# Modelling Heat Transfer and Heterocyclic Amines Formation in Meat Patties during Frying

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## ABSTRACT

The formation of heterocyclic amines (HAs) namely IQx (2-amino-3-methylimidazo[4,5-*f*]quinoxaline), MeIQx (2-amino-3,8-dimethylimidazo[4,5-*f*]quinoxaline) in meat patties during one sided pan-frying was modeled using combined classical heat transfer and kinetic equations. The heat conduction equations and associated boundary conditions in meat patties during the one sided pan frying were solved using the Galerkin finite element method. Temperature profiles at the surface and center areas in a ground beef patty during the cooking process were predicted and used to estimate HAs formation during frying. The model predictions were validated with experimental results.

**Keywords:** Frying, heat transfer, modeling, kinetics, heterocyclic amines

## 1. INTRODUCTION

Heterocyclic amines (HAs) have been shown to induce various kinds of cancers in mice, and monkeys. There is a strong possibility that the compounds can also cause cancers in humans (Ohgaki *et al.*, 1991; Sugimura, 1997). HAs are easily formed and have been found in cooked meat products processed using common household cooking methods such as pan-frying and barbecuing. Furthermore, the compounds have also been found in meats cooked in restaurants and fast food outlets (Balogh *et al.*, 2000; Knize *et al.*, 1998; Chiu *et al.*, 1997).

Time and temperature have the major influence in the formation of HAs in a product. HAs are generally formed at temperatures above 150$^{o}$C (Tran *et al.*, 2002; Hwang and Ngadi, 2002a and 2002b; Arvidsson *et al.*, 1997 and 1999; Jackson and Hargraves, 1995) and their concentrations increase with increasing cooking time and temperature. However, the compounds may degrade after attaining maximum concentration during prolonged cooking time. Other factors that influence HAs formation include fat content, sugars, various amino acids and antioxidants, water, and cooking methods (Tsen *et al.*, 2006; Ahn and Grun, 2005; Hwang and Ngadi 2003; Basira 1998; Jagerstad *et al.*, 1991; Taylor *et al.*, 1983).

Formation of HAs in model systems or meat juices with controlled mixture of possible precursors (amino acids, creatine, and with or without sugars) and at controlled temperatures has been studied by different authors. These studies have been useful in advancing understanding of HAs formation in foods. However, cooking of real meat systems may present different challenges due to their peculiar heat and mass transfer characteristics. Non-uniform temperature profiles develop in meat patties during pan-frying. A thin layer of crust forms at the surface and advances into the product with increasing frying times (Singh *et al.*, 1997). Inside the crust, a water evaporation zone moves inwards, while water and juices, which are released through protein denaturation and shrinkage, move outwards (Hallstrom and Skjolderand, 1983). Higher HAs concentrations have been found in the crust part of cooked meat and in pan residues during pan-frying (Jagerstad *et al.*, 1991; Holtz *et al.*, 1985; Johansson and Jagerstad, 1994 and 1995; Pais *et al.*, 1999). There is scarce information on the influence of these factors on the formation of HAs during pan-frying of meat patties.

In contact cooking such as pan-frying, heat transfer occurs largely by conduction and it may also occur by convection depending on process conditions (Singh *et al.*, 1997). A number of models have been developed to predict temperature profiles in beef patties cooked with pan-frying (Dagerskog, 1979a; Ikediala *et al.*, 1996; Singh *et al.*, 1997; Pan *et al.*, 2000). Ikediala *et al.* (1996) developed a two-dimensional model to predict heat transfer in beef patties using the finite element method. A model was proposed for heat and mass transfer in beef patties during double-sided contact frying by Dagerskog (1979a). Heating temperature, heat transfer coefficient, and patty thickness were determined to be the major factors that influence the patty center temperature profiles (Pan *et al.*, 2000).

The objectives of this study were to model temperature development in the crust and center of ground beef patties during pan frying; predict formation of HAs using the kinetic modeling approach; combine the heat transfer with reaction kinetic models to predict formation of HAs at the surface of fried meat patties and to validate the model predictions.

## 2. MATERIALS AND METHODS

### 2.1 Chemicals

All chemicals and solvents used in the study were for the solid-phase extraction and HPLC analysis. The solvents, acetonitrile, methanol, and dichrometane were purchased from Fisher Scientific Inc. (Nepean, ON). Materials for the solid-phase extraction namely Chem-Elut diatomaceouse earth, Bon-Elut propylsulfonic silica (PRS)(500 mg) cartridges, C-18 (100 mg) cartridges, cartridge coupling adaptors and vacuum manifold were purchased from Varian Inc (Harbor city, CA). Standards for the HAs namely IQ (2-amino-3-methylimidazo-[4,5-*f*]quinoline), MeIQ (2-amino-3,4-dimethylimidazo[4,5-*f*]quinoline), IQx (2-amino-3-methylimidazo[4,5-*f*]quinoxaline), MeIQx (2-amino-3,8-dimethylimidazo[4,5-*f*]quinoxaline), 4,8-DiMeIQx (2-amino-3,4,8-trimethylimidazo[4,5-*f*]quinoxaline), 7,8-DiMeIQx (2-amino-3,7,8-timethylimidazo[4,5-*f*]quinoxaline), and PhIP (2-amino-1-methy-6-phenylimidazo[4,5-*f*]pyridine) were from Toronto Research Chemicals (Toronto, ON). Deionized water was used throughout the study.

### 2.2 Sample Preparation

Extra lean ground beef with 6% of fat composition was purchased from a nearby grocery store. The Soxhlet solvent extraction method (Ngadi *et al.*, 2000) was used to verify the fat content of the samples. Cylindrical patties were made using 130 g of ground beef. The dimensions of the patties were 90 mm in diameter and 20 mm in thickness.

A commercial-scale, one-sided, pan-fryer (Moffat, Specialites Cuisine Inc, Montreal, QC) was used in the study. Before using the pan fryer, surface temperature distribution on the pan-fryer was determined at two set temperatures, 180 and 200$^{o}$C using the self-adhesive thermocouples (SA1-T) from Omega Inc. (Montreal, QC). The surface temperature was 199.67± 3.19 and 177.7±3.88 when fryer was set at 200 and 180$^{o}$C, respectively. Cooking times of 2.5, 5, 7.5 and 10 min per side were used at the 2 set temperatures. Triplicate experiments were conducted at each time and temperature combination. Cooked samples were individually packed in waterproof sealed plastic bags and frozen until analyzed for HAs concentration.

To measure temperatures during frying, holes were made horizontally from the side to the geometric center and to locations close to the top and bottom surface of meat patties using a 1 mm glass stick. T-type thermocouples were inserted into these holes to measure temperatures at the center and at the locations close to the surface. Once the thermocouples were inserted, the meat sample settled and established contact with the thermocouple wire. Several center temperatures and several surface temperatures were obtained during 20 min cooking time at the different set temperatures. The experimental temperatures were used to verify the predicted temperature profiles obtained by the mathematical model.

Since HAs was expected to be formed mainly at the product surface, the samples used for HAs analysis were very thin (about 1 mm) slices cut from the top and bottom sides of meat patties cooked at the different frying times and temperatures. Hwang and Ngadi (2002a) have described in detail, the protocols for extraction and purification of heterocyclic amines in cooked meat samples. A reversed-phase HPLC (Pro Star, Varian Inc., Habor City, CA), which was equipped with a photodiode array and a programmable fluorescence detector, was used to separate and quantify the HAs.

## 3. MODEL DEVELOPMENT

### 3.1 Heat Transfer

The mathematical model and finite element computer program developed by Ikediala *et al.* (1996) was modified and used to predict temperature profiles in the beef patties during pan frying. The governing Equation 1 was derived for the two dimensional axisymmetrical problem. Assumptions made in developing the model were as follows: heat was transferred inside the patty by conduction while heat transfer occurred between the surface of the pan and the beef patty by convection through a thin film of air, oil, and moisture; there was no heat generation in the beef patty.

$$\frac{\partial\left(\rho c_p T\right)}{\partial T} = \frac{1}{r}\frac{\partial}{\partial r}\left(kr\frac{\partial T}{\partial r}\right) + \frac{\partial}{\partial z}\left(k\frac{\partial T}{\partial z}\right) + L\rho\frac{d\overline{m}}{dt} \tag{1}$$

where T is temperature (°C), t is time (s), $\overline{m}$ is average moisture content (g/g wb), and r and z are directional coordinates. The product properties namely density (kg/m$^3$), heat capacity (J/kg°C) and thermal conductivity (W/m°C) are represented by ρ, $c_p$ and k, respectively. The initial and boundary conditions for the beef patty sample are given as follows (schematic of patty shown in Figure 1):

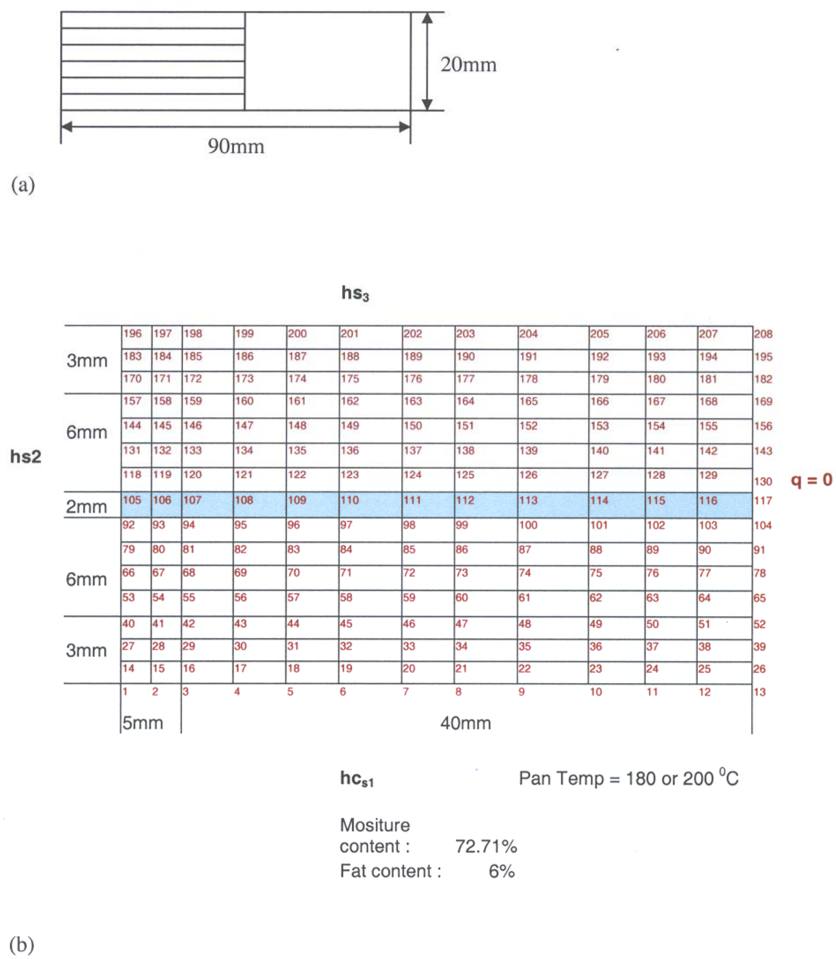



Figure 1. (a) Diameter and thickness of a ground beef patty (b) a symmetric half and discretization into 208 nodes and 180 elements

$$T\left(r,z,t=0\right) = \overline{T}_0 \tag{2}$$

$$M\left(r,z,t=0\right) = \overline{M}_0 \tag{3}$$

$$- k\frac{\partial T}{\partial z} = h_{s1}(T_p - T_{s1}) \qquad \text{at } z = 0 \qquad (4)$$

$$- k\frac{\partial T}{\partial r} = h_{s2}(T_{s2} - T_\infty) \qquad \text{at } r = R \qquad (5)$$

$$- k\frac{\partial T}{\partial z} = h_{s3}(T_{s3} - T_\infty) \qquad \text{at } z = Z \qquad (6)$$

Where $h_{s1}$, $h_{s2}$ and $h_{s3}$ are heat transfer coefficients at sides 1, 2 and 3, respectively.

$$\frac{\partial T}{\partial r} = 0 \qquad \text{at } r = 0 \qquad (7)$$

The weighted residual method based on the Galekin approximation was used to solve the equations. The finite element transformation of the governing heat transfer equation with initial and boundary condition was resolved as following:

$$[M]\dot{T} + [K]T = \{F\} \qquad (8)$$

Solutions of the governing equations with initial and boundary conditions were implemented by using a finite element computer program written in the FORTRAN language. Four node quadrilaterals of 208 nodes and 180 elements were used for the meat patty discretization as shown in Figure 1.

### 3.2 HAs Formation

The formation of HAs was predicted based on the kinetic modeling approach developed by Hwang and Ngadi (2002a). The formation of HAs was determined to be a first order reaction as shown in Equation 9.

$$\frac{dc}{d(t - t_o)} = -k_r(c - c_{\max}) \qquad (9)$$

where

$$k_r = k_0 e^{\left(-E/RT\right)} \qquad (10)$$

where $k_r$ is the kinetic reaction rate for HAs formation ($\text{min}^{-1}$) as the function of absolute temperature T, $t_o$ is the lag time of formation (min.) below which no HAs forms, $k_o$ is a constant, E is activation energy, R is gas constant, c is the concentration of HAs (ng/g), $c_{max}$ is the maximum concentration (ng/g). The values of the kinetic parameters were obtained from Hwang and Ngadi (2002a). The mass averaged predicted temperature profiles from the nodes 1 to 13, corresponding to the bottom side of the sample and the nodes 196 to 208, corresponding to the top side of the sample was used to predict formation of HAs. Since temperature of the product was changing during the frying process, a step wise cumulative transformation of the kinetic equation was applied to predict HAs at each time step. The prediction of the HAs formation was compared with the experimental data.

## 4. RESULTS AND DISCUSSION

### 4.1 Formation of HAs

Heterocyclic amines namely IQx, MeIQx, and PhIP were identified at the surface of the cooked samples for the various cooking times at the temperatures of 180 and 200$^{o}$C. The concentrations obtained for the compounds are shown in Table 1. IQx was only detected after 15 min cooking time at 200$^{o}$C. PhIP was detected at all the cooking times at both temperatures. MeIQx was identified only after 15 and 10 min cooking time at the 180 and 200$^{o}$C, respectively. The concentrations of these compounds in the cooked beef patties agree with those reported by Balogh *et al.* (2000) and Knize *et al.* (1998). PhIP was formed in the highest amount followed by MeIQx and IQx.

Table 1. Heterocyclic amines concentration in fried ground beef patties for various cooking times and two different temperatures with one turn-over

| Time per side (min) | Temperature (°C) | IQx (ng/g) | MeIQx (ng/g) | PhIP (ng/g) |
|---|---|---|---|---|
| 5 | 200 | ND* | ND | 0.67±0.11 |
| | 180 | ND | ND | 0.32±0.01 |
| 10 | 200 | ND | 1.65±0.08 | 2.25±0.77 |
| | 180 | ND | ND | 1.09±0.67 |
| 15 | 200 | 0.38±0.10 | 2.70±0.61 | 8.74±1.92 |
| | 180 | ND | 2.17±1.47 | 3.12±2.15 |
| 20 | 200 | 0.33±0.20 | 5.58±0.79 | 24.29±3.38 |
| | 180 | ND | 3.66±0.64 | 5.39±0.14 |

* ND means not detected

As expected, there was a pronounced increase in the HAs formation as cooking time and temperature increased. PhIP developed early during the cooking process; starting after 5 min of cooking (2.5 min of cooking each side) at 180$^{o}$C while there was yet no formation of IQx and

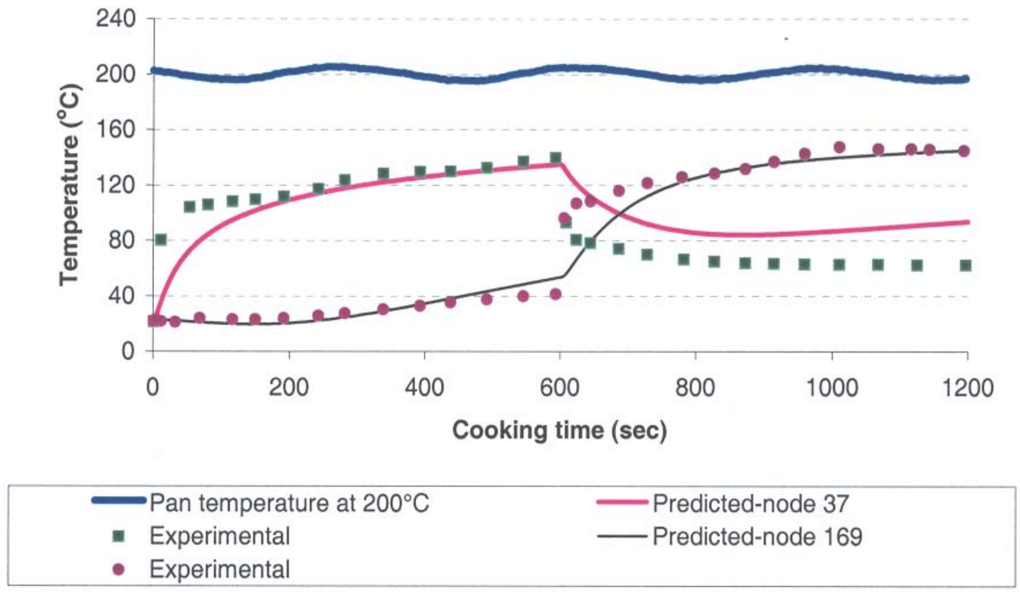
MeIQx. The concentration of PhIP was up to 4 times higher at 200°C than at 180°C. IQx was only detected at the higher 200°C and at the longer frying times. Other authors have also reported increase in HAs concentrations in ground beef patties with increasing cooking temperatures and times (Balogh *et al.*, 2000; Skog *et al.*, 1997, and Knize *et al.*, 1998). The amounts of PhIP obtained in this study were much higher (six times higher) than the values reported by Hwang and Ngadi (2002a) for meat emulsion sample heated isothermally in a closed system. The surface crust part of meat patty was dry resulting from moisture evaporation during pan frying. In comparison, there was no moisture evaporation in a closed system resulting in high moisture retention during heating. Therefore, results indicate that formation of PhIP maybe more favorable in drier conditions. Borgen *et al.* (2001) reported similar results that PhIP was detected at markedly higher level in dry heated sample. The amount of PhIP in dry heated meat juice from chicken breast was about 10 fold higher than in other samples (Skog *et al.*, 2000). The kinetics of formation of PhIP in an open system such as pan frying may not be the same as in closed systems such as reported by Hwang and Ngadi (2002a). The amounts of MeIQx and IQx obtained in this study were slightly lower than the data reported for isothermally heated closed system (Hwang and Ngadi, 2002a) presumably due to concentration of precursors. Therefore the kinetic modeling parameters were applied only for MeIQx and IQx and not for PhIP.

### 4.2 Model Simulation and Validation

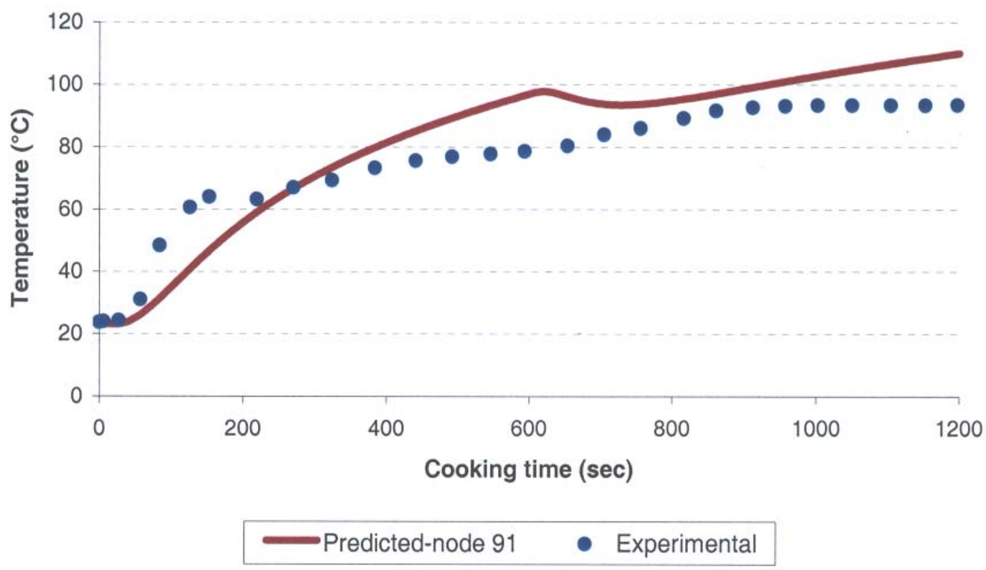
Values of thermal properties namely density, thermal conductivity and heat capacity of ground beef patties as reported by Dagerskog (1979a), was used for the study. The heat transfer coefficient values used in this study were optimized for the surface temperatures in order to minimize underestimation of temperatures at the nodes closest to the bottom and top sides of the meat patty and also avoid rapid cooling effect on the top side after a turn-over. The contact heat transfer coefficient on the side of the patty that was directly in contact with heating pan, $h_{s1,}$ was obtained as 300 W/(m$^{2o}$C) whereas the coefficients for the sides and top of the patty $h_{s2}$ and $h_{s3}$ were 10 and 60 W/(m$^2$C), respectively. These values were higher than those used by Ikediala *et al.* (1996), Dagerskog *et al.* (1979b) and Housova and Topinka (1983). The predicted temperature profiles in the crust and the center of the patty after 20 min of cooking time with one turn-over (performed at half time through the cooking process) at 180 and 200$^o$C of cooking temperatures were validated with the experimental data. For the surface and center temperatures, nodal points from the model and thermocouple positions in the observed profiles were matched since it was difficult to place the thermocouples at exactly the same positions during replication and also there was possibility for slight displacement of the thermocouples in the beef patty during the cooking process.

Comparison of the predicted beef patty temperatures and the experimental temperature profiles at the set pan temperatures, 180 and 200$^o$C as shown in Figure 2 and 3, respectively. Before turn-over, there was a marked increase in the temperature of the patty at the bottom side as expected whereas the center temperature gradually increased; the temperature of the top side showed a small decrease, before it increased slowly during the early stages of cooking. The experimental data shows a sudden and sharp increase in the temperature of the bottom side close to the surface of the pan immediately after the start of heating. However, the model prediction for the temperature increase was more gradual compared to the experimental data. This could be attributed to various factors including actual position of thermocouples, and the complicated relationship between heat transfer coefficient and heating time. The mathematical model used in this study assumed constant heat transfer coefficients.

After the turn-over, the previous bottom temperature decreased while the previous top temperature increased as expected. As seen in the early stage of the cooking, a sudden and sharp increase in the temperature of the new bottom side was also observed at the 180 and 200$^{o}$C of the cooking temperatures from the experimental data. This result was different from the report by Ikediala *et al.*, 1996. However, Dagerskog (1997b) showed a steeper change in temperature in nodes close to the surface of the pan. Possible explanation for this observation could be that the implanted thermocouples could have moved very slightly toward the surface of the pan. However, it was checked and confirmed that it was not touching the pan. The center temperature of the patty was not affected by the turn-over. The center temperature increased continuously and it tended to level off. Experimental temperature profiles for the new bottom side decreased more rapidly than the predicted temperature. This may be attributed a strong cooling effect from a fan over the fan fryer.

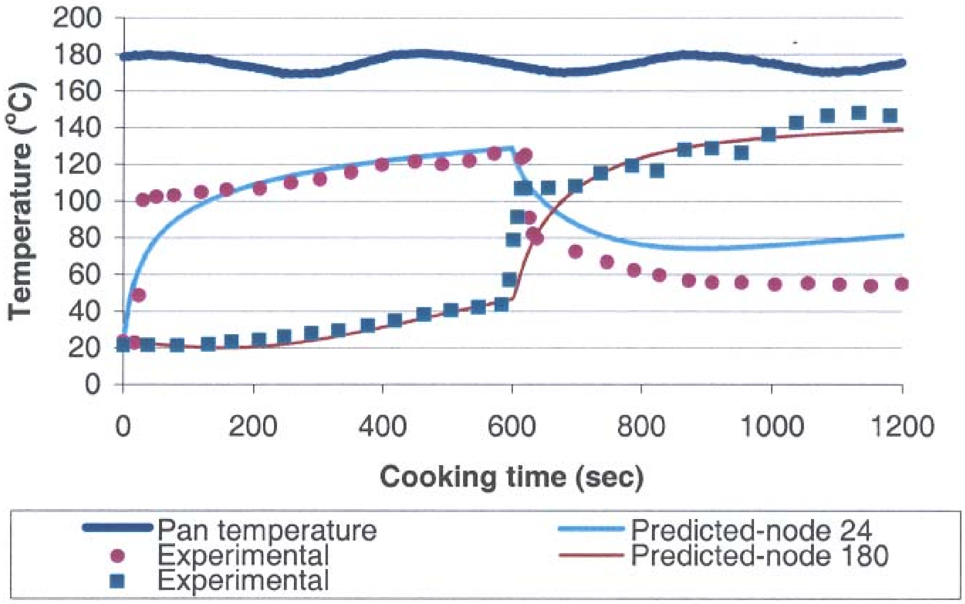



(a)

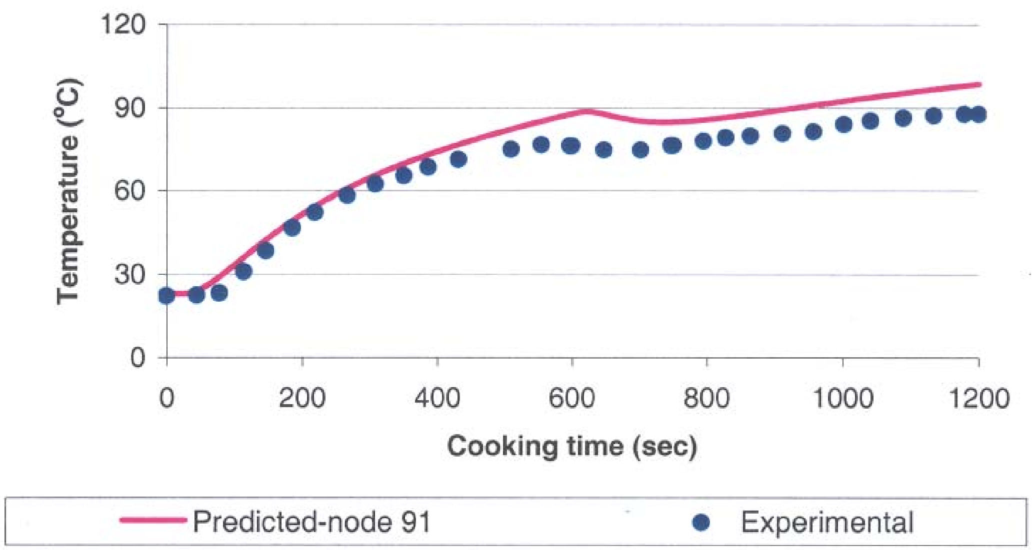



(b)

Figure 2. Predicted and experimental temperatures in ground beef patty cooked at the set pan temperature of 200$^{o}$C. (a) surface area for the top and bottom (b) near center temperature




(a)




(b)

Figure 3 Predicted and experimental temperatures in ground beef patty cooked at the set pan temperature of 180°C. (a) surface area for the top and bottom (b) near the center temperature

The mean (and standard deviation) ranges of the absolute error between the predicted and observed temperatures at nodes from near the center, the top and the bottom at the 180°C frying temperature were 7.00 (± 3.12), 4.96 (± 6.82), and 12.88 (± 8.59), respectively. Similarly at the 200°C, the mean (and standard deviation) ranges of the absolute error between the predicted and observed temperatures at nodes from near the center, the top and the bottom at the 200°C frying temperature were 9.77 ± 5.36, 5.72 ± 7.78, and 16.89 ± 11.83, respectively. The highest absolute error was obtained for the bottom side due to possible under estimation of surface parameters. However, the maximum temperature for both bottom and topside nodes showed that the model was overall adequate to predict temperature profiles in meat patty.

### 4.3 Prediction of HAs

The formation of HAs was expected to be mostly limited to the surface area (crust) and in the pan-residues during the cooking process (Jagerstad *et al.*, 1991; Holtz *et al.*, 1985; Johasson and Jagerstad, 1994 and 1995; Pais *et al.*, 1999). Furthermore, HAs are usually formed at higher cooking temperatures, 150 to 200$^o$C although trace amounts may be detected at temperatures between 100 and 150$^o$C after prolonged cooking time (Gross and Gruter, 1992; Arivdsson *et al.*, 1997 and 1999; Jackson and Hargrave, 1995; Hwang and Ngadi, 2002a). The surface of a hamburger patty in contact with the heating surface usually reaches a temperature higher than 100$^o$C during cooking (Singh *et al.*, 1997). Therefore, in this study only the surface of the cooked ground beef patty (the top and bottom side) was considered for HAs analysis. Model prediction was also concentrated at this region. It was assumed that there was no HAs formation in the inner part of the cooked meat. The formation of HAs namely IQx and MeIQx were predicted with the mean value of the predicted temperatures obtained from the nodes 1 to 13 and the nodes 196 to 208.

Prediction of MeIQx formation at the surface of meat patty during pan frying at 200°C is shown in Figure 4. Before the turn-over, the formation of MeIQx on the top side of the meat patty gradually increased as the temperature increased, while no formation of IQx and MeIQx were predicted at the top side as the temperature remained at low levels as expected. After, the turn-over, higher concentrations of MeIQx were predicted on the new bottom side of the beef patty due to the fact that the new bottom side increased to a higher temperature.

Predicted and experimental concentrations of IQx and MeIQx for 5, 10, 15, and 20 min at 180 and 200$^o$C are compared as shown in Figure 5. Lower concentration of MeIQx was predicted than the actual concentrations formed in the cooked ground beef patty and a similar concentration of IQx was predicted at cooking temperature 200$^o$C. The difference between the predicted and experimental values could be due to the under prediction of the temperature profiles in the nodes close to the surface of the pan. In addition, higher temperatures at the surface of the beef patty were also observed compared to the predicted temperatures. The results show HAs in meat patties during cooking may be closely predicted using mathematical modeling approach. The model prediction can be improved by implementing moving boundary models or by using verified varying heat transfer coefficients that will improve closer predictions of the surface temperatures.

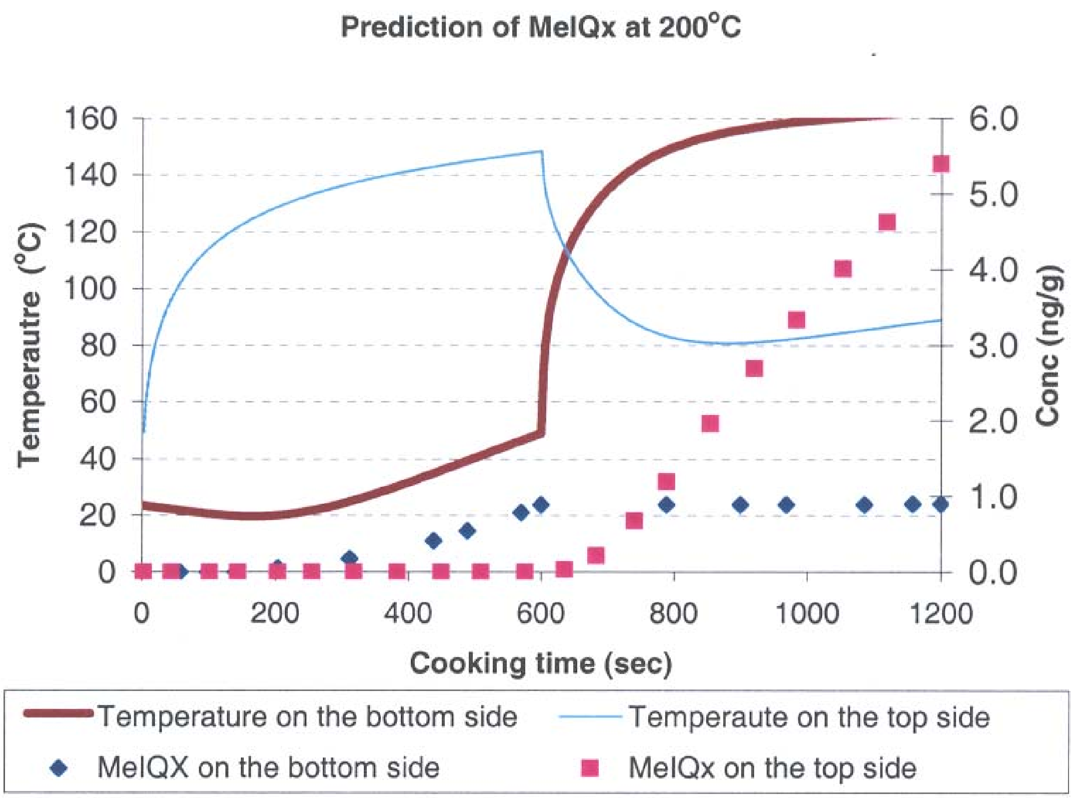
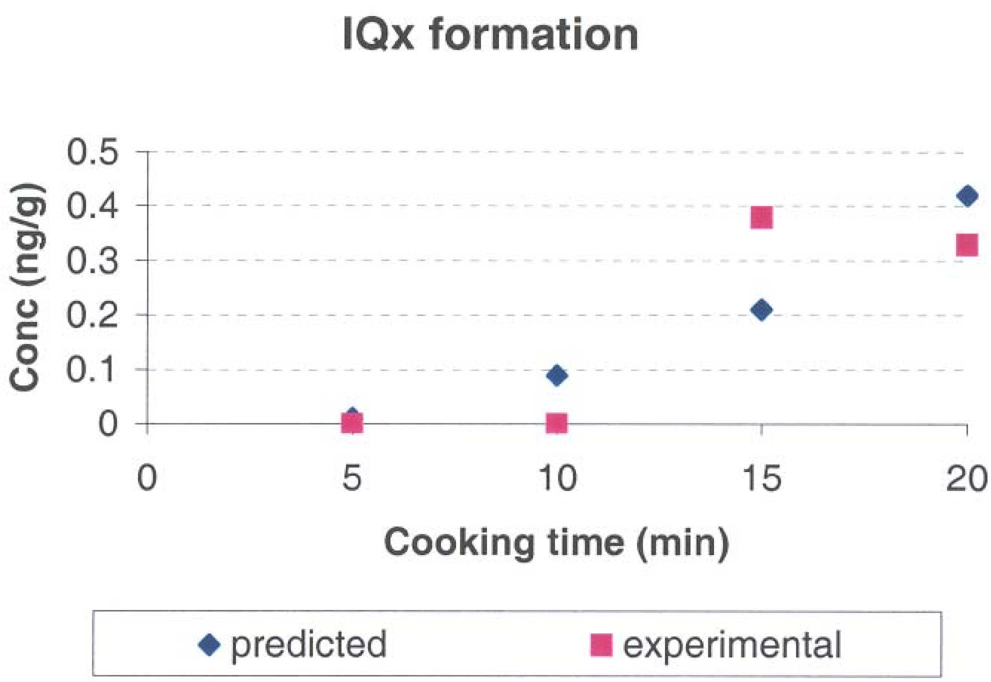



Figure 4 Prediction of MeIQx at the surface area of beef patty fried at 200°C with one turn-over.



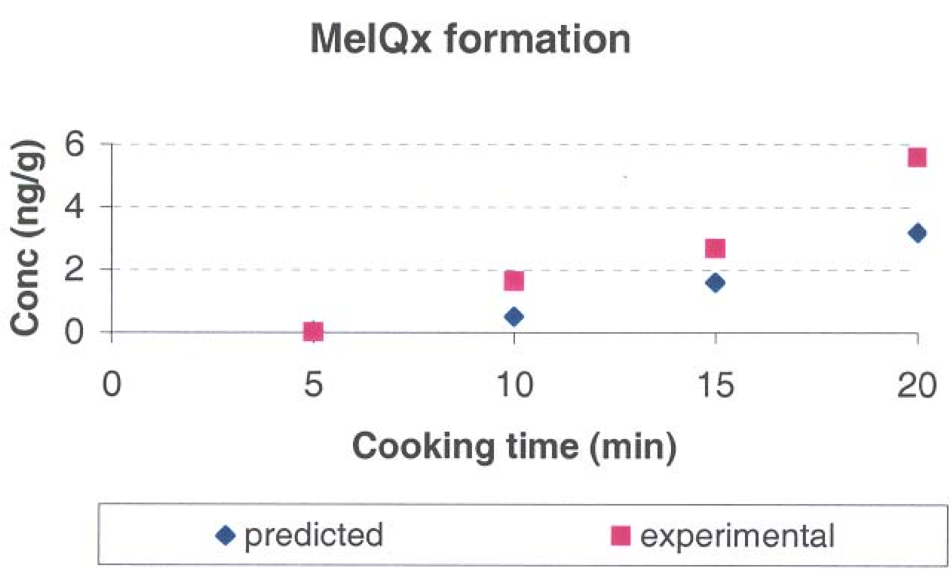




Figure 5 Comparison of the predicted and experimental values of IQx and MeIQx formed at

during pan frying at 200°C. There was one turn-over at the middle of the cooking time.

## 5. CONCLUSIONS

The formation of the HAs was predicted with actual temperature development during cooking process using a normal cooking method, a one-side pan-frying. As expected, the formation of HAs is strongly dependant on the temperature and the cooking time. The different concentrations of the HAs were predicted and could be formed on each side of the beef patty due to the different temperature development. The model prediction of HAs was close to experimental values. Prediction could be improved by improving predictions of surface temperatures. The developed mathematical modeling approach could be used to estimate HAs in cooked patty without necessarily resorting to laboratory analysis.

## 6. ACKNOWLEDGEMENT

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