Steady-state and stability analysis of a population balance based nonlinear ice cream crystallization model

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Abstract— The process of crystallization can be modelled by a population balance equation coupled with an energy balance equation. Such models are highly complex to study due to the infinite dimensional and nonlinear characteristics, especially when all the phenomena of nucleation, growth and breakage are considered. In the present paper, we have performed the stability analysis on a reduced order model obtained by the method of moments, which remains still highly complex. The considered model has been developed by the Cemagref and validated on experimental data. After computation, we get a scalar equation whose solutions correspond to the equilibrium points of the system. This equation is finally solved numerically for a concrete physical configuration of the crystallizer. We show that in most instances, there is only one steady state. The possibility of multiple steady-states is discussed.

I. INTRODUCTION

Crystallization (e.g. [1]) is encountered in many processes, in particular in the pharmaceutical industry and the food industry [2]. In crystallization processes, an important challenge is to control the quality of the product while minimizing the energy consumption of the system. To achieve this goal, it is important to rely on a model that adequately describes the key phenomena of the crystallization process. In ice cream crystallization, it is well known that the quality of the product, that is the hardness and the texture of the ice cream, depends on the ice crystal size distribution (CSD). Indeed, the smaller the crystals are, the smoother the ice cream is. It is therefore of importance to consider a model that describes the evolution of the CSD : this can be achieved for instance by considering a population balance equation (PBE) [3]. This model is coupled with an energy balance equation which can either be expressed as an equation of the volumetric internal energy or of the temperature. To control such a system, one possible solution consists in designing a control law on the basis of a reduced order model (*late lumping*). This one may be obtained by applying the method of moments, which transforms the PBE in a set of ordinary

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differential equations (ODEs). As the first four moment equations are independent of the ones of lower order, and as the energy balance equation only involves moments of order 3 or less, the system we consider is reduced to a set of 5 ODEs. As a preliminary step before the control design, we first analyse the process model in order to emphasize the key dynamical properties of the model and to evaluate if the reduced model is sufficiently reliable and precise to design efficient control laws. The analysis of such a reduced system has already been made in some simplified cases. In [4] for example, the authors consider the isothermal case and assume that there is no breakage, and show that apart from the trivial equilibrium point, there is only one steady-state.

In this paper, we consider a complete mathematical model described in [5], [6]. This model has been developed by a research team of the Cemagref and has been validated on experimental data obtained from a pilot plant located at Cemagref. In this paper, we analyse the stability of the corresponding reduced model. This work was conducted as a part of the European CAFE project (Computer-Aided Food processes for control Engineering) in which four case studies are considered among them the one of the ice crystallization process. The final objective is to design some efficient laws to control the quality (texture, viscosity) of the ice cream at the outlet of the freezer.

The paper is organized as follows. In section II the crystallization model we consider is described. Then, we show in section III how the computation of the equilibrium points can be reduced to the resolution of a scalar equation. In section IV, the equation is solved numerically for a concrete physical configuration of the crystallizer and the results are commented.

II. MODEL DESCRIPTION

The model we consider in this paper is the one described in [5], [6]. The ice cream crystallizer is a scraped surface heat exchanger which is assumed to behave as a plug flow reactor. The population balance of the crystal size distribution considers transport, crystal growth, nucleation, breakage and possible radial diffusion. If the radial diffusion is assumed to be negligible, and if the plug flow reactor is approximated, from an input-output point of view, by a Continuous Stirred-Tank Reactor (CSTR) with a transport delay (to account for the fluid transport in the freezer), then we get the following simplified model :

$$\frac{\partial \Psi}{\partial t} = \underbrace{-\frac{q}{V}\Psi}_{\text{transport}} \underbrace{- \frac{\partial(G\Psi)}{\partial L}}_{\text{growth}} + \underbrace{N\delta_{(L-L_c)}}_{\text{nucleation}} + \underbrace{B_b}_{\text{breakage}} \quad (1)$$

where Ψ is the number of crystals per meter (of the freezer) per cubic meter of the solution at the outlet of the freezer, t is the time variable, L and L_c are the crystal length variable and the initial crystal length, q is the inlet flow rate, V is the volume of the freezer, and G, N, B_b are the growth rate, nucleation rate, and net increase of crystals number by breakage, respectively. δ denotes the Dirac function.

The growth and nucleation rates are expressed by[1] [6]:

$$G = \beta(T_{\text{sat}} - T), \text{ and } N = \alpha S \left(T_{\text{sat}} - T_e\right)^2, \quad (2)$$

where T_{sat} is the saturation temperature, and α, β are some kinetic parameters.

Because of the scraper, the crystals can also be broken. We assume that a particle of size L' is broken into two particles of the same length L. The volume of ice is considered unchanged by the fragmentation and a spherical shape is assumed (as in [6]). Under this assumption, the net increase of particles by breakage B_b, can be expressed as[2] [6]:

$$B_b = \epsilon N_{\text{scrap}} \phi_i^\nu \left(2\, 2^{2/3}\, L\Psi(\sqrt[3]{2}\, L) - L\Psi(L)\right), \quad (3)$$

with N_{scrap} the dasher rotation speed, ϵ a breakage coefficient, ϕ_i the ice fraction and ν the breakage power coefficient which is taken equal to 0, as in [7].

Under the same hypotheses, the energy balance equation is written as follows :

$$\frac{dU}{dt} = \underbrace{-\frac{q}{V}(U - U_0)}_{\text{transport}} + \underbrace{h_e S(T_e - T)}_{\text{wall heat transfer}} + \underbrace{\mu\dot{\gamma}^2}_{\text{viscous dissipation}} \quad (4)$$

where U and T are the respective volumetric internal energy and temperature at the outlet of the freezer, U_0 is the volumetric temperature at the inlet of the freezer, T_e is the evaporation temperature, h_e is the convective heat transfer coefficient and μ is the viscosity. The effective shear rate $\dot{\gamma}$ is given by $\dot{\gamma} = 2\pi\chi N_{\text{scrap}}$ with χ the viscous dissipation coefficient. The quantity $S = \frac{2R_e}{R_e^2 - R_i^2}$ is the ratio of the circumference over the surface of the section of the freezer, R_e and R_i denoting the maximum and minimum diameters of the cylindrical freezer respectively.

Applying the method of moments[3] to equation (1), we get, for all $j \geqslant 0$ [7] :

$$\frac{dM_j}{dt} = -\frac{q}{V}M_j + j\, G\, M_{j-1} + N\, L_c^j + B\left(2^{1-\frac{j}{3}} - 1\right)M_{j+1}$$

where the jth order moment M_j is given by :

$$M_j(t) = \int_0^\infty L^j\, \Psi(L, t)dL \quad (5)$$

and $B = \epsilon N_{\text{scrap}}\phi_i^\nu$.

The saturation temperature $T_{\text{sat}} = T_{\text{sat}}(M_3)$ is supposed to depend only on M_3. We so have: $G = G(M_3, T)$ and $N = N(M_3, T_e)$.

[1]Only heterogeneous nucleation at the freezer wall ($r = R_e$) is considered here.

[2]Under these assumptions, the relation between L' and L is given by $L' = 2^{1/3}L$.

[3]The method of moments consists in multiplying the population balance equation by L^j and then integrating it from $L = 0$ to $L = \infty$.

If we consider the ice crystals as spherical particles (as in [6]), then we have $\phi_i = \frac{\pi}{6}M_3$ and equation (4) can be rewritten with the temperature T as the state variable by using the following relation :

$$U = -\Delta H \rho_i \phi_i + \rho_s \left(\omega_0 C_s + (1 - \omega_0)C_w\right)T \quad (6)$$

where $\Delta H, \omega_0, C_s, C_w, \rho_i$ and ρ_s are the specific fusion latent heat, the initial mass fraction of solute, the solute and water specific heat capacities, and the mass densities of ice and solution, respectively.

After computation, we finally get :

$$\frac{dT}{dt} = D\left(T_0 - T\right) + K_2\left(T_e - T\right) + \mu K_3 + K_1\left(3GM_2 + NL_c^3\right) \quad (7)$$

with the following quantities :

$$D = \frac{q}{V},\ K_0 = \rho_s\left(\omega_0\, C_s + (1 - \omega_0)C_w\right), \quad (8)$$

$$K_1 = \frac{\pi}{6}\frac{\Delta H\, \rho_i}{K_0},\ K_2 = \frac{h_e S}{K_0},\ K_3 = \frac{\dot{\gamma}^2}{K_0}. \quad (9)$$

If the viscosity μ is assumed to depend only on the third moment M_3 and the temperature T, then the system composed of the first four moment equations and the temperature equation is closed. In fact all the dynamic quantities of this system are functions of the moments M_0, M_1, M_2, M_3, the temperature T and the possible control variables T_e, q and N_{scrap}. In the sequel we shall therefore consider the following reduced model :

$$\frac{dM_0}{dt} = -DM_0 + N + BM_1 \quad (10)$$

$$\frac{dM_1}{dt} = -DM_1 + GM_0 + NL_c + c_1BM_2 \quad (11)$$

$$\frac{dM_2}{dt} = -DM_2 + 2GM_1 + NL_c^2 + c_2BM_3 \quad (12)$$

$$\frac{dM_3}{dt} = -DM_3 + 3GM_2 + NL_c^3 \quad (13)$$

$$\frac{dT}{dt} = D\left(T_0 - T\right) + K_2\left(T_e - T\right) + \mu K_3 + K_1\left(3GM_2 + NL_c^3\right) \quad (14)$$

with $\mu = \mu(M_3, T)$ and the following constants :

$$c_1 = 2^{\frac{2}{3}} - 1,\ c_2 = 2^{\frac{1}{3}} - 1. \quad (15)$$

III. STEADY STATES

In this section we concentrate on the determination of the equilibrium points of the system (10)-(14), that is the points such that $\frac{dM_i}{dt} = 0$, $i = 0:3$ and $\frac{dT}{dt} = 0$. We only consider here the steady states which verify the following physical conditions:

$$\phi_i \in [0, 1] \Leftrightarrow M_3 \in [0, \tfrac{6}{\pi}]$$
$$M_0, M_1, M_2, M_3 \geqslant 0 \text{ and } T \geqslant -273. \quad (16)$$

A. Expression of M_0, M_1 and M_2

From (10-14), $\frac{dM_1}{dt} = 0$, $\frac{dM_2}{dt} = 0$ and $\frac{dM_3}{dt} = 0$ lead, under the assumption that $G \neq 0$, to :

$$M_0 = \frac{1}{G}\left(-NL_c + DM_1 - c_1 BM_2\right)$$

$$M_1 = \frac{1}{2G}\left(-NL_c^2 - c_2 BM_3 + DM_2\right)$$

$$M_2 = \frac{1}{3G}\left(DM_3 - NL_c^3\right)$$

that is :

$$
\begin{aligned}
M_0 =\ & -\frac{1}{6G^2}\left(3D(NL_c^2 + c_2 BM_3) + 2c_1 B(DM_3 - NL_c^3)\right) \\
& + \frac{1}{6G^3}\left(-6NL_c G^2 + D^2(DM_3 - NL_c^3)\right) \\
M_1 =\ & \frac{1}{6G^2}\left(-3G\left(NL_c^2 + c_2 BM_3\right) + D\left(DM_3 - NL_c^3\right)\right) \\
M_2 =\ & \frac{1}{3G}\left(DM_3 - NL_c^3\right).
\end{aligned}
$$

$$(17)$$

As said previously, all the moments M_i are positive variables. We deduce from this positivity some conditions on G :

- the positivity of M_1 can be written as follows (under the assumption that $(NL_c^2 + c_2 BM_3) \neq 0$) :

$$\frac{D\left(DM_3 - NL_c^3\right)}{3\left(NL_c^2 + c_2 BM_3\right)} > G \; ; \qquad (18)$$

- the positivity of M_2 leads to:

$$sign(G) = sign(DM_3 - NL_c^3). \qquad (19)$$

B. Expression of G

Using (17), $\frac{dM_0}{dt} = 0$ gives :

$$-DM_0 + N + BM_1 = 0 \qquad (20)$$

$$\Leftrightarrow \quad \underbrace{a(M_3)G^3 + b(M_3)G^2 + c(M_3)G + d(M_3)}_{P(G)} = 0 \quad (21)$$

with:
$$a(M_3) = 6N, \qquad (22)$$
$$b(M_3) = 6DNL_c - 3B(NL_c^2 + c_2 BM_3), \qquad (23)$$
$$\begin{aligned} c(M_3) =\ & 3D^2(NL_c^2 + c_2 BM_3) \\ & + DB(1 + 2c_1)(DM_3 - NL_c^3), \end{aligned} \qquad (24)$$
$$d(M_3) = -D^3(DM_3 - NL_c^3) \qquad (25)$$

Let us denote G_1, G_2 and G_3 the 3 roots of P. We have under the assumption that $N \neq 0$:

$$-G_1 G_2 G_3 = \frac{d}{a} = \frac{-D^3\left(DM_3 - NL_c^3\right)}{6N}$$

and so : $\quad sign(G_1 G_2 G_3) = sign(DM_3 - NL_c^3). \quad (26)$

Consequently, either 1 or 3 of the roots can fulfil the condition (19).

The kind of roots depends on the sign of the quantity Δ defined by :

$$\Delta = 4\frac{p(M_3)^3}{27} + q(M_3)^2 \qquad (27)$$

with $p(M_3) = \frac{c}{a} - \frac{b^2}{3a^2} \qquad (28)$

and $q(M_3) = \frac{d}{a} - \frac{bc}{3a^2} + \frac{2b^3}{27a^3}. \qquad (29)$

Let us consider the different possible cases :

- **Case 1**, $\Delta > 0$ **:** then we have 1 real root G_1 and 2 complex roots G_2 and G_3. The expression of G_1 is given here after :

$$G_1 = -\frac{b}{3a} + (s_1)^{1/3} + (s_2)^{1/3}, \qquad (30)$$

with

$$s_1 = \frac{-q(M_3) + \sqrt{\Delta}}{2} \text{ and } s_2 = \frac{-q(M_3) - \sqrt{\Delta}}{2}. \qquad (31)$$

As the coefficients of the polynomial are real, the two complex roots are necessarily conjugate that is : $G_2 = \overline{G_3}$, and so $G_1 G_2 G_3 = G_1|G_2|^2$. From (26), it can then be shown that the root G_1 verifies the condition (19).

- **Case 2**, $\Delta \leqslant 0$**:** then we have 3 real roots G_1, G_2, G_3 given by :

$$G_1 = -\frac{b}{3a} - \sqrt{3}Im(s) - Re(s), \qquad (32)$$

$$G_2 = -\frac{b}{3a} + \sqrt{3}Im(s) - Re(s), \qquad (33)$$

$$G_3 = -\frac{b}{3a} + 2Re(s), \qquad (34)$$

with:

$$s = \left(\frac{-q(M_3) + i\sqrt{-\Delta}}{2}\right)^{1/3}. \qquad (35)$$

We can show that (the proofs are given in appendix VI) :

Proposition 1: $G_1 < G_1' < G_2 < G_2' < G_3$ with :

$$G_1' = \frac{-2b - \sqrt{\Delta'}}{6a} \text{ and } G_2' = \frac{-2b + \sqrt{\Delta'}}{6a} \qquad (36)$$

and $\Delta' = 4\left[b^2 - 3ac\right]$.

We also have :

Proposition 2: $D - 2c_1 BL_c > 0 \Rightarrow G_1, G_2, G_3 > 0$.

Remark 3: The condition $D - 2c_1 BL_c > 0$ is not very restrictive. It is indeed often verified in practice (see section IV).

Consequently, the three roots fulfil the condition (19). Nevertheless, it can be shown that there is at the most only one of the 3 roots which verify the condition (18). More precisely, we have :

Proposition 4: G_2 and G_3 do not fulfil the condition (18).

Finally, only G_1 can be a possible value of G at equilibrium.

As a conclusion, we have :

$$G = \begin{cases} -\frac{b}{3a} + (s_1)^{1/3} + (s_2)^{1/3} & \text{when } \Delta > 0 \\ -\frac{b}{3a} - \sqrt{3}Im(s) - Re(s) & \text{when } \Delta \leqslant 0 \end{cases} \qquad (37)$$

with s_1, s_2 and s expressed by (31) and (35), respectively.

Remark 5: This expression of G guarantees the positivity of M_2 but not necessarily the one of the other moments, which will have to be tested numerically.

Remark 6: The continuity of G, as a function of M_3, can be shown.

C. Expressions of T and M_3

After having computed the possible values of G, we can deduce the value of T from (2) :

$$T = T_{sat}(M_3) - \frac{1}{\beta}G \qquad (38)$$

Then, by replacing T by (38) in $\frac{dT}{dt} = 0$ and because $\frac{dM_3}{dt} = 0 \Leftrightarrow 3GM_2 + NL_c^3 = DM_3$, we finally get a scalar equation that only depends on M_3 :

$$
\begin{aligned}
&F(M_3) = 0 \\
\text{with } F(M_3) = \quad & D\left(T_0 - T(M_3) + K_1 M_3\right) \\
& + K_2\left(T_e - T(M_3)\right) + \mu(M_3, T(M_3))K_3.
\end{aligned}
\qquad (39)
$$

The number of physical equilibrium points of the system equals to the number of solutions of (39) which verify the physical conditions (16). Due to the complexity of the equation and because it depends on the expressions of T_{sat} and μ, neither the values nor the number of equilibrium points can be analytically computed.

IV. NUMERICAL SIMULATIONS

In this section, we compute numerically the solutions of equation (39) for a particular case of model (10)-(14). The values of the parameters correspond to an pilot plant ice cream crystallizer located at Cemagref and described in [6]. The crystallizer is a cylinder with a $0.40\,\text{m}$ length and whose maximum and minimum diameters are respectively given by: $R_e = 25\,\text{mm}$ and $R_i = 16\,\text{mm}$. The values of L_c, α, β, ϵ, χ, ξ and h_e are the same as the ones considered in [6]. For the others parameters, we have taken the following values :

$$
\begin{aligned}
\omega_0 &= 0.3, \rho_i = 1000 \text{kg}\,\text{m}^{-3}, \ \rho_s = 1120\,\text{kg}\,\text{m}^{-3} \\
C_s &= 1676\,\text{J}\,\text{kg}^{-1}\,{}^\circ\text{C}^{-1}, \ C_w = 4187\,\text{J}\,\text{kg}^{-1}\,{}^\circ\text{C}^{-1} \\
\Delta H &= 336.6\,10^3\,\text{J}\,\text{kg}^{-1}, \ T_0 = 5{}^\circ C
\end{aligned}
$$

The expression of T_{sat} is given by (see [7]) :

$$T_{\text{sat}} = -7.683\omega + 8.64\omega^2 - 70.1\omega^3 \text{ with } \omega = \frac{\omega_0}{1 - \frac{\rho_i}{\rho_s}\phi_i}.$$

It is obtained by interpolation of experimental data and depends on the commercial mix under consideration.
The viscosity μ of the sorbet is expressed by [6] :

$$\mu = \mu_{\text{mix}}\left(1 + 2.5\,\phi_i + 10.05\,\phi_i^2 + 0.00273\,\xi\,e^{16.6\,\phi_i}\right),$$

where μ_{mix}, the viscosity of the continuous phase of sucrose in water solution, is provided by experiments and written as follows [7] :

$$\mu_{\text{mix}} = 39.02\times10^{-9}\times\dot\gamma^{0.600-1}e^{\frac{2242.38}{T+273}}\times(100\,\omega)^{2.557}. \quad (40)$$

In Figure 1 we can see the values of G (expression (37)) and F (expression (39)) as functions of the variable M_3, plotted on $[0, \frac{6}{\pi}]$, in the case of an inlet mass flow rate[4] mfr

[4] We have: $q = \frac{\text{mfr}}{\rho_{\text{sol}}}$ where ρ_{sol} is the volumetric mass of the solution.

of $100\,\text{kg.h}^{-1}$, a scraper rotation speed of $300\,\text{rpm}$ and an evaporation temperature of $-20{}^\circ\text{C}$. In that case, we note that there is only one solution of $F(M_3) = 0$. In fact we can verify numerically that it is the case for all the admissible values of mfr, N_{scrap} and T_e [6], that is: $25\,\text{kg.h}^{-1} < \text{mfr} < 100\,\text{kg.h}^{-1}$, $300\,\text{rpm} < N_{\text{scrap}} < 1000\,\text{rpm}$ and $-25\,\text{C}{}^\circ < T_e < -10\,\text{C}{}^\circ$. We also verify that the equilibrium point is always Lyapunov stable.

We also verify the coherent respective influences of mfr, N_{scrap} and T_e on the value of the equilibrium point. Indeed, the values of the moments at equilibrium decrease with mfr, T_e and N_{scrap}, whereas the temperature increases with these three quantities. These behaviours are coherent with the physical signification of the moments; indeed M_0 is the number of particles per cubic meter, M_1 is the sum of characteristic lengths per cubic meter, and M_2 and M_3 are the images of the total area and volume of crystals per cubic meter.

In the configuration of the system considered here, equation $F(M_3) = 0$ admits only one solution. Nevertheless, we note that function F is not monotonic. There is a peak, visible in figure 1 (top and bottom), which could enable, under particular values of parameters, to get configurations for which there would be three equilibrium points[5]. This peak is located around the value of M_3 such that $T_{\text{sat}}(M_3) = T_e$, which corresponds to the case where $a(M_3) = 0 \Leftrightarrow N(M_3, T_e) = 0$ (see (22)). Suppose there exists a configuration of the system for which the axis $y = 0$ cut the curve of F three times, in the neighbourhood of the peak. The first intersection point is necessarily located before the peak, whereas the two other ones are located after. So it implies that the two corresponding equilibrium points are such that $T_e > T_{\text{sat}}$. On the other hand, we can easily deduce from the expression of F (formula (39)), that any solution of equation $F(M_3) = 0$ are such that $T > T_e$. Indeed, $K_2(T_e - T)$ is the only term of $F(M_3)$ which can take negative values. Finally the two equilibrium points located after the peak are such that $T > T_e > T_{\text{sat}}$, which implies that $G < 0$. So these equilibrium points would correspond to cases where the crystals are melting.

V. CONCLUSION

This paper focused on an ice cream crystallisation model, composed of a population balance equation describing the evolution of the crystal size distribution in the freezer, coupled with an energy balance equation. The key phenomena of the crystallisation process, that is the nucleation, the growth and the breakage have been taken into account, leading to a rather complex model. We have studied the stability of a differential equations model directly deduced from the preceding one by the method of moments. The five state variables were the first four moments and the temperature of the ice cream at the outlet of the freezer. We show that the

[5] Such configurations have been observed, in simulation, but only for parameter values which are not physically realistic (very low value of T_0 for instance). But the existence of other realistic configurations remains possible.
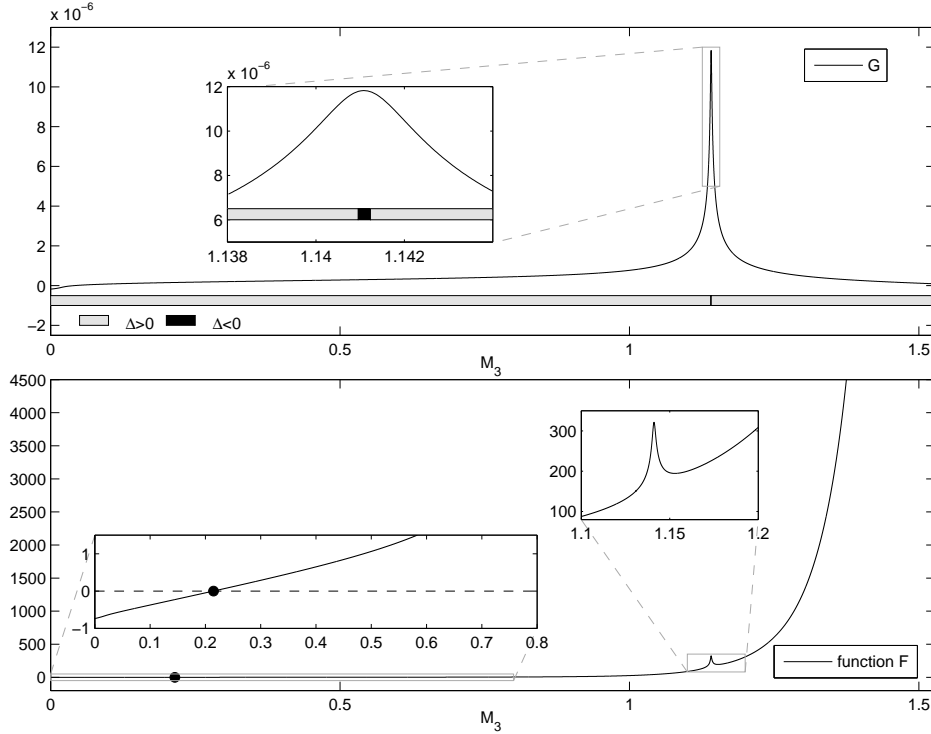
Fig. 1. Values of G (top) and F (bottom) as function of M_3, for an inlet mass flow rate of $100\,\text{kg.h}^{-1}$, a scraper rotation speed of $300\,\text{rpm}$ and an evaporation temperature of $-20°\text{C}$.

computation of equilibrium points of the system is reduced to the resolution of a scalar equation. This equation has been solved numerically, for a set of parameters corresponding to a concrete physical configuration of the freezer. In that case, only one stable steady state has been found. Nevertheless, in the mathematical point of view, the existence of 3 steady states remains possible. We note that, in that case, only one of the 3 steady states would correspond to a situation where the crystals are growing. A realistic set of parameters leading to such a situation has not been found yet.

This work is a preliminary step before the control design. The objective is to control the quality (texture, viscosity) of the ice cream at the outlet of the freeezer. This quality can directly be evaluated from quantities which can be expressed as functions of the temperature and the first four moments, which makes the study of the moments equations important.

VI. APPENDIX

In this appendix are detailed the proofs of all the results of the paper.

Proof of proposition 1

The quantity s defined by (35) is such that $Im\left(s^3\right) = \frac{\sqrt{-\Delta}}{2} > 0$. We so have:

$$Im(s) > 0, \ Re(s) > 0 \text{ and } \arg(s) \in [0, \tfrac{\pi}{3}].$$

As a consequence, $2Re(s) > -\sqrt{3}Im(s) - Re(s)$ and $\sqrt{3}Im(s) > -\sqrt{3}Im(s)$, and so $G_3 > G_1$ and $G_2 > G_1$.

Furthermore:

$$\arg(s) \in [0, \tfrac{\pi}{3}] \Longrightarrow 0 \leqslant \frac{Im(s)}{Re(s)} \leqslant \tan(\tfrac{\pi}{3}) = \sqrt{3}$$

As $Re(s) > 0$, then we have $\sqrt{3}Im(s) - Re(s) < 2Re(s)$ that is $G_2 < G_3$. Finally, $G_1 < G_2 < G_3$.

Let us study the function P. We have $P'(G) = 3aG^2 + 2bG + c$. The number of roots of $P'(G)$ depends on the sign of the following quantity $\Delta' = 4\left[b^2 - 3ac\right]$. We are in the case where $\Delta \leqslant 0$, which implies:

$$4\frac{p(M_3)^3}{27} + q(M_3)^2 \leqslant 0 \quad \Rightarrow p(M_3) \leqslant 0$$
$$\Leftrightarrow 3ac - b^2 \leqslant 0. \quad (41)$$

We so have $\Delta' \geqslant 0$. Consequently, $P'(G)$ has two real roots G_1' and G_2' expressed by:

$$G_1' = \frac{-2b - \sqrt{\Delta'}}{6a} \text{ and } G_2' = \frac{-2b + \sqrt{\Delta'}}{6a}$$

and such that $G_1' \leqslant G_2'$. As $a \geq 0$, then we have:

$$P'(G) \underset{-\infty}{\longrightarrow} -\infty \text{ and } P'(G) \underset{+\infty}{\longrightarrow} +\infty.$$

Moreover, as there are 3 real roots, we necessarily have $P(G_1') > 0$ and $P(G_2') < 0$, and, because $G_1 < G_2 < G_3$:

$$G_1 < G_1' < G_2 < G_2' < G_3.$$

$\square$

Proof of proposition 2

As shown previously (see (41)), we have $\Delta \leqslant 0 \Rightarrow 3ac - b^2 \leqslant 0$. After computations, we get:

$$3ac - b^2 = k_1 M_3^2 + k_2 M_3 N + k_3 N^2,$$

with $k_1 = -9B^4 c_2^2$,

$$k_2 = 9B \left(2D^2(1 + 2c_1 + 3c_2) + 2c_2 BL_c D - c_2 B^2 L_c^2 \right),$$
$$k_3 = 9L_c^2 \left(2D^2 + 2BL_c D(1 - 2c_1) - B^2 L_c^2 \right).$$

Assuming that $D - 2c_1 BL_c > 0$, we can show that $k_1 < 0$, $k_2 > 0$ and $k_3 > 0$.

Let us compute the discriminant of this polynomial in M_3, that is the quantity $\delta = (Nk_2)^2 - 4k_1 k_3 N^2$. After computations, we get $\delta = 9^2 N^2 B^2 \delta_2$ with:

$$\delta_2 = 4D^3(1 + 2c_1 + 3c_2)(D(1 + 2c_1 + 3c_2) + 2c_2 BL_c)$$
$$+ c_2 B^2 L_c(4D^2(2 - 2c_1 - 3c_2) - 3B^2 L_c^2 + 4DBL_c(1 - 4c_1))$$

As $k_1 < 0$ and $k_2, k_3 > 0$, we can show that $\delta_2 > 0$. The 2 real roots of the polynomial then write:

$$\frac{-Nk_2 \pm \sqrt{\delta}}{2k_1} = \frac{N(k_{2,2} \mp \sqrt{\delta_2})}{2B^3 c_2^2}$$

with $k_{2,2} = 2D^2(1 + 2c_1 + 3c_2) + c_2 BL_c(2D - BL_c) > 0$. As $k_1 < 0$ and $k_2, k_3 > 0$, then $\sqrt{\delta} > Nk_2$ and so $\frac{N(k_{2,2} - \sqrt{\delta_2})}{2B^3 c_2^2} < 0$ and $\frac{N(k_{2,2} + \sqrt{\delta_2})}{2B^3 c_2^2} > 0$. We then deduce:

$$3ac - b^2 \leqslant 0 \text{ with } M_3 > 0 \Rightarrow M_3 > \frac{N(k_{2,2} + \sqrt{\delta_2})}{2B^3 c_2^2}. \quad (42)$$

We also have, from (23), $b(M_3) = 3NL_c(2D - BL_c) - 3c_2 B^2 M_3$. As $2D - BL_c > 0$ and by use of (42), we get:

$$b < 3L_c(2D - BL_c)\frac{2B^3 c_2^2}{(k_{2,2} + \sqrt{\delta_2})}M_3 - 3c_2 B^2 M_3$$

$$= \frac{3B^2 c_2}{(k_{2,2} + \sqrt{\delta_2})}M_3 \left(2c_2 BL_c(2D - BL_c) - k_{2,2} - \sqrt{\delta_2} \right)$$

$$= -\frac{3B^2 c_2}{(k_{2,2} + \sqrt{\delta_2})}M_3 \left[2D^2(1 + 2c_1) + 2c_2 D(3D - 2BL_c) \right.$$
$$\left. + 2c_2 B^2 L_c^2 + \sqrt{\delta_2} \right] < 0.$$

We also have $d > 0 \Leftrightarrow DM_3 < NL_c^3$ and so:

$$k_1 M_3^2 + k_2 M_3 N + k_3 N^2 > k_1 \frac{NM_3 L_c^3}{D} + k_2 M_3 N + k_3 \frac{DNM_3}{L_c^3}$$

$$= \frac{NM_3}{DL_c^3}(k_1 L_c^6 + k_2 DL_c^3 + k_3 D^2) > 0$$

under the assumption that $D - 2c1BL_c > 0$. Finally:

$$d > 0 \Rightarrow 3ac - b^2 > 0 \Rightarrow p(M_3) > 0 \Rightarrow \Delta > 0.$$

As $\Delta \leqslant 0$, we so have $d < 0$.

We have: $G_1' = \frac{1}{6a}\left(-2b - \sqrt{\Delta'} \right) = \frac{1}{3a}\left(-b - \sqrt{b^2 - 3ac} \right)$. However:

$$c = 3D^2(NL_c^2 + c_2 BM_3) + DB(1 + 2c_1)(DM_3 - NL_c^3)$$
$$= NDL_c^2(3D - (1 + 2c_1)BL_c) + D^2 BM_3(1 + 2c_1 + 3c_2).$$

So, under the assumption that $D - 2c_1 BL_c > 0$, we have $c > 0$. As $a > 0$, we then have $0 \leqslant b^2 - 3ac < b^2$, which, because $b < 0$, gives:

$$0 \leqslant \sqrt{b^2 - 3ac} < -b \Leftrightarrow G_1' > 0 \Rightarrow 0 < G_2 < G_3.$$

As a conclusion, when $\Delta \leqslant 0$, we have $0 < G_2 < G_3$ and $G_1 G_2 G_3 = -\frac{d}{a} > 0$ and so $G_1 > 0$. $\square$

Proof of proposition 4

Recall that $a > 0$, $b < 0$, $c > 0$ and $d < 0$ (see proof of proposition 2). We will show that:

$$G_3 > G_2 \quad > \frac{-2b - \sqrt{\Delta'}}{6a} = G_1' > \frac{D\left(DM_3 - NL_c^3 \right)}{3(NL_c^2 + c_2 BM_3)},$$
$$\text{that is:} \quad \sqrt{\Delta'}Q_1 < -2aDQ_2 - 2bQ_1, \quad (43)$$

with $Q_1 = NL_c^2 + c_2 BM_3$ and $Q_2 = DM_3 - NL_c^3$.
The two members of the inequality (43) are positive. Indeed, the right member $-2aDQ_2 - 2bQ_1$ gives, after multiplication by $\frac{3B}{2} > 0$:

$$-3aBDQ_2 - 3BbQ_1$$
$$= -3aBDQ_2 + b^2 - 6DNL_c b \quad \text{(because of (23))}$$
$$> -3aBDQ_2 + 3ac - 6DNL_c b \quad \text{(because of (41))}$$
$$= 3a \left[3D^2 Q_1 + 2c_1 BDQ_2 \right] - 6DNL_c b > 0$$
$$\text{(because } a > 0 \text{ and } b < 0 \text{)}.$$

As a consequence, inequality (43) is equivalent to:

$$\Delta' Q_1^2 < (2aDQ_2 + 2bQ_1)^2$$
$$\Leftrightarrow b^2 Q_1^2 - 3acQ_1^2 < a^2 D^2 Q_2^2 + b^2 Q_1^2 + 2abDQ_2 Q_1$$
$$\Leftrightarrow 0 < aD^2 Q_2^2 + 2bDQ_2 Q_1 + 3cQ_1^2$$
$$\Leftrightarrow 0 < 6ND^2 Q_2^2 + 12D^2 NL_c Q_2 Q_1$$
$$+ 9D^2 Q_1^3 + 3DB(2c_1 - 1)Q_2 Q_1^2$$

This inequality is verified because $(2c_1 - 1) > 0$ and $Q_2 = -\frac{d}{D^3} > 0$, and so the four terms of the right member are positive. $\square$

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