Thermal pressure forming of Starch-Based Materials:

A Simplified Thermodynamic Model

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Abstract

A thermodynamic model describing the thermal pressure forming of starch based products is reported with the aim of explaining the surprising non-monotonic temperature and pressure time trends observed experimentally when heating a starch suspension in a closed mould. An explanation is based on an idea that the temperature of the sample is governed by the steam pressure drop during the free water evaporation phase. As opposed to the more complicated three layer core, crust and crumb model, Žitný (1999), the heated sample is considered here as a homogeneous fixed body having a uniform temperature and constant permeability. In spite of its simplicity (the model has only four parameters) not only the temperature, but also the pressure trends can be reproduced with reasonable accuracy.

1. Introduction

Biodegradable materials based on starch can be used for various packaging products, e.g. trays, dishes, cups, etc. They are manufactured in a thermal forming process, where a starch suspension is heated inside a closed space of the mould, e.g. between two parallel plates of an electrical heater. Even in a simple mould geometry, the process is complicated, mainly because of the complex nature of starch which changes its structure and properties with temperature and time. Information about the process is incomplete, but interesting: measurement at a center of circular sample reveals non-monotonic trends of temperature and pressure with time, see Fig.1, Tsiapouris (2001). Heated samples of potato starch suspension expand significantly, volumetric expansion reaches 270%, see Tsiapouris (2001).
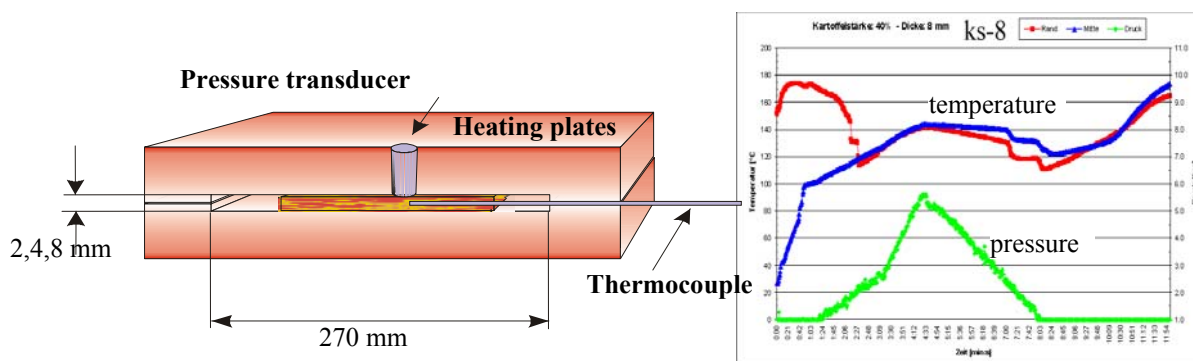

Fig.1 Experimental setup. Measurement of temperatures and pressure in thermal pressure forming of thin plates from potato starch, Tsiapouris (2001).

2. The Model

Mathematical models describing heat and mass transfer during frying or roasting of meat, and baking bread or biscuit were presented by Farkas, Singh & Rumsey (1996), Õzilgen & Heil (1994), Singh (1995), or Zanoni, Pierucci & Peri (1994). There are some common features in these models, usually the assumption of two regions - dried crust and moisten core - with a moving interface between them. The integral model suggested by Žitný et at. (1999) considers a thin circular sample

with three parallel layers: inner liquid core, porous crumb and a compact surface crust. Thickness of these layers is gradually changing during the second stage of the process when free water is evaporated and at the same time the whole sample expands and fills up the closed mould space. The model mentioned assumes that the formation of layers and expansion of the whole sample is governed by the pressure of steam flowing through the porous crumb and through a narrow gap between the surface of sample and heater walls. The pressure drop of steam is changing not only because the mass flow-rate of steam is changing but also because the flow resistance is not constant. Prediction of this friction resistance is rather complicated if we take into account radial flows through the crumb and along the wall and the cross-flow between these streams through the crust. Developing pressure profiles were described by Žitný et at. (2002) using modified Bessel functions. In this model the complexity is further increased by simultaneous consideration of the sample expansion. The model has many parameters characterizing properties of crumb, formation of crust, rheology of expanding liquid core, etc, and only a few of them can be directly measured. This paper is an attempt to simplify the model and to reduce the number of its parameters. This model is more empirical, preserving only the basic features of the preceding one. No layers are considered, the sample is considered as a homogeneous body, having a mass M and temperature T.

The first phase of process is essentially only heating without phase changes, the enthalpy balance can be described by equation (M_0 denotes initial mass and S is the heat transfer surface of the sample),

$$M_0 c \frac{dT}{dt} = k_1 S(T_w - T) . \tag{1}$$

The overall heat transfer coefficient k_1 depends first of all upon the thickness of sample; the higher the thickness (i.e. the higher the mass of the sample M_0), the lower the heat transfer coefficient k_1 and we shall assume that the product $M_0 k_1$ is approximately constant. Therefore we rewrite Eq.(1) in the form

$$M_0 \frac{dT}{dt} = \frac{S_1^*}{M_0}(T_w - T) , \qquad S_1^* = \frac{M_0 k_1 S}{c} . \tag{2}$$

Assuming a constant wall temperature T_w, and constant physical properties (specific heat capacity c), the mean temperature increases exponentially

$$T = (T_0 - T_w)e^{-tS_1^*/M_0^2} + T_w , \tag{3}$$

where S_1^* is a model parameter, presumably independent of the sample mass.

The second phase starts when temperature exceeds the boiling point of water at atmospheric pressure, 100^0C (this is confirmed experimentally because measured pressure starts increasing just at this temperature). We suppose that the core temperature is equal to the boiling point temperature and that the enthalpy balance can be described by the differential equation

$$Mc \frac{dT}{dt} = k_2 S(T_w - T) - r\Psi[(\frac{T}{100})^4 - 1], \tag{4}$$

where the last term on the right hand side represents the enthalpy of steam (r is latent heat) escaping from mould with the mass flow-rate dM/dt. We assume that this flow-rate is proportional to the pressure drop of steam, i.e. to the difference between the pressure of saturated vapour p'' and atmospheric pressure (approximately 1 bar). The pressure p'' can be expressed as a function of temperature e.g. using Antoine's equation or by an empirical correlation

$$p'' = (\frac{T}{100})^4 , \tag{5}$$

where p'' is substituted in bars and temperature in ^{0}C. Therefore, the term p''-1 is the pressure drop of steam and the mass flow-rate can be expressed as

$$\frac{dM}{dt} = -\Psi[(\frac{T}{100})^4 - 1] \tag{6}$$

where Ψ is overall permeability of sample. A rather general expression for permeability Ψ can be derived from the idea of two parallel radial flows of steam (through the crumb and at the heater wall), connected by the cross-flow through a gradually formed crust, see Žitný (2002). However, we shall consider permeability as a constant – another parameter of the integral model.

The enthalpy balance Eq.(4) can be rearranged in a similar way as the balance (1),

$$M\frac{dT}{dt} = \frac{S_2^*}{M_0}(T_w - T) - r^*\Psi[(\frac{T}{100})^4 - 1], \qquad S_2^* = \frac{M_0 k_2 S}{c}, \qquad r^* = \frac{r}{c}. \tag{7}$$

introducing two additional model parameters S_2^* and r^*.

Bound water is only evaporated in the third phase of the process and the enthalpy balance was formed in a similar way as that in the second phase

$$M\frac{dT}{dt} = \frac{S_2^*}{M_0}(T_w - T) - r^*\varphi M_b. \tag{8}$$

Eq.(8) follows from the assumption, that the mass flow-rate of evaporated water is proportional to the mass of the remaining bound water M_b, i.e. $dM/dt = - \varphi M_b$. The proportionality coefficient φ seems to be another independent parameter of model, but it is not so. Its value follows from the commonly used rule that the mass flow-rate of steam is a continuous function, i.e. that the mass flow-rate calculated from Eq.(6) in the end of the second phase equals the mass flow-rate calculated as φM_b in the beginning of the third phase. This requirement of continuity results in the following equation for φ

$$\varphi = \frac{\Psi}{M_b}[(\frac{T}{100})^4 - 1], \tag{9}$$

evaluated during the last time step of the second phase. Pressure drop of steam during the third phase follows from

$$\Delta p = \frac{\varphi M_b}{\Psi}, \tag{10}$$

assuming the same permeability Ψ as in the second phase.

The model describing the change of temperature and mass of the sample has therefore only four parameters: S_1^*, S_2^*, Ψ and r^*. Equation (8) can be solved numerically by time steps (for details see Appendix) and an example of the temperature profiles is given in Fig.2.
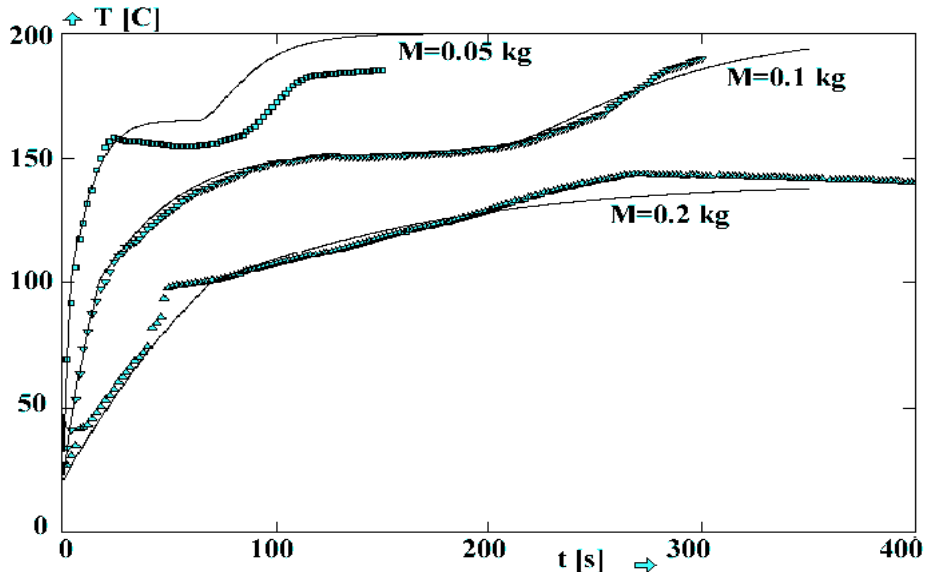

Fig.2 Time trends of temperature predicted by model (3,7,8,9) for parameters: mass fraction of water ω_W=0.6, ratio of bound to free water M_b/M_f=0.44, T_0=20 ^{0}C, T_w=200 ^{0}C, S_1^*=0.00034, S_2^*=0.000135 kg^2/s, Ψ=0.000073 kg.bar^{-1}.s^{-1}, r^*=201 ^{0}C and masses M_0=0.05, 0.1, 0.2 kg. Comparison with experimental data for plates manufactured from potato starch without additives. Thickness of plates 2,4, and 8 mm.

At the beginning of the second phase of the process, the last term on the right hand side of Eq.(7) is small (it equals zero for T=100 ^{0}C), and therefore the solution of Eq.(7) would be a smooth continuation of Eq.(1) if the heat transfer coefficient k_2 were the same as in the first phase. Experiment indicates that this is not quite so, see Fig.2, and the slope of temperature increase is lower in the

second phase. This decrease in heat transfer coefficient cannot be explained by a low thermal diffusivity of the crumb because the crumb does not exist at the beginning of the second phase; a possible explanation could be the thermal resistance of the gap between the sample surface and heater walls. Respecting this, a higher value of S_1^*=0.00034 was used in the first phase, while S_2^*=0.000135 in the second phase. A more or less constant temperature is characteristic for the second phase of the process and its value follows from Eq.(7), where the time derivative of temperature is neglected,

$$T = T_w - \frac{r^* M_0 \Psi}{S_2^*}[(\frac{T}{100})^4 - 1].$$

(11)

Therefore the temperature of this plateau depends only upon a single parameter $r^* M_0 \Psi / S_2^*$ for a given temperature of wall and this relationship, which is a solution of the algebraic equation (11), is shown in Fig.3,
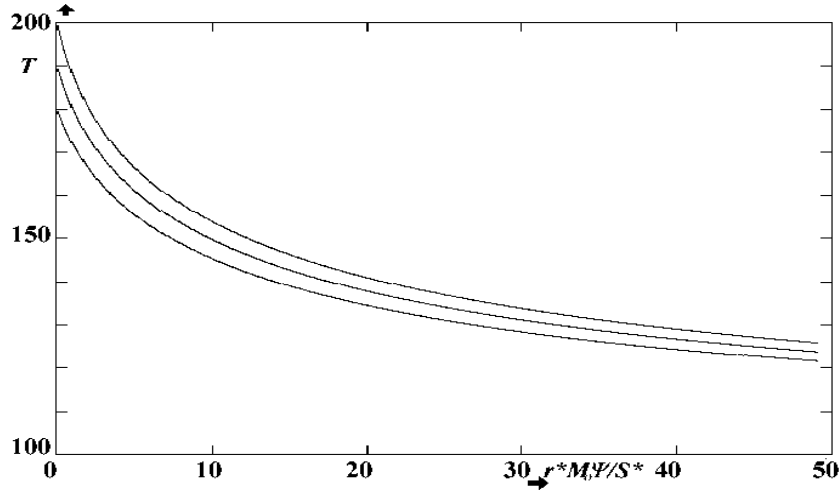

Fig.3 Temperature of plateau during the second phase of processing (free water evaporation, constant permeability) as a function of the $r^* M_0 \Psi / S_2^*$ group for temperatures of heater wall T_w=180,190,200 ^{0}C.

This graph facilitates an initial estimate of the model parameter from experimental data, more specifically from a time temperature trend $T(t)$ measured in the center of a sample of mass M_0. Thus, the recommended procedure is as follows:

- First phase of heating is characterized by a mean slope dT/dt which can be used to estimate S_1^*.
- Slope of dT/dt at the beginning of the second phase is used to estimate S_2^*.
- Knowing the amount of free water in the sample and duration time of the second phase, the permeability Ψ can be estimated according to Eq.(6).
- Temperature of the plateau T gives the value of $r^* M_0 \Psi / S_2^*$ using Fig.3.
- Values of Ψ and S_2^* can be used to estimate r^* using the $r^* M_0 \Psi / S_2^*$.

For example: Let us consider an experimentally determined temperature profile for the sample M_0=0.1 kg in Fig.2. Its slope dT/dt during the initial phase is approximately 5 ^{0}C/s, temperature difference T_w-T ~ 150 ^{0}C, therefore S_1^*~(dT/dt) $M^2/(T_w$-$T)$ = 5·0.1^2/150=0.00034 kg^2/s. The slope dT/dt at the beginning of the second phase, i.e. at 100 ^{0}C, is approximately 1.3 ^{0}C/s, T_w-T ~ 100 ^{0}C, giving S_2^*~1.3 0.1^2/100= 0.00013 kg^2/s. Amount of free water in the sample is 0.04 kg, (this value follows from the known amount of water 0.06 kg and from an estimated ratio of bound water in the sample), duration of the second phase is approximately 130 s, giving Ψ~$\Delta M/[t((T/100)^4$-1)]=0.04/[130·4]=0.000077 kg. bar^{-1}.s^{-1}. Temperature of plateau is 150 ^{0}C and from Fig.3 we read the value of 12 of $r^{\cdot} M_0 \Psi / S_2^{\cdot}$. Therefore r^*~13$S_2^*/(M_0\Psi)$ =12·0.00013/(0.1·0.000077)=202 ^{0}C.

Steam pressure is expressed by Eqs.(5) and (10) in the second and the third phase respectively. Figure 4 presents the predicted time trends of pressure for the same case as previously. The experimental trends were measured by a piezoresistive pressure transducer ISI 0170, flush mounted at the center of the heater wall. Even if there were problems with the measurement of pressure and data

could not be considered as fully reliable, the agreement between prediction and experiment is fairly good (with regard to the fact that no information concerning pressure was used for model identification). Differences are caused probably by the assumption of constant permeability, which neglects crust formation.
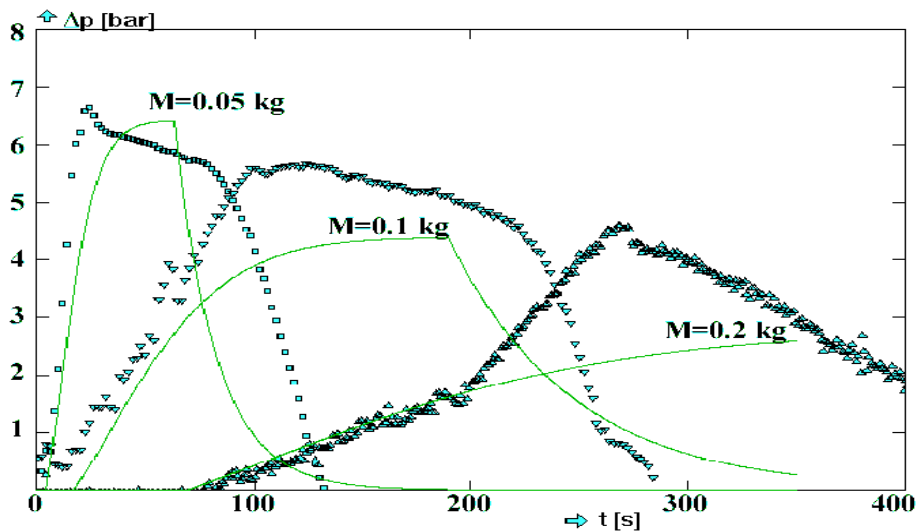

Fig.4 Overpressure measured at center of wall by pressure transducer, Tsiapouris (2002), and pressures calculated from Eqs.(5) and (10) for the same parameters of model as in Fig.2, S_1^*=0.00034, S_2^*=0.000135, Ψ=0.000073, r^*=201, masses M_0=0.05, 0.1, 0.2 kg.

3. Conclusions

An integral model of the thermal pressure forming process has been developed from a much more complicated core, crumb and crust model, which respects volumetric expansion of the processed material, Žitný (1999). Surprisingly this extremely simple model, which does not take into account all the features above and assumes a homogeneous sample, constant thermophysical properties, constant permeability, no expansion of material, etc, is capable of describing the experimentally determined variation of temperature and pressure with time to an acceptable accuracy. A great advantage of the reported model is it's reliability and easy identification of its parameters. Of course, there are restrictions as well: the model cannot describe the phenomenon of the temporary temperature decrease which is probably caused by a break-through a porous structure of crumb and consequently by increase of permeability (the breaking-through of dried faces of cavities by steam pressure has been sometimes manifested acoustically).

While the mean pressures are predicted fairly well, the model fails to describe correct shapes of pressure trends, sudden changes of slopes dp/dt, location of maxima and so on. These features are probably related to the formation of crust accompanied by a gradually decreasing permeability and by constrained expansion of the heated material which are ignored by the presented model.

Acknowledgements

Appendix

Solution of Eq.(7) for temperature during the second phase (free water evaporation) can be obtained numerically in time steps; let us suppose that the temperature T_0 at the beginning of a time step is known. If this applies the nonlinear term T^4 can be linearized,

$$T^4 = T_0^4 (\frac{4T}{T_0} - 3),$$ (12)

and Eq.(7) after substitution Eq.(12) may be linearized as well,

$$M\frac{dT}{dt} = \frac{S_2^*}{M_0}T_w + r^*\Psi[3(\frac{T_0}{100})^4 + 1] - T[\frac{S_2^*}{M_0} + \frac{4}{T_0}r^*\Psi(\frac{T_0}{100})^4].$$ (13)

Solution of Eq.(13) can be expressed in an analytical (exponential) form for temporary constant mass of sample M,

$$T = \frac{\frac{S_2^*}{M_0}T_w + r^*\Psi[3(\frac{T_0}{100})^4 + 1] + \{\frac{S_2^*}{M_0}(T_0 - T_w) + r^*\Psi[(\frac{T_0}{100})^4 - 1]\}e^{-[\frac{S_2^*}{M_0} + \frac{4}{T_0}r^*\Psi(\frac{T_0}{100})^4]\frac{t-t_0}{M}}}{\frac{S_2^*}{M_0} + \frac{4}{T_0}r^*\Psi(\frac{T_0}{100})^4}$$ (14)

Temperature of sample during the last phase of bound water evaporation can be derived in a similar way from Eq.(8),

$$T = (T_0 - T_w + \frac{r^*\varphi M_b M_0}{S_2^*})e^{-\frac{S_2^* t}{M_0 M}} + T_w - \frac{r^*\varphi M_b M_0}{S_2^*}.$$ (15)

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