH FAIRINGS REMOVED
Fig. 6. SIMPLIFIED MODEL OF THE DISTORTED DROPLET.

The left side of the picture gives the case for \( h > b \), while
the right side of the picture gives the case for \( h < b \). For \( h > 2r \)
the droplet is entirely beneath the flat Formvar surface, \( c = 0 \),
and the droplet remains spherical so \( a = b = r \). The dashed lines
show a more realistic shape of the droplet and Formvar solution,
but a shape which is more difficult to treat mathematically.
Example:
\[ \frac{\delta F_A}{\delta F_t} = 0.3 \]
and \( \frac{h}{2a} = 0.35 \)

Fig. 7. SIMPLIFIED THEORETICAL DROPLET DISTORTION NOMOGRAPH.
**Fig. 8. DISTORTION CALIBRATION WITH ASPERGILLUS NIGER SPORES.**
Fig. 9. CALIBRATION FACTOR RELATED TO DRY FORMVAR THICKNESS.

\[ \frac{a}{r} = \text{replica radius} \]
\[ r = \text{droplet radius} \]
\[ h_f = \text{final dry formvar thickness} \]
For convenience, certain properties of the materials involved in the continuous particle sampler are collected here. The data sources are primarily Lange's "Handbook of Chemistry" and "Properties and Uses of Butvar and Formvar", Shawinigan Resins Corp., 1963 (Springfield 1, Mass.). Figure A 1 summarizes the vapor pressure-temperature curves for chloroform, ethylene dichloride, and water. The curves emphasize the fact that the chloroform is much more volatile than the ethylene dichloride (by a factor of $2-1/2$ or 3), a fact which is particularly important in the evaporation of solvent at cold temperatures.

<table>
<thead>
<tr>
<th></th>
<th>Chloroform ($\text{C}<em>\text{H}</em>\text{Cl}_3$)</th>
<th>Ethylene Dichloride ($\text{C}_\text{2H}_4\text{Cl}_2$)</th>
<th>Water ($\text{H}_2\text{O}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melting Point</td>
<td>-63.5°C</td>
<td>-35.5°C</td>
<td></td>
</tr>
<tr>
<td>Boiling Point</td>
<td>61.26°C</td>
<td>83.5°C</td>
<td></td>
</tr>
<tr>
<td>Density (Room Temp.)</td>
<td>1.47</td>
<td>1.25</td>
<td>1.00</td>
</tr>
<tr>
<td>Specific Heat (cal/gm)</td>
<td>0.232 at 0°C</td>
<td>0.301 at 20°C</td>
<td>1.00 at 15°C</td>
</tr>
<tr>
<td></td>
<td>0.226 at 15°C</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.234 at 20°C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Heat of Vaporization/Condensation (gm cal/gm)</td>
<td>64.7 at 0°C</td>
<td>65.3 at 0°C</td>
<td>584.9 at 20°C</td>
</tr>
<tr>
<td>Solubility (in 100 gm of water at 15°C)</td>
<td>1.0 cc</td>
<td>0.92 cc</td>
<td></td>
</tr>
<tr>
<td>Viscosity (millipoise)</td>
<td>7.86 at -10°C</td>
<td>11.322 at 0°C</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6.99 at 0°C</td>
<td>8.385 at 20°C</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.63 at 20°C</td>
<td>6.523 at 40°C</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4.64 at 40°C</td>
<td>4.357 at 80°C</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3.89 at 60°C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Surface Tension (dyne/cm) of Material-Air Interface</td>
<td>28.5 at 10°C</td>
<td>33.6 at 10°C</td>
<td></td>
</tr>
<tr>
<td></td>
<td>27.1 at 20°C</td>
<td>32.2 at 20°C</td>
<td></td>
</tr>
<tr>
<td></td>
<td>21.7 at 60°C</td>
<td>24.0 at 80°C</td>
<td></td>
</tr>
</tbody>
</table>

Viscosity and surface tension of the mixtures of Formvar in the two solvents are discussed in other appendices.
The leader film on which the Formvar solution is placed presumably has a specific heat in the 0.3 to 0.5 cal/gm range. Other properties are listed in the main body of the report. This material is Cronar, P-40A Leader, Polyester Photographic Film Base, obtained from duPont, Wilmington, Delaware.

The Formvar used for the collector is type 15-95E. Specific gravity is listed at 1.23 (MRI measurements, 1.25). Viscosity of 5 per cent solution of Formvar-ethylene dichloride at 20°C was determined by Shawinigan Resins as 40 - 60 centipoise with an Oswald Viscometer.
Fig. A1. VAPOR PRESSURES.
SURFACE TENSIONS

The surface tension of the liquid water droplet to the Formvar solution, and the surface tension of the interface of air to the Formvar solution are both fundamental quantities in determining the final distortion shape of the droplet and hence the replica diameter. Since these quantities were not available from literature, experiments were set up for their measurement. It turns out that the most pertinent information for droplet distortion comes from measurements at high concentrations of the Formvar-solvent solutions, but the measurements are difficult at the high viscosities encountered. The results given here are for 10 per cent and thinner concentrations.

The air-Formvar surface tension was obtained by the standard Du NoUy ring method. The details of the method are described in "Interfacial Phenomena" by J. T. Davies and E. K. Rideal, Academic Press, 1961.

The variation of surface tension with temperature was experimentally verified as being small (the temperature variation of surface tension for the solvent without Formvar to air is shown in Appendix A). Therefore, for convenience, all the surface tension tests were conducted at normal temperatures (20°C to 25°C). Each reading is the average of five tests. Reproducibility was good.

Table I lists the values of \( \gamma_{F/A} \) (surface tension in dyne/cm for the Formvar-chloroform and Formvar-ethylene dichloride solution to air interface).

<table>
<thead>
<tr>
<th>Per Cent Solution</th>
<th>Chloroform ( \gamma_{F/A} ) dyne/cm</th>
<th>Ethylene Dichloride ( \gamma_{F/A} ) dyne/cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>0%</td>
<td>27.25</td>
<td>31.2</td>
</tr>
<tr>
<td>1</td>
<td>27.35</td>
<td>30.8</td>
</tr>
<tr>
<td>2</td>
<td>27.49</td>
<td>31.5</td>
</tr>
<tr>
<td>3</td>
<td>27.77</td>
<td>31.4</td>
</tr>
<tr>
<td>5</td>
<td>27.31</td>
<td>31.8</td>
</tr>
<tr>
<td>10</td>
<td>29.46</td>
<td>32.0</td>
</tr>
</tbody>
</table>

A plot of the above data will show slope changes which would be somewhat unexpected in natural data, and one might assume these changes to be...
related to the experimental technique. In any case, the surface tension is very constant over the range of solutions. The values are in close agreement with the 0 per cent values reported for 20°C in Lange's "Handbook of Chemistry": 27.1 dyne/cm for chloroform, vs. 32.2 dyne/cm for ethylene dichloride.

The water-Formvar solution surface tension was obtained by the drop rise method, the upward analog to the downward drop-weight method as described in Davies' and Rideal's "Interfacial Phenomena". Table II lists $\gamma_{F/W}$, the results for the water to Formvar-chloroform and Formvar-ethylene dichloride solution interface.

**TABLE II**

<table>
<thead>
<tr>
<th>Per Cent Solution</th>
<th>Chloroform $\gamma_{F/W}$ dyne/cm</th>
<th>Ethylene Dichloride $\gamma_{F/W}$ dyne/cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>0%</td>
<td>32.7</td>
<td>30.6</td>
</tr>
<tr>
<td>1</td>
<td>20.6</td>
<td>16.3</td>
</tr>
<tr>
<td>2</td>
<td>19.9</td>
<td>16.1</td>
</tr>
<tr>
<td>3</td>
<td>21.3</td>
<td>18.0</td>
</tr>
<tr>
<td>5</td>
<td>30.2</td>
<td>20.1</td>
</tr>
<tr>
<td>10</td>
<td>63.6</td>
<td>41.5</td>
</tr>
</tbody>
</table>

The same measurement was attempted with the Du Nolyl ring method, with water overlying the Formvar solution, but this technique proved impossible to make sufficiently quantitative. The tests for chloroform did duplicate the minimum for the 2 per cent solution, but gave values which were between 10 and 80 per cent too high.

The surface tension values are involved in computations in terms of the ratio of surface tensions,

$$\frac{\gamma_{F/A}}{\gamma_{A/W}}$$

Values of this ratio are plotted on Fig. B1.
Fig. B1. SURFACE TENSION RATIOS
APPENDIX C

VIScosity MEASUREMENTS AT VARIOUS CONCENTRATIONS

Introduction

The viscosity of the Formvar solution is an important factor in permitting or preventing flocculation, in minimizing spreading or blow-off of the Formvar film material, in determining the thickness of the film deriving from a particular coating wheel configuration, and in determining the solution concentration at which the droplet distortion becomes fixed. Therefore measurements of viscosity for various solutions and temperatures were made. None of these data were available from literature.

Measurements with Ethylene Dichloride

The viscosities of several concentrations of Formvar dissolved in ethylene dichloride were measured at various temperatures using Cannon-Finske viscometers per ASTM Method D455-60. The Cannon-Finske viscometer consists of a glass bulb reservoir connected to a glass capillary tube. The time required for the reservoir to empty through the capillary tube is proportional to the viscosity of the liquid. Quantitative results are obtained by determining the calibration constant of the viscometer and using equation (1).

$$\text{Kinematic viscosity (in centistokes, cs)} = Ct$$

- $C = \text{calibration constant in cs/sec}$
- $t = \text{efflux time in sec}$

Kinematic viscosity values can be converted to Saybolt Universal Seconds as shown in Table I.

TABLE I

CONVERSION OF KINEMATIC VISCOSITIES TO SAYBOLT UNIVERSAL SECONDS AT 100°F

<table>
<thead>
<tr>
<th>Kinematic Viscosity (cs)</th>
<th>Saybolt Universal Seconds</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>45.6</td>
</tr>
<tr>
<td>8</td>
<td>52.1</td>
</tr>
<tr>
<td>10</td>
<td>58.9</td>
</tr>
<tr>
<td>20</td>
<td>97.6</td>
</tr>
<tr>
<td>30</td>
<td>141.3</td>
</tr>
<tr>
<td>40</td>
<td>186.3</td>
</tr>
<tr>
<td>50</td>
<td>232.1</td>
</tr>
<tr>
<td>60</td>
<td>278.3</td>
</tr>
<tr>
<td>70</td>
<td>324.4</td>
</tr>
</tbody>
</table>
Note: Values between those listed can be obtained by linear interpolation.

For kinematic viscosities above 70 cs, equation (2) can be used at 100°F.

\[
\text{Saybolt universal seconds} = \text{kinematic viscosity (cs)} \times 4.635 \tag{2}
\]

The constant multiplier has the value 4.667 at 210°F. Since the variation with temperature is small, the values listed in Table I and the constant of equation (2) will be used at all temperatures in the range -4°F to 120°F.

An uncalibrated viscometer can be calibrated by comparing with a calibrated unit at a given temperature and fluid. Both units are filled with the same fluid and placed in the same water bath and the individual efflux times measured. The unknown calibration constant is given by equation (3).

\[
C_x = \frac{t_c C_c}{t_x} \tag{3}
\]

- \(C_x\) = constant of uncalibrated viscometer
- \(t_x\) = efflux time of uncalibrated viscometer
- \(C_c\) = constant of calibrated viscometer
- \(t_c\) = efflux time of calibrated viscometer

One calibrated viscometer was purchased (Viscometer No. 200/L134, Cannon-Fenske-Ostwald Type) with which to calibrate three unknown units. The constant varies slightly with temperature, e.g., 0.0923 cs/sec at 100°F and 0.0919 cs/sec at 210°F. This variation was neglected as too small to affect significantly our results and, therefore, calibration data were taken at various temperatures in the range 50°F to 110°F without correction for temperature.

The efflux time should always be \(\geq 200\) sec to avoid errors introduced by timing of the meniscus passage past the fiducial marks and turbulence of the fluid in the capillary tube. This condition was not always met since the number of viscometers was limited to four (4) to cover the whole viscosity range of interest. However, all data were consistent and produced smooth curves indicating negligible error was introduced by using efflux times less than 200 sec.

Density measurements were made at 70°F for the different concentrations of Formvar-ethylene dichloride. For solutions from 0 per cent to 7 per cent the density is 1.25 gm/cc; for the 10 per cent solution the density is 1.26 gm/cc.

The viscosity for various concentrations and various temperatures was measured with the four viscometers after the calibration constants had been established. The results in three units are given in Table II.
### TABLE II

<table>
<thead>
<tr>
<th>Formvar Concentration (%)</th>
<th>Temp. (°F)</th>
<th>Centistokes</th>
<th>Centipoises</th>
<th>Saybolt Universal Seconds</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>56.0</td>
<td>0.73</td>
<td>0.91</td>
<td>~28.8</td>
</tr>
<tr>
<td>1</td>
<td>40.0</td>
<td>2.95</td>
<td>3.68</td>
<td>36.0</td>
</tr>
<tr>
<td>1</td>
<td>64.0</td>
<td>2.21</td>
<td>2.76</td>
<td>33.6</td>
</tr>
<tr>
<td>1</td>
<td>82.5</td>
<td>1.87</td>
<td>2.34</td>
<td>~32.5</td>
</tr>
<tr>
<td>1</td>
<td>94.4</td>
<td>1.41</td>
<td>1.76</td>
<td>~31.1</td>
</tr>
<tr>
<td>2</td>
<td>39.0</td>
<td>8.05</td>
<td>10.1</td>
<td>52.2</td>
</tr>
<tr>
<td>2</td>
<td>70.0</td>
<td>5.49</td>
<td>6.86</td>
<td>44.0</td>
</tr>
<tr>
<td>2</td>
<td>109.5</td>
<td>3.86</td>
<td>4.83</td>
<td>38.9</td>
</tr>
<tr>
<td>3</td>
<td>32.0</td>
<td>30.2</td>
<td>37.8</td>
<td>141.0</td>
</tr>
<tr>
<td>3</td>
<td>45.0</td>
<td>22.4</td>
<td>28.0</td>
<td>109.0</td>
</tr>
<tr>
<td>3</td>
<td>70.0</td>
<td>15.2</td>
<td>19.0</td>
<td>77.5</td>
</tr>
<tr>
<td>3</td>
<td>94.0</td>
<td>11.4</td>
<td>14.3</td>
<td>64.1</td>
</tr>
<tr>
<td>3</td>
<td>109.5</td>
<td>9.6</td>
<td>12.0</td>
<td>56.9</td>
</tr>
<tr>
<td>5</td>
<td>41.0</td>
<td>113.0</td>
<td>141.0</td>
<td>524.0</td>
</tr>
<tr>
<td>5</td>
<td>64.0</td>
<td>69.6</td>
<td>87.0</td>
<td>323.0</td>
</tr>
<tr>
<td>5</td>
<td>81.9</td>
<td>51.4</td>
<td>64.2</td>
<td>239.0</td>
</tr>
<tr>
<td>5</td>
<td>118.5</td>
<td>30.4</td>
<td>38.0</td>
<td>142.0</td>
</tr>
<tr>
<td>7</td>
<td>54.0</td>
<td>232.0</td>
<td>292.0</td>
<td>1080.0</td>
</tr>
<tr>
<td>7</td>
<td>74.1</td>
<td>157.0</td>
<td>196.0</td>
<td>729.0</td>
</tr>
<tr>
<td>7</td>
<td>86.0</td>
<td>126.0</td>
<td>157.0</td>
<td>585.0</td>
</tr>
<tr>
<td>7</td>
<td>99.0</td>
<td>101.0</td>
<td>126.0</td>
<td>469.0</td>
</tr>
<tr>
<td>10</td>
<td>45.0</td>
<td>1730.0</td>
<td>2160.0</td>
<td>8020.0</td>
</tr>
<tr>
<td>10</td>
<td>63.0</td>
<td>992.0</td>
<td>1240.0</td>
<td>4600.0</td>
</tr>
<tr>
<td>10</td>
<td>81.0</td>
<td>703.0</td>
<td>878.0</td>
<td>3260.0</td>
</tr>
<tr>
<td>10</td>
<td>115.7</td>
<td>357.0</td>
<td>446.0</td>
<td>1660.0</td>
</tr>
</tbody>
</table>

These results, in Saybolt Universal Seconds units, are presented on Fig. C1. Figure C1 is plotted on ASTM Viscosity-Temperature graph paper (see ASTM Standards, Part 7, Standard Viscosity-Temperature Charts for Liquid Petroleum Products, D341-43). The coordinate lines are arranged such that petroleum oils appear as straight lines, thus allowing one to obtain a complete curve from only two viscosity measurements at two different temperatures. All of the Formvar data points lie along straight lines, therefore the Formvar solutions are assumed to "act" like petroleum oils over a limited temperature range, e.g., 0°F to 130°F. Measured points and the resulting straight lines in which Formvar concentration is a parameter are shown in Fig. C1. Using these curves it is possible to produce a second set of data points and curves in which temperature is the parameter. This has been done, and the results are shown in Fig. C2.
Shawinigan Resins Corp. lists the viscosity of 5 per cent Formvar-ethylene dichloride at 20°C as 40 - 60 centipoise. Their 5 per cent solution is actually 5/105 or 4.75 per cent. From Fig. C2, and the conversion of Saybolt Universal Seconds to centipoise, the present measurements give 58 centipoise, within the Shawinigan range. Lange (1956) lists viscosity for ethylene dichloride alone, and the measured value in Table II is in agreement. The Shawinigan manual plots viscosity variations for various concentrations of Formvar - 60/40 Toluene-Ethanol (95 per cent). The Shawinigan curves and Fig. C2 both show the viscosity increasing by a factor of 10 from 5 to 10 per cent concentrations.

Chloroform Solutions

The viscosity of Formvar-chloroform solutions was not measured because it was felt that to the accuracy required this could be estimated from available data. As shown in Appendix A, the viscosity of chloroform is about two-thirds that of ethylene dichloride over a sizeable temperature range. For Formvar-chloroform solutions, therefore, it is assumed that the viscosity is somewhere between two-thirds and one times the value for the same Formvar-ethylene dichloride concentration as shown on Fig. C1 or C2.

The density of dilute chloroform solutions is 1.47, lowering to 1.45 for a 10 per cent solution.

Reference

Fig. C 1. VISCOSITY VS. TEMPERATURE FOR VARIOUS FORMVAR CONCENTRATIONS IN ETHYLENE DICHLORIDE.
Fig. C.2. FORMVAR CONCENTRATION VS. VISCOSITY.
Introduction

One approach to the continuous particle collector is to use precoated film and soften the Formvar coating by passing the film through a chamber containing solvent vapor. This method would be the continuous analog to the vapor method presented by Schaefer (1962). Hindman (1963) has developed the continuous vapor method to capture particles settling by gravity. The expected virtues of the system are a) the method avoids bulk liquids in the softening area and so should minimize the presence of bubbles and also decrease the sticking of the film from solvent-Formvar dripping, and b) the technique should aid in making the Formvar softening more quantitative and controllable. The main disadvantage might be the difficulty of getting the Formvar softened quickly enough for a continuously operating system. To examine this potential problem, some crude experiments were set up to measure the rate at which chloroform (the fastest acting of the available solvents) would soften the precoated Formvar film. The experimental setup also afforded the opportunity for measuring (again crudely) the rate at which the solvent evaporates from the film, and so these tests were also made.

Measurement Techniques

The apparatus consisted of a sensitive analytical balance to measure the amount of vapor in grams, a cold chamber to sample or expose the different temperatures, and a vapor chamber (a container with chloroform-soaked blotters) to keep the environment saturated with respect to CHCl₃ vapor. The vapor chamber was a pint thermos bottle, with a centimeter of liquid chloroform in the bottom and corrugated cardboard up the inside to conduct the solvent throughout the chamber. The experimental technique was finally standardized to the following:

1. The Formvar is applied onto 16 mm film as uniformly as possible.

2. The film is weighed before and after the Formvar is applied in order to determine the amount of Formvar. (All the tests are done by weight, so film length is immaterial. Film lengths of about 3 cm were used.)

3. The exposing of the film to vapor is executed by bringing the vapor chamber up under the film and then noting the rate of
absorption by keeping the balance at zero by adding weight. The best way to note the absorption rate is to set the balance 0.002 gm too heavy and note the time for this weight to be attained. This pre-weighting is continued by adding weight in 0.002 gm units until equilibrium is reached. This will then tell how much vapor was absorbed and how long it took.

The evaporation rate of the vapor can be determined by setting the scale 0.002 gm too light, withdrawing the vapor chamber, and noting the time for that weight of solvent to be evaporated. Continue lightening the weight by 0.002 gm amounts until equilibrium is reached. Equilibrium should be near the initial weight of the Formvar (dry) plus film. The plotted results for evaporation show considerable scatter, but they are sufficiently consistent for meeting the aims of the experiment.

The absorption tests might more properly be called condensation tests. The solvent does end up within the Formvar material, but whether it gets there directly from the vapor form or first condenses on the surface is not known. Presumably the mechanism involves condensation when the film is relatively cold and the vapor is removed from the air quickly.

Results

Figures D1, D2, and D3 give the results of the measurements. They all pertain to precoated film made with a coating wheel and a Formvar-ethylene dichloride solution. Figure D1 is for a one per cent solution, while D2 is for a 5 per cent solution. The actual dry Formvar thickness varies somewhat with the solution viscosity during coating, and so varies nonlinearly with the percentage solution, and varies with the temperature because of the viscosity-temperature effect. Typically, a 5 per cent solution will dry to a thickness on the order of 10µ. The maximum vapor absorbed has a weight of about three or four times the dry Formvar. For the experiments discussed here, short lengths of film were used (bare uncoated film weight about 0.21 gm). The one per cent coating was about 0.0035 gm, and absorbed a maximum of about 0.013 gm of chloroform vapor (see Fig. D1); the 5 per cent coating was three times as thick, about 0.01 gm and absorbed about 0.03 gm (see Fig. D2).

The absorption tests involved surrounding the dry film, which had reached 20°C in the cold box, with a solvent vapor atmosphere at another warmer temperature, say 0°C (for the evaporation tests the vapor chamber was removed and the soft coated film exposed again to the ambient cold box temperature). During the first few moments the temperature difference, 20°C in this case, would cause rapid condensation (or absorption) but then the film would heat (due both to condensation and conduction and perhaps some slight convection) until the absorption rate became slow. The initial rate can
be estimated from Figs. D1 and D2, especially well from D1. Referring to Fig. D1, presumably the heat of condensation provides the major limitation in the first 30 seconds or so. The rate is roughly linear to that point. The initial rates are not directly proportional to the vapor pressure difference between the ambient and film temperature. For example, the rate for the 12°C chamber (ΔT = 32°C) is only twice the rate for the -11°C chamber (ΔT = 9°C). After about 30 seconds the rates of absorption are linear, with a different slope, and are about the same for all starting temperatures. This implies that the films are then all at the same temperature, say on the order of a degree warmer than the ambient vapor. As might be expected, the initial rates for the 5 per cent Formvar coating of Fig. D2 are only slightly greater than the corresponding rates for the one per cent Formvar coating of Fig. D1. For the 5 per cent case the total absorbed is of course greater than for the thinner film. On Figs. D1 and D2 the "Average Percentage Solution Throughout Film" is given as an alternate ordinate scale. It can be noted that the thin film, Fig. D1, gets much more quickly to a particular average thinness of concentration.

Thus, in terms of softening, the film from the one per cent Formvar softens more quickly than the thicker film from the 5 per cent Formvar, an effect which has been noted qualitatively in tests with droplet collection using vapor softening of Formvar.

The vapor method of softening presumably provides a gradation of percentage solution across the Formvar film, with softer (or more liquid) film in the outer layers. For the Formvar solution to flow over a droplet, the solution may have to be as dilute as 15 or 20 per cent, which dilution must therefore be exceeded at least on the outer layer of the Formvar.

In summary, vapor softening seems to be feasible in some cases, particularly with very thin Formvar coatings. These thin coatings can be expected to give nicely detailed replicas. The vapor source must be kept considerably warmer than the film in order to have sufficiently rapid absorption. If the dry Formvar is 1 μ thick, for a steady 30°C temperature differential, perhaps 30 seconds of absorption would be adequate (the average concentration might be about 25 per cent, but the surface might be sufficiently soft). This is an inconveniently long time for a continuous airborne sampler, but not impossible. A thinner Formvar coating would soften more quickly, but would also harden so quickly in the air blast during collection that it might prove impractical. Absorption would be fastest with a high absolute temperature for both film and solvent vapor, and would presumably be speeded by a forced draft.

Figure D3 shows the Formvar hardening (solvent evaporation) from the same films used in the absorption tests. The most interesting part of the curves is the portion pertaining to short time intervals. Unfortunately there are no data at short time intervals. However, the evaporation setup was an exact reverse analog to the adsorption experiment, and the curves on Fig. D3
are almost mirror images of Figs. D1 and D2. Thus the short period data of the absorption curves can logically be applied to the evaporation estimates. For the thicker Formvar, the total evaporation is greater, but the relative hardening is slower.

In conclusion, for a continuous airborne collector some heating appears necessary for operation at cold temperatures to harden the film before it reaches the take-up reel. Forced ventilation would speed the hardening. The vapor must be removed continually to afford maximum hardening.

All these tests were made with chloroform. With ethylene dichloride the absorption and evaporation would all proceed much slower.

References


Fig. D1. CH\textsubscript{3}Cl \textit{VAPOR ABSORPTION INTO FORMVAR (1%)}
Fig. D2. CHCl₃ VAPOR ABSORPTION INTO FORMVAR (5%)
WICK DESIGN CRITERIA

This appendix summarizes investigations which were carried out in support of a wick coating system. A large portion of the following information was taken from the "American Institute of Physics Handbook" (1957). The subject covered is fluid-flow properties of porous media.

List of symbols:

- \( P \) Percentage porosity
- \( K \) Permeability
- \( v \) Volume of fluid crossing unit area per unit time
- \( \eta \) Shear viscosity
- \( K' \) \( K/\eta \)
- \( R \) Reynolds number
- \( p \) Fluid pressure

Percentage porosity \( P \) is a measure of the fluid capacity of a porous medium. It is defined as the percentage volume of voids per unit total volume. In dealing with the flow properties of a porous medium one is concerned with the percentage porosity actually available during flow, i.e., the relative amount of interconnected pore space. Therefore, the available porosity (Table I) may be somewhat less than the total porosity calculated from the density of the medium.

Permeability \( K \) is a measure of the ease with which a fluid flows through a porous medium under the influence of a pressure gradient. It is defined from the empirical relation known as Darcy's Law in the following way: If \( v \) is the volume of fluid crossing unit area per unit time under the pressure gradient \( \frac{dP}{dx} \), for small values of \( v \), one finds empirically that

\[
K' \frac{dP}{dx} = v
\]  

(1)

where \( K' \) is a constant dependent on both the fluid and the medium. It is found further that the constant \( K' \) can be written as \( K' = K/\eta \), where \( \eta \) is the coefficient of shear viscosity of the fluid, and \( K \) is by definition of the permeability. Defined in this way, \( K \) is practically independent of the properties of the fluid and depends only upon the character of the porous structure.
If a Reynolds number is defined as \( R = \frac{\nu L}{\mu} \) (where \( \mu \) is the fluid density and \( L \) is a length characteristic of the porous structure, such as the average pore size, it is found empirically that Darcy's Law as given above holds for \( R \) less than about 5.

Many attempts have been made to calculate permeabilities in terms of more fundamental properties of the medium, but in general this has not met with success, and \( K \) is usually looked upon as a parameter which can be known only by direct measurement. Consequently a porous structure will have a permeability of one darcy if for fluid of one centipoise viscosity the volume flow is 1 cc per square centimeter area under a pressure gradient of 1 atmosphere per centimeter. Permeability measured in darcys can be converted to a self-consistent cgs system of units by the relation

\[
1 \text{ darcy} = 9.8697 \times 10^{-9} \frac{\text{poise}}{\text{(centimeter per second)}} = \frac{\text{poise}}{\text{(dynes per cm}^2\text{)}}.
\]

### TABLE I

**TYPICAL FLOW PARAMETERS OF SOME POROUS MATERIALS**

<table>
<thead>
<tr>
<th>Material</th>
<th>% Porosity</th>
<th>Permeability, darcys</th>
</tr>
</thead>
<tbody>
<tr>
<td>Graded sand: 30-40 mesh</td>
<td>40</td>
<td>345</td>
</tr>
<tr>
<td>40-50 mesh</td>
<td>40</td>
<td>66</td>
</tr>
<tr>
<td>50-60 mesh</td>
<td>40</td>
<td>44</td>
</tr>
<tr>
<td>60-70 mesh</td>
<td>40</td>
<td>31</td>
</tr>
<tr>
<td>70-80 mesh</td>
<td>40</td>
<td>26</td>
</tr>
<tr>
<td>Fine heterogeneous sands</td>
<td>30 - 35</td>
<td>1 - 10</td>
</tr>
<tr>
<td>Silts</td>
<td>35 - 45</td>
<td>5 - 180</td>
</tr>
<tr>
<td>Fine powders</td>
<td>35 - 70</td>
<td>0.01 - 0.1</td>
</tr>
<tr>
<td>Sandstones</td>
<td>10 - 20</td>
<td>0.01 - 1.0</td>
</tr>
<tr>
<td>Acoustic absorbing materials</td>
<td>90 - 95</td>
<td>35 - 180</td>
</tr>
<tr>
<td>Hair felt</td>
<td>95</td>
<td>900</td>
</tr>
</tbody>
</table>

Note: The permeability of a given type of porous substance varies widely depending upon such factors as the degree of cementation and the nature of the interconnections between pores, and so the values quoted above represent only typical values that have been reported in the literature.

Equation (1) can be used to obtain the pressure required to produce the desired flow. It is assumed that a coating 10 microns thick by 1 cm wide is applied to the 16 mm film base at a linear speed of 7.62 cm/sec (3 in/sec). This requires a volume flow of 7.62 \( \times 10^{-3} \) cc/sec. Assuming a wick cross
section of 1 cm x 0.6 cm provides a wick volume flow (v) of 3.81 x 10⁻⁴ cc/cm²/sec at room temperature and pressure. The required pressure is given by

\[ dP = \frac{dx \cdot v}{K} \]  \hspace{1cm} (2)

\[ dP = \frac{dx \cdot \eta \cdot v}{K} \]  \hspace{1cm} (3)

substituting

\[ dx = 1 \text{ cm} \]
\[ \eta = 0.5 \text{ cp (1\% Formvar solution at stp)} \]
\[ v = 3.8 \times 10^{-4} \text{ cc/cm}^2/\text{sec} \]
\[ K = 900 \text{ darcys (Table I)} \]

gives

\[ dP = 21 \text{ dynes/cm}^2 \]  \hspace{1cm} (3.1 \times 10^{-4} \text{ psi}).

This, using a density of 1.25 gm/ml for a 1 per cent Formvar solution, converts to a pressure head height of 0.017 cm. Pressure head heights required to maintain a constant flow of 3.8 x 10⁻³ cc/cm²/sec for various Formvar concentrations at stp are listed in Table II.

<table>
<thead>
<tr>
<th>Formvar Concentration (wt)</th>
<th>Viscosity (cp)</th>
<th>( dP ) (atmos.)</th>
<th>Height Solution (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1%</td>
<td>0.5</td>
<td>2.1 x 10⁻⁵</td>
<td>0.017</td>
</tr>
<tr>
<td>5</td>
<td>30</td>
<td>1.3 x 10⁻³</td>
<td>1.0</td>
</tr>
<tr>
<td>10</td>
<td>300</td>
<td>1.3 x 10⁻³</td>
<td>10</td>
</tr>
</tbody>
</table>

If the viscosity can be maintained at a level less than 30 cp, then a reasonable head can be maintained which will insure an adequate flow through the felt wick. Since the thickness of the coating applied to the film is essentially a function of the viscosity of the Formvar solution, only then a wick with a sufficient pressure head for the most viscous solution (less than 30 cp) may suffice for all lower viscosities, since surface tension may be able to retain the solution at the interface between the felt and the air when not in contact with the film. Experimental tests could be conducted to determine the leak rate as a function of pressure head to verify this assumption. In operation then it is assumed that the coating thickness within limits can be made independent of wick size and porosity, the contact pressure.
between wick and the film and the film speed. The coating thickness would then be a first order function of the viscosity of the Formvar solution only.

The approximate size of the felt hairs is 0.0025 cm, which gives a characteristic length of the porous structure of approximately 0.001 cm. This value when combined with other parameters indicated below

\[ a = 0.001 \text{ cm} \]
\[ \nu = 3.8 \times 10^{-2} \text{ cc/cm}^2/\text{sec} \]
\[ \rho = 1.25 \text{ gm/ml} \]
\[ \eta = 0.5 \text{ cp} \]

results in a Reynolds number from equation (2) of

\[ R = \frac{a \nu \rho}{\eta} = 0.5 \times 10^{-6} \]

which is many orders of magnitude lower than the upper limit of 5. Thus equation (1) is applicable to this flow problem over any realistic felt porosity, coating speed, solution density, and solution viscosity.
Fig. D3. CHCl₃ EVAPORATION FROM FORMVAR SOLUTIONS