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Operational models for ice crystal formation in highly concentrated systems

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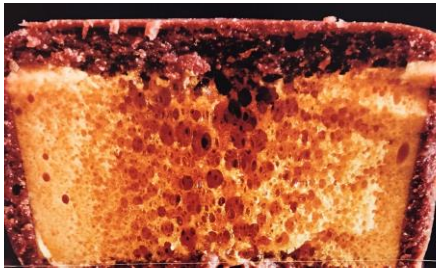
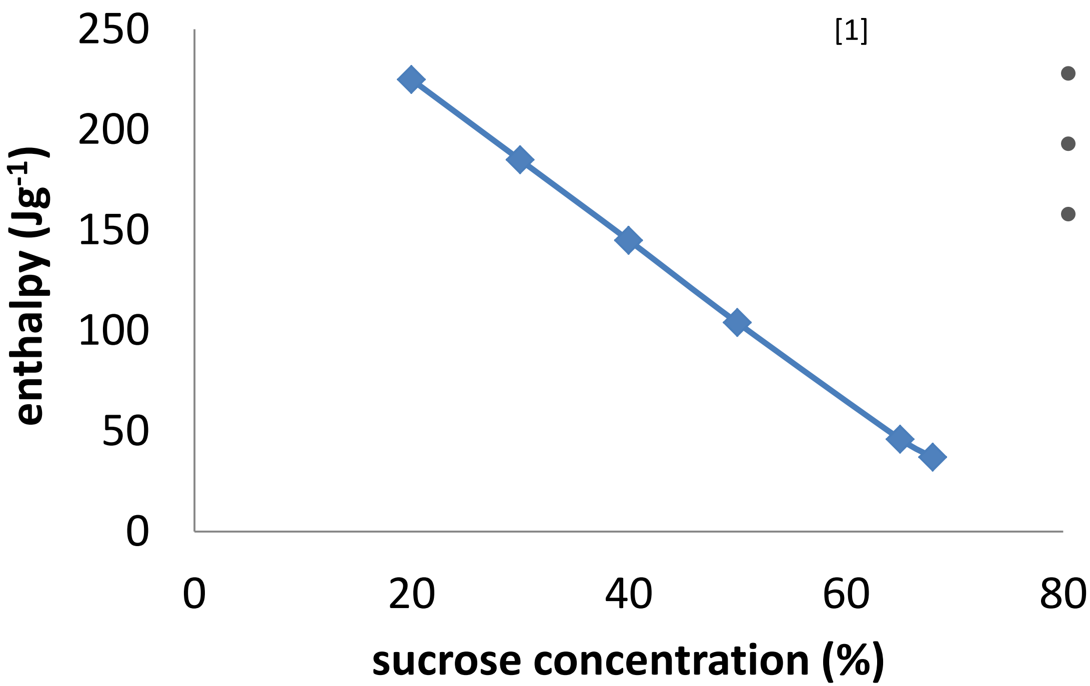
Crystallisation

- Important in food processing ⟶ freeze-drying & freezing
- Determinant in creating microstructure ⟶ properties & texture



Crystallisation
- Nucleation
  - Primary
  - Secondary (seeding)
- +
- Growth

Highly concentrated systems



- Experimentally challenging
- Available water?
- Difficult crystallisation

LESS WATER

↓

LESS ENERGY

Modelling: **understanding and control** (Quality, process-ability)

[1] Roos and Karel (1991), *International Journal of Food Science and Technology*, 26, pp. 553-566.

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Modelling

- Liquid into solid ⟶ Phase change

Moving boundary problem

Primary[2]

Kinetics:
DSC experiments

crystal fraction, T

Heat transfer: Fourier
+ phase change heat

Fixed domain

Secondary[3]

Mass transfer:
Fick

Stefan problem
Front-tracking

Heat transfer:
Fourier

T, C, freezing front

Growth rate
↓
Mean crystal size

[2] Chégnimonhan *et al*. (2010). *International Journal of Refrigeration* 33, 1559-1568.
[3] Crank (1984). *Free and moving boundary problems*, Clarendon Press, Oxford, UK.

Primary crystallisation: model

- Governing equations

$$\rho_m C_{p_m} \frac{\partial T_m}{\partial t} = \nabla\left(k_m \nabla T_m\right) + \rho_s \Delta H \frac{\partial \alpha}{\partial t}$$

Crystal fraction

- External boundary conditions

$$T_s\left(0,t\right) = T_c < T_{freezing} \; ; \; \frac{\partial T_l}{\partial r}\left(L,t\right) = 0$$

- Continuous material properties

Air fraction

$$k_m = \varepsilon k_{air} + \left(1-\varepsilon\right)\left(\alpha k_s + [1-\alpha]k_l\right)$$

$$C_{p_m} = \varepsilon C_{p_{air}} + \left(1-\varepsilon\right)\left(\alpha C_{p_s} + [1-\alpha]C_{p_l}\right)$$

$$\rho_m = \varepsilon \rho_{air} + \left(1-\varepsilon\right)\left(\alpha \rho_s + [1-\alpha]\rho_l\right)$$

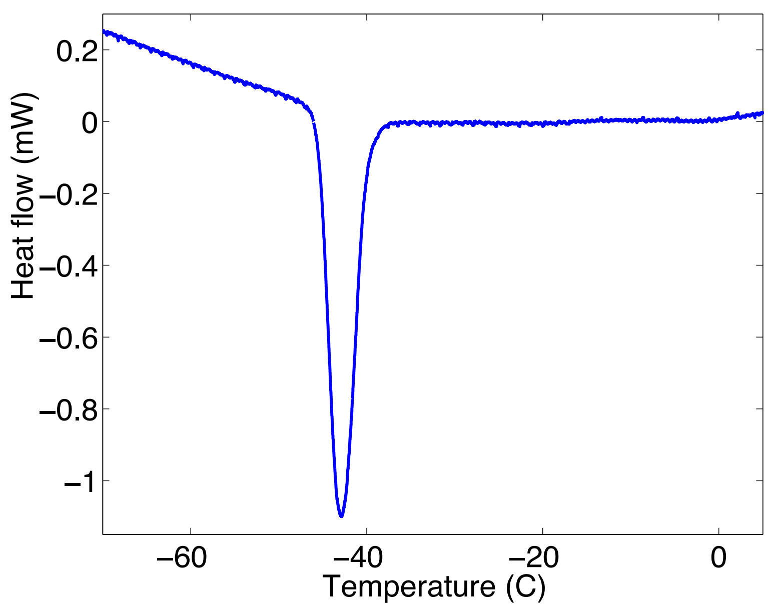
Primary crystallisation: model

- Crystallisation kinetics

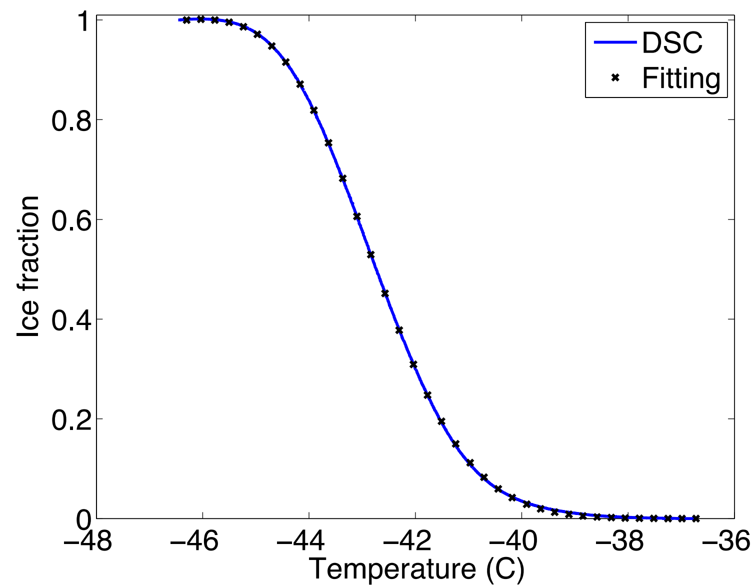
$$\alpha(T) = \frac{1}{\Delta H \dot{q}} \int \phi \, dT$$

DSC experiments → α - Ice crystal fraction

[4]



Non-isothermal DSC thermogram for the 60% (w/w) sucrose solution

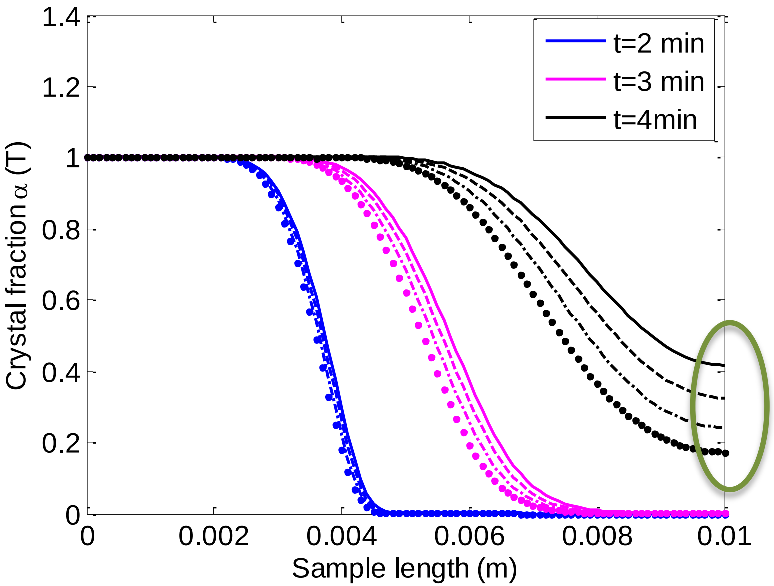


Crystallisation rate for the 60% (w/w) sucrose solution
RMSE = 0.001945

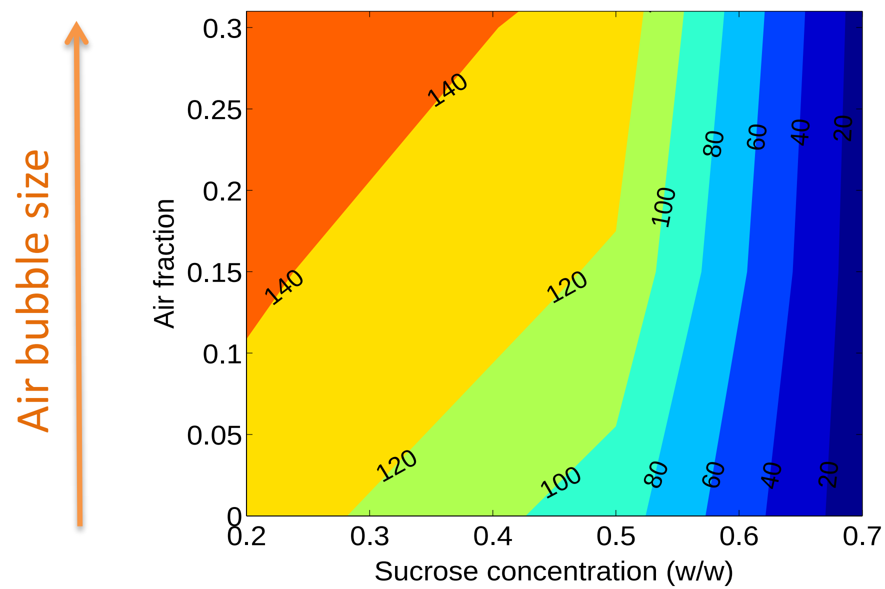
[4] Málek, J. (2000). *Thermochimica Acta* 335, 239-253.

Primary crystallisation: results

- 20-70% sucrose solution
- FEM in COMSOL
- 101 nodes, tol=10^{-6}, L= 1cm
- Air fraction ε = [0, 0.1, 0.2, 0.3]

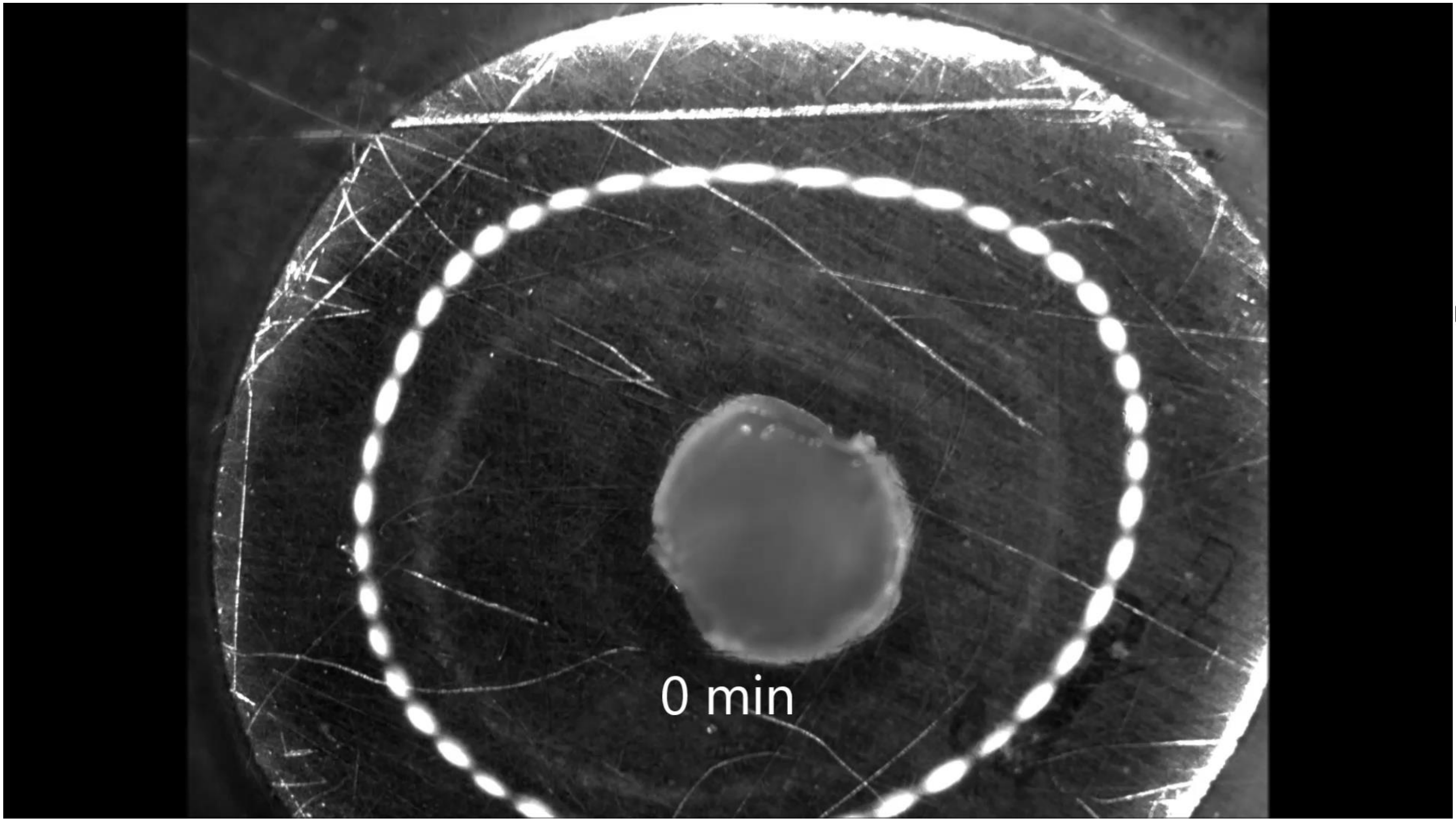


Crystal fraction the for the % 60 non-aerated sample (solid), and the aerated system with air fractions ε=0.1 (dashed), ε=0.2 (dashed-dot) and ε=0.3 (dot)



Effect of product formulation and aeration on the mean ice crystal size (μm).

Secondary crystallisation: experiments



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Secondary crystallisation: model

- Governing equations

$$\frac{\partial c_i}{\partial t} = \nabla\left(D_i \nabla c_i\right), \quad i = l,s \qquad r_i C_{p_i} \frac{\partial T_i}{\partial t} = \nabla\left(k_i \nabla T_i\right), \quad i = l,s$$

- Moving front boundary conditions

$$\left[c\,\big|_{S(t)^+} - c\,\big|_{S(t)^-} \right] \frac{\partial S}{\partial t} = D_l \left.\frac{\partial c}{\partial x}\right|_{S(t)^+} - D_s \left.\frac{\partial c}{\partial x}\right|_{S(t)^-}$$

Freezing depression [5]

$$DH r_s \frac{\partial S}{\partial t} = - k_l \left.\frac{\partial T}{\partial x}\right|_{S(t)^+} + k_s \left.\frac{\partial T}{\partial x}\right|_{S(t)^-} ;\ T_i\left(S(t),t\right) = T_f^* - DT$$

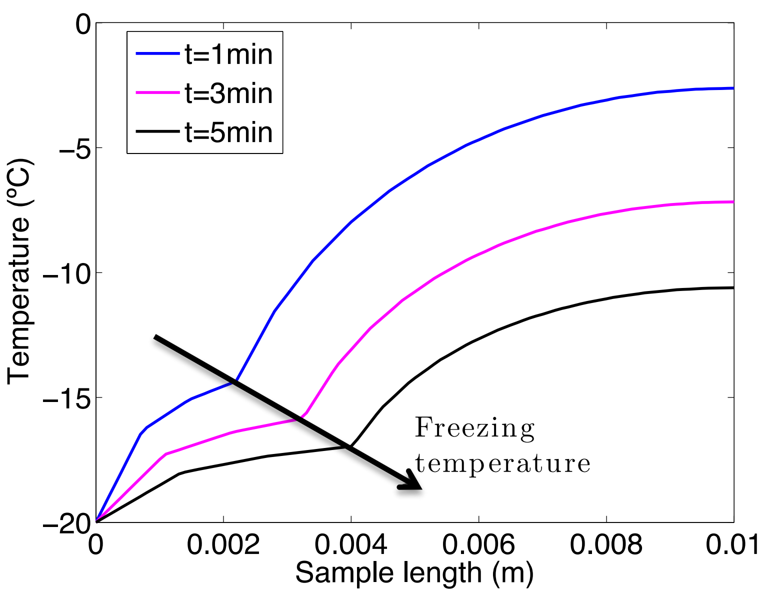
- External boundary conditions

$$T_s\left(0,t\right) = T_c < T_i\left(S(t),t\right);\ \frac{¶T_l}{¶r}\left(R,t\right) = 0 \qquad c_s\left(0,t\right) = c_{seed}\ ;\ \frac{¶c_l}{¶r}\left(R,t\right) = 0$$

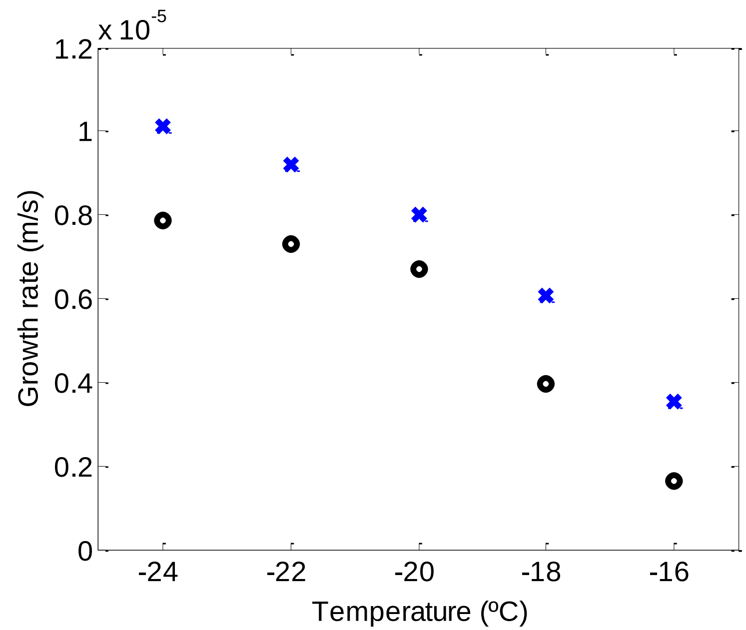
[5] Ross, Y. (2005). *Phase transitions in Foods*. Academic Press, London.

Secondary crystallisation: results

- 60% sucrose solution
- FEM+ALE (adaptive mesh) in COMSOL
- 101 nodes, tol=10^{-6}, R=1 cm



Temperature distribution along the sample during the seeding simulation with T_c = −20 ºC .



Comparison of experimental (o) and simulated (x) growth crystal rates for different cooling conditions

Conclusions

- First approach to modelling of high concentrated systems

- Overall good agreement between models and experiments:

  - seeding model overestimates growth rates.

  - aeration affects heat transfer delaying ice crystal formation.

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