Formation of amorphous sugar in the syrup film – a key factor in modeling of industrial sugar drying

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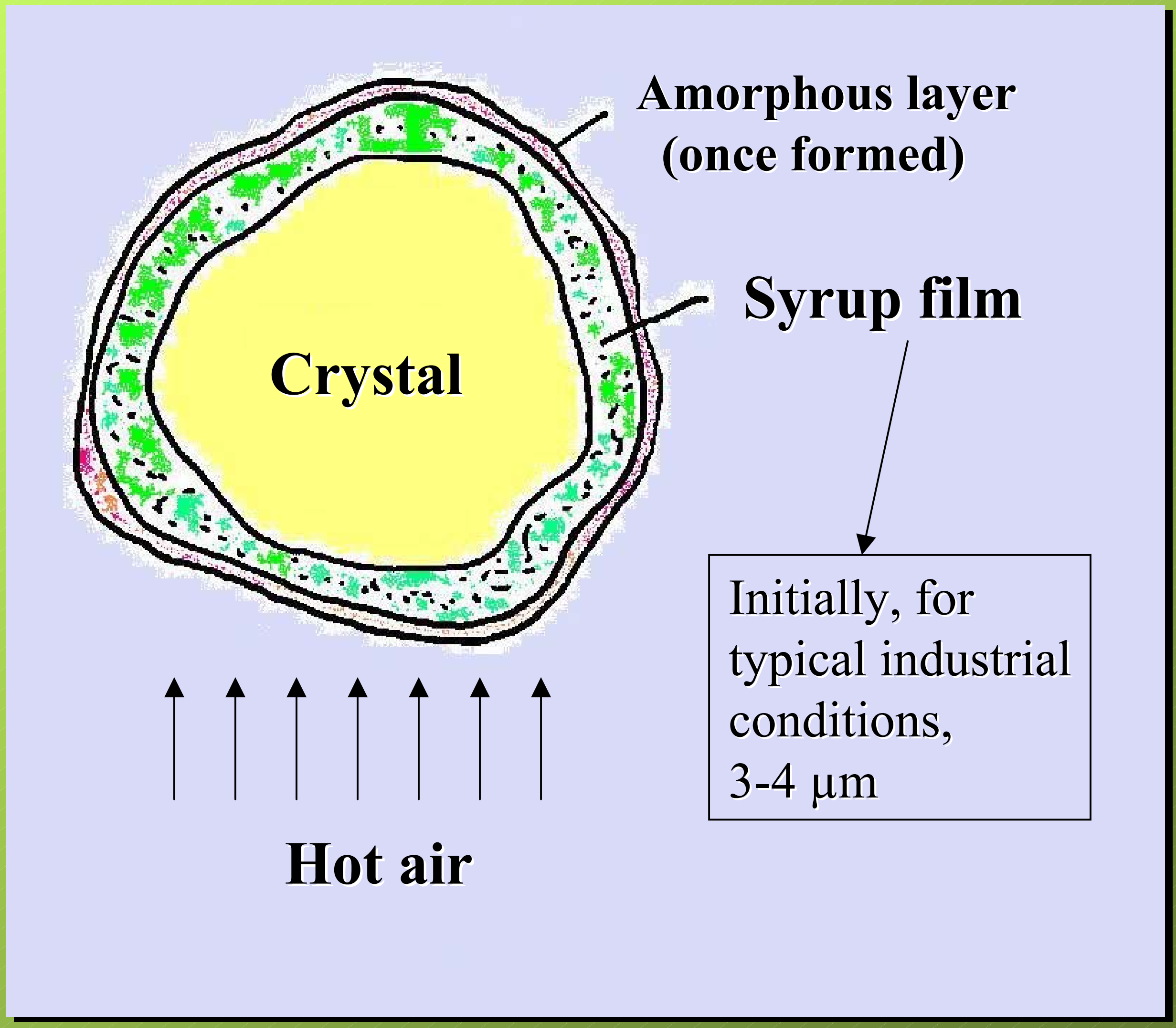
Outline

1. Sugar crystal, syrup film and amorphous layer
2. Key features of sugar drying
3. Cross-flow rotary dryer operation
4. Model development
5. Numerical simulation vs industrial measurements
6. Conclusions

Crystal of sugar and syrup film



What makes the process of sugar drying different from water evaporation from a film of pure water?

1. **Partial pressure of water above the syrup film depends on syrup composition (variable due to drying!)**

2. **Mass transfer resistance of the syrup film (absent for pure water!)**

3. **Crystallization of sugar from syrup**

4. **Possibility of formation of amorphous sugar.**

Formation of amorphous sugar crust

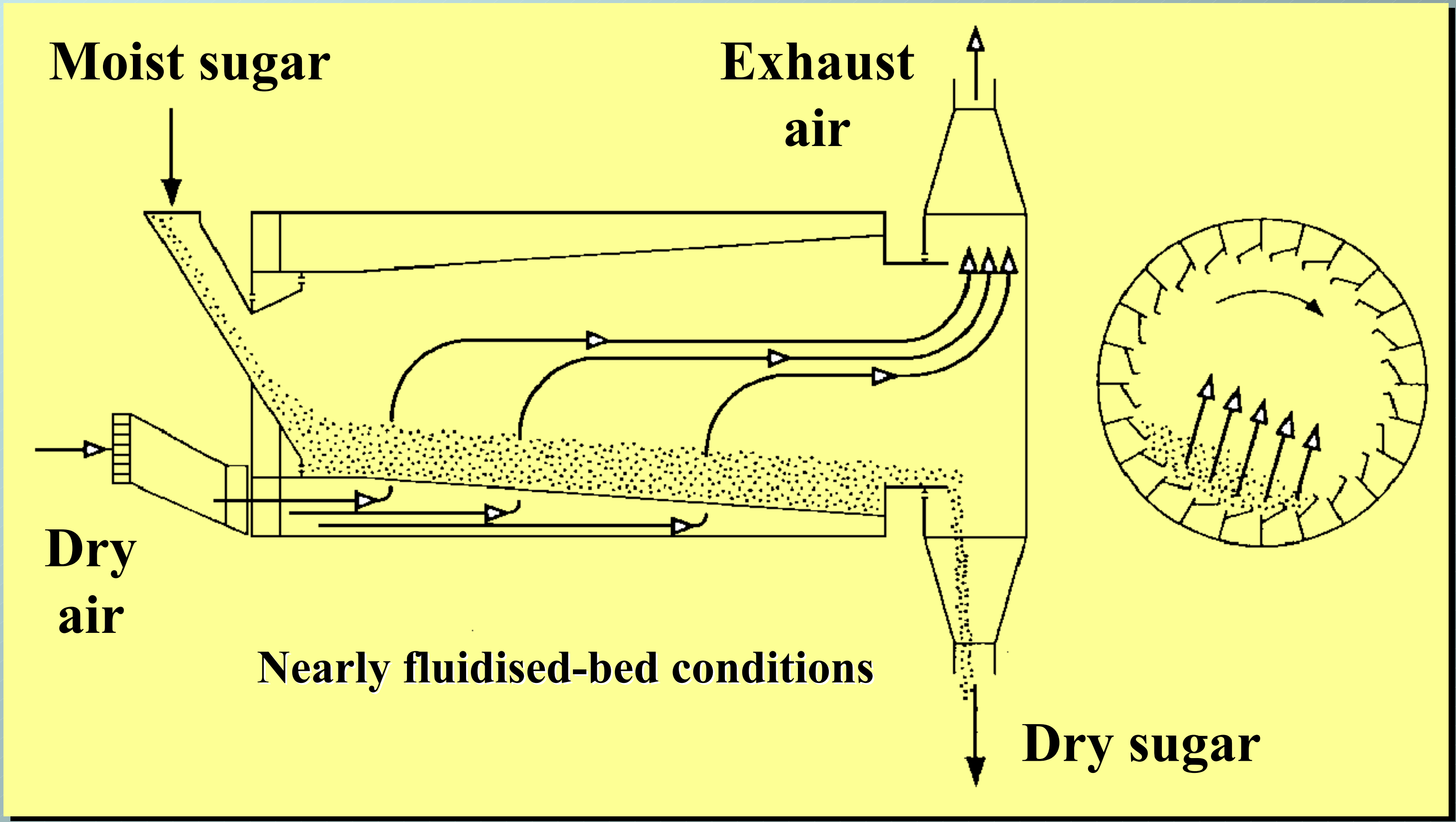
MECHANISM

- **Rapid evaporation from the outer layers of the syrup film**
- **Insufficient rate of water diffusion through the inner layers of the syrup film**

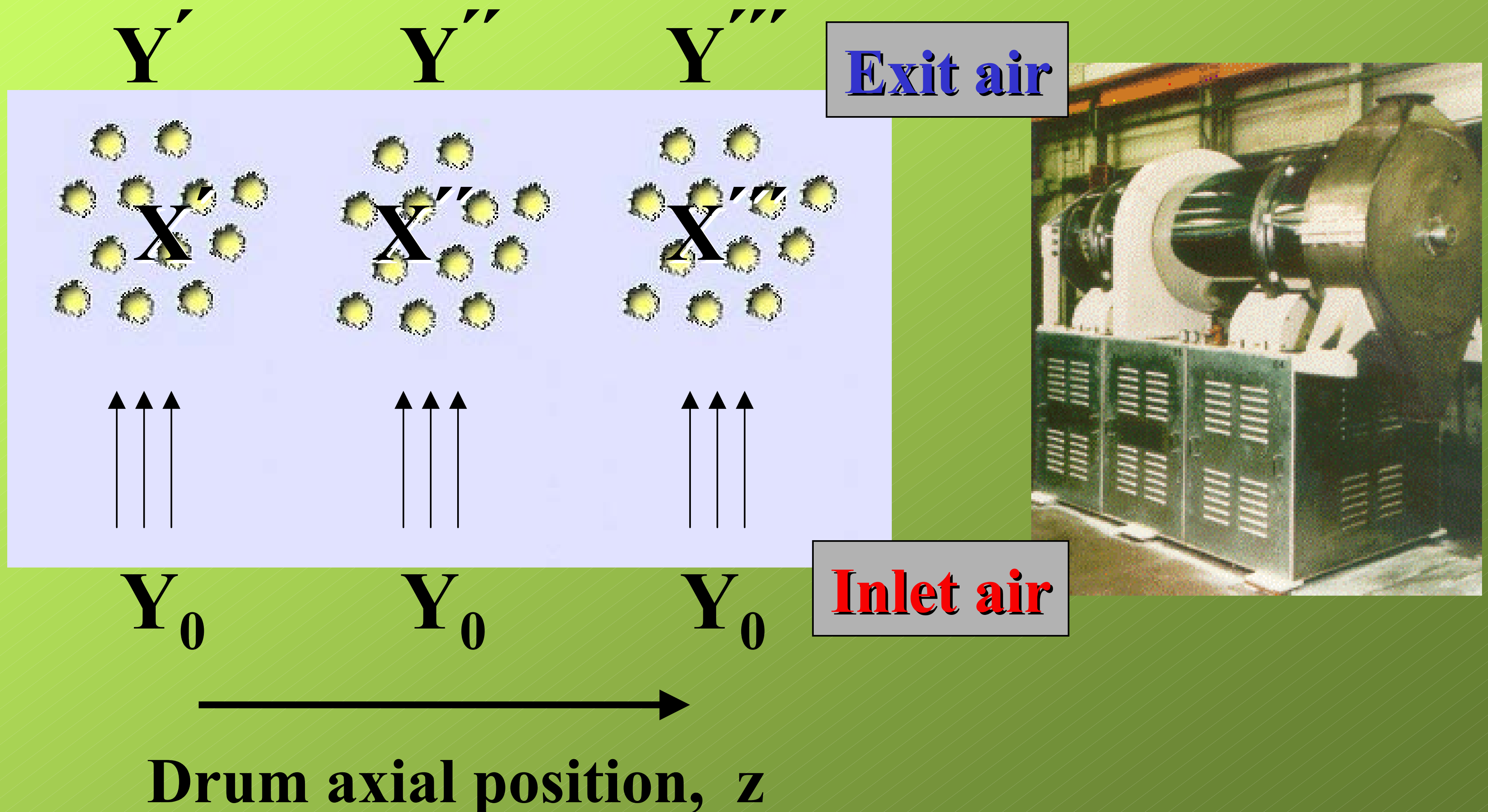
CONSEQUENCES

- **High supersaturation of syrup near the interface**
- **Formation of an amorphous sugar crust at the interface**
- **Water trapped within the syrup film**
- **A dramatic decline of the rate of drying**

Rotary Louvre dryer -
- principle of operation



Varying conditions of evaporation



Model development
Basic assumptions I

GENERAL

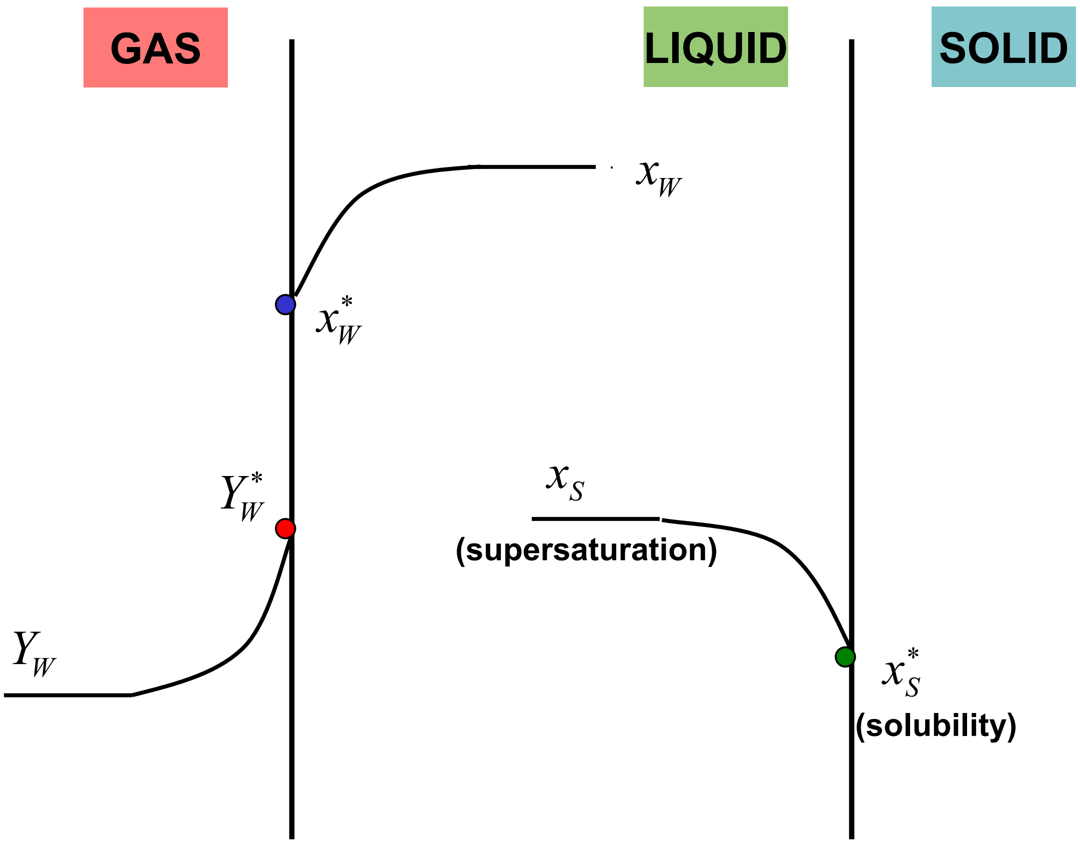
- **three-phase system**
  (air, syrup, crystals+amorphous sugar)
- **syrup contains impurities**
- **perfect mixing model for cross-flowing air**
- **plug flow model for crystals and moisture**
- **uniform crystal size distribution**
- **steady-state & non-adiabatic operation**

Model development
Basic assumptions II

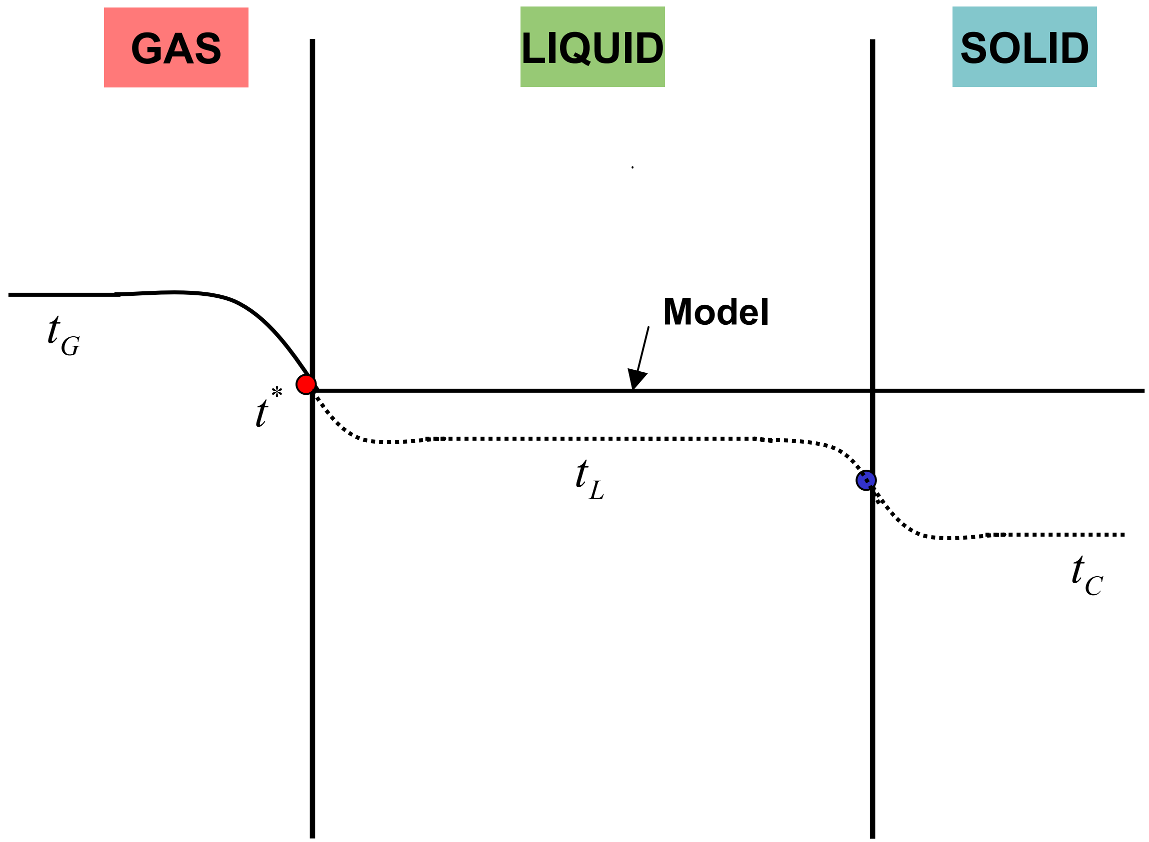
HEAT & MASS TRANSFER

- **two-film model**
- **heat & mass transfer analogy**
- **syrup and crystals in thermal equilibrium**
- **all properties composition & temperature dependent (incl. partial pressure of water)**

Water and sucrose concentration profiles across interfaces



Temperature profiles across interfaces

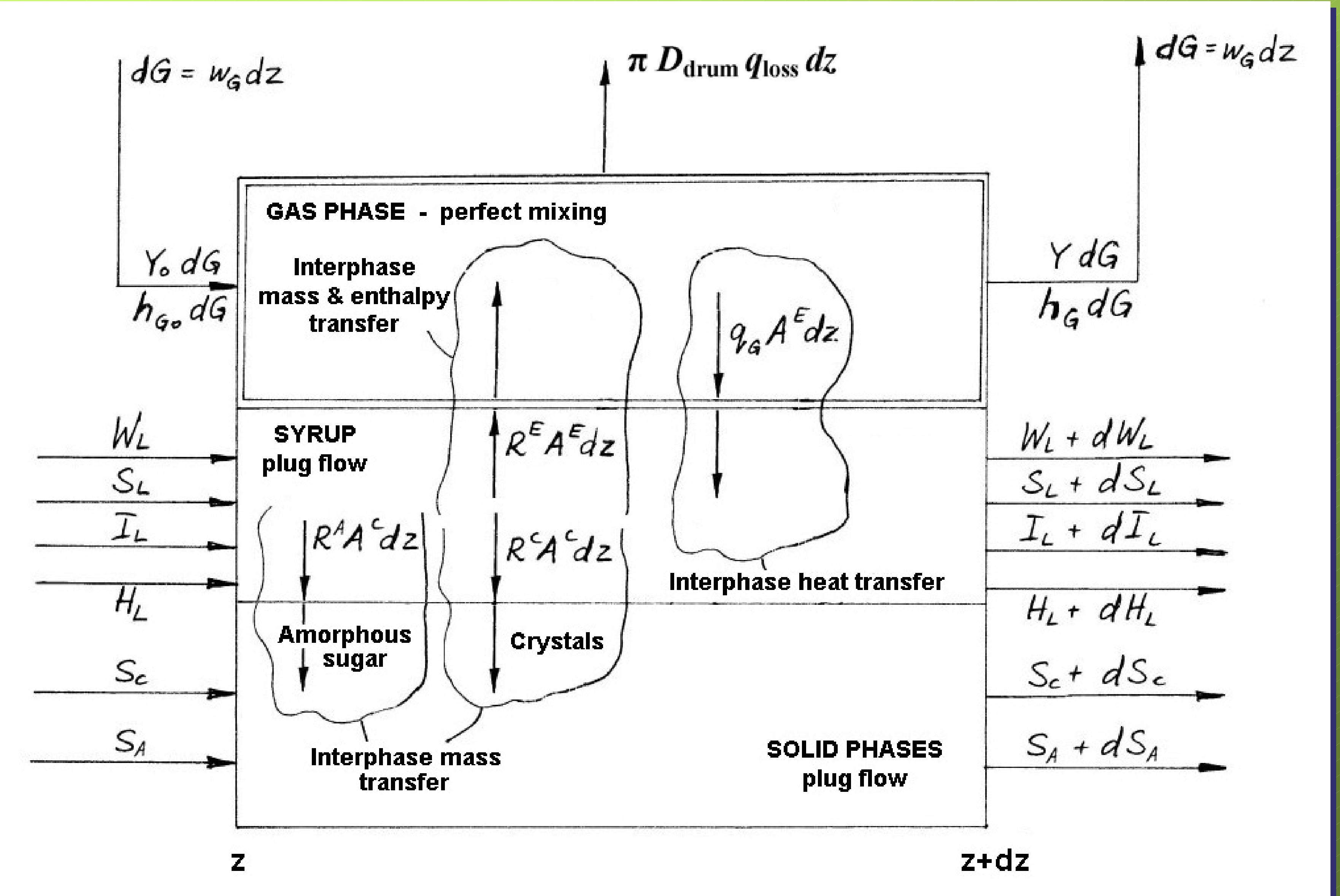


Model development
Basic assumptions III

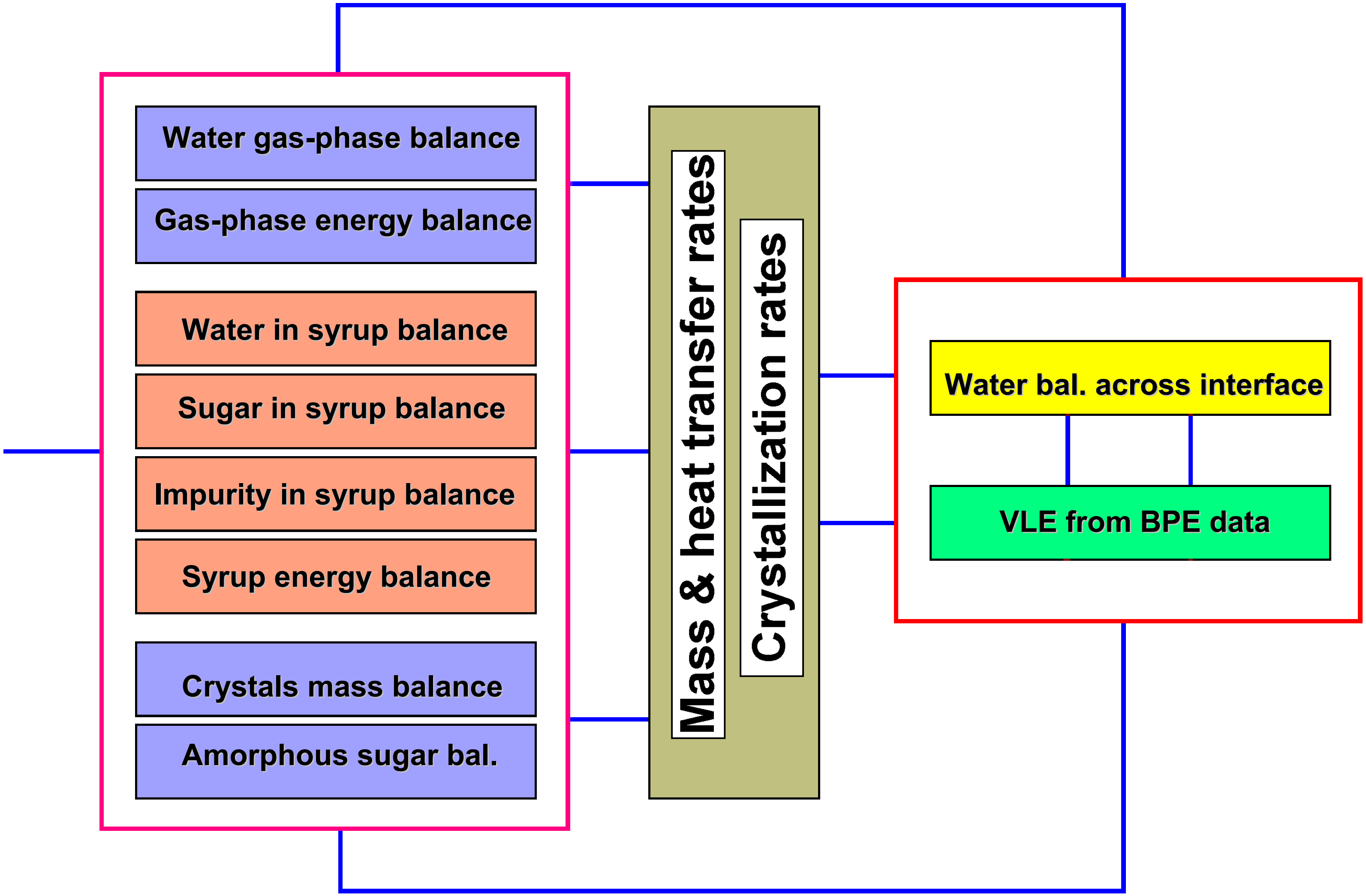
CRYSTALLIZATION / AMORPHIZATION

- sucrose transferred from syrup to:
  - crystals via normal crystal growth (within the metastable zone)
  - an outer amorphous layer via spontaneous agglomeration of sucrose clusters (above the metastable zone limit)
- negligible rate of impurity crystallization
- no inherent moisture in crystals

Differential element of the dryer



Basic equations of the model



SYRUP-AIR INTERPHASE MASS TRANSFER

Rate of evaporation

$$R^E = k_G(Y^* - Y) = M_W K(x_W - x_W^*)$$

Overall liquid/solid phase mass transfer coefficient

$$\frac{1}{K} = \frac{x_{Sm}\delta_L}{(1-\beta)C_L D_L} + \frac{\delta_A}{C_A D_A}$$

HEAT & MASS TRANSFER KINETICS

Gas-phase heat transfer

$$Nu_G = 0.88\,Re_G^{0.47}Pr_G^{1/3}$$

Gas-phase mass transfer

$$Sh_G = 0.88\,Re_G^{0.47}Sc_G^{1/3}$$

PHASE EQUILIBRIA

Gas-liquid
(boiling point elevation – Bubnik et al., 1995)

$$BPE = f\left(t_0(P), Brix, Purity\right)$$

Liquid-solid (sucrose solubility - Vavrinecz, 1962;
saturation coefficient – Tait et al., 1994)

$$Brix^*(t^*) = 64.447 + 0.08222\,t^* + \;......$$

$$SC = 1 - 0.088\left(c_I^* / c_W^*\right) \approx 1 - 0.088\left(I_L / W_L\right)$$

Rate of sucrose crystallization [kg/m²s]

Shardlow et al., 1996

Rate of sucrose crystallization

$$R^C = 0.00131\times(\sigma - 1.0046)\exp\left[-\frac{E(T)}{R}\left(\frac{1}{T}-\frac{1}{333}\right)-1.75\frac{I_L}{W_L}\right]$$

Supersaturation ratio

$$\sigma = \frac{Purity \times Brix}{100 - Brix}\,\frac{100 - Brix^*}{SC \times Brix^*}$$

SPECIFIC MASS & HEAT TRANSFER AREA

$$A\,[\mathrm{m}^2/\mathrm{m}] = \frac{6}{d_p}\frac{S_C\tau}{\rho_S Z}$$

NUMERICAL SIMULATION
Operating conditions

Total pressure:	**1 bar**
Sugar residence time:	**20 min**
Hot section:	**7.4 m**
Cool section:	**1.5 m**
Av. crystal size:	**0.55 mm**

NUMERICAL SIMULATION
Basic stream parameters

***Inlet hot air*:**

- **Flow rate of dry air:** **14 tons dry air/hour**
- **Absolute humidity:** **0.015 kg water/kg dry air**
- **Temperature:** **80°C**

***Inlet cool air*:**

- **Flow rate of dry air:** **11.6 tons dry air/hour**
- **Absolute humidity:** **0.015 kg water/kg dry air**
- **Temperature:** **30°C**

***Inlet moist sugar*:**

- **Throughput:** **25 tons/hour**
- **Sugar moisture:** **0.75 % wt.**
- **Sugar purity:** **0.995 kg tot. sucrose/kg tot. solids**
- **Temperature:** **65°C**
- **Supersaturation of syrup film:** $\sigma = 1$

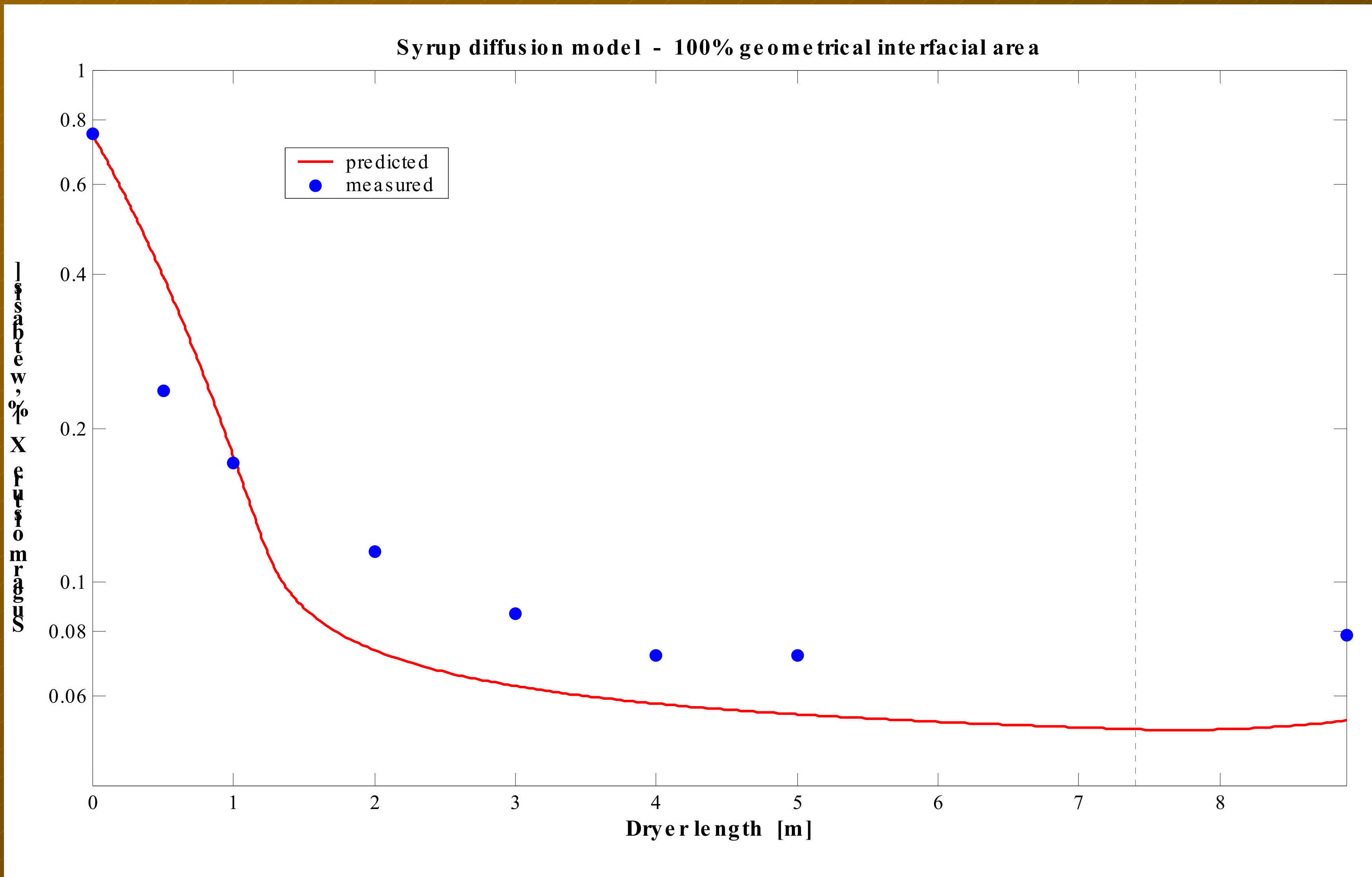
SIMULATION No. 1

Diffusion across the syrup film
Mutual diffusivity coefficient –
composition and temperature dependent
Non-equimolar diffusion

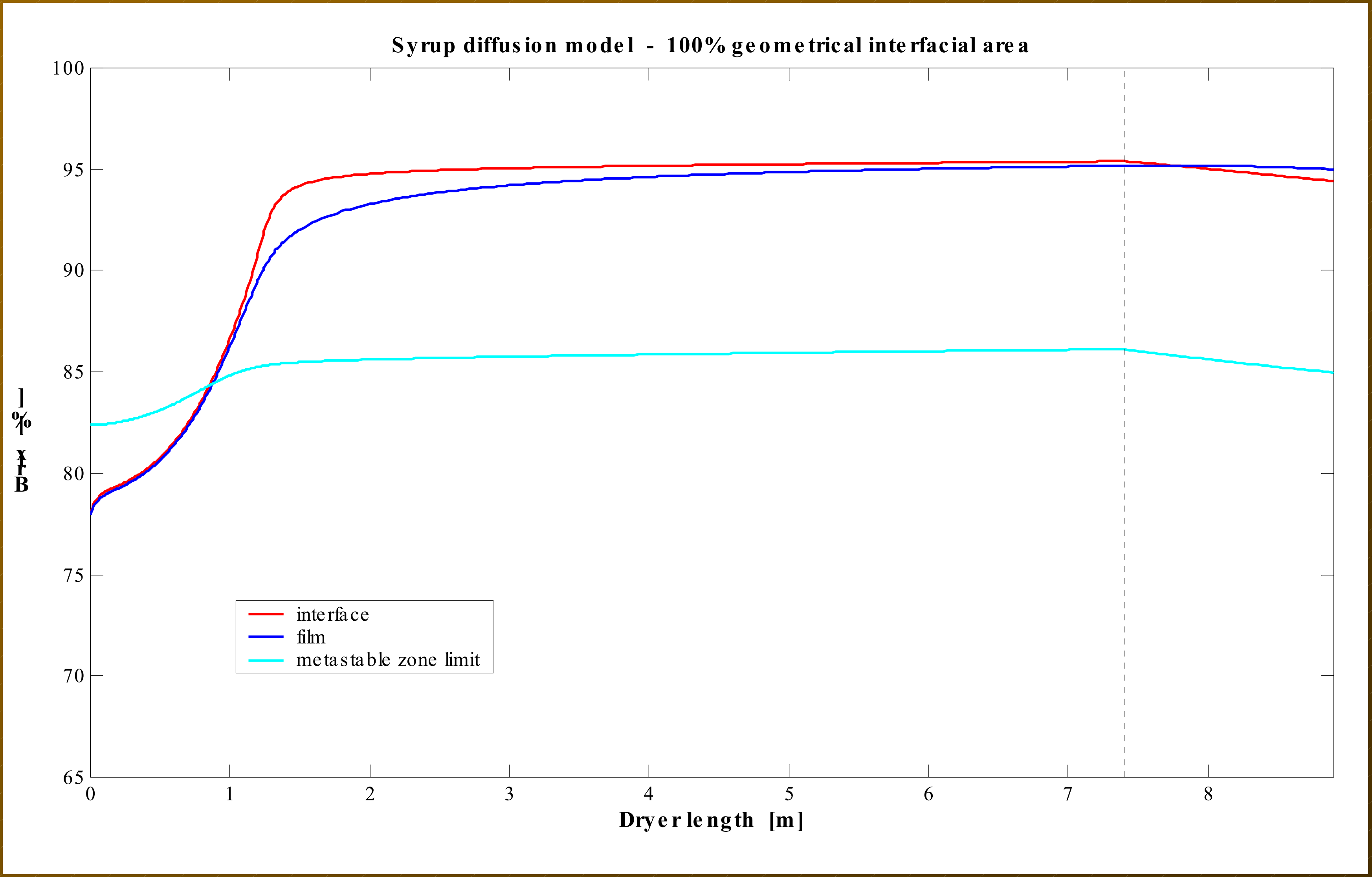
Amorphous layer not formed

Entire geometrical surface area used in
mass and heat transfer

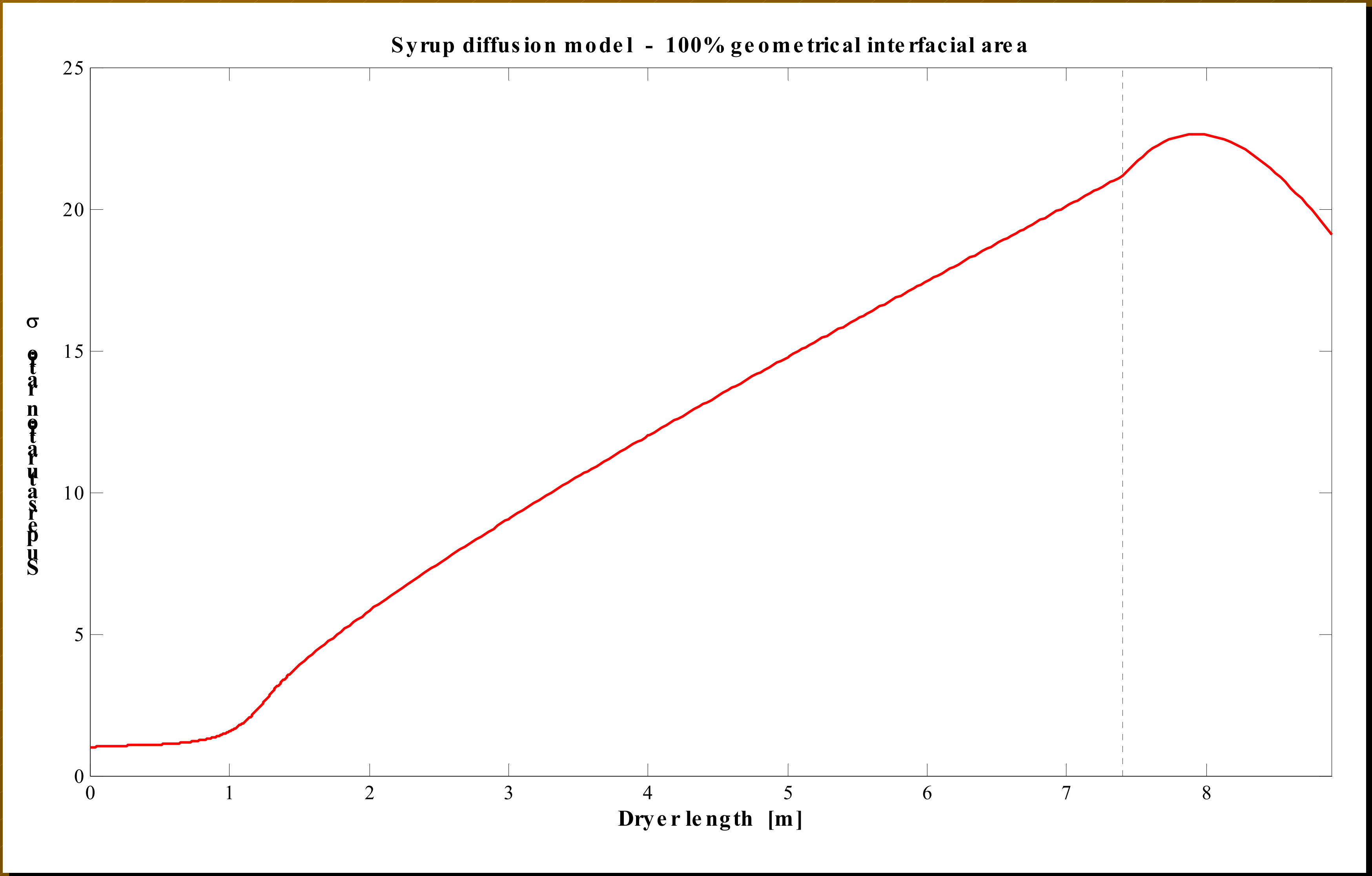
Moisture profile



Brix profiles



Supersaturation ratio profile



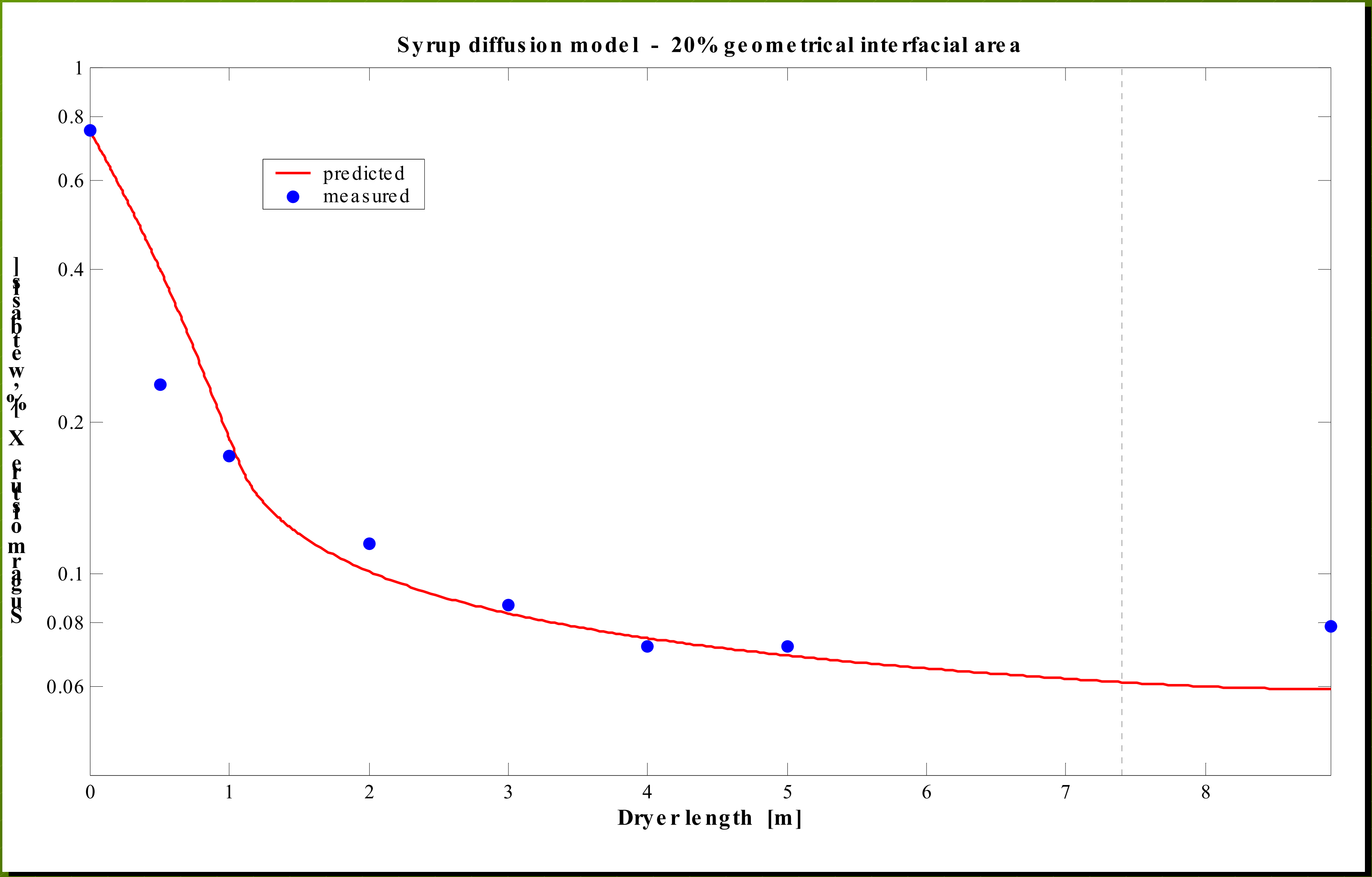
SIMULATION No. 2

Diffusion across the syrup film
Mutual diffusivity coefficient –
composition and temperature dependent
Non-equimolar diffusion

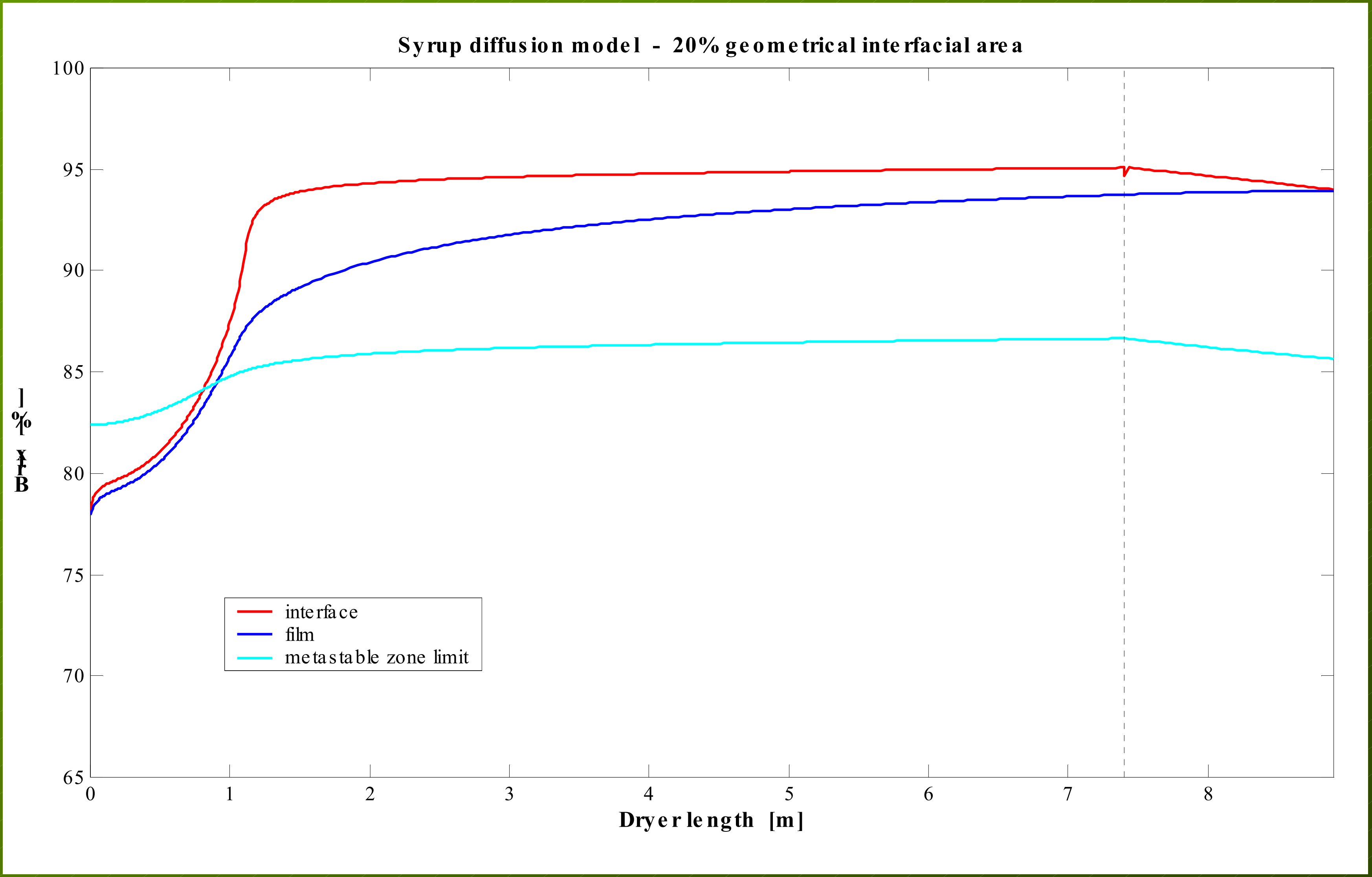
Amorphous layer not formed

Only 20% of geometrical surface area
used in mass and heat transfer

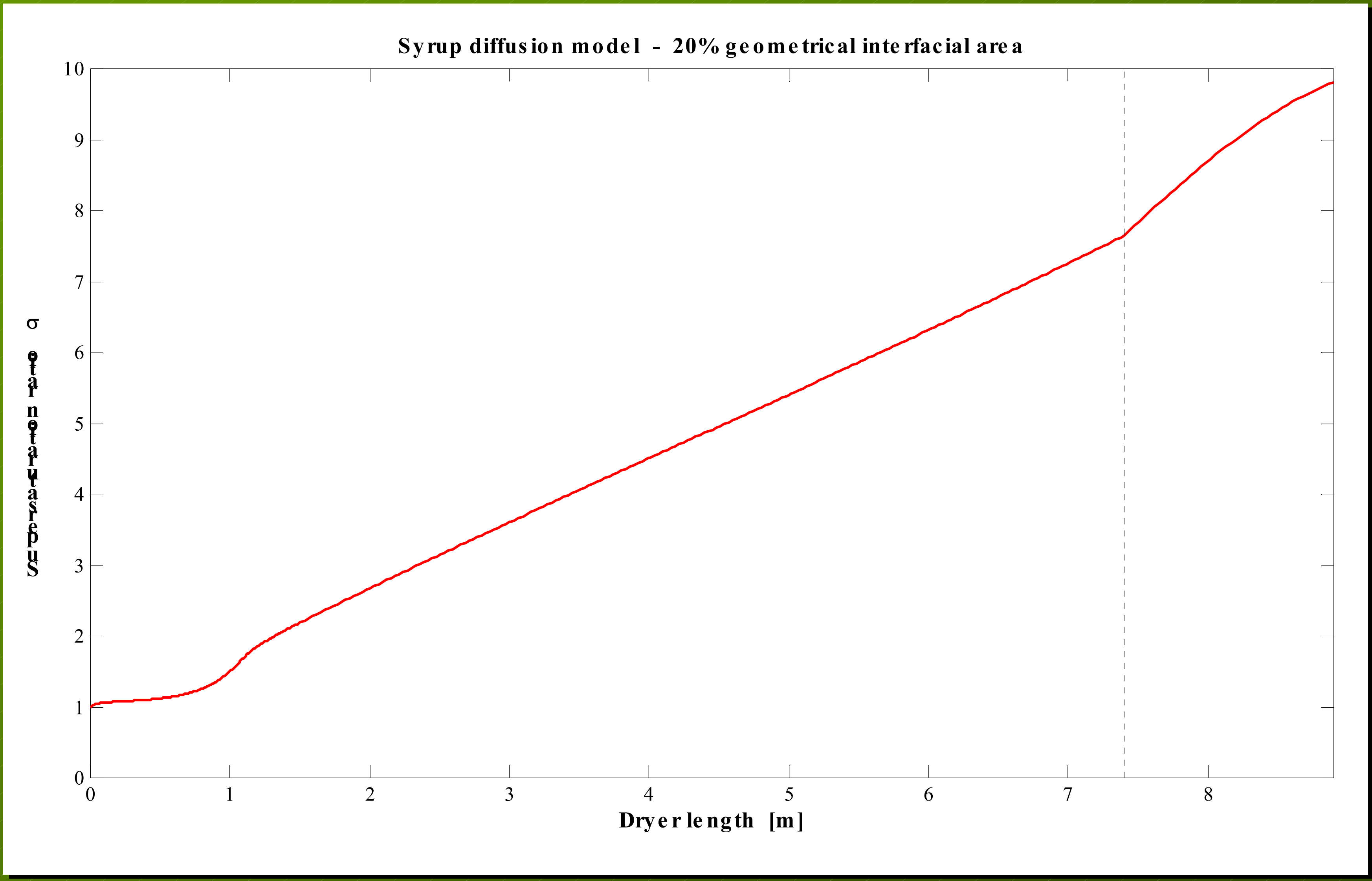
Moisture profile



Brix profiles



Supersaturation ratio profile



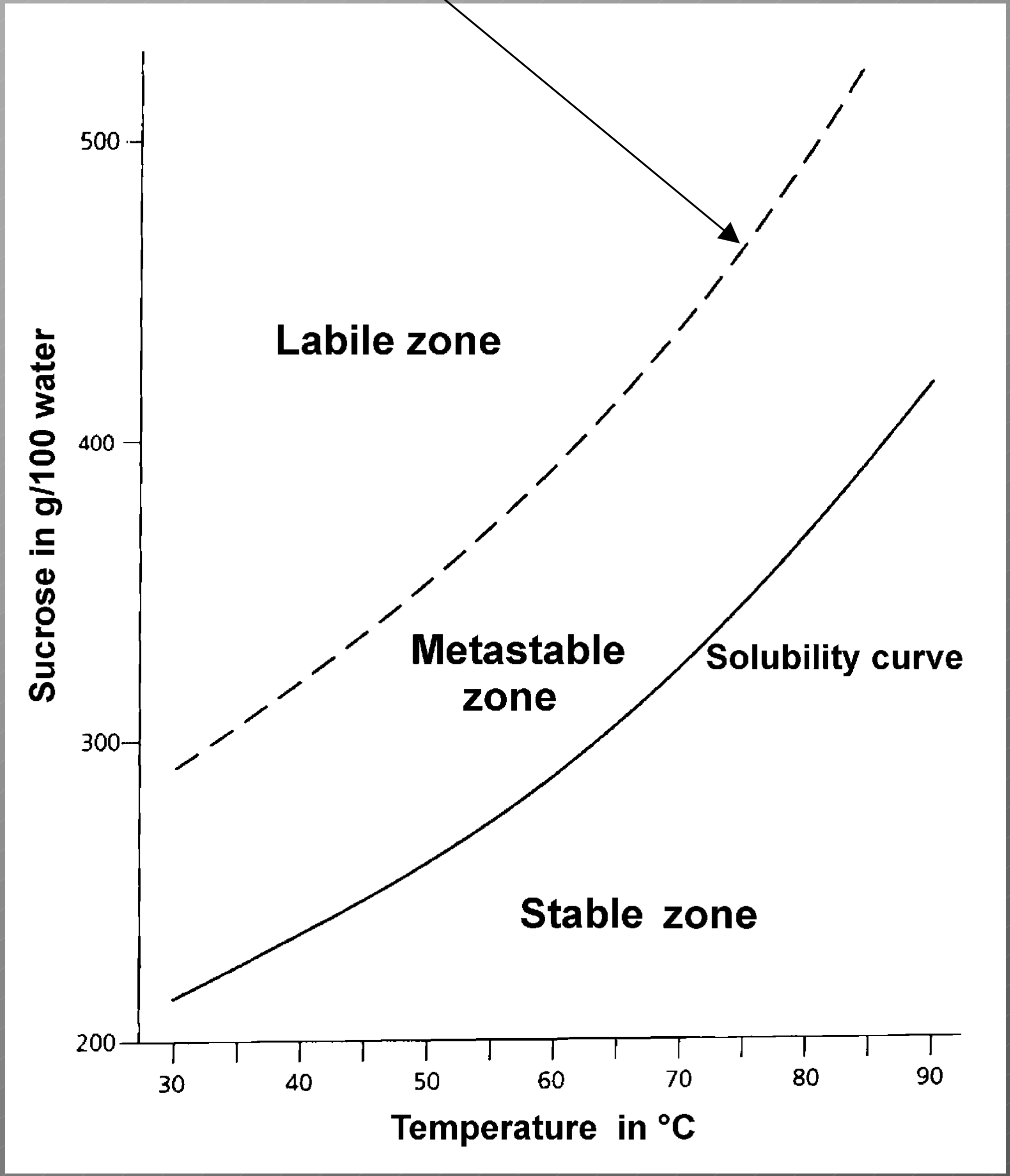
Estimation of water diffusivity in amorphous sugar

Fitting industrial data:

rotary Louvre dryer – Umfolozi Mill
sugar moisture by Karl Fischer titration

water diffusivity through amorphous sugar
assumed to be temperature independent

Onset of amorphous sugar formation



Rate of amorphous sugar formation ???

Model assumptions:

- amorphous sugar is formed in the labile zone
- the process is spontaneous, possibly by massive agglomeration of small sucrose clusters
- however, its rate is restricted by the labile zone conditions, i.e. any excess of sugar in the syrup film above the metastable zone limit is instantaneously converted to amorphous sugar

Rate of amorphous sugar formation - mathematical formulation

$$Brix\,(syrup\;\;film) \quad = \quad Brix_{crit}\,(metastable\;zone\;limit)$$

$$\frac{d(Brix)}{dz} \quad = \quad \frac{d(Brix_{crit})}{dz}$$

The above conditions allow for the rigorous determination of the rate of amorphous sugar formation (involves tedious algebra).

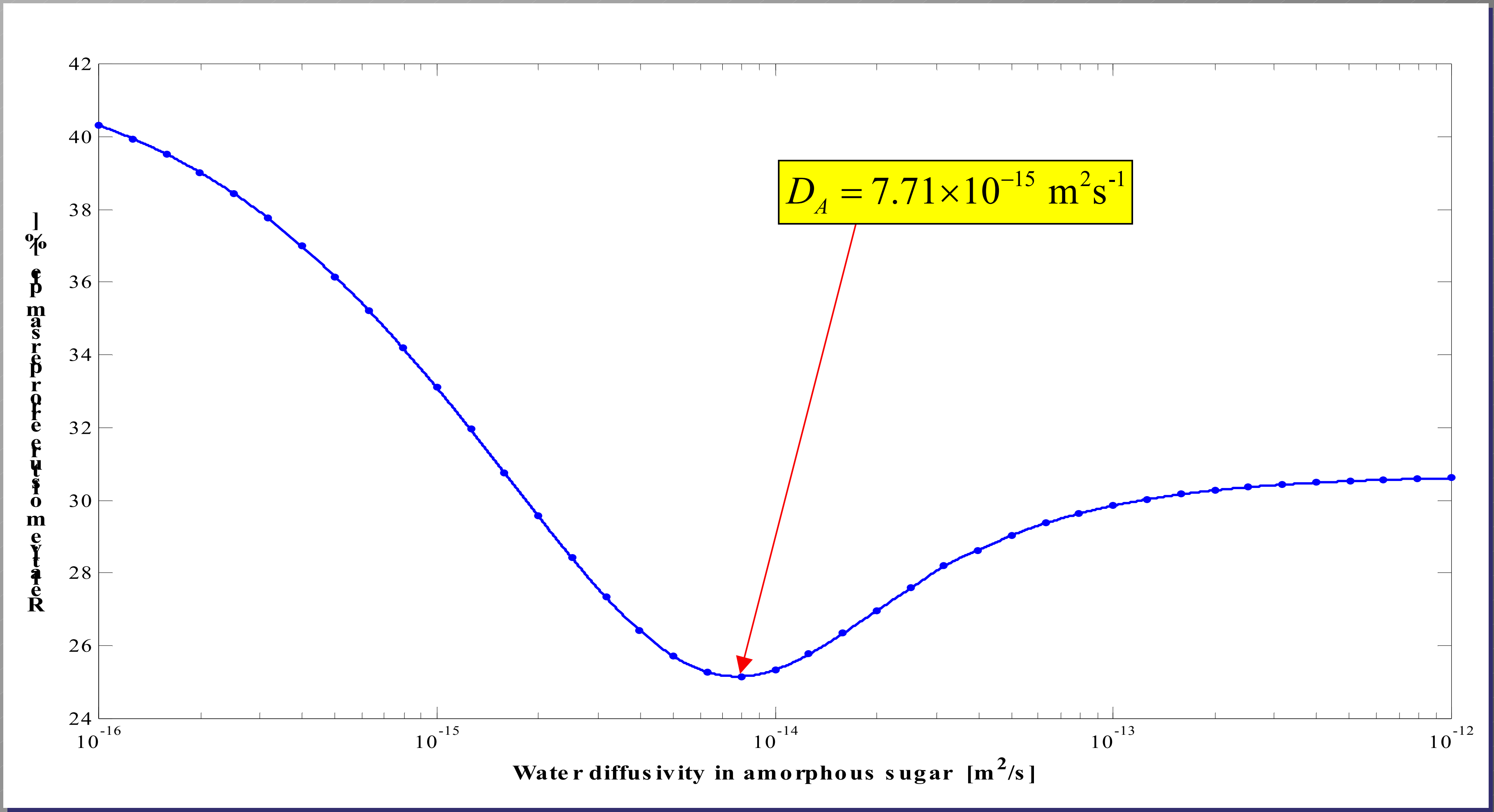
SIMULATION No. 3

Diffusion across the syrup film
Mutual diffusivity coefficient –
composition and temperature dependent
Non-equimolar diffusion

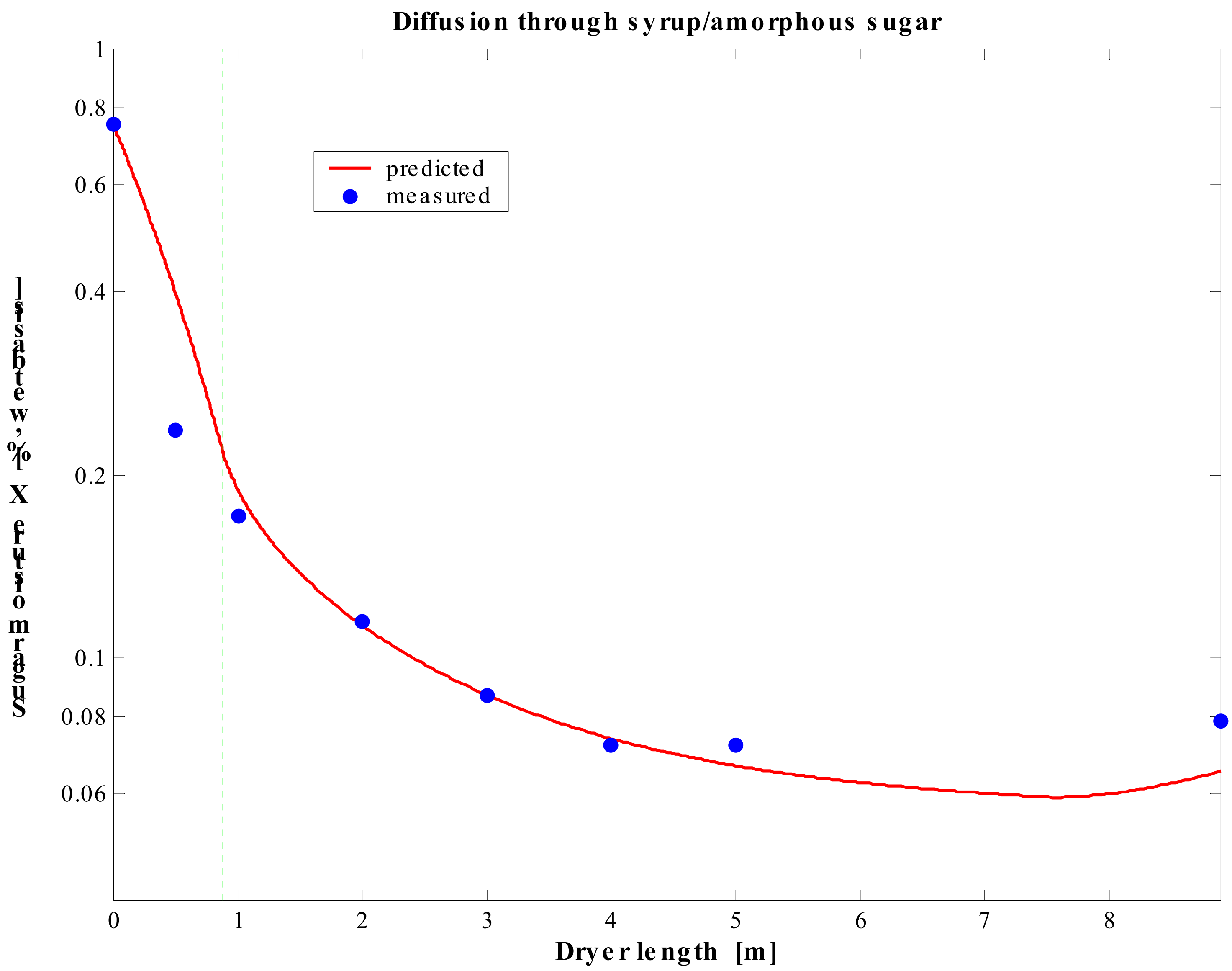
Entire geometrical surface area used in
mass & heat transfer

Formation of amorphous sugar
Constant water diffusivity

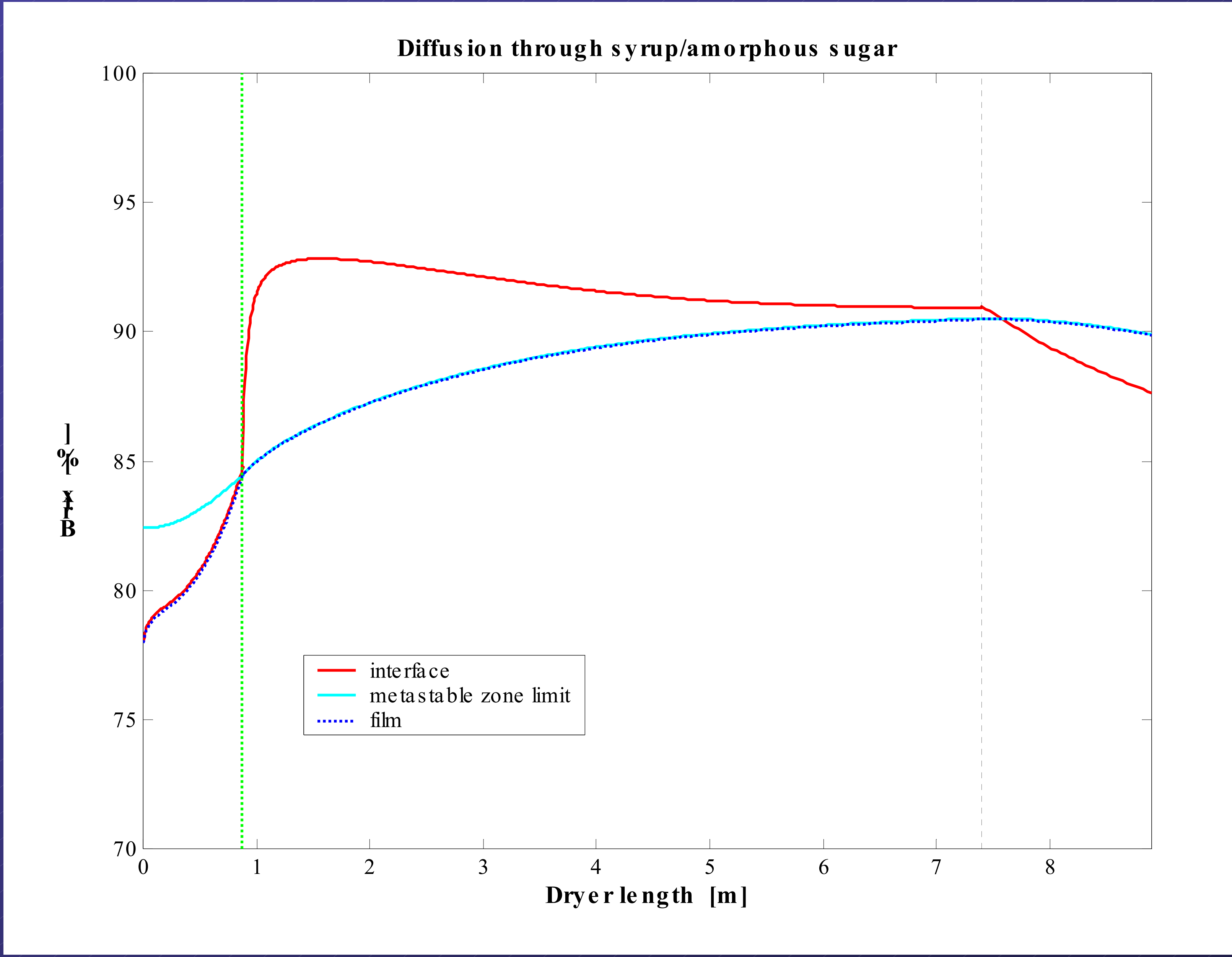
Estimation of water diffusivity in amorphous sugar



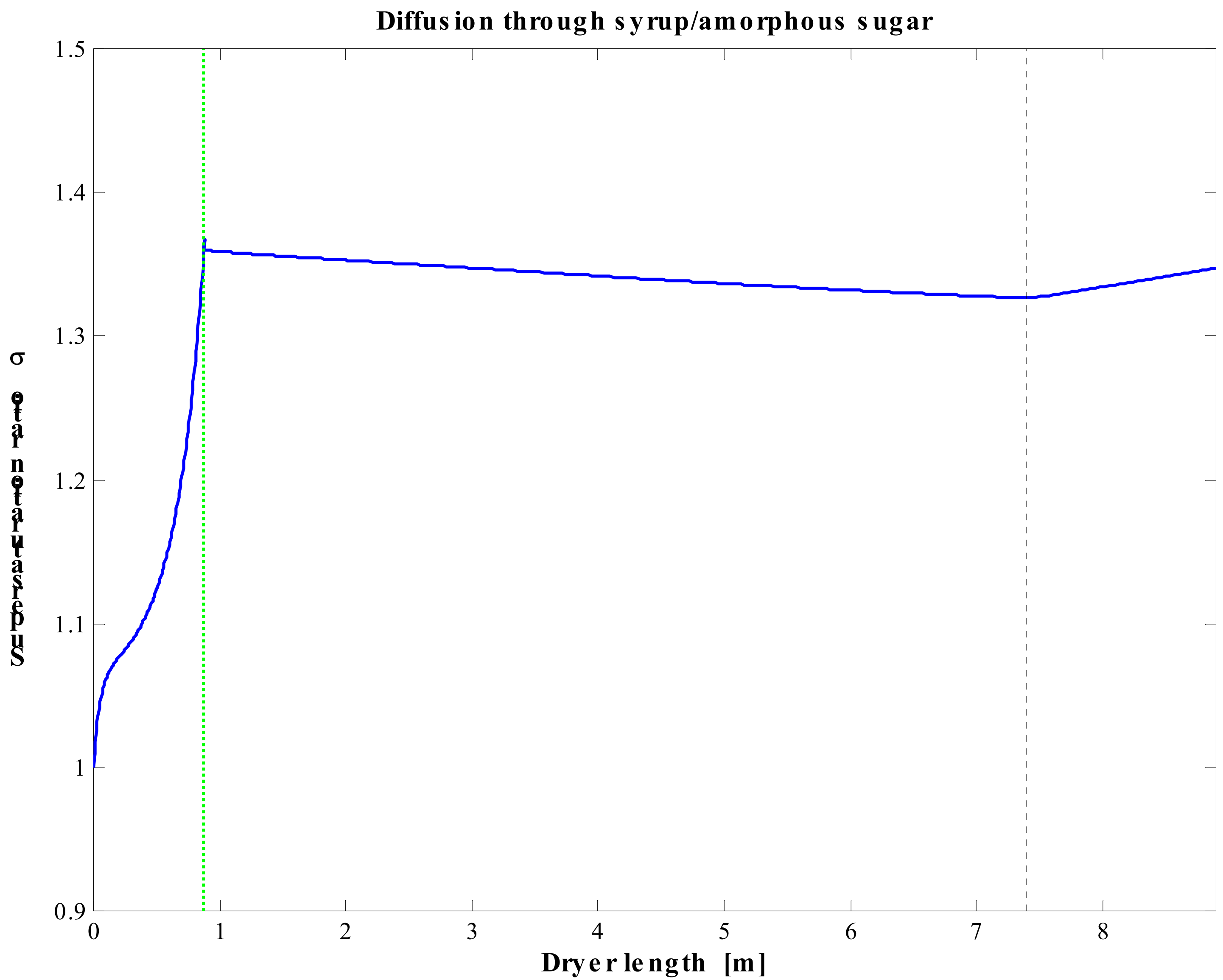
Moisture profile



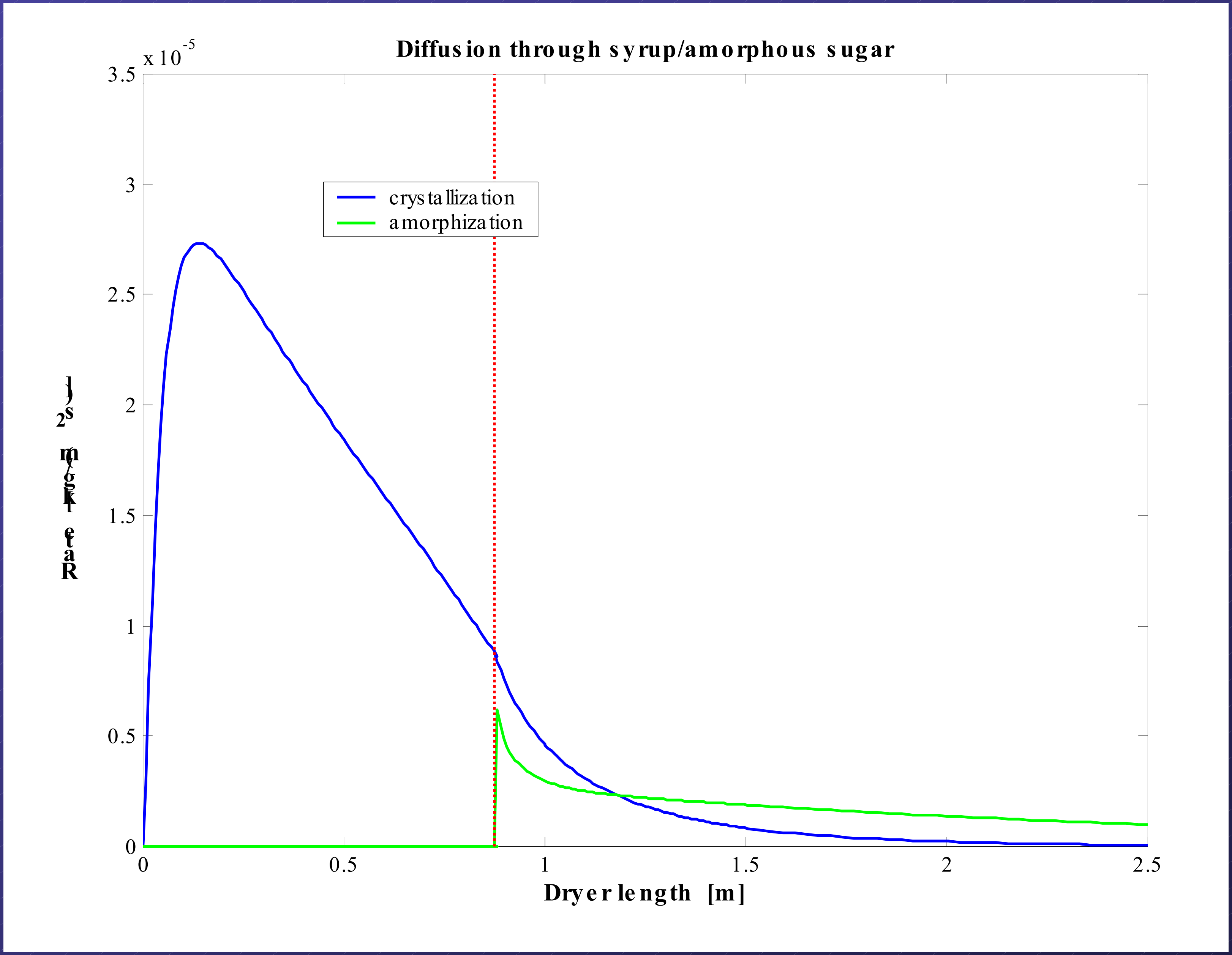
Brix profiles



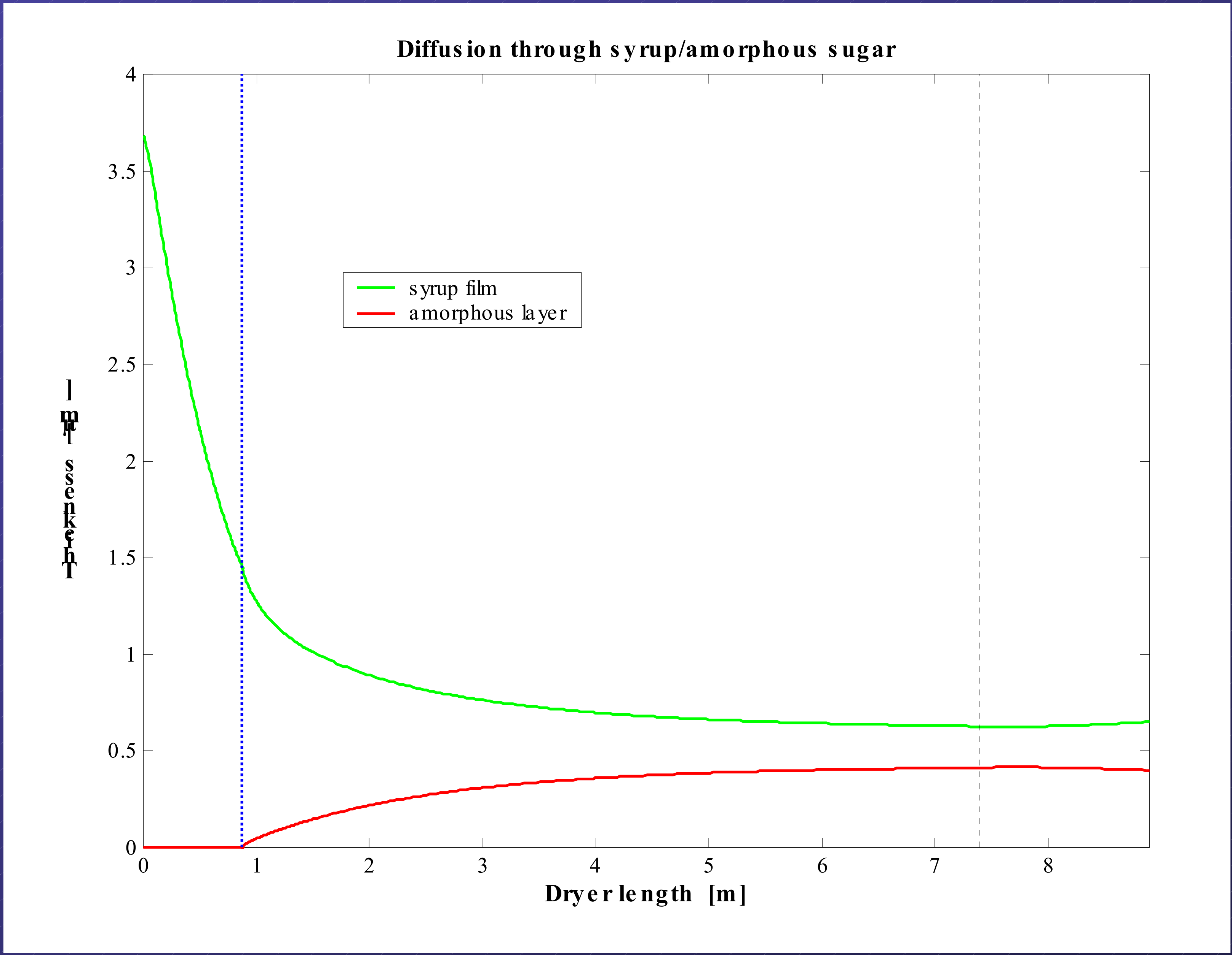
Supersaturation ratio profile



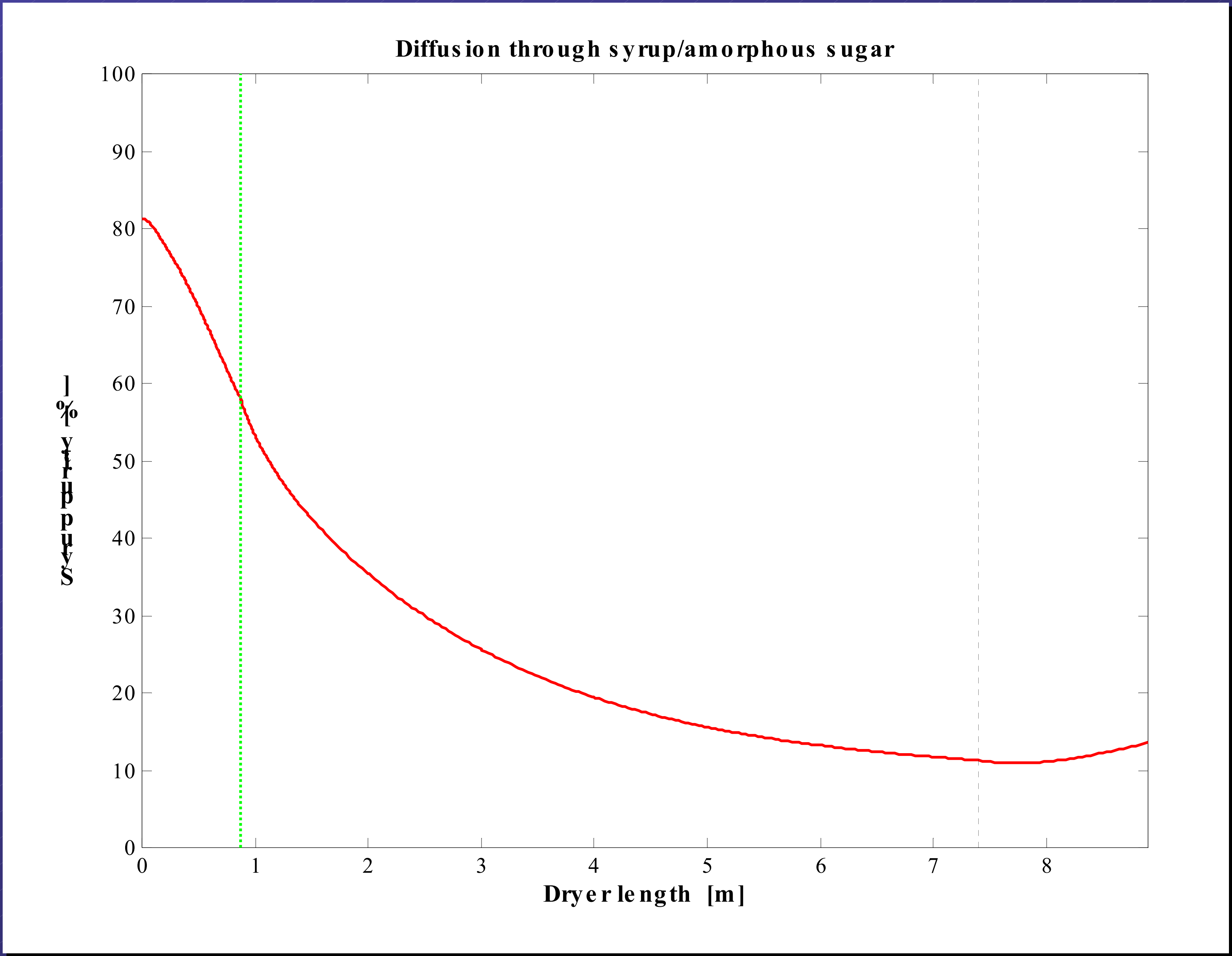
Crystallization & amorphization rate profiles



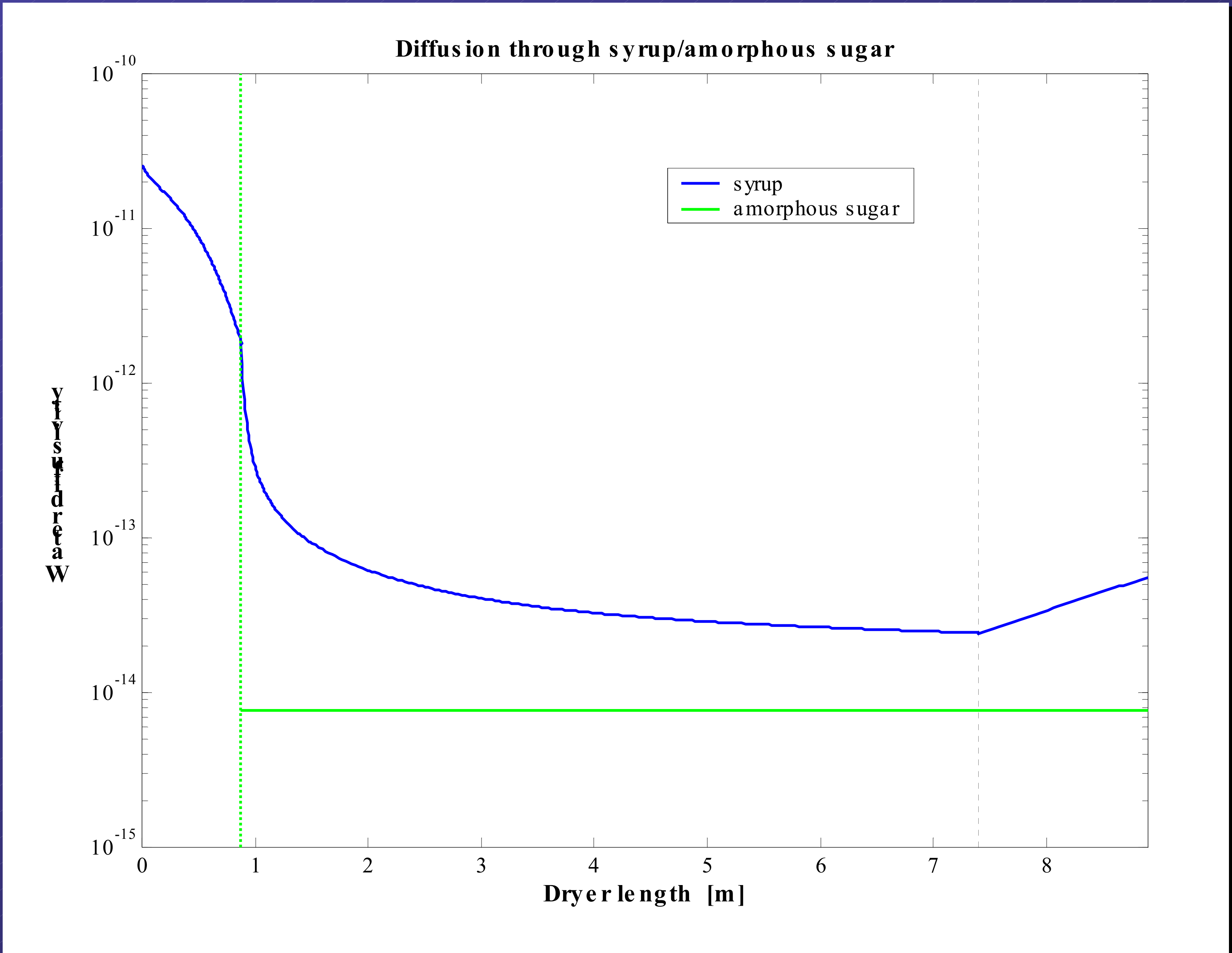
Syrup film & amorphous layer thickness



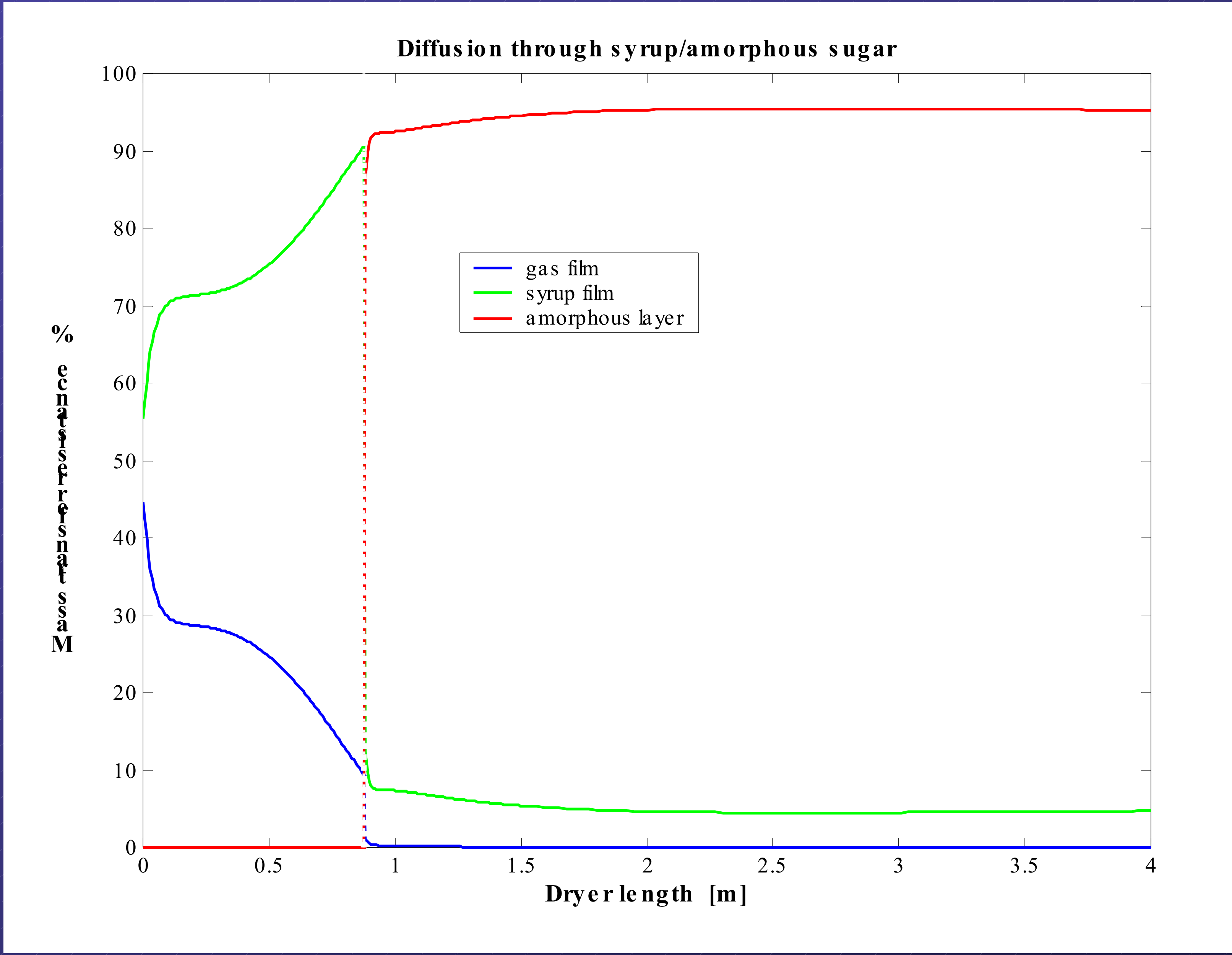
Syrup purity profile



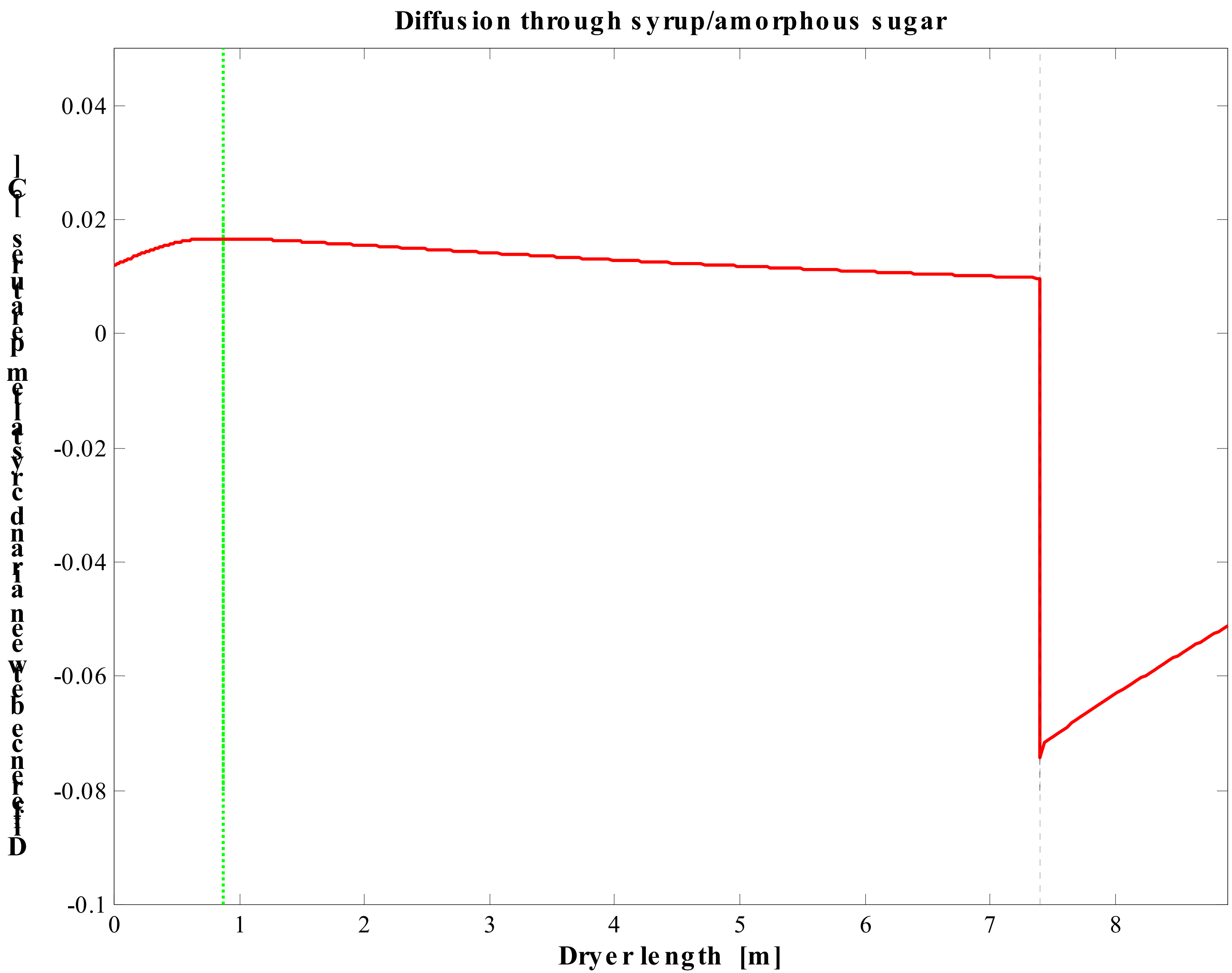
Water diffusivity profiles



Mass transfer resistance



Air-crystal temperature difference

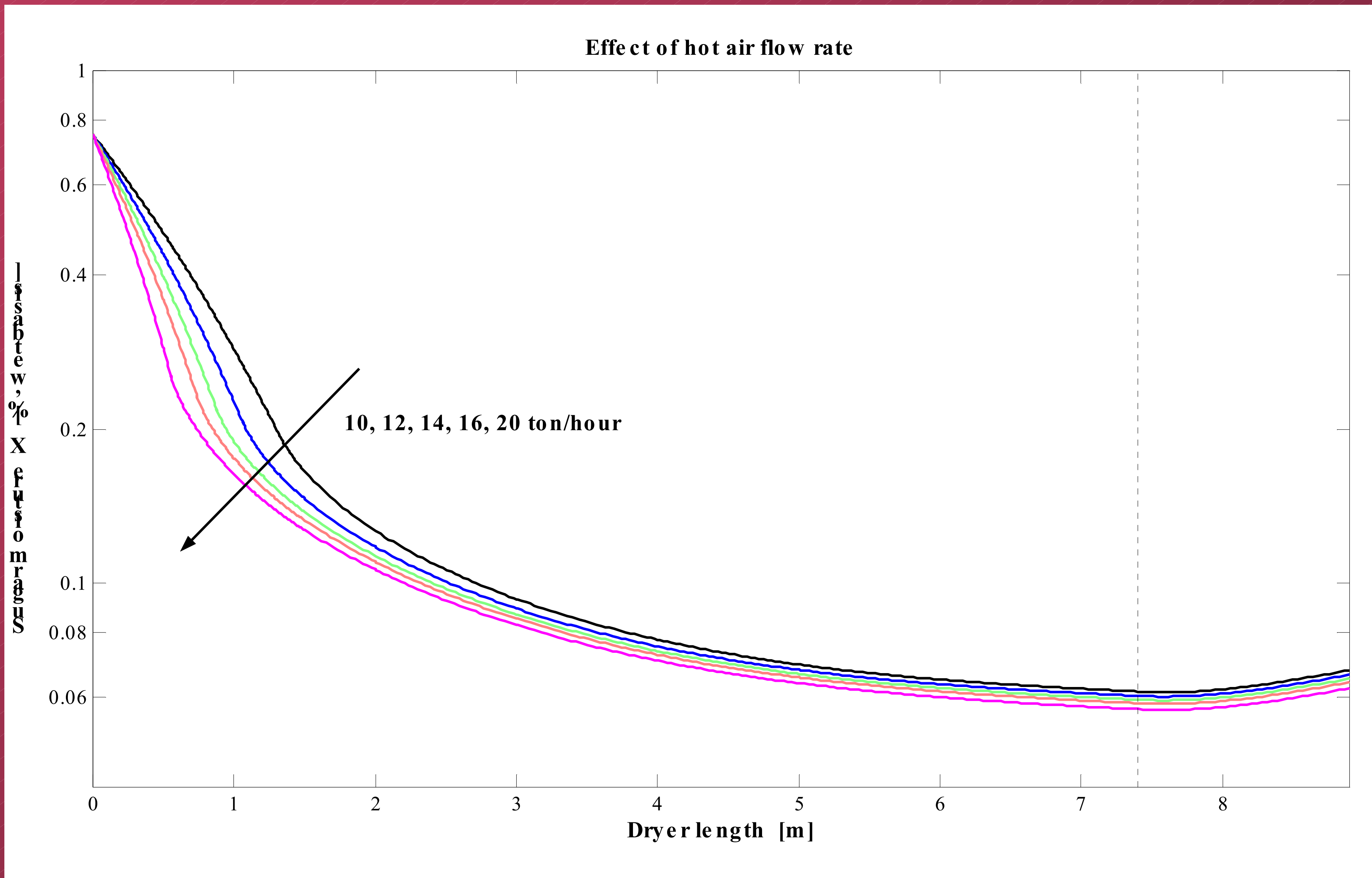


SIMULATION No. 4

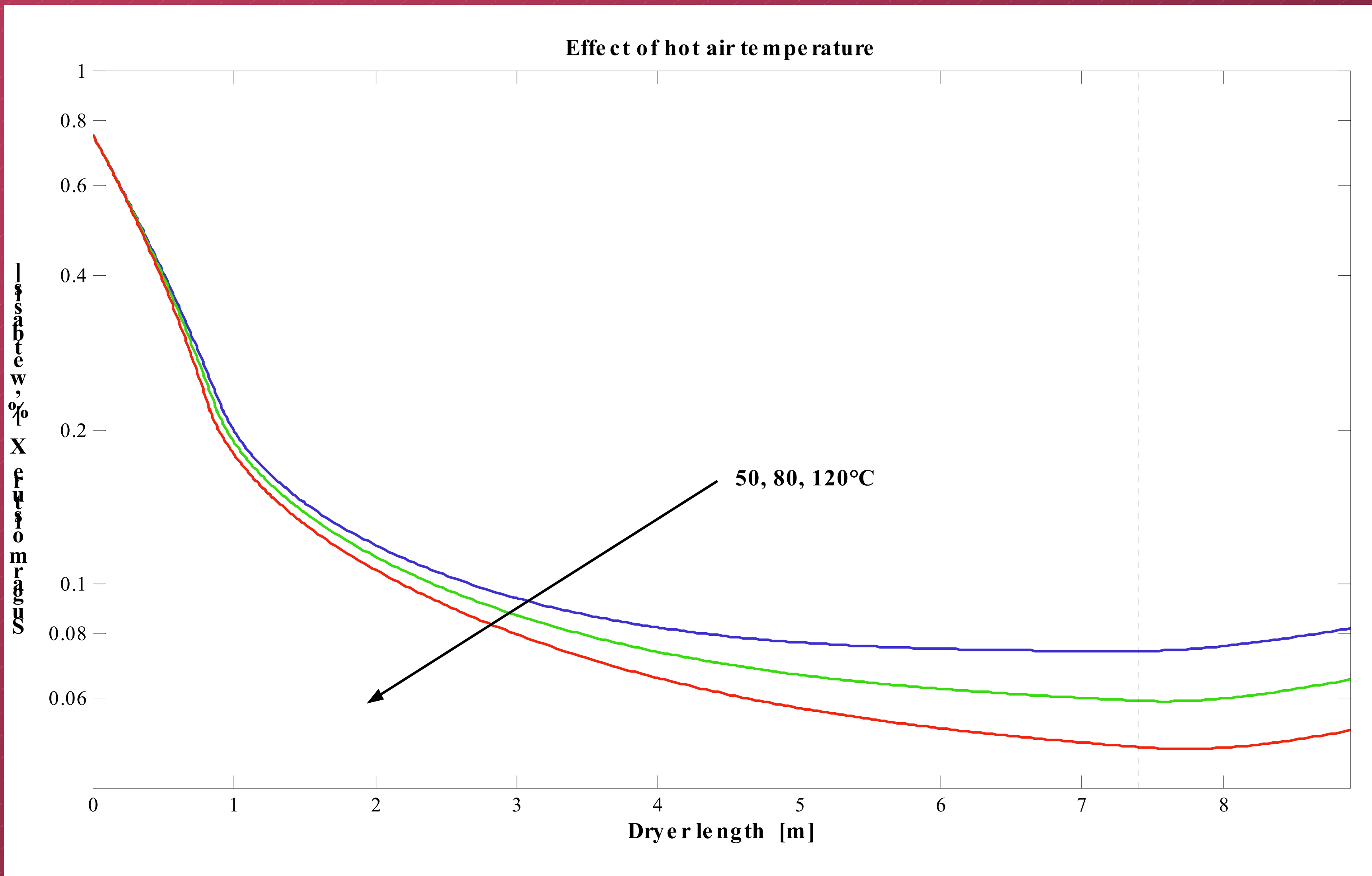
Studying the effect of various process parameters:

- **hot air flow rate**
- **hot air temperature**
- **crystal size**

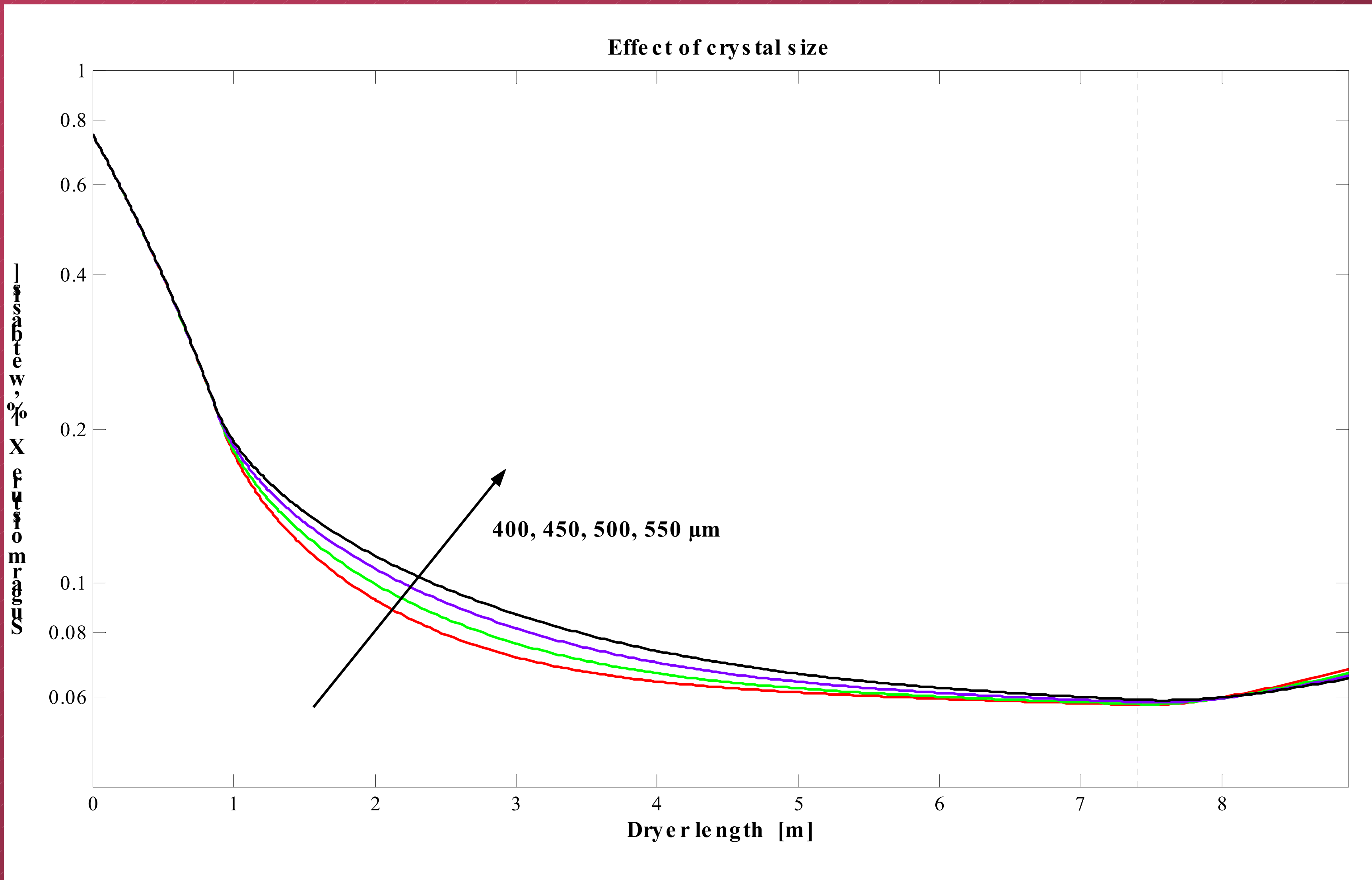
Hot air flow rate



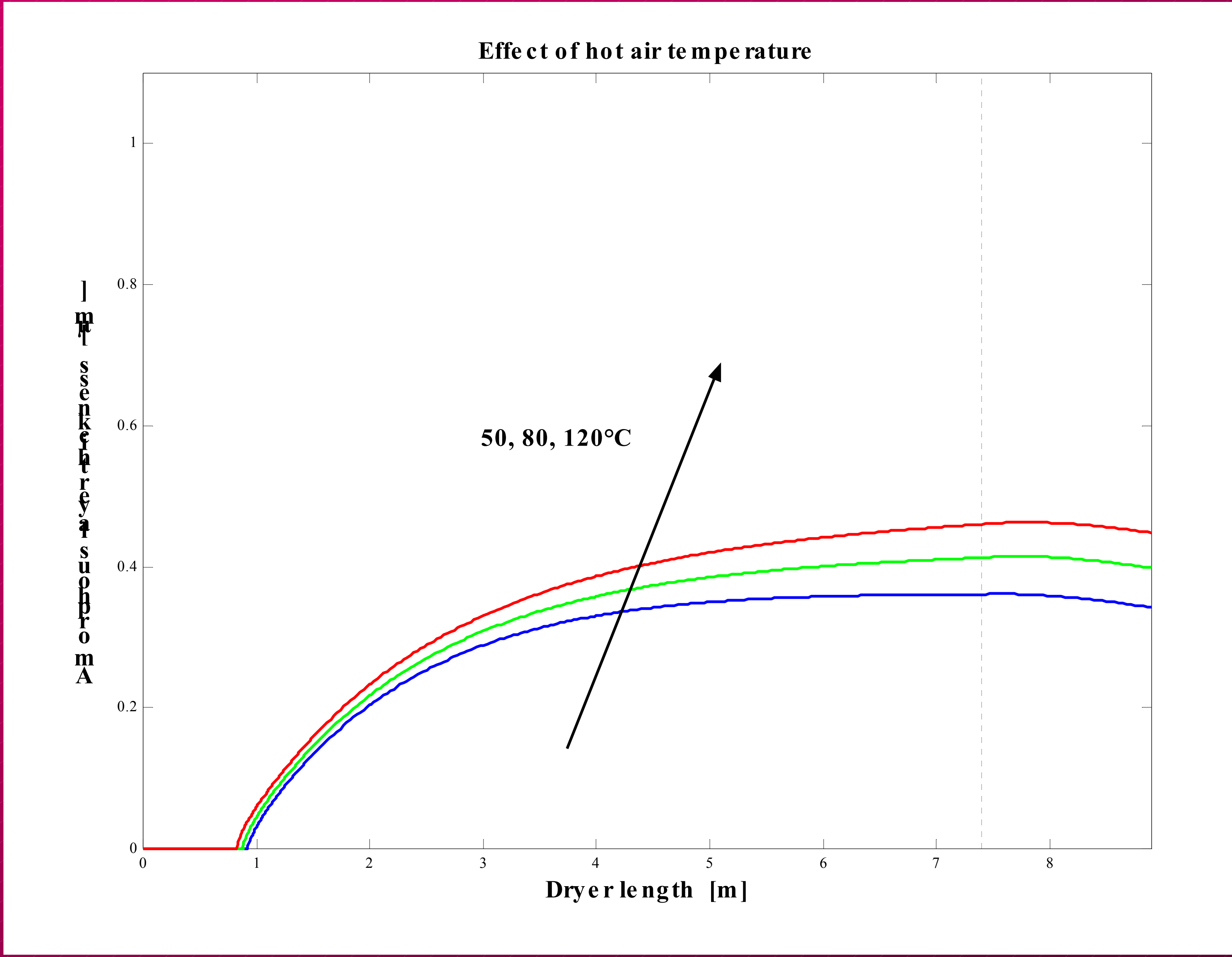
Hot air temperature



Crystal size



Amorphous layer *vs* air temperature



CONCLUSIONS

1. A model of the rotary cross-flow dryer for sugar was proposed.
2. Drying of sugar is mass-transfer controlled.
3. In the first period of drying, the major part of mass-transfer resistance is in the syrup film.
4. In the second period of drying, an external amorphous sugar layer is formed which slows the drying rate dramatically.
5. The temperature difference between the sugar crystal and the surrounding air is negligible.
6. Simulation results compare very well with sugar moisture measurements from an industrial cross-flow dryer.

Acknowledgements

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