Influence of temperature dependence of viscosity on thermal process establishment using CMC-based liquid food model

B Chuenchumsap1, S Asavasanti2 and C Tangduangdee1*

1 Department of Food Engineering, Faculty of Engineering, King Mongkut’s University of Technology Thonburi, Bangkok, Thailand
2 Food Technology and Engineering Laboratory, Pilot Plant Development and Training Institute, King Mongkut’s University of Technology Thonburi, Bangkok, Thailand
E-mail: chairath.tan@kmutt.ac.th

Abstract. Since determination of the slowest heating zone (SHZ) is a tedious task in thermal process establishment, mathematical simulation has been introduced. In this research, a numerical study on 2-dimensional natural convection in a cylindrical can during sterilization of liquid food (1% Carboxy-methyl cellulose, CMC) was performed to simulate the flow pattern and temperature distribution. SHZ and sterilizing value ($F_0$) were also determined. Temperature dependent viscosity and density were assumed, while heat capacity and thermal conductivity were kept constant. To represent the actual mechanisms of heating in an overpressure retort, non-isothermal boundary condition (B.C.) was applied; the results were compared with isothermal B.C. The non-isothermal B.C. resulted in a slower heating rate and the simulated temperature profiles that agreed well with the experimental data (2.6% RMSE). Simulated SHZ was forced to move downwards from the geometric centre to the bottom of the can and stayed at about 10% of the can height from the bottom. SHZ moved closer to the wall ($r = R/2$) as a result of the secondary flow due to natural convection. An increase in viscosity and thermal conductivity and a decrease in specific heat and density of higher than 5% resulted in a significantly lower $F_0$-value.

1. Introduction
Canning is one of the widely used food preservation techniques. Thermal treatment aims at destroying both spoilage and pathogenic microorganisms. The primary concern of establishing the optimal heat treatment for package food sterilization is food safety. Therefore, the process lethality ($F$-value) needs to be established.

For a canned thin liquid food, the heat transfer in the food is dominated by natural convection, whereas heat conduction is dominated for a canned thick liquid food. However, both heat transfer mechanisms coexist in some liquid foods [1]. Thus, the rheological property of a liquid food is of important for determination of the $F$-value of sterilizing. In other words, viscosity is one of the critical factors that the manufacturer has to know and strictly control. The viscosity of liquid food often increases or decreases when heated depending on the molecular structure of the fluid. The change in viscosity certainly results in the change of heat transfer mechanism.

To determine the $F$-value of a package food, slowest heating zone (SHZ) inside the can needs to be located. In practice, a series of experiment have to be conducted by inserting a set of thermocouples at various locations to determine the point that gives the slowest heating rate. It was reported that the SHZ in a can of viscous sodium carboxy-methyl cellulose (CMC) suspension is not a stationary region as it...
underwent convection heating [2]. As heating progress, the SHZ moved from the geometric center of the can to the region close to the sidewall, and stayed at about 10 – 12% of the can height from the bottom. The simulated flow pattern of water filled in a can undergoing heating indicated that the cluster of slowest heating points is located away from the sidewall and the center line to form a donut-shaped region near the bottom of the can [3]. The significant differences between the slowest heating temperature and the temperature at other points might be observed in a more viscous liquid and a larger size can. In addition, placement of thermocouple probes at various positions can disturb flow pattern of the moving liquid [4].

To overcome these difficulties, a computational fluid dynamics (CFD) is introduced to describe a fluid flow and heat transfer in geometry. It consists of the solution of partial differential of Navier-Stokes equations, consisting of conservation of mass, momentum and energy equations [5].

Previous studies are limited to using CFD for simulating the flow pattern and the temperature isotherms [2-4]. Although the SHZ can be simulated using CFD, it has not been validated with experimental data. The validation was usually performed to examine only a specific point to check the goodness of fit between the observed data and the predicted temperature. Therefore, the objective of this research was to investigate the effect of temperature dependence of viscosity on thermal process establishment, flow pattern and the SHZ of liquid model food. The SHZ will be validated with experimental data.

2. Materials and methods

2.1. CMC suspension preparation
Carboxy-methyl cellulose (CMC) with the molecular weight of 700,000 g mol⁻¹ (food grade) was purchased from Chemipan Corporation Co., Ltd, Thailand. The aqueous solution of CMC was prepared by dissolving 1% (w/w) CMC powder in distilled water at room temperature and homogenized using a handheld homogenizer (IKA, T-25 ULTRA-TURREX, Germany) at the speed of 17,000 rpm. The sample was left for at least 24 h to release air bubbles before used.

2.2. Rheological measurement
The apparent viscosity was measured by a rheometer (HAAKE MARS III, Germany). One mL of sample was put between parallel plate sensors (P35 Ti L) at 60, 70 and 80 °C. Shear stress was measured and recorded versus shear rates in the range of 10⁻³ to 10² s⁻¹ by RheoWin 4 software. At very low shear rate range, a shear-rate independent viscosity can be assumed; the model foods will behave as Newtonian fluids, although the real model foods are non-Newtonian fluids [3]. Thus, the apparent viscosity was selected at the shear rate of 0.1 s⁻¹ to plot against temperatures. The temperature-viscosity plot was then fitted with the second order polynomial equation:

\[ \mu = a + bT + cT^2 \]  

where \( \mu \) is the apparent viscosity (Pa·s), \( T \) is temperature (K), \( a \), \( b \) and \( c \) are constant values that obtained from the curve fitting of viscosity versus temperature.

2.3. Mathematical model
To simulate flow and temperature of CMC-based model food during heating, simple governing equations of 2-dimentional axisymmetric was developed. The dimensions of the metal can were 8.1 cm in diameter and 11.1 cm in height.

2.3.1. Assumptions
- Fluid movement occurs in 2-dimentional cylindrical space with axisymmetric.
- Heat generation due to viscous dissipation is trivial.
- Bossiness approximation is valid.
- Thermal properties are constant, except viscosity.
• No-slip condition is valid.
• The outer surface temperature is constant in the case of isothermal boundary and are varied with time in the case of non-isothermal boundary.
• Thermal resistance of the can wall is negligible.

2.3.2. Governing equations. The governing partial differential equations for natural convection motion of a liquid food in a cylindrical space are given by Navier-Stokes’ equations in cylindrical coordinates as follows [6].

- Continuity equation:
  \[
  \frac{1}{r} \frac{\partial}{\partial r} \left( r \rho v \right) + \frac{\partial}{\partial z} \left( \rho u \right) = 0
  \]  
  (2)

- Momentum equation in the vertical direction:
  \[
  \rho \left( \frac{\partial u}{\partial t} + v \frac{\partial u}{\partial r} + u \frac{\partial u}{\partial z} \right) = -\frac{\partial p}{\partial z} + \mu \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial u}{\partial r} \right) + \frac{\partial^2 u}{\partial z^2} \right] + \rho g
  \]  
  (3)

- Momentum equation in the radial direction:
  \[
  \rho \left( \frac{\partial v}{\partial t} + v \frac{\partial v}{\partial r} + u \frac{\partial v}{\partial z} \right) = -\frac{\partial p}{\partial r} + \mu \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial v}{\partial r} \right) \right] + \frac{\partial^2 v}{\partial z^2}
  \]  
  (4)

- Energy conservation:
  \[
  \frac{\partial T}{\partial t} + v \frac{\partial T}{\partial r} + u \frac{\partial T}{\partial z} = \frac{k}{\rho C_p} \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) + \frac{\partial^2 T}{\partial z^2} \right]
  \]  
  (5)

To verify the calculation algorithm, a previous result of simulation using water was used; the properties of water and CMC are presented in table 1.

In addition, Boussinesq Approximation (Eq. 6) was applied to the buoyancy term in the momentum equation.

\[
\rho = \rho_{ref} \left[ 1 - \beta (T - T_{ref}) \right]
\]  
(6)

Initial condition, the liquid food is at the stationary with uniform temperature.

- \( T = T_i, u = 0, v = 0 \) at \( 0 < r < R, 0 < z < H \)

Boundary conditions: At the wall of the can, \( r = R \)

| Table 1. The properties of water and CMC used in the simulation [2]. |
|-----------------------------|-------|-------|
| Property                    | Water | CMC   |
| Initial temperature, \( T_i \) (K) | 303.15 | 320.15 |
| Density, \( \rho \) (kg·m\(^{-3}\)) | 998   | 980*  |
| Specific heat, \( C_p \) (J·kg\(^{-1}\)·K\(^{-1}\)) | 4187  | 20    |
| Thermal conductivity, \( k \) (W·m\(^{-1}\)·K\(^{-1}\)) | 0.6   | 0.7   |
| Volume expansion coefficient, \( \beta \) (K\(^{-1}\)) | 0.0002 | 0.0002 |

*measured
2.3.3. Model implementation. The governing equations depending on the initial and boundary conditions were solved using COMSOL Multiphysics® version 5.3.

2.4. Heat treatment
To evaluate the temperature profile at various locations: 10, 15, 20, 25, 30, 35, 40, 45 and 50% of can height from the bottom, a thermocouple was inserted to record the temperature during the process at a given location for each can. Next, 400 g of sample was filled in each can, pre-heated until the temperature reaches 80 °C and hermetically sealed by a can seamer. The cans were placed on the tray and loaded into a water-spray retort (SRA TECH, Model 1150, Thailand). The data acquisition system (CALSoft software, USA) was used to record temperature at every 15-s interval. The retort was automatically operated by programming 10-working steps in PLC controller (UNITRONICS, Model Vision 280, Israel) with 11-min come-up time and 30-min cooking time at 2 bars.

2.5. Data analysis
Validated with the experimental data, the efficiency of the model prediction was evaluated by root mean square error (RMSE) values (Eq. 7).

\[
\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (T - T_{\text{simulated}})^2}
\]  

(7)

3. Results and discussion

3.1. Effect of temperature on apparent viscosity
The apparent viscosities (\(\mu\)) of CMC was measured at 60, 70, 80 °C, and \(10^2\) to \(10^2\) s\(^{-1}\) shear rates. Due to a low shear rate, the viscosity of the fluid may be assumed to be independent of shear rate and will behave as Newtonian fluid [3]. Thus, the \(\mu\) at 0.1 s\(^{-1}\) was chosen to correlate with the temperatures and given by:

\[
\mu = 111.03 - 0.5816T +0.00087T^2
\]  

(8)

3.2. Flow pattern and temperature simulation
To ensure that the mathematical model is able to predict the transient temperature of the liquid in a can with the radius of 0.0365 m and height of 0.105 m, the simulated results were verified with the work of Ghani, et al. [3]. From figure 1, the temperature profile of water obtained from this work was slightly different from the reference result [3]. Nevertheless, the published result has not been validated through experimental data. The discrepancy could be attributed to properties of water that were not included in the literature or the computing algorithm in different software. COMSOL® software used in the present study is based on the finite element method, whereas PHEONICS® software the finite volume. However, the simulation results are reasonable. It implies that this mathematical protocol can be further used to simulate the flow pattern and transient temperature during heating of other liquid foods.
3.2.1. Comparison of different boundary conditions. An isothermal boundary condition (B.C.) at the wall, i.e. constant temperature, was used in the mathematical simulation of many previous studies [3, 5, 7, 8]. This is likely valid only in case of steam retort in which the saturated steam is used. However, in the case of water spray retort, the temperature of the medium gradually rises at the early stage until reaches the target temperature of 121 °C as represented by the broken line in figure 2.

The temperature equations of the retort are:

For $0 \leq t < 300$, $T = 304.22 + 0.2165t - (7 \times 10^{-5})t^2$ \hspace{1cm} (9a)

For $300 \leq t < 650$, $T = 324.3 + 0.146t - (6 \times 10^{-5})t^2$ \hspace{1cm} (9b)

For $t \geq 650$, $T = 394.15$ \hspace{1cm} (9c)

where $t$ is heating time (s) and $T$ is temperature (K).

3.2.2. Influence of boundary conditions on transport phenomena of CMC during heating. For isothermal B.C., transient flow pattern and temperature distribution were simulated with an assumed constant wall temperature (121 °C) as shown in figure 3.

The flow was driven by natural convection. The liquid close to the side wall, the top and bottom of the can receive heat from the medium. As the liquid is heated, it expands and becomes lighter. The buoyancy force created by the change of density due to temperature variation causes the upward flow near the wall. The heated liquid then radially moves towards the core when rising up to the top wall, whereas the heavier liquid at the core moves towards near the centre line, thereby developing the recirculating flow. At 1800 s, the highest velocity of CMC is 0.14 mm s$^{-1}$. Heat conducts to the fluid near the bottom, meanwhile the colder fluid coming in contact with the heated bottom gives rise to
Benard convective cells [3], resulting in the subsequent secondary flow occurrence that pushes the SHZ closer to the wall as shown in figure 3b. It is obvious that the secondary flow affects the location of SHZ by pushing the SHZ deviating from the centre-line of the can.

For non-isothermal B.C., the actual temperature of the heating medium (hot water) was used instead. From figure 4, although it shows similar flow pattern and heat transfer mechanism to figure 3, the difference between isothermal and non-isothermal boundary is the surface temperature progression in the first stage of heating. The actual temperature at the outer surface gradually increased until reaching 121 °C at about 650 s. As the liquid temperature increased with a slow heat rate, a small temperature difference attributed low velocity of the liquid circulation. It appears that the liquid velocity increases from the maximum of 2.5×10^{-2} mm s^{-1} at 120 s to 5.0 mm s^{-1} at 600 s, and then steadily decreases until reaches the same order of the magnitude at the initial heating period as a result of the change in temperature difference.

The secondary flow disappears at 2400 s. The lower part of figure 4 shows the movement of the SHZ, which keeps moving from the centre line to the bottom of the can. After 600 s, the SHZ is influenced by the secondary flow and moves closer to the wall (R/2). After the secondary flow disappears at 2400 s, the SHZ finally moves to the centre axis at 10% of the can height from the bottom. It was found that the isothermal B.C. gives rise to higher heating rate, thus being caution when applying B.C. in the model.

3.3. Model validation
Experimental data showed that, during 8–9 min of the heating period, the temperature fluctuated, likely as a result of the secondary flow, which was similar to the simulation result but with different degree. Temperature fluctuation in the simulation is higher than that of the experiment, resulting in higher \( F_0 \).
The influences of boundary conditions and measuring locations on $F_0$ are presented in table 2. The assumption of the isothermal B.C. resulted in about two folds higher $F_0$-values than that of the non-isothermal B.C. The efficiency of the prediction was represented by Root Mean Square Error (RMSE); it is apparent that the lowest RMSE of 2.6% was attained when the $F_0$ was calculated from the data at the simulated SHZ at $r = R/2$. Thus, for the food safety aspect, the temperature measurement should be conducted at $r = R/2$ rather than at the centreline because there was no temperature fluctuation in this region. The sensitivity analysis revealed that increasing viscosity and thermal conductivity, and decreasing specific heat and density more than 5% resulted in significantly lower $F_0$-value from 5 to 4 min.

Table 2. Sterilizing value ($F_0$) of CMC at 10% from the bottom of the can height.

| Data Source                  | $F_0$-value (min) | $F_0$\@$r=0$ | $F_0$\@$r=R/2$ |
|------------------------------|------------------|--------------|----------------|
| Experiment                   | 6.4              | 6.5          | 1.0            |
| Simulation (Isothermal)      | 14.5             | 11.4         | 1.3            |
| Simulation (Non-isothermal)  | 7.3              | 4.6          | 1.6            |

4. Conclusions
Viscosity is a critical factor that has to be strictly controlled because it affects heat transfer mechanism and the SHZ of liquid food in a container. The mathematical model using CFD commercial software can effectively predict the SHZ and estimate the $F_0$-value when the viscosity changes with temperature so that the number of experiments to determine the cold point of a packaged product can be reduced. When the secondary flow is created, the SHZ will migrate closer to the wall (at $r = R/2$) rather than at the centre line (at $r = 0$). Thus, measuring the temperature at improper position can cause an unsafe food product. The non-isothermal boundary condition should be applied when hot water was used as the heating medium instead of saturated steam.

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