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RÉSUMÉ

La sublimation de la glace dans les milieux poreux pose des problèmes dans un certain nombre de domaines de l'ingénierie. Ce document expose les fondements théoriques nécessaires pour décrire mathématiquement les phénomènes en jeu, fournit des données sur les vitesses de sublimation de la glace pure et décrit brièvement un modèle numérique de calcul des vitesses de sublimation dans des conditions non isothermes.

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Experimental measurements and a numerical method for ice sublimation

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Abstract

Problems involving sublimation of ice in porous media arise in a number of engineering contexts. This paper reviews the theoretical basis necessary for a mathematical description of the phenomena concerned and presents data on sublimation rates for pure ice. A numerical model for computing sublimation rates in non-isothermal conditions is described briefly.

Introduction

In response to concerns of the Food Engineering Industry, on one hand, and Energy storage and Artic Engineering interest, on the other hand, during the last five years the "Laboratoire d'Aérothermique du C.N.R.S" has studied sublimation problems in ice and in frozen dispersed media.

In fact, sublimation studies carried out to date have been largely concerned with biological porous media in the context of freeze drying. Consequently, research has, for the most part, concentrated on sublimation at low pressures |1,2| But in order to reduce the high cost associated with such drying procedures new techniques are constantly being sought, and a potentially interesting avenue is offered by freeze drying at atmospheric pressures.

Moreover, sublimation of free ice or ice contained in porous media arises in certain Civil Engineering problems. In particular, a problem of present day interest is that of liquid gas storage at atmospheric pressure in underground cavities where the thermal and mechanical behaviour of the frozen cavity walls, constituted by an ice layer on frozen soil or rock, requires the study of the sublimation process acting upon the surfaces exposed above the liquid gas in the reservoir during very long periods |3|.

Sublimation problems also arise in the context of the dehydration of stored frozen food-stuffs |4| of medical substances, the thermal balance and thermal regime in permafrost and at natural ice surfaces, the thermomechanical behaviour of structures in arctic conditions, the behaviour of interstellar ice, etc.

A considerable amount of experimental data has already been obtained and we now can provide information concerning the fundamental knowledge of ice as well as certain aspects useful for pratical applications.

Starting from general physical and mathematical models of the sublimation process, we suggest in this paper simplifications and adaptations required to interpret the experimental data and we indicate the most urgent, problems remaining to be investigated. A numerical model for general sublimation problems is presented and, finally, a new experimental device conceived to study ice-sublimation phenomenon in the absence of convection and under very low temperatures and relative humidities is described briefly.

Experimental study

In order to study the sublimation problem in the laboratory, an insulated tunnel was constructed where temperature T, velocity V and relative humidity RH of the cooled air flowing in a closed circuit could be regulated.

The tunnel is composed of four linear sections placed in a vertical plane. The 20cm x 20cm test section is located in the upper horizontal portion and is preceded and followed by a convergent and a divergent system respectively, used to ensure suitable air flow conditions around a vertical cylindrical sample undergoing sublimation. The samples of approximate dimensions 7cm high by 2,6cm diameter are placed between two insulated metallic supports, one of which is removable to permit the sample to be weighed periodically in order to follow the dehydration process.

Details of the experimental device are given in previous papers |5,6|. The range of conditions which can be explored is as follows : $0,5 < V < 4\ m\ s^{-1}$, $243\,K < T < 264K$, $30 < RH < 90\%$.

Ice, vegetal and animal tissues (potatoes, meat) and a biological simulation substance (methyl-cellulose) have been studied so far |6,7|. Experimental data is recorded and reduced to various types of diagrams the most useful of which give the sublimation flux vs.velocity, relative humidity and temperature. An example is shown in figure 1 which corresponds to a set of tests made on pure ice.

A discussion of various fundamental and applied aspects of the previous research is contained in several published papers |8,9|. The information obtained so far, for pure ice makes it possible to establish an empirical relation ship between the sublimation flux and the air velocity and temperature. Calculations were made for several values of relative humidity but here we mention only the results for 70% RH.

To illustrate the type of relation ship obtained using the well known method of separation of variables, the data were fitted to curves of the form

$$i = A + B.V \tag{1}$$

where $A = \exp -(24.2932 + 0.0989\ T)$
and $B = 1.013^{T}.(-1.2689 + 5.4138.10^{-3}\ T)$
This equation correlates quite well with the experimental data. Most of the results lie within a few percent of the corresponding curves (Fig. 1). The values for 258K diverge, however, by as much as 18% and appear to be anomalous |10|.

Theoretical

The mechanism governing the sublimation of ice in frozen dispersed media have been fully discussed elsewhere |7,8| and will be only briefly described here. Physically, the sublimation process arises when molecules within a solid arrive at the surface with a thermal energy sufficient to allow their departure to the surrounding medium, a balance being established between molecules departing and those returning. Sublimation is than on-going process which can occur at any temperature below the melting point of the solid. The vapour produced is removed by vapour transfer processes such as diffusion and convection while the rate of vapour production is conditionned by the thermal conditions imposed. The process is thus one of simultaneous heat and mass transfer in the presence of a non constant space and time dependent vapour source which consumes part or all of the solid ice phase. Mass transfer rates are determined by moisture and temperature gradients as well as by the thermophysical properties of the dispersed medium.

While, in frozen soils or biological materials, the fact that the ice present is dispersed throughout the material matrix modifies somewhat the final result, nevertheless the case of sublimation from a pure ice surface is fundamental to understanding the more complex problem and its study may be justified on these grounds alone, aside from its interest for the applications already noted.

Mathematical statement

A three-zone general model for sublimation within frozen dispersed media has been proposed earlier |8,9|. This model is governed by simultaneous heat and mass transfer with a moving transition zone separating the dehydrated from the non affected zone. The equations have poorly known non-linear thermophysical coefficients and source and sink terms which make the model difficult to apply. In this paper we study the case where the sublimation transition zone can be reduced to a moving front and we consider the extreme case of dispersed media, namely that of pure ice. Assuming the diffusion of vapour and heat are governed respectively by Fick and Fourier's laws, the set of equations can be written for the ambient medium :

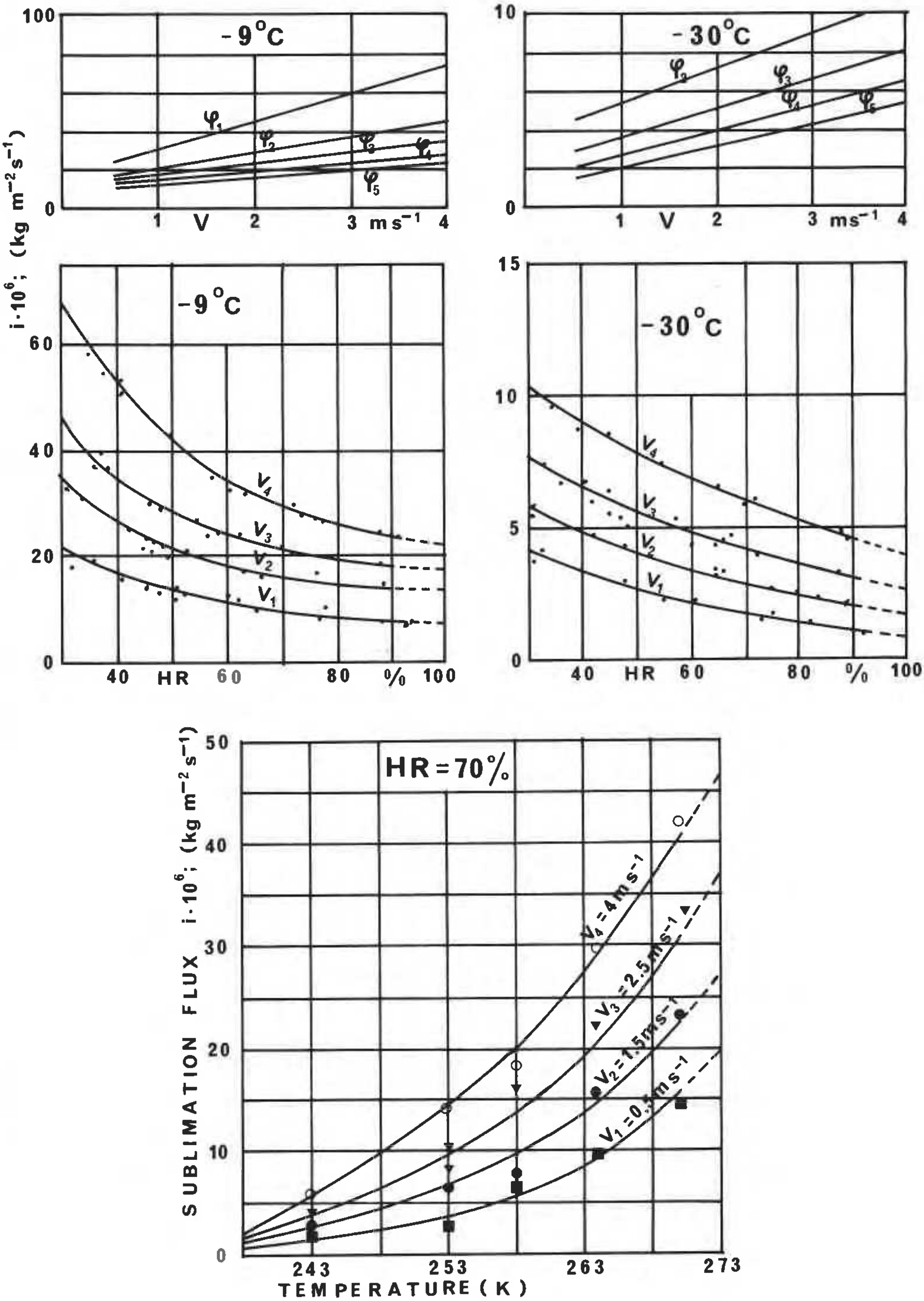

Figure 1 : Ice sublimation flux vs velocity, relative humidity and temperature of the air

$$\rho_a c_a \frac{\partial T}{\partial t} - \frac{\partial}{\partial x}[\lambda_a \frac{\partial T}{\partial x}] = 0 \quad , \qquad (2)$$

$$\rho_a \frac{\partial \omega}{\partial t} - \frac{\partial}{\partial x}[D_{va} \frac{\partial \omega}{\partial x}] = 0 \quad , \qquad (3)$$

and for ice :

$$\rho_i \, c_i \frac{\partial T}{\partial t} - \frac{\partial}{\partial x}[\lambda_i \frac{\partial T}{\partial x}] = 0 \quad , \qquad (4)$$

$$\rho_i \frac{\partial \omega_i}{\partial t} = 0 \quad , \text{(no vapor transfer)}, \qquad (5)$$

where ρ_a, ρ_i and λ_a, λ_i are the density and thermal conductivity of air and ice respectively, D_{va} the coefficient of vapour diffusion in air, c_a and c_i the heat capacity of air and ice respectively, ω the mass per unit volume of moist air, T the temperature, x the space coordinate and t the time.

The model does not formally take into account forced convection at the interface. This can be done, however, using the characteristics of the diffusion boundary layer corresponding to the velocity considered.

At the moving interface, $x = S(t)$, vapor is produced at a rate given by

$$I = (\rho_i - \omega_F) \frac{dS}{dt} \qquad (6)$$

where ρ_i and ω_F are the mass per unit volume of ice and water vapour at the interface.

In general, the sublimation flux term contains contributions arising from simple diffusion in air I_d, from convective processes I_c, as well as a term I_f to account for possible ablimation, and we have

$$I = I_d + I_c - I_f \qquad (7)$$

Straight forward mass and heat balance considerations at the interface yield :

$$D_{va} \left. \frac{\partial \omega}{\partial x} \right|_{x = S} = (\rho_i - \omega_F) \frac{dS}{dt} \qquad (8)$$

and

$$\lambda_i \left. \frac{\partial T}{\partial x} \right|_{x = S} - \lambda_a \left. \frac{\partial T}{\partial x} \right|_{x = S} = L_{i/v} . I \; (9)$$

In addition, vapor saturation is assumed to prevail at the interface and a relation between saturation vapor content and temperature is assumed known in order to completely specify the problem. Diffusion is taken to be through a stagnant air film constituting the diffusion boundary layer. The mixture of dry air and water vapour is assumed to obey the ideal gas law.

Numerical method

Equations (2) to (5) can be discretized using either finite element or finite difference procedures. Taking linear basis functions with lumped capacitance matrix terms in the finite element formulation or considering the heat balance over a finite volume surrounding a node in the finite difference formulation, leads to the following set of nodal equations for the regions above and below the moving interface :

$$\frac{1}{2}.(x_{i+1} - x_{i-1}).(W_i^{n+1} - W_i^n) =$$

$$D_{m-1}^n.[\theta.(W_{i-1}^{n+1} - W_i^{n+1}) + (1-\theta).(W_{i-1}^n - W_i^n)] \quad (9a)$$

$$D_m^n.[\theta(W_{i+1}^{n+1} - W_i^{n+1}) + (1-\theta).(W_{i+1}^n - W_i^n)] \quad (9a)$$

for nodes $i \leqslant iu-1$

and

$$\frac{1}{2}.(S_{m-1}^n + S_m^n).(T_i^{n+1} - T_i^n) =$$

$$K_{m-1}^n.[\theta.(T_{i-1}^{n+1} - T_i^{n+1}) + (1\ \theta).(T_{i-1}^n - T_i^n)] +$$

$$K_m^n.\ [\theta.(T_{i+1}^{n+1} - T_i^n) + (1-\theta).(T_{i+1}^n - T_i^n)] \quad (9b)$$

for nodes $i \leqslant iu-1$ and $i \geqslant id+1$, where iu and id are the nodes delimiting the element m_f containing the moving sublimation front, W_i^n and T_i^n are the nodal values of moisture concentration and temperature respectively at time-level n, and the remaining material and mesh parameters are defined in figure 2.

In eqns (9a) (9b) material properties may be evaluated at time-level n to avoid nodal iterations, this being adequate in most cases. Time-stepping is controlled by the parameter θ where :

$$\frac{1}{2} \leqslant \theta \leqslant 1$$

The moving interface is treated by a local moving-mesh method in which the element containing the moving interface is temporarily subdivided into two sub-elements with distinct material properties. Geometrical non-linearly is confined to the moving interface element only and the original mesh definition is retained throughout the calculation.

$X_{iu-1}, H_{iu-1}, T_{iu-1}$ ———————————— iu-1

$$S_{mf-1} = c.(x_{iu}-x_{iu-1})$$

$$K_{mf-1} = \Delta t.k/(x_{iu}-x_{iu-1}) \qquad \text{element } m < m_f$$

$$D_{mf-1} = \Delta t.D/(x_{iu}-x_{iu-1})$$

X_{iu}, H_{iu}, T_{iu} ———————————— iu

$$S_{mfu} = c_u.(x_{id}-x_{iu})$$

$$g = \frac{x_f-x_{iu}}{x_{id}-x_{iu}} \qquad K_{mfu} = \Delta t.k_u/(x_{id}-x_{iu})$$

$$D_{mfu} = \Delta t.D_u/\ (x_{id}-x_{iu})$$

X_f, H_f, T_f - element m_f

$$S_{mfd} = c_d.(x_{id}-x_{iu})$$

$$K_{mfd} = \Delta t.kd/\ (x_{id}-x_{iu})$$

X_{id}, H_{id}, T_{id} ———————————— id

$$S_{mf+1} = c.(x_{id+1}-x_{id})$$

$$K_{mf+1} = \Delta t.k/(x_{id+1}-x_{id}) \qquad \text{element } m > m_f$$

$X_{id+1}, H_{id+1}, T_{id+1}$ ———————————— id+1

Figure 2 : Définition of mesh parameters

Similarly to equations (9a) (9b), heat and mass balance equations at nodes iu and id can be written :

$$\frac{1}{2}[x_{iu}-x_{iu-1}+g^{n+1}.(x_{id}-x_{iu})].(W_{iu}^{n+1}-W_{iu}^{n}) =$$

$$D_{mf-1}.(W_{iu-1}^{n+1}-W_{iu}^{n+1}) + D_{mfu}.(W_f^{n+1}-W_{iu}^{n+1})/g^{n+1} \tag{10a}$$

$$\frac{1}{2}[S_{mf-1}+g^{n+1}.S_{mfu}]\ .(T_{iu}^{n+1}-T_{iu}^{n}) =$$

$$K_{mf-1}.(T_{iu-1}^{n+1}-T_{iu}^{n+1}) + K_{mfu}.(T_f^{n+1}-T_{iu}^{n+1})/g^{n+1} \tag{10b}$$

and $\frac{1}{2}\ [S_{mf+1}+(1-g^{n+1}).S_{mfd}]\ .(T_{id}^{n+1}-T_{id}^{n}) =$

$$K_{mfd}(T_f^{n+1}-T_{id}^{n+1})/(1-g^{n+1})+K_{mf\ +1}.(T_{id+1}^{n+1}-T_{id}^{n+1}) \tag{10c}$$

where backward time differences have been used for simplicity.

At the moving interface, equations (8) and (9) correspond to :

$$(x_{id}-x_{iu})(W_o-W_f)(g^{n+1}-g^n) =$$

$$D_{mfu}.\frac{(W_{iu}^{n+1}-\ W_f^{n})}{g^{n+1}} \tag{11a}$$

and

$$(x_{id}-x_{iu}).W_o.L.(g^{n+1}-g^n) =$$

$$K_{mfu}.\frac{(T_{iu}^{n+1}-T_f^{n+1})}{g^{n+1}} + K_{mfd}.\frac{T_{id}^{n+1}-T_f^{n+1}}{1-g^{n+1}} \tag{11b}$$

Equations (10) and (11) form a simple type of local moving mesh method in which the mesh deformation is confined to a single element. A solution technique originally devised for the heat solution equation with discrete phase change |11| can, with suitable modifications, be extended to the present case. This technique involves the simultaneous use of both forward and backward Gaussian elimination formulations for tridiagonal matrices, given a relation between ω_f and T_f, equs (10) and (11) can then be expressed entirely in terms of known coefficients. The primary non-linearity of the problem is thus isolated and, for constant or sufficiently slowly varying material properties, an iterative solution technique is required only for the interface element equations. Having computed g^{n+1}, T_f^{n+1}, W_{iu}^{n+1}, T_{iu}^{n+1} and T_{id}^{n+1} the remaining nodal temperatures and moisture contents can be calculated directly by back substitution using the appropriate Gaussian elimination coefficients. As a result, the method is very efficient and, because it is a moving mesh formulation, the accuracy is excellent and is relatively insensitive to the size of mesh spacing used, in contrast with the more usual methods.

A complete description of this method is given in a separate paper |12| which also includes a number of general results obtained with the model. As an indication of the numerical accuracy of the technique, Table 1 compares the surface position as a function of time computed by the model, with the analytic solution for a simple Neumann problem. For an initial concentration ρ_i, the surface $x = 0$ being held at concentration ω ambient, the progression of the sublimation front is given by the dimensionless equation :

$$\frac{S}{L} = 2\lambda \sqrt{F_o}$$

where λ is the root of

$$\sqrt{\pi}\cdot\Omega\cdot\lambda\cdot e^{\lambda^2}\cdot\mathrm{erf}\ (\lambda) + 1 = 0$$

and F_o is the Fourier number, $F_o = \dfrac{D_{va}.t.}{L^2}$.

L is a scale length and $\Omega = (\rho_i - \omega_f)/(\omega\ \text{ambient} - \omega_f)$

Table 1 was calculated for the following values $\Delta x = 0.01$, $\Delta F_o = 1.0 \times 10^{-4}$, $\Omega = 1$.

the exception of a small initial error the values are nearly indistinguishable for pratical purposes.

Discussion

In order to analyse sublimation problems in general we require information about the diffusion conditions at the surface, and the experiments carried out to date can provide some insight. Using the measured average sublimation flux and supposing a linear distribution of vapour concentration near the surface the diffusive boundary layer thickness δ_{diff}, can be estimated from Ficks law.

From hydrodynamic theory |13| we can also calculate a thickness for the dynamic boundary layer, δ_{dyn}. From the experimental data, for velocities ranging from 1.5 to 4 $m.s^{-1}$, the ratio $\delta_{dyn}/\delta_{diff}$ was found to vary linearly between 3.3 and 4.4 for temtemperature -30°C and between 2.3 and 2.7 for temperature -9°C. Until more thorough studies can be carried out which take account of local sublimation rates and the real geometry of the specimens, we suggest that these ratios can be used to estimate the thickness of the diffusion boundary layer from the dynamic boundary layer thickness for other sublimation problems in the range of temperatures and velocities studied. This should make it possible to exploit more fully the numerical model presented for more general cases of porous media and non-isothermal conditions.

Conclusion

In this article we have presented an experimental study of the sublimation of pure ice and have proposed a numerical model to solve the system of equations which describe sublimation phenomena in a more general context. It is however necessary to verify the model by comparison with non-isothermal experiments which are more elaborate than those

TABLE 1 : Comparison of numerical and analytical solutions for sublimation depth.

F_o (Fourier N°)	S/L calculated	S/L analytic	error	% error
1×10^{-4}	0.114×10^{-1}	0.124×10^{-1}	-1.05×10^{-3}	- 8.5
1×10^{-3}	0.383×10^{-1}	0.392×10^{-1}	-9.37×10^{-4}	- 2.4
1×10^{-2}	0.124	0.124	$+3.54 \times 10^{-5}$	+ 0.0
5×10^{-2}	0.278	0.277	$+8.39 \times 10^{-4}$	+ 0.3

carried out to date. At the same time it is also necessary to study the range of conditions most important for modern engineering concerns, namely very low temperatures with relative humidities near zero. To this end we have recently designed a new experimental system. This consists of an experimental cell in which the diffusion of the sublimated vapour takes place in a nearly dry nitrogen atmosphere and under a one-dimensional temperature gradient directed upwards to avoid natural convection.

In addition it is necessary to have at hand a more thorough knowledge of the diffusive boundary layer. This will require further experimental work with the wind tunnel, measuring local sublimation rates on cylindrical specimens suitably arranged, for example, to allow control of the zone exposed to sublimation.

Along with this experimental work, a theoretical study of thermal, diffusive and dynamic boundary layers around a cylinder is also envisaged.

References

1 D. Simatos, G. Blond et P. Dauvois 1974 La lyophilisation, A.N.R.T, Paris
2 A.V. Lykov, and D.P. Lebedev 1973 Study of the ice sublimation process. Int. J. of Heat and Mass Transfer, 16, 1087-1096.
3 A. Boulanger et W. Luyten 1983 Le stockage souterrrain à basse température et ses applications. C.R. XVI° Cong. Int. du Froid Paris ,1, 365-370.
4 T.J.R. Cooper 1970 Control of weigh losses during drilling, freezing, storage and transport of pig meat. Bull. de IIF/IIR, Annexe 1970-3, 175-181, 187-190.
5 G. Lambrinos, J. Aguirre-Puente et M. Giat 1983 Une soufflerie pour l'étude expérimentale des milieux dispersés soumis à des températures négatives. Int. J. of Refrig., 6, 1.
6 G. Lambrinos 1982 Deshydratation des milieux dispersés pendant leur congélation et leur conservation à l'état gelé. Thèse Ing- Doct, Paris VI.
7 R.N. Sukhwal 1984 Physique de la sublimation en milieu dispersé et expérimentation sur la viande et la tylose. Thèse Ing-Doct, Paris VI.
8 J. Aguirre-Puente and R.N. Sukhwal 1984 Sublimation of ice in frozen dispersed media. 3rd Int. Symp. on Offshore Mech. and Artic Engng, New Orleans, U.S.A.
9 J. Aguirre-Puente, G. Lambrinos and M. Sakly 1984 Sublimation of ice in frozen dispersed media : Physics phenomena, equations and experimental study. Proc. of the Coll. Int. "Problèmes à frontières libres" Maubuisson-Carcans , France to be pub. by Pitman.
10 J. Aguirre-Puente, G. Lambrinos and M. Sakly 1984 Experimental research of the sublimation rate of ice samples. 10th Annual Meeting of European Geophys. Soc. to be pub. in Ann. Geophysicae.
11 L.E. Goodrich 1978 Efficient numerical technique for one-dimensional thermal problems with phase change. Int. J. of Heat and Mass Transfer, 21, 615-621.
12 L.E. Goodrich et al. Anumerical model for heat and mass diffusion with a moving sublimation front.(in preparation).
13 H. Schlichting 1955 Boundary layer theory Pergamon Pr London.