Calcium film indicator of moisture associated with noctilucent cloud particles

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ABSTRACT

The techniques for the preparation, testing and handling of protected calcium films for use as an indicator of moisture associated with noctilucent cloud particles are described. Study of the exposed and unexposed films flown in the rocket sampling experiments has revealed evidence for the presence of moisture.

The rocket sampling technique previously described (Ref. A) has provided an opportunity to obtain direct experimental evidence of the presence of water in nontilucent clouds. The detector chosen for this task was a thin calcium film protected from atmospheric moisture. The calcium films were designed to be sufficiently thin to allow transmission of light through spots or optical "holes" produced by the reaction with water to form calcium hydroxide.

In order to prevent the film from deteriorating in air, the calcium was sandwiched between several protective films. As shown in Fig. 1, a total of six films were applied to a lucite mounting surface. The lucite was first cleaned by a replica stripping technique, and then baked at 200°C for twenty minutes to remove moisture from the surface of the plastic. While still hot, the lucite was transferred into vacuum and allowed to cool for several hours. To seal the lucite surface, a relatively thick film ($\sim$1000 Å) of lithium fluoride and a thin film ($\sim$50 Å) of aluminum were applied using standard vacuum deposition techniques. The calcium was then deposited over these substrates and for protection against atmospheric moisture, another thin film of aluminum was evaporated over the calcium. For further protection, the vacuum system was filled to atmospheric pressure with argon, and the composite film was placed on a water-cooled surface in a hot chamber of argon gas saturated with paraffin vapor. The paraffin was allowed to condense on the cooled calcium surface for ten minutes and finally, the whole surface was coated with a thin layer of dehydrated silicone oil. This served to both seal the surface against water vapor and entrap cloud particles upon collection. The uncertainties involved in the vacuum deposition process resulted in a variation in thickness of the calcium films. As a consequence, some of the samples became slightly translucent.

Several experiments were performed to simulate the collection conditions and determine the stability of the protected calcium surfaces. First, ice crystals with a velocity of 800 meters per second were made to impact on the calcium surface in vacuum. Afterwards, transparent spots or optical "holes" could be seen in the film with an optical microscope. These holes ranged from a half-micron to several millimeters in diameter. Spraying water on the surface also produced holes down to one micron in diameter. Thus these calcium surfaces should have been able to detect ice particles at least as small as one-half micron in diameter. Metallic dust and abrasive particles whose sizes were 0.1 micron and larger were also fired at 800 meters per second at a protected calcium surface. Inspection revealed that only the particles greater than ten microns caused any noticeable ruptures in the films.

The stability of the protected calcium surface to atmospheric moisture was tested by exposing the film to a high humidity environment. The time for the reaction spots to develop was a good criterion for measuring the extent of film deterioration. This development time was about fifteen minutes for a freshly prepared detector. After one month's exposure to a high humidity

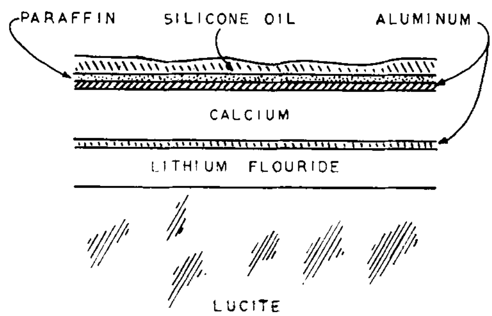

FIG. 1. Cross-section of calcium film moisture indicator.

environment, the development time increased to three hours, and half-micron holes could still be produced by ice particle impact. It is believed that the detector films had adequate sensitivity and stability for their task since they were stored for about a month in a low humidity environment prior to flight use.

Four calcium surfaces were flown in the two successful flights, two in the presence of clouds and two when no clouds were visible. A control area on each sample was obtained by shielding with an undercut section of the plastic sample clamps. After the flights, the calcium surfaces were placed in small, sealed casettes designed to permit high magnification microscopic analysis and act as desiccators. Then optical micrographs were made using transmitted light from all areas of the samples. The number of holes per unit area of control surface was determined to establish a background density (i.e., holes and ruptures due to flaws in the calcium surface). Next, the number of holes per unit area of exposed surface was found along with any significant changes in surface structure.

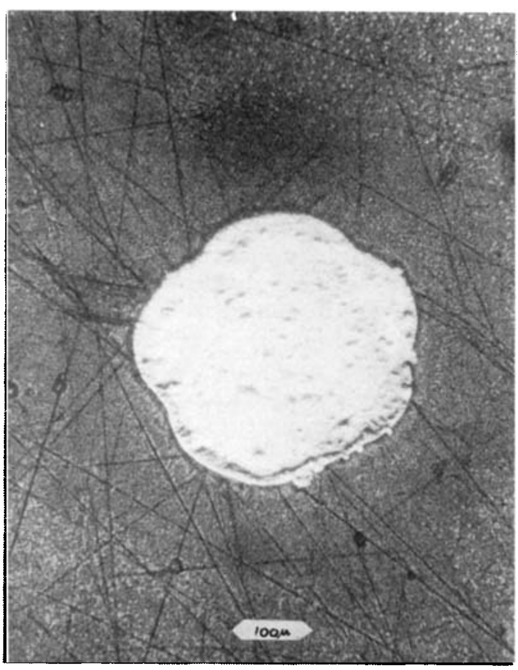

FIG. 2. Reflected-light micrograph of large reaction spot found on surface from box 9 (no-cloud flight).

The samples flown when no noctilucent clouds were observed showed a fairly uniform density of holes over the entire surface. In box 5, this density was negligible, (~ 10 holes/cm^2) and in box 9 the density was high ($\sim 1.1 \times 10^5$ holes/cm^2). The much higher density of the sample from box 9 appears to have resulted from too thin a film of either lithium fluoride or aluminum being deposited during the preparation of the detectors. This large background density was also found in the control area of the sample from box 11.

Fig. 2 shows the one outstanding feature in the control flight samples. This was a large hole, 300 microns in diameter, found on the exposed surface of the sample from box 9. The micrograph shown in Fig. 2 was taken with reflected light, consequently small holes cannot be readily seen. This hole appears to have been caused by a water droplet between 20 and 30 microns in diameter. No similar feature was found on any of the other samples.

The control area of the sample from box 11 of the flight through the noctilucent cloud showed a high background density of 1.8×10^5 holes/cm^2. However, although the hole density on the exposed areas fluctuated, it was significantly greater than that on the control. The average value of this density was 6.3×10^5 holes/cm^2 and indicated that approximately 4.5×10^5 holes/cm^2 were present, presumably caused by the reaction of the calcium film with the cloud particles. Figures 3 and 4 show portions of control and exposed areas of the sample from box 11.

The sample from box 14 of the noctilucent cloud flight was slightly translucent, thus the distribution and nature of holes on its surface were difficult to discern with standard light microscopy. The study of this sample was best

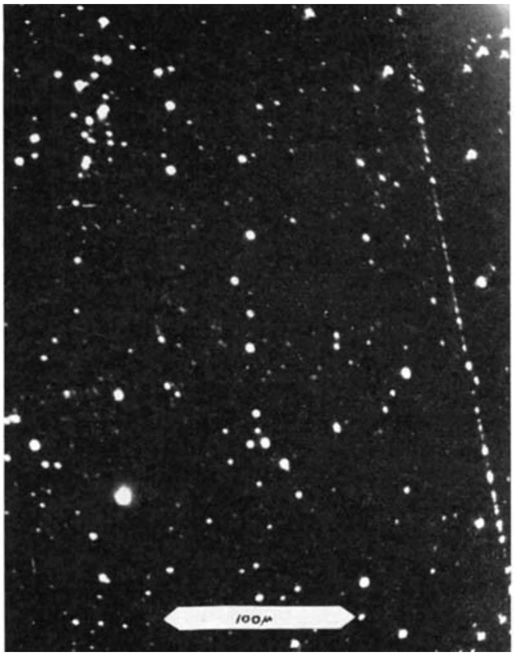

FIG. 3. Micrograph showing portion of control area (unexposed) from noctilucent cloud flight. (Box 11.)

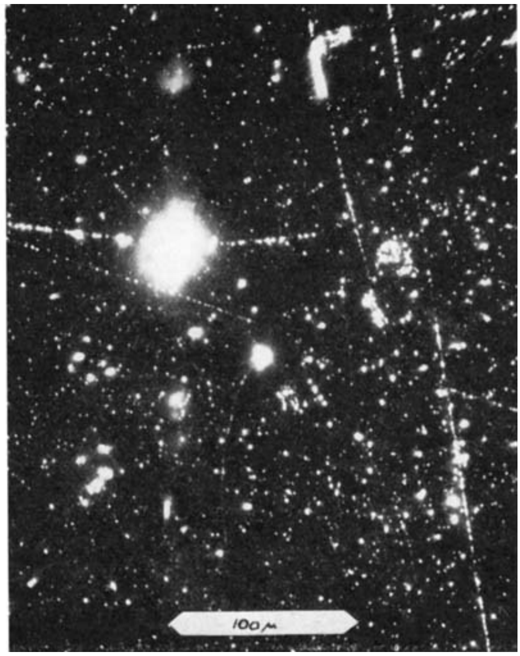

FIG. 4. Portion of exposed area from the same sample as in Fig. 3.

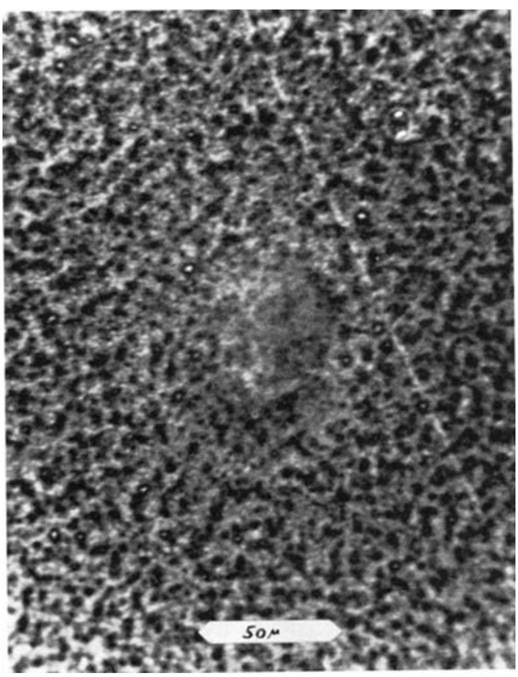

FIG. 5. Phase-contrast micrograph of a portion of the exposed area on the sample from box 14. (Noctilucent cloud flight).

accomplished by the technique of phase-contrast microscopy. Using this method, slight changes in thickness or optical characteristics of a translucent sample produce contrast in the image. With this technique, the calcium film appeared to have a texture or grain, which was much coarser on the exposed area than on the control. The exposed areas exhibited an additional feature not found on the control. In the center of many of the granules were small holes whose density was measured to be about 4.5×10^5 holes/cm^2. The numerical agreement between this density and the excess density from box 11 may be fortuitous. Since no similar structure was observed on the control area, this value was taken as the hole density produced by the cloud particles. Fig. 5 is a phase-contrast micrograph of a portion of the exposed area on the sample from box 14.

The optical transmission and reflection characteristics of these calcium films were studied with a sensitive photometer coupled to a metallograph. The intensity of white light transmitted and reflected at normal incidence was measured using a scanning beam of about 0.75 mm^2 cross-sectional area. The samples from the control flight (boxes 5 and 9) showed marginal increases in transmission and reflection of the exposed areas compared to the control areas. However, those from the noctilucent cloud flight (boxes 11 and 14) showed decreases of about 5 % in both transmission and reflection.

To estimate the amount of water necessary to produce these decreases, a series of simulation tests were conducted. Known amounts of water were sprayed on calcium films, prepared in the same manner as the flight samples, and the corresponding changes in reflected and transmitted light measured. These experiments were conducted in a chamber maintained at a temperature sufficiently low that the relative humidity was essentially 100 %, while the absolute humidity was nearly zero. This chamber included an air lock for inserting the films and an atomizer for projecting microscopic water droplets into the calcium surfaces. The minute quantities of moisture deposited per unit area were measured by a radioactive tracer technique. This series of tests indicated that the samples from the flight through the noctilucent cloud gained approximately 1.5 micrograms of moisture per square centimeter.

The density of "haloed" particles on the nitrocellulose films (see Ref. B) was found to be close to the number of holes per unit area observed on the calcium films ($\sim 4.5 \times 10^5$ holes/cm^2). This suggests that an average haloed noctilucent cloud particle had approximately 3×10^{-12} grams of water associated with it.

The results of these studies of the calcium films, when compared with other investigations (Refs. B and E), indicate that moisture is a constituent of the haloed-type noctilucent cloud particles.

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