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**THIN ACCRETIONS GROWN ON AN ICE SUBSTRATE : EXPERIMENTAL STUDY AND NUMERICAL SIMULATION**

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**RESUME.** On étudie la structure de couches minces de glace, formées sur un substrat basal et prismatique. On observe que, à toutes les températures au-dessus d'une température de transition, les nouveaux cristaux se développent uniquement sur le substrat prismatique. Au-dessous de la température de transition ( $T_a \sim T_s \sim -18^\circ\text{C}$ ) beaucoup de petits cristaux se forment sur les deux substrats. Un modèle de simulation numérique, utilisant comme paramètres les probabilités de nucléation de nouvelles orientations dans les gouttes et les vitesses de croissance latérales des cristaux a été proposé. Il permet de décrire les variations avec la température de ces paramètres et de la structure des dépôts.

**ABSTRACT.** The structure of thin sheets of ice accreted on a basal and prism substrate is studied. It is observed that, at all temperatures above a transition value, new crystals develop only on the prism substrate. Below the transition temperature ( $T_a \sim T_s \sim -18^\circ\text{C}$ ) many small new crystals are formed on both substrates. A numerical simulation model is build up, where the nucleation probability in the droplets and the lateral crystal growth speeds are used as parameters and a good correlation is found between the variations with the temperature of these parameters and of the deposit structure.

## 1 - Introduction

In a previous work<sup>(1)</sup>, thin accretions formed by riming on a prismatic ice substrate were studied, in order to obtain a direct evidence of the progressive changes that occur in the accretion structure at the beginning of the process. On the basis of the results obtained a qualitative explanation of the structure of wind tunnel ice accretions grown in dry regime, was given. Considering that a more quantitative analysis of the phenomenon could be of interest, in the present work, the structure of thin accretions has been more completely studied, using prismatic and basal ice substrates.

The results obtained have been applied to formulate a computational model where the phenomenon is simulated on the basis of assumptions about the processes of crystal nucleation and growth derived from experimental data. Several simulation experiments, corresponding to different growth conditions, have been carried out.

## 2 - Experimental methods and results

The samples were single-crystalline ice sheets, about 1mm thick and 1x1cm in section, cut according to a plane normal to or containing the c-axis. They were introduced in the working section of the icing wind tunnel (Institute FISBAT-C.N.R., Italy)<sup>(2)</sup>, supported by a sample holder, which could be electrically warmed up, so to obtain higher values of the surface than of the air temperature, i.e.  $T_s > T_a$ . The droplets injected in the tunnel had a mean volume diameter  $d=30\mu\text{m}$  and the water received on the collector surface corresponded to a flux of  $N_g$  droplets with  $N_g = 800\text{mm}^{-2} \text{ s}^{-1}$ . The time  $t$  during which an experiment was carried out was varied between 1s and 60s, so obtaining rime thicknesses in the range from  $15\mu\text{m}$  to 1mm (about 5 to 300 droplet layers).

Formvar replicas of the accreted surface, obtained immediately after each experiment, were analysed in the microscope. Replica analysis was used to determine: 1) the

number of crystals per unit area with an orientation different from that of the substrate ( $N$ ), from which the mean number of such crystals per droplet,  $N_c = N/tNg$  was derived; 2) the mean crystal area  $\sigma$ ; 3) the fraction  $H$  of the sample surface covered by the new crystals. When thermal etch pits were well defined, as it usually occurred for crystals more than  $100\mu\text{m}$  in size, the measurements included those of crystal orientation.

Notice that, if all the crystals created along the process would grow up, reaching the surface,  $N_c$  would be constant as far as the conditions are not altered. In most cases, however, while the deposit thickness increases, a fraction of the crystals created below the final surface disappears, hindered by superposed nucleations and by lateral crystal growth. In this case,  $N_c$  would usually decrease with increasing time, varying as  $1/t$  if the disappearance and creation of new crystals compensate.

Representative results are plotted in Fig.1 and 2. The diagrams in Fig.1a and 2a show the behaviour on the prismatic substrate. Three different cases are considered. Case 1 corresponds to  $T_a \sim -16^\circ\text{C}$ ,  $T_s \sim -4^\circ\text{C}$ . At these relatively high temperatures  $N_c$  is always small ( $N_c \leq 0.01$ ). Notwithstanding, after 15s, it is found  $H=100\%$ , this effect

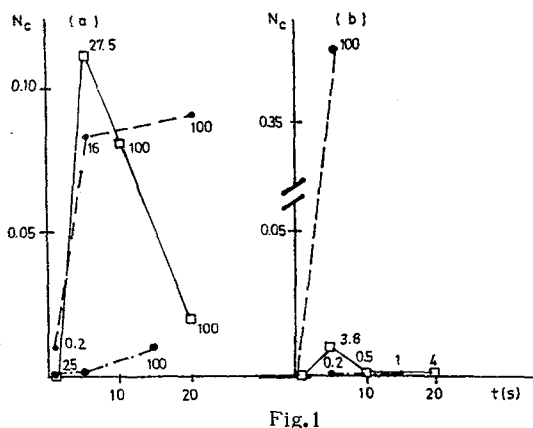


Fig.1 New Crystal number per droplet as a function of  $t$ .

---  $T_a = -16 \pm 1^\circ\text{C}$   $T_s = -4 \pm 1^\circ\text{C}$  —  $T_a = -20 \pm 1^\circ\text{C}$   $T_s = -3 \pm 1^\circ\text{C}$  - - -  $T_a = -18 \pm 1^\circ\text{C}$   $T_s = -17 \pm 1^\circ\text{C}$   
At the side of each point  $H\%$  is indicated.

(a) Prism substrate

(b) Basal substrate

being due to the rapid lateral growth of the new crystals, as shown by the corresponding curve of  $\sigma$  in Fig.2a. Case 2, corresponding to  $T_a \sim -20^\circ\text{C}$ ,  $T_s \sim -3^\circ\text{C}$ , differs from case 1 mainly because of the lower value of  $T_a$ . It is characterized by an initial larger value of  $N_c$ , being  $N_c \sim 0.1$  at  $t=5s$ . For larger accretion times, however, the value of  $N_c$  decreases according to a trend which, for  $t > 10s$ , has a larger slope than  $1/t$ . In fact, during the last part of the process, the number  $N$  of crystals on the surface decreases, as it results from the increase of their area occurring, as shown in Fig.2a, even after the condition  $H=100\%$  is reached. From the values of  $\sigma$  at  $t=5s$  it may be derived that, in case 1 and 2, the lateral crystal growth varied between 10 and  $20\mu\text{m/s}$ . Case 3, corresponding to  $T_a \sim -18^\circ\text{C}$ ,  $T_s \sim -17^\circ\text{C}$ , differs from previous ones mainly for the low value of  $T_s$ . In this condition, it is found  $N_c \sim 0.1$  at  $t=5s$ , as in case 2, but the decrease of  $N_c$  in the subsequent 15s time interval is not observed. This behaviour indicates that the process now controlling the phenomenon is the continuous formation of new crystals, which progressively cover the whole surface, despite their low development in size, shown in Fig.2a.

A quite different behaviour was observed for deposits grown on the basal substrate as shown in Fig.1b and 2b. It may be seen in Fig.1b that, when on this substrate the temperature conditions are the same as in cases 1 and 2 described for the prism substrate, both  $N_c$  and  $H$  remain small at all times. An abrupt change occurs, however, in the conditions of case 3, where  $N_c$  reaches, during the first 5s, a value markedly above that on the prism substrate. The values of  $\sigma$  indicated in Fig.2b show that, on

this substrate the crystal area usually did not increase with  $t$ , remaining in most cases near  $500\mu\text{m}$ . An exceptionally large mean area is reached in case 2 after 20s. It must be noted, however, that the few large crystals observed in this case were oriented at small angle to the substrate crystal and only covered a 4% of the substrate surface.

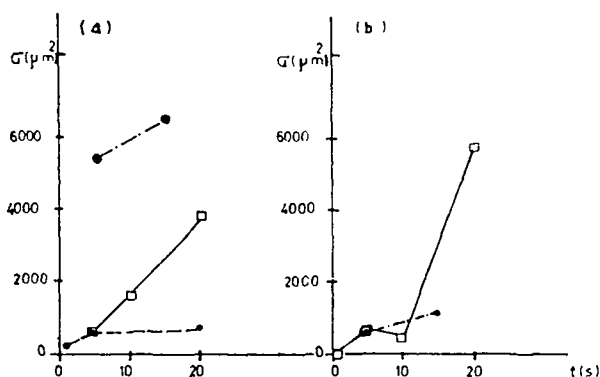


Fig.2

New crystal mean area  $\sigma$  as a function of  $t$ ;  $T_a$  and  $T_s$  and  $H^\circ/\%$  as in Fig.1.  
(a) Prism substrate (b) Basal substrate

### 3 - Numerical simulation model

The results described in the previous section have been used to build up a numerical model where the evolution of the accretions during their initial growth is simulated.

The numerical simulation was performed assuming the following hypothesis: 1) the droplets impinge on the substrate forming successive uniform layers, once each  $t_p$  seconds; 2) at the initiation of each  $t_p$  interval, the nucleation process may occur inside the droplets; this is followed by the solidification process, where adjacent crystals may change its transverse section according to their relative growth rate.

In order to perform the calculations, the substrate was represented by a grid of  $70 \times 70$  meshes, each mesh representing an elemental crystal of  $15\mu\text{m}$  side. To each mesh two numbers were assigned, representing the crystal orientation and its stay time in this orientation. In this way two different matrices were defined. The first one gives the structure configuration, where each mesh corresponds to a given orientation and, consequently, to given values of the nucleation probabilities  $P_i$  and of the lateral growth rates  $v_i$ . The second one permits to establish when a grain boundary can move from one to an other mesh, according to the lateral growth rate relative to the adjacent crystal. When a new crystal is nucleated, its orientation, represented by the angle  $\Psi$  between its c-axis and the growth direction, is selected at random assuming a senoidal random distribution.

The crystal were grouped in 5 classes, according to the value of  $\Psi$ . The interval  $0^\circ \leq \Psi \leq 60^\circ$  was divided into 4 classes, each  $15^\circ$  wide. Taking into account that the experimental error in the evaluation of  $\Psi$  increases with the angle, being of the order of  $10^\circ$  for  $\Psi > 60^\circ$ , in class 5 the whole  $60^\circ$ - $90^\circ$  interval was included. Notice that, in the conditions considered here, most crystals generated in this last class disappeared during growth, so that, in the results, only classes 1 to 4 will be usually indicated.

The program permitted to follow the evolution with time of the number of new crystals belonging to each class. It was executed by assigning to the nucleation probabilities  $P_i$  per elemental crystal and to the lateral growth rates  $v_i$  values compatible with the results in section 2. The values of the  $P_i$  were derived from those of the  $N_c$ . The evaluation of  $v_i$  was performed by deriving a mean value  $\bar{v}$  from the experimental results and by assuming that the maximum growth rate  $v_m$  occurred in the direction of dendritic growth. Thus, the different  $v_i$  were obtained as the projections of  $v_m$  on

each crystal surface. The crystallographic orientation of ice dendrites was selected by taking into account the fact that the angle  $\varphi'$  between the dendrites and the basal plane varies inside the interval  $0^\circ$ - $25^\circ$  with increasing supercooling (3,4). Some examples of the results obtained on the prism substrate are given in Fig.3, 4.

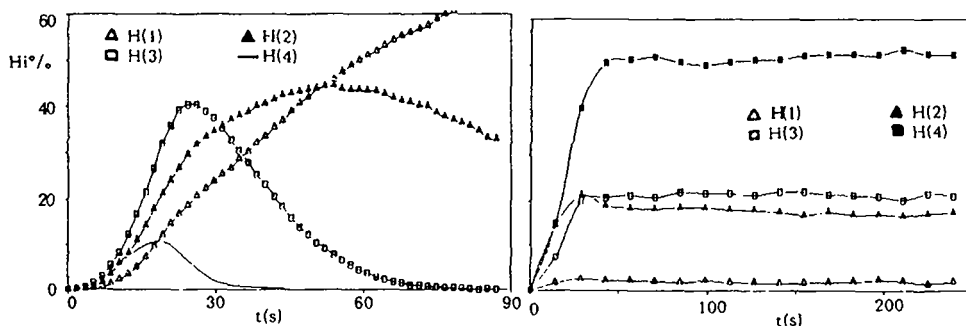


Fig.3

Curves of the  $H_i(t)$  for: a)  $P_i=0.0009$ ,  $v=15\mu\text{m/s}$ ,  $\Psi'=0^\circ$ ; b)  $P_1=0.09$ ,  $P_2=0.07$ ,  $P_3=0.05$ ,  $P_4=0.01$ ,  $P_5=0.02$ ,  $v=5\mu\text{m/s}$ ,  $\Psi'=25^\circ$

Fig.3 gives the evolution of the area covered by new crystals of classes 1 to 4, in two different conditions. Fig.3a corresponds to  $\bar{v}=15\mu\text{m/s}$  and  $\Psi'=0^\circ$ . The probabilities  $P_i$  are small ( $\sim 10^{-4}$ ) and do not vary with the class. This example may be considered to correspond to case 1 in Fig.1 and 2; it represents the phenomenon as it occurs at relatively low  $\Delta T$ . It may be seen that crystals of all orientations appear at the beginning but that, after 60s, those having the maximum lateral growth rate (class 1) predominate. During the last period, crystals of this class increase at the expenses of crystals of class 2. Fig.3b corresponds to  $\bar{v}=5\mu\text{m/s}$ ,  $\Psi'=25^\circ$ . The probabilities  $P_i$  have been assumed to be in the average nearly two orders of magnitude above that in the previous example and to vary with the class as it occurred in case 3, where  $N_c$  was larger on the basal than on the prism substrate ( $P_1 > P_5$ ). In the simulation experiment, the maximum value has been assigned to  $P_1$  and the minimum one to  $P_4$ , that is to the class containing the crystals with  $\varphi=45^\circ$ . In this way, previous results have been taken into account, according to which the lowest value of the transition temperature where the probability of nucleation per droplet increases rapidly with  $\Delta T$ , would be that corresponding to a substrate with this orientation. It may be seen in the figure that, in these conditions, class 4 rapidly predominates, despite the relatively small value of  $v_4$ . Actually the development of crystals with this orientation is not hindered by the high frequency of new nucleations as it occurs for crystals with other orientations. The lateral development of crystals, in the case of Fig.3a, is shown in Fig.4, where the printed matrix of the corresponding structure configuration is given at the beginning of the process and a few seconds later.

The examples given in Fig.3a and b represent two extreme cases respectively corresponding to relatively high temperatures, where the  $P_i$  are small and the dendrites are basal (case 1) and to relatively low temperatures, where the  $P_i$  are large and the dendrites depart markedly from the basal plane.

Other simulation experiments were performed for values of the  $P_i$  between  $10^{-2}$ ,  $10^{-3}$  and  $\Psi' < 20^\circ$ . In general, the  $H$  curves, though similar to those in Fig.3a, were characterized by larger values of the time  $t_e$  where the final prevailing orientation (class 1) was established. This is shown in Table 1, where  $t_e$  varies from about 2 minutes to about 7 minutes for  $\Psi'$  increasing from  $0^\circ$  to  $15^\circ$ .

Table 1

Time  $t_e$ , for  $P_1=0.02$ ,  $P_2=0.01$ ,  $P_3=0.009$ ,  $P_4=0.008$ ,  $P_5=0.009$ ,  $v=5\mu\text{m/s}$  and different values of  $\psi'$ .

$\psi' (^{\circ})$	0	10	15
$t_e$ (min)	1.8	4.9	7.

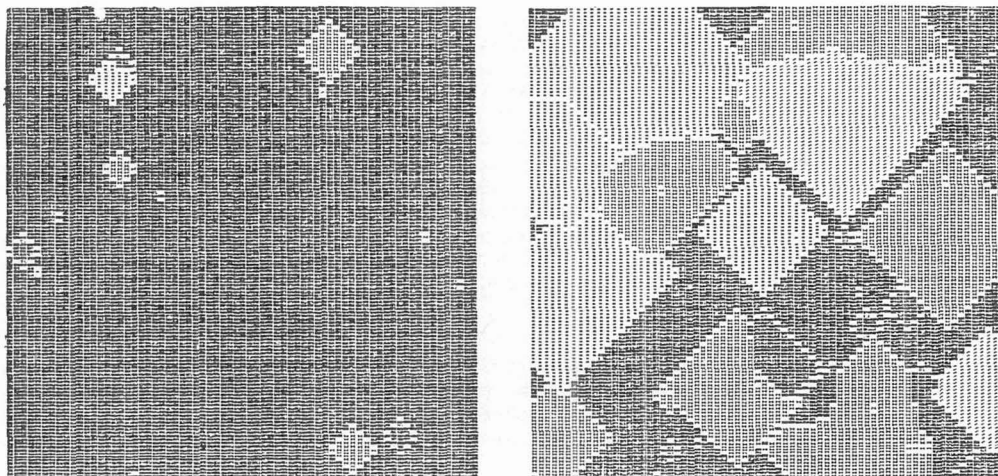


Fig.4  
Configuration structure, for the example in Fig.3a,  
at  $t=2.5\text{s}$  and  $t=12.4\text{s}$

Notice that the cases considered in columns 2 and 3 of the table could correspond to the conditions that determined case 2 in Fig.1,2. In this case, most growth could have occurred in conditions where the difference between  $H_1$  and  $H_2$  was small so that the maximum of the  $\Psi$  distribution could have been located in one or other of the corresponding classes.

Simulation experiments were also performed assuming a basal substrate. In this case, when the conditions were as in Fig.3a the development of new crystals was strongly hindered by the lateral growth of the substrate itself. For conditions similar to those in Fig.3b the effect of the initial substrate orientation in the structure evolution was small, due to the large value of the nucleation probabilities  $P_i$ , so that basal and prism substrates could not give a markedly different behaviour.

#### 4 - Discussion and conclusions

From the analysis performed in the present work, values have been found for the probability of nucleation of new crystals in the freezing droplets, which are in satisfactory agreement with those obtained previously for droplets colliding separately on similar substrates (5). It could be noted, on the contrary, that the values found for the lateral crystal growth rate are about two orders of magnitude smaller than those which could be derived from dendritic growth occurring at similar water supercoolings. One of the causes of this may be the fact that we have treated here the process of lateral growth as if it were a continuous process, while, when accretion occurs in dry regime, dendritic growth, which would be the responsible of the phenomenon, would only take place during initial droplet freezing (6) which is a very small fraction of the total time of droplet freezing.

It is also interesting to observe that the application of the simulation model gives the possibility of including in the discussion the characteristics of large accretions which have been grown on rotating and fixed cylinders to simulate hail growth and other glaciation phenomena (7). At this respect, the results obtained here suggest the convenience of considering separately three different temperature ranges:

1) The range of relatively high temperatures (approximate limits  $T_a > -18^\circ\text{C}$ ,  $T_s \geq -5^\circ\text{C}$ ) where the mechanism that mainly determines the crystal structure is the lateral crystal growth, with maximum rate in the basal plane. In these conditions the crystals which form the deposit may be several millimeters in size and are mainly oriented with their c-axes in the growth direction.

2) A range of intermediate temperatures (approximate limits  $-22 \leq T_a \leq -18^\circ\text{C}$ ,  $-10 < T_s < -5^\circ\text{C}$ ) where the values of the nucleation probabilities increase slightly and the predominant role played by the basal orientation becomes less pronounced. In these conditions the crystal size decreases and the maximum of the distributions for the angle  $\varphi$  is not well defined because the difference between class 1 and class 2 is small.

3) A range of low temperatures (approximate limits  $-27 \leq T_a \leq -20^\circ\text{C}$ ,  $T_s \leq -10^\circ\text{C}$ ) where the nucleation probabilities increase rapidly, at first on the basal substrate and, at slightly lower temperatures, on substrates with other orientations. Since the transition temperature where the nucleation probability increases sharply is lower for a substrate at  $45^\circ$  to the basal substrate than for other orientations, a temperature interval exists where only this substrate is not rapidly covered by a large number of new crystals. In these conditions the crystals are small (50-100  $\mu\text{m}$  mean size) and the frequency distributions of  $\varphi$  present a well defined maximum at about  $\varphi = 45^\circ$ .

It may be expected that, at lower temperatures, both the nucleation probabilities and the lateral growth rates will become independent of the crystal orientation, so that the crystal orientation would remain at random.

## 5 - Acknowledgements

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## COMMENTS

T. KURODA

Could you explain the difference of lateral growth velocity between basal and prismatic surfaces in terms of the difference of contact angles on both faces ?

Answer :

The lateral growth speed is assumed to be maximum when the surface of the crystals coincides with the place of dendrites. In other directions a projection is done of the velocity in the dendrite plane.