Properties of ice accreted in two-stage growth*

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SUMMARY

The hypothesis that hailstones may grow in two stages by the soaking and refreezing of a previously formed porous structure, has been explored by obtaining artificial accretions on rotating cylinders in a wind tunnel, in controlled laboratory conditions. Once the first stage of growth was obtained, the inner plastic embryo was extracted and slices were cut for a series of analyses; the remaining pieces of the deposit were rejoined and a new layer was grown onto it or, alternatively, the deposit was partially melted in a flow of warm air, and other slices were sawn for another set of analyses.

Changes in accreted ice characteristics due to soaking have been determined for local density, crystallographic parameters and morphology.

While deposits obtained at high updraught velocity (26 m s\(^{-1}\)) were already at a high density in the first stage, the lower density of deposits grown at lower impact velocities (10 m s\(^{-1}\) or less) was increased, due to water penetration, to values similar to those observed in natural hailstones. The local density measurements were obtained by X-ray contact micrography. This technique enabled us to record even minor density variations crucial for correct interpretation and analysis.

All crystallographic parameters: the average grain size, \(\bar{V}\), the mean maximum crystal length, \(\bar{l}\), and the mean maximum crystal width, \(\bar{w}\), showed an increase after penetration due to both annealing during the time spent at 0°C and the filling of voids.

In deposits grown at impact speeds \(\leq 10\) m s\(^{-1}\) the appearance and shape of the first stage deposits are considerably changed as a result of soaking, and the changes introduced are more important the lower the density of the first stage. The dry growth lobes are smoothed and the ice made translucent, radial lines of bubbles are formed within the channels between lobes, and sometimes features are propagated to the external layer. Features similar to those found in natural hailstones have been observed. The lobe structure defined in the first stage, though modified by soaking, retains its general characteristics.

Evidence of two-stage growth in natural hailstones is given and the origin of features such as cuspidated lobes, radial lines of bubbles and rounded air bubbles is explained.

The evidence that two-stage growth may be a frequent occurrence in hailstones is changing the criteria for natural hailstone interpretation by introducing new criteria and suggesting modifications to those currently adopted.

1. INTRODUCTION

The impulse towards laboratory investigation to define the criteria for an unequivocal interpretation of hailstone features in terms of the growth conditions in the parent cloud has slowed down considerably in recent years. The reason for this is the increasing awareness that features of natural hailstones are difficult to reproduce with artificial accretions, and, conversely, the criteria derived from artificially accreted ice are hard to apply to natural hailstones. This lack of success has been attributed by List (1977) to the large number of parameters or processes affecting hailstone growth (size distribution and concentration of droplets, air and deposit temperatures, updraught velocity, dynamic variables such as rotation rate and position of hailstones during fall, microphysical-dynamical feed-back, rain-hail connection, etc.) which are very difficult to reproduce in all their possible combinations in the hail cloud, so as to find criteria applicable with an acceptable degree of generality.

None the less, important steps have been made in the right direction, narrowing the gap between artificial accretions and natural hailstones. We mention, in this regard, the studies of the crystallographic parameters in relation to growth conditions, and of the annealing effects which allow the changes in hailstone structure (during fall, before

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collection, and during storage) to be properly taken into account (Levi and Prodi 1978; Prodi and Levi 1980a; Prodi et al. 1982; McCappin and Macklin 1984a, b).

Moreover, a promising line of investigation which has been little explored is based on the hypothesis that the whole hailstone or certain hailstone layers may grow in two stages by the soaking and refreezing of a previously formed porous structure. The possibility of a two-stage growth in hailstones has been neglected probably because of the work of Browning et al. (1963) which indicated that in the conditions of the hail cloud only densities very close to the maximum value of 0.917 g cm\(^{-3}\) could be produced; according to this assumption, the ice fabric is completely defined in the first accretion phase without significant effects thereafter. Notwithstanding this, initial experimental evidence of the possibility of two-stage growth in hailstones was provided by Prodi (1970), using local density measurements performed by contact X-ray micrography on both artificial and natural hailstones. Artificial accretions of porous ice were shown to change density due to water penetration from a subsequent wet growth stage, while in natural hailstones a decreasing density towards the centre and other features favouring the two-stage interpretation were observed. Previous to this, Kidder and Carte (1964) had made qualitative observations of soaking effects on accreted structures, but it was not until Pfleum (1980) that this specific growth condition was investigated and deemed probable on the basis of model calculations and other supportive evidence. The present investigation has been prompted by the need to extend Prodi's (1970) initial results into more systematic investigations of the characteristics and properties of ice accreted in two stages. This type of growth could explain the discrepancies between artificial accretions and natural hailstones and may leave unequivocal indications for their interpretation inside the hailstone structure.

Subsequent to the completion of experiments for our present work, a paper by Pfleum (1984) was published reporting experiments along similar lines. However, the experimental conditions tested and the analyses performed are quite different in our own work, and the technique of local density measurement used here produces additional quantitative information and conclusions. Moreover, the experimental procedure followed allows a complete set of analyses to be performed on the first stage growth, before the superposition of the second stage.

Preliminary to our investigation of the two-stage growth, we decided to improve the contact micrography technique to be used for local density measurements. In so doing, we realized that the entire problem of accreted ice density should be re-examined, starting from Macklin's (1962) paper, which was based only on volume and weight measurements of the deposit. The results obtained here on first stage rotating cylinders as well as additional results on fixed cylinders form the basis of a separate work, devoted to the density of one-stage accreted ice (Prodi et al. 1986a).

2. EXPERIMENTAL PART

(a) Wind tunnel and growth conditions

The experiments were conducted on rotating cylinders in a vertical wind tunnel alongside a cold room, already described in previous papers (Prodi 1975; Levi and Prodi 1978). Here, we describe only the main experimental details; the other papers can be referred to for any points left unspecified.

The square section vertical wind tunnel (2 m high, 30 cm wide) is connected through the lower and upper ends with the cold room of 13 m\(^2\). In the working section, at about 1.6 m from the floor, the tunnel is reduced to 7 cm per side. To grow accreted ice of
various densities, different impact velocities of the impinging droplets were used, by operating the fan at the upper end of the tunnel so as to obtain four main velocities in the working section: 26 m s\(^{-1}\) (with a few deposits grown at 29 m s\(^{-1}\)), 10, 5 and 2.5 m s\(^{-1}\). The velocities were measured by a hot-wire and Venturi-type anemometers in the working section with the growing deposit removed. The velocity profile we found is in agreement with that expected for square section ducts. On the other hand, the regular shape of the deposits (see Figs. 11(a), (b)) ensures the homogeneity of the droplet supply in the tunnel.

Two types of sprayer were used, one with the pressure directly on the water, for the high liquid water contents, and the other on the air of the water bottle, for lower liquid water contents. In order to keep the deposit temperature, \(T_0\), constant, the pressure to the spray nozzles, or the number of droplets entering the tunnel intake, was regulated, thus introducing small variations in the droplet size distributions, and, as a consequence, in the mean and median volume radii during individual growth tests. The droplet spectra were measured after successive series of a few runs by counting and sizing the droplet imprints on a glass slide 4 mm wide, covered with gelatine and briefly exposed to the flow, by means of a spring-release device. Typical droplet spectra are reported in Fig. 1 together with the mean and median volume radii of the distributions. Mean volume radii were obtained in the range 12–16 \(\mu\)m, and median volume radii in the range 14–21 \(\mu\)m.

![Graph](image)

**Figure 1.** Two examples of impinging droplet spectra used in the experiments. Mean, \(\bar{r}\), and median, \(r_m\), volume radii are indicated.
The mean volume diameter was thought to be more appropriate to the physical nature of the dry growth accretion process. The ice fabric being defined by the freezing of individual droplets, we thought that the droplets having the mean volume could better represent the individual accretion process than droplets having the median volume radius. However, since other authors use the median volume diameter (e.g. Macklin 1962) we also made calculations of the latter parameter for comparison, in a companion paper (Prodi et al. 1986a), with results similar to those obtained previously.

The liquid water content, \( W \), during the first stage growth has been evaluated from the growth time and the final size of the accreted deposits, using the experimental efficiencies, \( E \), of cylindrical collectors and the relationship

\[
W = (\pi \rho / t) \int_{R_0}^{R} dR / Ev
\]

where \( \rho \) is the ice density, \( R_0 \) and \( R \) the initial and final accretion radius and \( v \) the airspeed. The value of \( W \) varied between 0.2 and 3 g m\(^{-3} \) for the different experiments, and reached 6 g m\(^{-3} \) during the second stage growth. The measurements of \( W \) for the second stage growth were performed in the same way after the first stage was completely soaked. An independent measurement of \( W \) was performed, for some tests, by the same glass slide we exposed to the flow to measure droplet spectra, knowing the time spent by the slide in the droplet flow. Agreement between the two quite different methods was within 20%.

The rotating cylinder embryo on which the accretions were grown was a 6 mm diameter plastic rod on which a bulk ice layer was previously frozen from distilled water, giving an initial effective diameter of 1 cm for all deposits.

A platinum resistance thermometer, embedded in the outer surface of the plastic rod, measured the temperature of the deposits and of the airstream (before the injection of the water droplets). The output signal was fed into a recorder, thus providing the continuous track needed for growth-time computations and for the adjustment of the water supply from the nozzles during the accretion experiments. The temperature of the airstream fluctuated within 1°C due to the temperature control and the compressor cycle.

The rotation rate of the cylinders is also an important parameter to be specified in accretion experiments (Kidder and Carte 1964). In the present experiments a rotation rate of 4 Hz was maintained for all deposits, high enough to make the surface features and the temperature uniform, and low enough to avoid centrifugal effects.

(b) Specific operations to obtain two-stage growth deposits

Once the first stage of growth was obtained, the inner plastic cylinder was extracted and three normal slices were cut out from the deposit's central region with a band saw, for the first series of analyses. The remaining pieces of the deposit were rejoined and replaced on the plastic cylinder, and a new layer was grown on it, or, alternatively, the deposit was partially melted in a flow of warm air. The melting process was achieved by injecting ambient air into the tunnel with a separate fan until water drops were observed to be shed. The deposit was then left to freeze inside the freezer unit. Finally the inner plastic cylinder was extracted again from the deposit and other slices were sawed out for analysis.

In order to minimize possible annealing effects on the accreted structure, the deposits were stored immediately in the deep-freezer unit at -17°C and worked within two to three hours. Quenching was not performed since for the first stage deposit, \( T_s \) is already well below 0°C and for the second stage the cooling time of the deposit without quenching
is in any case short when compared with the time spent at 0°C by the deposit during the soaking process.

(c) **Analyses performed on the deposits**

The slices were worked smooth with a microtome and prepared for the series of analyses. One slice was used for X-ray contact micrography. The second slice was photographed in reflected light and, having been reduced to about 300 μm thickness, in crossed polaroid light; normal and macrolenses (Zeiss Luminar) were used according to the features under observation, the latter giving a magnification up to 18×. The third slice was replicated with formvar (polyvinyl-formal) solution in ethylene dichloride following the technique of Aufdermaur *et al.* (1963). The crystal parameters and c-axis orientations revealed by the replicas were analysed by drawing the replica features (grain boundaries and etch pits) using a side projector applied to the optical microscope. Details of the crystallographic and photographic analyses have been reported in a series of papers (Levi and Prodi 1978, 1983; Prodi and Levi 1980a, b; Levi *et al.* 1980; Prodi *et al.* 1982).

The technique to measure the local density of ice by X-ray contact micrography was developed and described by Prodi (1970). A slice of about 1 mm uniform thickness is laid on an X-ray film or plate, separated only by a very thin mylar sheet, and exposed to an X-ray beam in a contact micrography apparatus. In these tests, the film used was Kodak M-type and the X-ray apparatus (Faxitron) was operated at 20 kV, with a film-to-source distance of about 45 cm. The ice density is derived from the optical density of the film measured at the densitometer. Some improvements have been introduced with respect to Prodi's (1970) work and details of the procedure will now be specified. Since the inner cylindrical embryo is of clear bulk ice its density is considered to be that of pure ice (0.917 g cm⁻³): thus the optical density corresponding to it is taken as the reference to evaluate the optical density and consequently the ice density in other areas of the film. Since it is important for the slice thickness to be uniform (otherwise thickness variations could erroneously be attributed to ice density variations), a device to work the slice with high precision has been designed. This is essentially an aspirating table where the slice is held in place while the microtome blade slides on the upper surface. Thickness variations in different zones of the slice were within 5 μm and the consequent error in density was lower than 0.5%.

The possibility of error arising from the non-uniformity of the X-ray was checked by measuring the optical density in different areas of a film exposed without the slice on it. No variations in optical density from point to point were observed in the region where the slice is normally located.

In order to fulfil the requirement of operating within the range of linear response of the film, the calibration curve of the film was verified by determining the optical density after different exposure times.

Using optical densities inside the linear range of the film the measurements are extremely accurate, thanks to the high precision of the optical densitometer. Error sources in this phase are: the densitometer reading on paper recorder, and minor departures from linearity in the response of the film. The total error in density measurements is estimated to be below 1%.

A specific test of the technique was performed on a bulk ice slice of stepped thickness. The linear relationship obtained in this test between optical density and ice thickness ensures the correct application of the technique.

In the optical densitometer, a rectangular light beam of variable size inspects the film at constant speed and the signal is recorded on paper. In a film from a specific slice of accreted ice deposit, the resulting density path (directly converted to an ice density
path, through the bulk ice reference value) depends on the beam shape and on the path followed, because of the presence of bubbles, channels and lobes. The resolution of the local density measurement is determined by the area of the beam. However, such measurement is not of practical use because of its great variability caused by air bubbles and channels between lobes. Therefore the choice of the beam shape and size, and the area over which to average are related to the degree of data smoothing required. We mainly used the largest available beam (140 × 640 μm) and made averages over five radial paths for each deposit. The measurement of density to the thousandth place refers to the individual measurement over the area inspected by the beam; the individual measurement is within ±0.005 g cm⁻³. We maintain three significant figures after the point in measurements referring to wider areas, where the local density value is an average over many individual measurements. Moreover, since in many cases the density decreases with radius, due to the development of deep channels between lobes, the radial tracts were separated, and averaged in two parts: one 2 mm-long tract from the inner embryo, and the second from there to the external part of the first stage growth. A typical example of a densitometric reading on a diameter path, together with the corresponding photograph obtained from the X-ray micrograph, is presented in Fig. 2.

3. EXPERIMENTAL RESULTS

(a) Local density measurements

The results on local density are grouped in Table 1 according to the updraught velocity, which is closely related to the droplet impact velocity. All density data are reported for the various deposits, together with the main data concerning the growth

![Figure 2](image-url)

Figure 2. Composite picture showing an X-ray micrograph of a first stage dry growth deposit (TS13) and the corresponding densitometric path. The first and second radial tracts averaged in density computations are also indicated. The inner bulk ice of 1 cm diameter serves as a scale in this and following pictures.
### Table 1. Values of Local Densities Measured in First Stage Growth and Their Change Due to a Subsequent Wet-Spongy Growth or Melting

<table>
<thead>
<tr>
<th>Deposit</th>
<th>Velocity, ( m/s )</th>
<th>( T_a ) (°C)</th>
<th>( T_d ) (°C)</th>
<th>( R_{vol} ) (μm)</th>
<th>( \rho_{01} )</th>
<th>( \rho_{1,1} )</th>
<th>( \rho_{1,2} )</th>
<th>( \rho_\text{I} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>TS19</td>
<td>26</td>
<td>-25</td>
<td>-21</td>
<td>&gt;0.905</td>
<td>0.905</td>
<td>0.905</td>
<td>0.911M</td>
<td></td>
</tr>
<tr>
<td>TS21</td>
<td>26</td>
<td>-25</td>
<td>-18</td>
<td>16</td>
<td>0.907</td>
<td>0.907</td>
<td>0.907</td>
<td>0.911W</td>
</tr>
<tr>
<td>TS14</td>
<td>26</td>
<td>-23</td>
<td>-14</td>
<td>12</td>
<td>&gt;0.905</td>
<td>0.905</td>
<td>0.905</td>
<td>&gt;0.910M</td>
</tr>
<tr>
<td>TS16</td>
<td>26</td>
<td>-21</td>
<td>-7</td>
<td>0.910</td>
<td>0.910</td>
<td></td>
<td></td>
<td>&gt;0.910M</td>
</tr>
<tr>
<td>TS6</td>
<td>29</td>
<td>-19</td>
<td>-5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>&gt;0.910W</td>
</tr>
<tr>
<td>TS5</td>
<td>29</td>
<td>-19</td>
<td>-5-0</td>
<td>&gt;0.91</td>
<td></td>
<td></td>
<td></td>
<td>0.96W</td>
</tr>
<tr>
<td>TS2</td>
<td>29</td>
<td>-27</td>
<td>-16</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.874W</td>
</tr>
<tr>
<td>TS5</td>
<td>10</td>
<td>-28</td>
<td>-16</td>
<td>0.835</td>
<td>0.843</td>
<td>0.828</td>
<td>0.871W</td>
<td></td>
</tr>
<tr>
<td>TS6</td>
<td>10</td>
<td>-25</td>
<td>-16</td>
<td>0.842</td>
<td>0.850</td>
<td>0.834</td>
<td>0.872W</td>
<td></td>
</tr>
<tr>
<td>TS7</td>
<td>10</td>
<td>-24</td>
<td>-16</td>
<td>0.846</td>
<td>0.863</td>
<td>0.828</td>
<td>0.871W</td>
<td></td>
</tr>
<tr>
<td>TS15</td>
<td>10</td>
<td>-25</td>
<td>-16</td>
<td>0.876</td>
<td>0.882</td>
<td>0.871</td>
<td>0.890W</td>
<td></td>
</tr>
<tr>
<td>TS22</td>
<td>10</td>
<td>-22</td>
<td>-16</td>
<td>15</td>
<td>0.863</td>
<td>0.896</td>
<td>0.830</td>
<td>0.888M</td>
</tr>
<tr>
<td>TS17</td>
<td>10</td>
<td>-22</td>
<td>-7</td>
<td>13</td>
<td>0.875</td>
<td>0.900</td>
<td>0.850</td>
<td>0.888M</td>
</tr>
<tr>
<td>TS4</td>
<td>10</td>
<td>-18</td>
<td>-12</td>
<td>0.884</td>
<td>0.892</td>
<td>0.873</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TSX</td>
<td>10</td>
<td>-23</td>
<td>-16</td>
<td>0.853</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>TS11</td>
<td>10</td>
<td>-22</td>
<td>-12</td>
<td>0.880</td>
<td></td>
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<tr>
<td>TS12</td>
<td>10</td>
<td>-22</td>
<td>-14</td>
<td>0.884</td>
<td>0.893</td>
<td>0.876</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TS18</td>
<td>10</td>
<td>-26</td>
<td>-22</td>
<td>13</td>
<td>0.826</td>
<td>0.844</td>
<td>0.809</td>
<td>0.860M</td>
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<tr>
<td>TS13</td>
<td>5</td>
<td>-26</td>
<td>-18</td>
<td>0.755</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TS9</td>
<td>5</td>
<td>-26</td>
<td>-12</td>
<td></td>
<td>0.749</td>
<td>0.660</td>
<td></td>
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</tr>
<tr>
<td>TS20</td>
<td>5</td>
<td>-23</td>
<td>-17</td>
<td>14</td>
<td>0.737</td>
<td>0.626</td>
<td></td>
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</tr>
<tr>
<td>TS8</td>
<td>5</td>
<td>-22</td>
<td>-12</td>
<td>0.650</td>
<td>0.706</td>
<td>0.595</td>
<td></td>
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</tr>
<tr>
<td>TS23</td>
<td>2.5</td>
<td>-22</td>
<td>-12</td>
<td>0.427</td>
<td></td>
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</tbody>
</table>

\( \rho_{01} \) is the average first stage density in the first radial tract; \( \rho_{1,2} \) is the average first stage density of the second radial tract; \( \rho_{0 \text{vol}} \) is the average of \( \rho_{01} \) and \( \rho_{1,2} \); \( \rho_0 \) is the density, along the radial line, of the first stage growth layer which has been modified by penetration of liquid water due to a wet-spongy growth (in this case the letter W follows the number) or due to melting (the letter M follows the number).

条件下。第一阶段生长的局部密度（第二阶段值）及其变化由于第二阶段的生长也在图 3 中作为参数 \(-rv_o/T_a\) 的函数给出，类似于 Macklin's (1962) 参数，但与 \( \bar{r} \)，即平均体积半径，而不是 \( r_m \)，即体积半径；\( v_o \) 是在 stagnation point 和 \( T_a \) 处的表面温度。实际使用的 Macklin 参数将使数据右移 1.37 在图 3 中。

(i) First stage values. It is immediately apparent that high updraught velocities (29 m s\(^{-1}\) and 26 m s\(^{-1}\)) cannot produce densities lower than 0.9 g cm\(^{-3}\); even deposit TS19, obtained at the lowest air and deposit temperatures (\( T_a = -25^\circ \text{C}, T_d = -21^\circ \text{C} \)), produced a milky opaque ice with a density of 0.903 g cm\(^{-3}\). Channels begin to be observed in deposits TS16, only towards the end of growth.

For deposits obtained at 10 m s\(^{-1}\) there is a pronounced departure from high density values. Only deposits TS12, TS17, TS4 and TS15 have first tract densities close to that of pure ice. At the same time, they present a radial decrease due to the presence of channels.

At 5 and 2.5 m s\(^{-1}\) the first stage density decreases proportionally, with mean values between 0.755 and 0.650 g cm\(^{-3}\) for 5 m s\(^{-1}\), and a mean value of 0.427 g cm\(^{-3}\) for the deposit grown at 2.5 m s\(^{-1}\), with the ice becoming more and more fragile.
Figure 3. Second stage local density values obtained by wet growth (open octagons) and by melting (open squares) for the deposits. The corresponding first stage lower density values are connected to them by arrows, and are presented as open rhombi, or directly given with their values when $\rho < 0.75 \text{ g cm}^{-3}$. Values are given as a function of a parameter similar to Macklin's parameter but with the mean radius in place of the median radius.

(ii) Second stage values. The density change in the layer produced by the superposition of second stage growth was tested for both wet growth and melting. In Table 1 the density variations show that, during the final freezing, the density of the deposit increases to values normally reported for natural hailstones. In the table the second stage density values are separated, indicating those due to melting by the letter M and those to wet growth by the letter W.

The deposits grown at 26 m s$^{-1}$ are, already in the first stage, at maximum density and obviously cannot increase their density further by liquid water penetration. In all other deposits, the second stage growth increases the density of the layers to high values, regardless of the original first stage value. In fact, even deposits grown at 5 and 2.5 m s$^{-1}$, whose original first stage density was quite low, increased their density to the same rather high values of deposits grown at 10 m s$^{-1}$, in several cases up to the maximum possible density (TS13 and TS9). The long growth time and the fragile fabric of low density deposits limit the extent of radial growth, thus easing the water penetration during the second stage. Moreover Fig. 3 reveals a feature of particular note. There is an indication that, after penetration, first stage deposits of high density reach a final density lower than that reached by others grown at lower first stage density. This appears to indicate that the latter have better protected air enclosures hindering water penetration; consequently,
TABLE 2. Crystallographic parameters

<table>
<thead>
<tr>
<th>Deposit and growth conditions</th>
<th>$\bar{\sigma}$ (mm)</th>
<th>$\bar{l}$ (mm)</th>
<th>$\bar{w}$ (mm)</th>
<th>$\bar{\eta}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>TS2 $T_s = -27 , ^\circ C$</td>
<td>I Stage 2.1 x 10^{-3}</td>
<td>0.062</td>
<td>0.040</td>
<td>47$^\circ$</td>
</tr>
<tr>
<td>$T_a = -16 , ^\circ C$</td>
<td>II Stage 5 x 10^{-3}</td>
<td>0.089</td>
<td>0.058</td>
<td>53$^\circ$</td>
</tr>
<tr>
<td>$\nu = 29 , \text{m s}^{-1}$</td>
<td>Ext. layer 6.4 x 10^{-3}</td>
<td>0.28</td>
<td>0.13</td>
<td></td>
</tr>
<tr>
<td>TS16 $T_s = -21 , ^\circ C$</td>
<td>I Stage 11 x 10^{-2}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$T_a = -7 , ^\circ C$</td>
<td>II Stage-M 12.3 x 10^{-2}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\nu = 26 , \text{m s}^{-1}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TS14 $T_s = -23 , ^\circ C$</td>
<td>I Stage 3.0 x 10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$T_a = -14 , ^\circ C$</td>
<td>II Stage-M 2.8 x 10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\nu = 26 , \text{m s}^{-1}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TS7 $T_s = -24 , ^\circ C$</td>
<td>I Stage 2.36 x 10^{-3}</td>
<td>0.051</td>
<td>0.044</td>
<td>41$^\circ$30$'$</td>
</tr>
<tr>
<td>$T_a = -16 , ^\circ C$</td>
<td>II Stage-W 6.27 x 10^{-3}</td>
<td>0.089</td>
<td>0.068</td>
<td>39$^\circ$30$'$</td>
</tr>
<tr>
<td>$\nu = 10 , \text{m s}^{-1}$</td>
<td>(Wet) 17.4 x 10^{-3}</td>
<td>0.61</td>
<td>0.38</td>
<td></td>
</tr>
<tr>
<td>TSX $T_s = -23 , ^\circ C$</td>
<td>I Stage 6 x 10^{-3}</td>
<td>9.74 x 10^{-2}</td>
<td>5.66 x 10^{-2}</td>
<td>39$^\circ$15$'$</td>
</tr>
<tr>
<td>$T_a = -16 , ^\circ C$</td>
<td>II Stage-M 8.2 x 10^{-3}</td>
<td>10.1 x 10^{-2}</td>
<td>8.7 x 10^{-2}</td>
<td>43$^\circ$30$'$</td>
</tr>
<tr>
<td>$\nu = 10 , \text{m s}^{-1}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TS3 $T_s = -21 , ^\circ C$</td>
<td>I Stage 5.5 x 10^{-4}</td>
<td>0.027</td>
<td>0.022</td>
<td></td>
</tr>
<tr>
<td>$T_a = -15 , ^\circ C$</td>
<td>II Stage-W 4.6 x 10^{-2}</td>
<td>0.23</td>
<td>0.18</td>
<td>47$^\circ$</td>
</tr>
<tr>
<td>$\nu = 5 , \text{m s}^{-1}$</td>
<td>Ext. layer 8.9 x 10^{-3}</td>
<td>0.3</td>
<td>0.2</td>
<td></td>
</tr>
</tbody>
</table>

$\bar{\sigma}$ is the average grain size; $\bar{l}$ the mean maximum length; $\bar{w}$ the mean maximum width; $\bar{\eta}$ the mean value of the angle $\eta$, in first stage growth and in subsequent wet-spongy growth or melting.

In TS2, TS7 and TS3 the crystal parameters of the external layer (the layer external to the two-stage layer) have been added. 'M' indicates a second stage by melting, 'W' by wet growth.

these deposits undergo only a minor increase in density due to the melting or penetration of the second stage (see the typical cases of TS5, TS6, TS7, TS15, TS17, TSX, TS18).

(b) Crystallographic parameters

The crystallographic parameters in the first stage growth layer, and in the same layer once subjected to the second-stage growth, are summarized in Table 2, where $\bar{\sigma}$ is the average grain size, $\bar{l}$ the mean maximum crystal length, $\bar{w}$ the mean maximum crystal width, and $\bar{\eta}$, the mean value of the angle between the radial direction and the projection of the c-axis onto the plane of the section. Due to the time-consuming procedure, only a few of the deposits have been considered for this type of analysis. The same parameters have been measured also for the external layer of wet growth, for checking and comparison with data obtained by Prodi et al. (1982).

All size parameters, $\bar{\sigma}, \bar{l}, \bar{w}$, for all deposits show an increase. This increase is small, and due only to the annealing during the 0$^\circ$C growth of the second stage, when the first stage is already at high density and no significant penetration of liquid water occurs (TS2, TS16); in TS14 no increase is observed at all, due to the relatively short melting period. The deposits which experienced penetration exhibit an increase in grain size (TS7 by a factor of 2.6; TSX by a factor 1.36) which seems related to both the amount of penetration (revealed by the change in density) and the time spent at 0$^\circ$C. For the same reason a
very sharp increase (of almost two orders of magnitude) is experienced by the deposit TS3.

In all cases when the second stage was obtained by wet growth, the size values of the external layers were never reached by the first stage deposit modified by penetration, indicating that the first fabric has a dominant role in defining the grain size.

The data on $l$ and $w$ do not seem to contribute any more information to that already provided by the grain size; furthermore the aspect ratio $\varepsilon = (1/N)\Sigma(l_i/w_i)$ does not change appreciably after penetration.

Measurements of $c$-axis orientation based on $\eta$ values and the histograms of $\eta$ distributions, not reported here, indicate that the first stage structure is not eliminated and that it regulates the orientation of penetrated water. The orientation distribution of the grown fabric was also observed to persist while investigating the annealing effects (Prodi and Levi 1980a).

(c) Morphology

The internal features are investigated in reflected light and in photographs taken from X-ray micrographs. A typical set of photographs is shown in Figs. 4 to 7, each with four sectors of the same deposit: reflected light pictures of first and second stage deposits and X-ray micrographs of first and second stage deposit.

In deposits obtained at 26 m s$^{-1}$ (Fig. 4), the features observed in the first stage confirm what has already been observed by Prodi and Levi (1980b) when investigating the hyperfine features in deposits grown at 29 m s$^{-1}$. The separation between the first stage of dry growth and the second stage of wet growth is very distinct since the first was already grown at high density and penetration was negligible. Incidentally we note that the X-ray densitometric path, not reported here, shows a very small but distinct step, passing from the first stage to the wet one, with density increasing from 0.907 to 0.911 g cm$^{-3}$, thus indicating that the air bubbles trapped during dry growth contribute, though very slightly, to lowering the density.

For deposits grown at 10 and 5 m s$^{-1}$ (Figs. 5 to 9) one observes that the appearance and shape of the first stage deposit is considerably changed as a result of soaking. The structure and external shape shows well-defined dry growth lobes separated by channels which depart into a very dense mesh of tiny minor channels, a structure well evidenced by the X-ray pictures. In Fig. 6 we observed a transformation from opaque to transparent ice of the first 2–3 mm (in the photograph) of the lobes, and it was noted how penetration decreases moving from the periphery towards the centre.

By further decreasing the airspeed, the channels become deeper, separating the lobes and considerably decreasing the densities. As a consequence, the changes introduced by the subsequent penetration are more important, the lower the density of the first stage. The dry growth lobes are smoothed and the ice made translucent, radial lines of bubbles are formed within the channels between lobes and, more surprisingly, features are sometimes propagated also in the wet layer, continuing the radial lines of bubbles, with rings (fronts) of bubbles following the shape of the lobes (see Figs. 5, 6 and 8).

The morphological appearance of the first stage dry growth region modified by penetration is also strongly dependent on the conditions in which penetration occurs in the very initial stage (afterwards it occurs at 0°C for all the deposits). This is well documented by a comparison between deposit TS20, in which the penetration initiated at $T_d$ near 0°C ($-5$ °C) (Fig. 6), and deposit TS3, in which $T_d$ was initially rather low ($-24$ °C) (Fig. 9). It is seen that, in the first case, the external part of the opaque dry growth lobes is converted into transparent ice, while in TS3 there is a gradual transition
from dry growth, evidenced by bubble rings following the shape of dry growth lobes, before the layer becomes definitely wet.

In reality the above mentioned features—radial chains of bubbles extending also in the transparent layer, rounded lobes of opaque to translucent appearance, rounded elongated air bubbles—are rather similar to features frequently found in natural hailstones, and confirm those already reported by Prodi (1970). The importance of dry growth lobes in shaping the observed internal structure in natural hailstones is confirmed by the deposit obtained at 2·5 m s⁻¹ (TS23 in Figs. 10 and 11(b)): here, the lobes become more separated and the channels between lobes cut deeper into the deposit.

The features specifically introduced by melting are shown in Fig. 7 (deposit TS18). It is observed that the shape of lobes changed. The rounding and smoothing effect is
more evident than in the case of penetration by superposed wet growth. The areas of translucent ice between lobes are a clear indication of the soaking process, while the air bubbles have a more rounded appearance.

A parameter to be considered for all the deposits is the sectorial concentration of lobes. A glance at the whole deposit external appearance (Fig. 11) and at sections in Figs. 4 to 7 immediately shows that the lobes are less pronounced and more sectorially frequent in the deposits grown at higher impact speed. At 26 m s\(^{-1}\) lobes are so sectorially frequent and ill-defined that they are hardly observable. This general qualitative observation has been quantified by counting the lobes observed in a 90° sector. The results are summarized in Fig. 12, which is limited to the experimental conditions tested, in which other possible relevant parameters such as droplet size distribution remained relatively unchanged. The sectorial concentration and depth of lobes appears to be related to the impact velocity, other conditions being constant. To discuss this effect it
Figure 6. As Fig. 4 for deposit TS20. First stage growth conditions: $T_s = -23^\circ C$, $T_d = -17^\circ C$, $v = 5 \text{ m s}^{-1}$; second stage obtained by wet-spongy growth.

Figure 7. As Fig. 4 for deposit TS18. First stage growth conditions: $T_s = -26^\circ C$, $T_d = -22^\circ C$, $v = 10 \text{ m s}^{-1}$; second stage obtained by melting.
Figure 8. Examples of the different penetration effects in a two-stage growth deposit for deposit TS7: $T_s = -24 \, ^\circ \text{C}$, $T_d = -16 \, ^\circ \text{C}$, $v = 10 \, \text{m s}^{-1}$ ($a'$ and $a''$ are the reflected light and X-ray micrograph respectively); and TS22: $T_s = -22 \, ^\circ \text{C}$, $T_d = -16 \, ^\circ \text{C}$, $v = 10 \, \text{m s}^{-1}$ ($b'$ and $b''$ are the reflected light and X-ray micrograph respectively). The enlargement in $b''$ shows radial lines of bubbles following the channels between lobes and the rings of bubbles which follow the profile of the lobes.

has been checked whether the rotation rate also affects the sectorial concentration of lobes and lobe formation. This has been done by growing deposits at lower rotation rates (0.5 Hz), without finding any appreciable difference. Clearly a feature like the radial concentration of lobes is an important parameter in the analysis and interpretation of natural hailstones.

4. DISCUSSION

The extensive investigation of the two-stage growth and use of X-ray micrography for local density measurements has produced a great variety of results and a large amount of fresh material for discussion and interpretation. In our discussion we will follow the order of the presentation of the results.
Figure 9. Features indicating soaking in a $5 \text{ m s}^{-1}$ growth deposit in which the gradual transition is evidenced by the bubble rings following the shape of the dry growth lobes before the layer becomes definitely wet. Reflected light and X-ray micrograph, (a), and detail in reflected light of the transition from two-stage growth to wet external layer, (b). First stage growth temperatures are $T_s = -21^\circ \text{C}$, $T_d = -15^\circ \text{C}$. 
Figure 10. Reflected light picture and X-ray micrograph of a slice of a first stage low density deposit TS23. Growth conditions $T_a = -22^\circ C$, $T_g = -12^\circ C$, $v = 2.5 \, m \, s^{-1}$.

Figure 11. Reflected light pictures of the whole deposit showing the lobe arrangement. Deposits are TS18 (first stage growth conditions: $T_a = -26^\circ C$, $T_g = -22^\circ C$, $v = 10 \, m \, s^{-1}$) (a); and TS23 (b). The latter is the same deposit as in Fig. 10.
Figure 12. Number of lobes counted in a 90° sector of the slice as a function of impact velocity for the grown deposits. Other possible relevant parameters affecting the sectorial frequency of lobes were practically constant during growth of the different deposits.

(a) Local density measurements

The systematic application of X-ray micrography to local density determination in a wide range of experimental conditions confirmed the potential of this technique in accreted ice investigations and in natural hailstone interpretation, as proposed in the original paper (Prodi 1970). The technique allows us to go into greater detail, beyond the observation of the important density increase of even low density deposits due to soaking and final freezing of liquid water. The second stage density behaviour, Fig. 3, demonstrates that the final density may be slightly lower for first layers, grown at higher original density, than for first layers grown at lower density. This apparently paradoxical result is explained by the fact that the higher density first stage deposit functions as an efficient obstacle to penetrating water.

Where penetration occurs, capillarity and freezing from outside is highly efficient in forcing water and confining the air in a few bubbles, probably at higher pressure, producing a final bubble volume much smaller than the one originally occupied by the air. It is also possible that a fraction of the trapped air can escape to the outside through free channels. Only specific experiments on the internal pressure of air bubbles can solve this question.

It can also be deduced that the mode of water penetration is of some importance in this process. If penetration is slow the water at the front has time to freeze, dissipating the heat of freezing to the deposit already grown, thus forming a more efficient barrier to subsequent penetration. Figure 8 (deposit TS7) shows an example in which the mesh of tiny channels of the first stage growth is maintained, except between lobes in the outer zone where air gathers in larger bubbles.

The X-ray micrographs are helping to provide detailed images of microchannels, air bubbles and air cavities. Whereas a reflected light picture will only give information regarding the layer in focus, X-ray micrography gives integrated information through the whole slice. The air bubble configurations as observed in X-ray micrographs are commented upon in the morphology section of the discussion.
(b) Crystallographic parameters

(i) Size of grains. The increase in grain size can be interpreted as a result of the annealing effect taking place during second stage growth (or melting) and of the penetrating water which fills up the air gaps. This second mechanism explains also the penetrative structures observed in the replica which reveal areas of small crystals within lobes, maintaining the original crystal fabric, and areas of larger crystals between lobes.

We can discuss whether it is possible to differentiate between grain growth behaviour caused by melting, and wet growth penetration. In so doing we have to take into account that in the grain growth mechanism the larger the initial grain size, the smaller the increase will be. By applying the relationship \( D^2 - D_0^2 = kt \), with \( D \) and \( D_0 \) the final and initial average grain size, and \( t \) the time, we found that there is no evidence to suggest that the effect of grain growth is different when the second stage is due to melting or when it is due to wet growth penetration. Moreover, we observed that grain growth was less compared with that in bulk ice. In brief, the effect of the second stage is not much different in the various cases considered: it is more pronounced when the initial grain size is small and presents a large scatter in the data, as usually happens in such types of phenomena. It is interesting to note that in natural hailstones we do not generally find layers with \( \delta < 10^{-2} \text{mm}^2 \) since even if the layer was originally formed by small grains, they usually have enough time to grow to a larger size before reaching the ground.

(ii) Grain orientation. The data on grain orientation seem to confirm that it is the first stage growth which determines the final fabric, since we noticed \( \overline{\eta} \) values to be approximately unchanged. We might expect some variation in deposit TS3, not shown in Table 1, where the original first stage density is low. However, in this case we have no information about the first stage since etch pits are difficult to obtain in these conditions.

(c) Morphology

Two-stage growth was not found to reproduce perfectly transparent ice except possibly when the first stage deposit is at \( T_d \) close to 0 °C and the fabric is completely rearranged with an almost total rejection of air bubbles (a narrow layer in deposit TS20 in Fig. 6). Kidder and Carte (1964) examined a case of transparent ice obtained by the soaking of very low density ice \( (<0.3 \text{ g cm}^{-3}) \); we, however, did not conduct tests in these experimental conditions. Had we done so, we would expect very large radial bubbles or cavities to be present. Under our own conditions, the penetration effect causes the ice to become more translucent, rather than transparent.

The mechanism of dry growth lobe formation is clarified both here and in a companion paper (Prodi et al. 1986b) which investigates ice accretion on fixed cylinders, and we have found that lobes are formed only on rotating cylinders, not on fixed ones. Here the main observation is that channels are deeper and the lobes less radially frequent the lower the impact speed, other parameters being constant. This suggests that a surface irregularity screens a surrounding area from impinging droplets while the impact velocity modulates the extent of this area and consequently the radial frequency of lobes. However, an additional important parameter affecting lobe formation is the droplet size spectrum. The dependency on this parameter has not been investigated in detail since our size spectra did not change much from one test to another. Under identical conditions of other parameters, variations in the size of cloud droplets can produce dramatic changes in the nature of the accretion, including the sectorial concentration of lobes and their depth (Pflaum 1984).

In two-stage growth, the observed radial bubbles are a remnant of the original channels between lobes. Large radially oriented bubbles cannot originate from water
penetration into a high density dry growth first stage; on the contrary they offer evidence of a low density first stage with large and pronounced channels between lobes.

(d) Application to natural hailstones analysis

The evidence that two-stage growth may be a frequent occurrence is changing the criteria of natural hailstone interpretation: it introduces new criteria based on specific measurements and suggests various modifications of the criteria currently adopted. Here, however, we simply wish to focus on the former of these in so far as it helps to identify the features which indicate two-stage growth in natural hailstones.

We obviously do not expect to find natural hailstones reproducing all the features presented by freshly grown first stage layers. This is because they are generally collected at the ground and are always affected by melt water penetration, even if no wet growth has been superposed.

The observed cuspidation of dry growth lobes is an indication of an original first stage dry growth. The lower the original first stage density the more accentuated this feature becomes. In particular a translucent lobe exterior indicates effective penetration of liquid water.

The sectorial frequency of lobes is expected to provide additional information on the growth conditions once the relationship with the other parameters, such as droplet size and surface temperature, is completely clarified. To reach a complete definition of this dependency, further experimental work is needed over a wide range of variability of $\hat{r}$ and $T_e$.

Furthermore, local density results can also be used to infer criteria for hailstone analysis. There may be several different situations:

(i) Layer is at maximum density ($\rho > 0.905$ g cm$^{-3}$).

(a) It is an unmodified first stage growth layer. In this case there are again two possibilities: the first stage was grown dry or wet-spongy. If it has grown dry the appearance shows a diffuse opacity, unless $T_d$ was very close to 0°C. Hyperfine regularly spaced rings of bubbles would form in relation to the rotation rate (Prodi and Levi 1980b). Large radial bubbles are absent. If the layer was wet or spongy the ice is transparent and rings of bubbles peculiar to wet or spongy growth should be visible (Prodi and Levi 1980b). Other features such as fan-like air bubbles may be found in association with wet growth layers. Radial lines of bubbles are still absent.

(b) The layer under examination originated from a first stage modified by soaking. In this case we have a variety of possibilities within two main situations: soaking of a dry first stage or melting of a layer already at relatively high density. In the former case, we do not observe transparent ice and the radial features of bubbles will reveal the penetration of liquid water into channels. The occurrence of transparent ice due to penetration into very low density layers ($\rho < 0.3$ g cm$^{-3}$) (Kidder and Carte 1964) would be revealed by large air bubbles or cavities. Melting of a layer already at relatively high density (0.80–0.85 g cm$^{-3}$) would not produce such high values, as the deposits grown at 10 m s$^{-1}$ in Table 1 indicate. Melting of a layer at relatively low density (e.g. TS13, TS9) would produce ice of opaque–translucent appearance, without radial bubbles.

(ii) Density between about 0.82 and 0.905 g cm$^{-3}$.

In this case wet or high density dry first stage growth can be excluded. Again, two possibilities remain.

(a) It is an unsoaked layer grown dry in the first stage (e.g. deposits grown at 10 m s$^{-1}$). The appearance is opaque and internal cavities are not rounded into bubbles; but, as mentioned before, it is almost impossible to find such layers in natural hailstones because
of melting effects. Only direct sampling inside clouds could provide evidence of this type of growth in natural hailstones.

(b) The layer is a first stage modified by a second stage (leading to a limited increase in density). In this case, the lobes have a translucent smoothed appearance.

(iii) Low density, \( < 0.82 \text{ g cm}^{-3} \).

These densities are rather rare in natural hailstones and the explanation for this is given in the present work: penetration and soaking, whether due to a wet growth or melting, are efficient in considerably increasing the density of even very low density ice. The trapped air can be compressed into high pressure bubbles or can escape via channels to the outside. Exceptions to this behaviour may result from situations where the transition to wet growth is so gradual that the communication channels are sealed off by the freezing of the advancing water, thus hindering further penetration. Such is the case of the low density embryo reported by Prodi (1970) in his Fig. 5, where an exceptionally low density, for natural hailstone, of \( 0.7 \text{ g cm}^{-3} \) was measured.

Crystallographic observations in this paper agree with those in previous work and can be used in combination with other criteria (morphological, local density, etc.) to allow cross-verification, with a consequent improvement of hailstone analysis. We also add that the penetration and soaking process influences two other methods of investigation: the analysis of soluble and insoluble contaminants and the isotopic analysis. The difficulties found up to now in applying these methods will probably be reduced if evidence of soaking is independently provided and all the criteria are used in combination.

To conclude this section we present in Fig. 13 two examples of natural hailstones. They clearly indicate the overall dry growth outside the embryo region. The dry growth lobes are seen to affect almost the whole of the hailstone, and the translucent appearance of ice and the radial lines of bubbles indicate a final soaking.

5. CONCLUSIONS

In this work, additional evidence has been presented that two-stage growth (Prodi 1970) is an important mode of hailstone growth. However, only a statistical analysis of the structure of natural hailstones could confirm the validity of this assumption.

The origin of features such as cuspidated lobes, radial lines of bubbles, internal cavities and rounded air bubbles has been confirmed.

The first documentation is presented that soaking and subsequent freezing increase the density of the first stage of growth, even when initial densities are in the \( 0.7-0.9 \text{ g cm}^{-3} \) range. In these cases the penetration due to the second stage growth is small, but still produces some modification of the first stage growth layer.

For ice with density near \( 0.9 \text{ g cm}^{-3} \) it appears possible to differentiate between one- and two-stage growth.

These conclusions have been reached on the basis of morphological and crystallographic observations and, above all, by local density measurements obtained by X-ray contact micrography. This technique enables us to record both the considerable modification of the fabric in artificially accreted ice as well as the minor density variations, close to the maximum value, in natural hailstones, crucial for their correct interpretation and analysis.

The experimental conditions tested in this work, though not completely covering all possible cloud conditions do cover the most common ones and lead to a number of unequivocal features and criteria for interpretation.

New criteria, mainly based on the local density determination in hailstone layers,
Figure 13. Examples of natural hailstones, collected from a storm near Verona, in the Po Valley of northern Italy, showing two-stage growth revealed by the lobe array, the translucent ice and the radial lines of bubbles. Scale is 1 cm for both photographs (inner tick marks 0.5 cm).
have been formulated in order to identify two-stage growth and, combined with other analytical methods, are of considerable help in hailstone interpretation.

Consequences of two-stage growth on hail cloud models and hail prevention have been outlined by Pflaum (1980) and we agree with them. Essentially, the indication (Prodi 1970) that two-stage growth lessens the updraught intensity needed to sustain the hailstone, due to both the lower density and higher drag because of the increased surface roughness, is at the root of all possible effects.

This area of research is open to further experimental analysis, in order to provide further details and quantify some observations, within the general framework described herein. The main points to be investigated appear to be:

The role of cloud droplet size distribution in determining the density of accreted ice should be investigated by performing additional experiments over a wider range of this parameter. We are even considering the possibility of accretion experiments with mono-disperse water droplets.

The role of actual soaking conditions of porous ice (mainly, the deposit temperature and the latent heat that can be immediately transferred to the hailstone structure). At deposit temperatures close to 0 °C, the soaked structure could completely redistribute the enclosed air causing transparent ice to form.

Possible annealing effects on air bubbles during storage, which could change the internal appearance and, if observed, should be taken into account in the analysis.

The pressure inside large bubbles: since the final freezing of a soaked structure originates from the outside, air could become trapped within the final air bubbles, causing pressure inside to rise. This process could also explain why cracks and internal fractures are so rarely observed in hailstones, in spite of the high pressure produced by the freezing of soaked water.

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