Sterilization of Foodstuffs Using Radio Frequency Heating

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ABSTRACT: A study was conducted with a pilot-scale sterilization system based on 27-MHz radio frequency (RF) energy to investigate the effectiveness in shortening process time and in improving quality for foods sealed in 6-pound military-ration polymeric trays. Chemical marker *M-1* was used to evaluate heating uniformity in 20% whey protein gels as a model food, and macaroni and cheese was processed to assess the influence of RF process on product quality. With the RF system, a lethality (F_0 = 10 min) was achieved in both model food and macaroni and cheese within 30 min with relative uniform heating, compared to a 90 min conventional retort process that delivered a similar lethality.

Keywords: dielectric heating, radio frequency, sterilization, chemical marker, military ration, poly tray food

Introduction

THE GOALS OF PASTEURIZATION AND STERILIZATION ARE TO REDUCE spoilage and eliminate pathogenic microorganisms in foods. However, concomitant with that is the simultaneous degradation of food quality (Lund 1988). Conventional retorting methods are time-consuming and often detrimental to the quality of heat-sensitive products. A comparison of *z* values (the increase in temperature required to reduce the thermal death time 10-fold) indicates that the rate of destruction of nutrients is less temperature-dependent than the rate of destruction of microbial spores (Lund 1977, 1986). Therefore, when compared with an equivalent process at lower temperatures, a process using short time at high temperatures can produce a better quality food product while ensuring food safety (Banwart 1989). High-temperature short-time processes for solid foods in commercial containers are, however, almost impossible with conventional heating methods using steam or heated water. In these processes, slow heat conduction from the heating medium to the cold-spot often results in a much more severe treatment of the material at the periphery of the container than that required to achieve commercial sterility (Meredith 1998). Consequently, the quality of the solid foods at the overheated corners and edges will be severally degraded (Priestley 1979).

Dielectric heating, that is, microwave and radio frequency (RF) heating, offers the possibility of fast heating in solid and semi-solid foods. Over the past 60 years, numerous studies have been done on microwave heating. An advantage of microwave heating over the conventional thermal processing is the rapid heating by direct interaction between microwaves and foods that are hermetically sealed in microwavable packages (Burfoot and others 1988; Harlfinger 1992). Thus, microwave pasteurization or sterilization can potentially improve the product quality (Ohlsson 1989; Giese 1992). But microwave heating is limited to small-sized food packages due to the relatively small penetration depth of microwaves in dielectric materials. This limitation can be overcome by using RF energy. The principles of RF heating are very similar to microwave heating. Heat is generated within dielectric materials, such as foods, when the electromagnetic field reverses the polarization of individual molecules or causes migration of ions within the material as it alternates at high frequency (Barker 1983). The main difference between RF and microwave is wavelength. The wavelength at the RF heating frequencies designated by the Federal Communication Commission (FCC) for industrial heating is 22 to 360 times as great as that of the 2 commonly used microwave frequencies, which allows RF energy to penetrate dielectric materials more deeply than microwaves. Radio frequency heating may be particularly useful when applied to institutional-sized packaged food products such as the 6-pound-army ration (contained in 24.5 × 23.5 × 4.5 cm trays)because of its deep penetration.

Cathcart and Park (1946) first studied the use of RF heating to thaw frozen eggs, fruits, vegetables, and fish. Radio frequency dielectric heating is now widely used in industrial applications such as drying textile products (spools, rovings, skeins), final drying of paper, final dehydration of biscuits at outlets of baking ovens, and melting honey (Barker 1983; Orfeuil 1987). However, possible non-uniform temperatures within products and difficulty in the application of overpressure during RF heating have previously stood as major barriers to developing applications of RF in sterilization and cooking foodstuffs.

The ultimate objective of this work was to develop high-temperature short-time sterilization (HTST) protocols for foodstuffs using RF dielectric heating. The specific object of the reported project was to develop and test a pilot-scale RF system to process large trays of food.

Materials and Methods

RF sterilization system

A pilot-scale 6-kW, 27.12-MHz RF system (COMBI 6-S; Strayfield Fastran, United Kingdom) with plate applicators was used as the source of RF energy in our study. The applicators and the tuning system were modified to improve the electromagnetic field uniformity between the 2 plate applicators. A pressurized vessel was developed at Washington State Univ. (Pullman, Wash., U.S.A.) to provide an overpressure of up to 0.276 MPa gauge (40 psig) that allows foods in large polymeric trays to be heated up to 135 °C without bursting (Figure 1). The vessel was constructed with 4 Ultem polyetherimide (PEI) plastic walls and 2 parallel aluminum plates as upper and lower lids. Several probe ports permitted (Figure 2) the insertion of fiber-optic sensors directly into food packages through sealed thermal wells. In addition, a custom-built water-condition-

ing system circulated temperature-controlled water of a particular conductivity through the vessel. This system consisted of 2 exchangers: one used steam for heating, and the other used tap water for cooling. A surge tank was used to help maintain an overpressure with compressed air. The circulating water temperature could be varied from 20 °C to 130 °C, an overpressure from 0.136 to 0.204 MPa gauge (20 to 30 psig).

To reduce fringe effect at the interface between the side of the food package and the air in the RF applicators, low-conductivity water was used to immerse the food tray to approximately match the dielectric properties of the food. Together, the water and package present a very flat surface to the imposed field, and push the boundary with air away from the food package. This is expected to reduce the effect of variations in package thickness that could lead to nonuniform heating in the food. Temperature-controlled water from water-conditioning system is required to match the temperature of the heated food to prevent cooling of the package surface. Use of immersion water may compromise energy efficiency in RF pasteurization and sterilization. However, the lost RF energy in the immersion water can be recovered by using it to preheat food trays in industrial applications. It is also possible to minimize the volume of immersion water by lining food packages in continuous systems.

Temperature acquisitions system and data analyses

Fiber-optic temperature sensors (UMI; FISO Technologies Inc, Quebec, Canada) were used to measure real-time temperatures at several different locations in the tested foodstuff, and a computer was used to record data that were acquired by the sensors. To permit the installation of the fiber probes that record time–temperature data during RF heating, 3 thin-walled polyimide tubes were preinserted into each tray with silicone fittings to provide a tight seal. The tubes were installed in the middle layer (25 mm from the bottom of the tray), and a plan view of locations of the tubes is shown in Figure 2. The selection of the locations was based on a previous chemical marker study, which located the least- and most-heated spots in the package. Since the tube (1.9-mm O.D. and 1.8-mm inner dia) and the fiber-optic sensor (<1.8-mm O.D.) are small compared with the volume of food and wavelength of RF in food (about 1200 mm) and made of electric insulating material, they do not interfere with the temperature profile of the surrounding food.

Retort heating was conducted to allow a comparison between conventional methods and the new RF method. Temperatures were measured using thermocouples placed in locations corresponding to the locations of fiber optic sensors in the food trays when heated in an RF system.

The acquired data were used to calculate cumulative thermal effect, F_0, on target bacterium in thermal processing (Stumbo 1973):

$$F_0 = \int_0^t 10^{\left[\frac{T-121.1}{z}\right]} dt \quad (1)$$

where F_0 is the process sterilizing value (min), T is temperature (°C), t is processing time (min), and z is a temperature-dependent factor in thermal inactivation kinetics, defined as temperature difference required for a 10-fold change in the thermal death time (°C) (Teixeira 1992). Eq. 1 was used to determine the cumulative thermal effect during the thermal processing. Commonly, a z value of 10 °C is used for *Clostridium botulinum* spores. F_0 is used to establish the sterilization value to ensure safety for low-acid (pH ≤ 4.6) canned food. F_0 for commercial sterilization of canned foods range from 2 to 15 min; most of them lie within 6 min (Toledo 1991; Teixeira 1992). In this study, the products processed to an F_0 of about 10 min followed immediately by cooling with 20 °C water in both RF and retort processes.

Some quality degradation occurs with the achievement of process sterilization values (F_0) during a thermal process. The relative thermal effect on food quality in a thermal process can be quantified by using the concept of cook value, which was first used by Mansfield (1962). Specifically, the cook value, C_{100}, relates the quality loss during a high-temperature thermal process to an equivalent cooking process at 100 °C (stovetop temperature) (Lund 1986). C_{100} can be calculated according to:

$$C_{100} = \int_0^t 10^{\left[\frac{T-100}{z}\right]} dt \quad (2)$$

The z value, ranging from 25 °C to 47 °C, corresponds to sensory attributes, texture softening, and color changes. A z value of 33 °C is often used to compute a cook value describing the overall quality loss (Lund 1986).

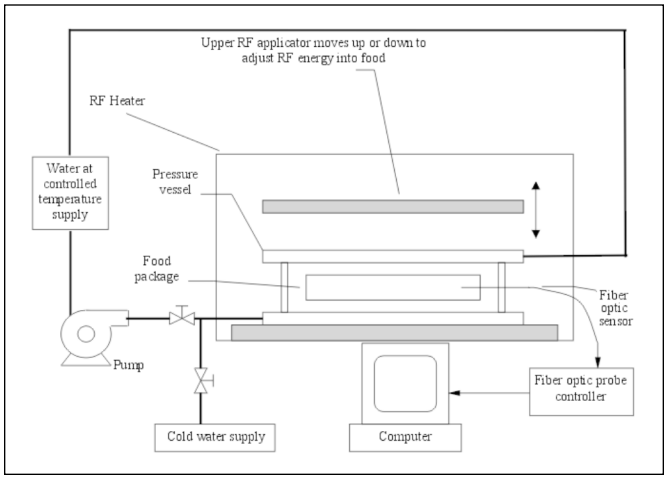

Figure 1—Simplified schematic diagram of the RF sterilization system developed at Washington State Univ. (Pullman, Wash., U.S.A.)

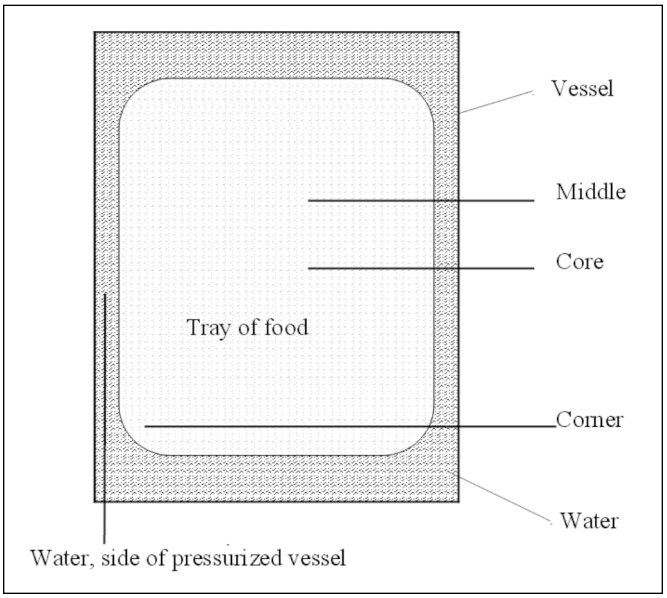

Figure 2—Placement of fiber-optic sensors in the middle layer of a sealed 6-pound military group ration tray

Chemical marker assay

In dielectric heating, the location of the cold spot is often different from that of the conventional retort. In conventional retort, the cold spot in a container with solid or semi-solid food is typically near the core of solid or semi-solid food in a package. With RF heating, the location of the cold spot is dependent on the many factors affecting the interaction between the electromagnetic field and the food, and is not easily predicted. While fiber-optic sensors provide valuable real-time temperature measurements, they can measure only a few discrete points within the food (Berek and Wickersheim 1988). The limited numbers of point temperature measurements in dielectric heating may not be adequate in evaluating the time–temperature distribution and locating the cold spot in the packed foods in our study. The Intrinsic Chemical Marker method, developed at the U.S. Army Natick Research labs by Kim and Taub, was used to assess heating uniformity and determine the least heat-treated locations in food.

The chemical marker method uses the yield of thermally produced compounds as a time–temperature integrator. One such compound, 2,3-dihydro-3,5-dihydroxy-6-methyl-(4H)-pyran-4-one, designated *M-1*, is formed in the reaction between D-glucose or D-fructose and amines through 2,3-enolization under weakly acidic or neutral conditions at sterilizing temperatures (Kim and others 1996). Summary of reaction pathways and construction of *M-1* is shown in Figure 3. The formation of *M-1* can be approximately estimated by the following empirical equation (Wang 2002):

$$M(t) = M_\infty - (M_\infty - M_0)\exp[\int_0^t -k_{ref}\, e^{\left(-\frac{E_a}{R}\left[\frac{1}{T}-\frac{1}{T_{ref}}\right]\right)}dt] \quad (3)$$

where M, M_0, and M_8 is marker yield at time zero, current time, and time infinite; t is time (min); k is reaction rate constant (0.0351/min); R is the universal gas constant (1.987 cal/mol K or 8.314 J/mol K); T is temperature (K); and E_a is the activation energy (24.51 kcal/mol). The saturation levels can vary from 1.667 to 1.854 peak area/g (Table 1) at sterilization temperatures.

In this study, a high-performance liquid chromatography (HPLC) system was used to analyze *M-1* chemical marker yields due to the thermal treatments in a whey protein gel used as a model food. The system consisted of a photodiode array detector (HP1040A; Hewlett Packard Co., Plainsboro, N.J., U.S.A.) and a solvent delivery system

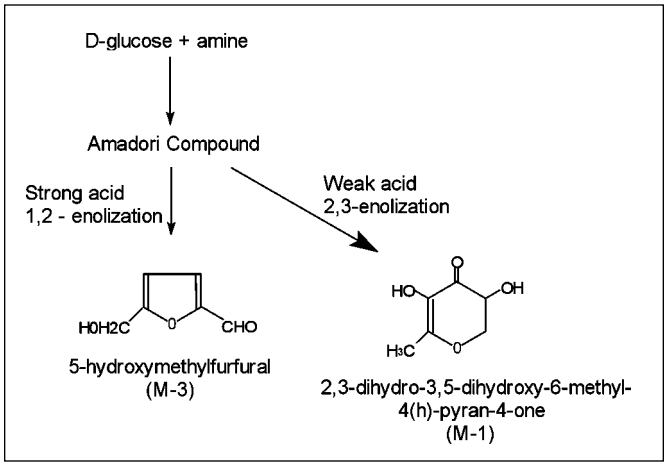

Figure 3—Summary of reaction pathways and construction of *M-1* (Kim and others 1996)

Table 1—The M_0 and M_∞ (saturation level) of *M-1* formation determined by nonlinear regression

T (°C)	M_0 (Peak area/g)	M_∞ (Peak area/g)	r^2
116	0.000 ± 0.091	1.667 ± 0.104	0.985
121	0.000 ± 0.072	1.750 ± 0.110	0.990
126	0.000 ± 0.089	1.758 ± 0.110	0.986
131	0.000 ± 0.075	1.854 ± 0.143	0.988

(ISCO model 2350; ISCO Inc., Lincoln, Nebr., U.S.A.) controlled by a desktop computer and connected to an integrator. To prepare for HPLC analysis, whey protein gels were sampled from a thermally processed tray and weighed. About 0.2 g of whey protein gel was then homogenized in 4 mL of 10 mM H_2SO_4, centrifuged and filtered through 0.45-µm nylon membrane filters. The filtered solutions were injected in an HPLC Fast Acid Analysis Column (Bio-RAD, Hercules, Calif., U.S.A.) equipped with an automatic injection system (HP1050; Hewlett Packard Co.). The mobile phase was 10 mM H_2SO_4 at a flow rate of 1 mL/min. Absorbance of the effluent was determined at 295 nm as per Kim and Taub (1993).

The model food was made of 20% Alacen 882 whey protein concentrate (New Zealand Milk Products, Santa Rosa, Calif., U.S.A.) containing 80% protein on dry basis, 2% glucose (Fisher Scientific, Fair Lawn, N.J., U.S.A.), 0.59% sodium chloride, with the remainder distilled water. The salt was added to closely match the sodium content of the packaged macaroni and cheese. The 6-pound capacity polymeric tray, 292 × 229 × 49 mm, with a 1.6-mm-thick wall (Rexam Containers, Union, Mo., U.S.A.) was filled with the whey liquid mixture and sealed using a laboratory vacuum tray sealer (Reynolds Metals Company, Richmond, Va., U.S.A.) with a 0.1-mm thick aluminum foil lid (Jefferson Smurfit, Dublin, Ireland). The sealed tray was immersed into a water bath at 80 °C for 40 min to form the gel.

Macaroni and cheese

Based on the success of the model food with RF, the sterilization of a real food, macaroni and cheese, was conducted. Retort heating was also conducted to allow a comparison between conventional methods and the new RF method. The RF heating system and temperature measurement instruments used were the same as those used for chemical marker studies.

To prepare the sample for sterilization, semolina noodles (IGA, Inc., Chicago, Ill., U.S.A.) were cooked for 6 min in boiling water. A sauce was made of whole milk, margarine (Imperial, Lipton, Englewood Inc., Cliffs, N.J., U.S.A.), cheese powder (Kraft Foods Inc., Northfield, Ill., U.S.A.), and water. The cheese sauce and drained noodles were combined before filling the trays. Each tray was filled with six pounds (2.7 kg). This precooking method was considered most suitable for RF heating after many tests were conducted.

A Minolta colorimeter (Minolta Spectrophotometer CM-2002; Minolta Camera Co., Ltd., Japan) was used to measure the color of RF-treated, conventional retorted and control products. Values of L*, a*, and b* were recorded, where L* is lightness or darkness, whose value varies from 100 for perfect white to zero for black; +a* is redness and –a* is greenness; +b* is yellowness and –b* is blueness (Francis 1998). The colorimeter was calibrated with a white tile before measurement. The food sample was placed in a glass tray with a plastic film cover in each measurement. The color measurement was performed in triplicate.

Results and Discussion

Time–temperature profile for the model food

Time–temperature profiles from the fiber-optic temperature

sensors indicated that using RF heating, 30 min were required for all 3 measured locations of the food to reach the target temperature of 121 °C. The temperature curves at the 3 locations followed almost the same trend during the heating period (Figure 4). The F_0 values were 9.4, 7.8, and 9.6 min for the core, corner, and middle (that is, midway between core and short side), respectively (Table 2). Using a conventional retort, 90 min were required for the core of the food to reach 121 °C, and the temperature curve of the bottom followed quite a different trend from those of the core and middle. The F_0 values were 10.6, 12.0, and 18.0 min for the core, middle, and bottom respectively. The cook values obtained using RF heating were 81, 57, and 84 min for core, corner, and middle, much lower than the corresponding values of 151, 159, and 189 min for core, middle, and bottom of conventionally retorted product (Table 2).

Table 2—Sterilizing values (F_0) and cook values for whey protein gel

Method	Location	F_0 (min)	C_{100} (min)
RF	Core	9.4	81
	Middle	9.6	84
	Corner	7.8	57
Retort	Core	10.6	151
	Middle	12.0	159
	Bottom	18.0	189

Chemical marker profile for the model food

Chemical marker *M-1* yields at different locations in 3 layers of the model food after RF heating were lower and relatively uniform with concentrations around 0.87 peak area/g and standard deviation of 0.07 peak area/g among 33 samples (Figure 5). For the conventional retort method, marker yields were lower at the core (1.18 peak area/g) and higher at the edges of the model food method (1.64 peak area/g) (Figure 6). The average concentration of *M-1* among all 33 samples was 1.39 peak area/g much higher than 0.87 peak area/g of treated sample with a standard deviation of 0.16 (Figure 7), again larger than 0.07 peak area/g in RF treated sample. It can therefore be concluded that for a thermal process to deliver the same F_0 in a 6-pound tray, RF heating has less adverse impact on product quality and is much more uniform than conventional retort.

Figure 7 shows that the top layer of RF heated model food yielded the least *M-1* yield, followed by the middle layer, and most yield in the bottom layer. This pattern reflects the feature of RF heating, the construct of the tray, and the nature of heat transfer. During RF heating, the RF field interacted directly with the food and generated relatively uniform heating. During cooling, the top layer directly under the thin aluminum foil lid cooled more rapidly, followed by the middle layer, with the bottom layer cooling the slowest because of the relative thick, poorly conducting polymeric material. This is supported by the information from the temperature sensors. During heating, the core, middle, and corner of the food were heated uniformly. During cooling, the corner was cooled down faster than core and middle (Figure 4), since the corner of the food faced the wall of the tray. The rapid cooling of the corner resulted in F_0 and C values that were lower than those of the core and middle. The top layer seemed to be the cold spot during RF process.

The current aluminum laminate film lid stock does not block the RF field from coupling in the food. This is because of polarization of positive and negative charges in metal film to form 2 pairs of electrodes: the first between the top plate of the pressurized vessel and the aluminum film; the second between the aluminum film and the bottom of the pressurized vessel. The food was between the second pair of the electrodes. There was no energy lost due to the

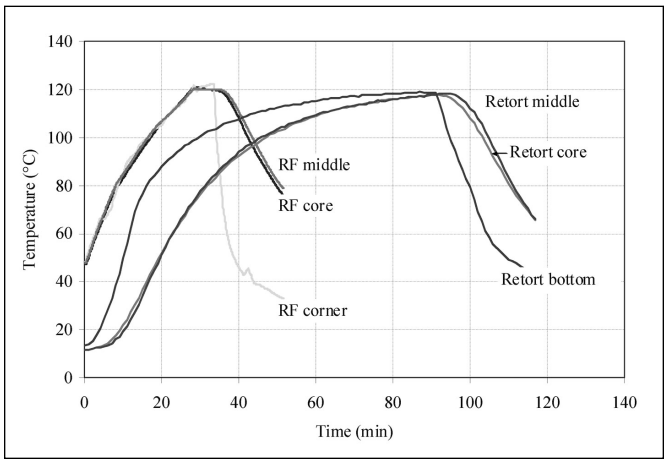

Figure 4—Time–temperature profile for whey protein gel thermally processed by radio frequency and conventional retort methods

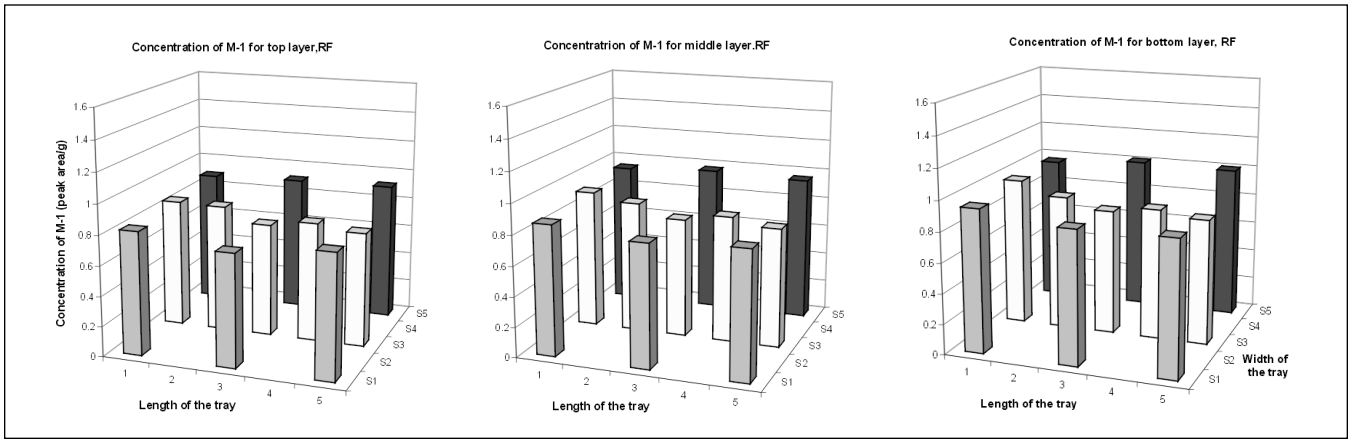

Figure 5—Concentrations of *M-1* yields at different locations in 3 layers of the model food after radio frequency treatment

presence of the metal film lid stock. This is significantly different from microwave heating, in which a metal film shields the food from the electromagnetic wave.

Figure 7 also shows that the bottom layer of the retorted model food yielded the most *M-1*, followed by the top layer, then the middle layer. Heat transfer in the retort method is governed by conduction. Slow heat transfer from the heating medium to the cold spot (normally in the core of a solid food) can cause the food product near the wall of the container to be significantly overcooked. To ensure that the cold spot (the center of the middle layer) reached its target heat treatment, both the top and bottom layers were thermally abused, though the top layer cooled faster than the other 2 layers. This resulted in *M-1* yields increasing from the middle, top, to bottom layer. This is supported by data from the temperature sensors. The bottom part received a larger value of F_0 than the core (Figure 4 and Table 2).

The predicted *M-1* yield was calculated by taking numerical integration of Eq. 3, using the time–temperature profile for whey protein gel thermally processed by RF and conventional retort methods (Figure 4). The values predicted by Eq. 3 agreed well with those obtained from experiments (Table 3) except the point of the corner of the tray for RF heating. This discrepancy might have been caused by slight differences in locations where temperature was measured by fiber-optic temperature sensor and where the sample was taken for chemical marker analyses. Specifically, the temperature was taken right next to the wall of the tray, which might have responded more closely to changes in circulating water temperature while the sample for chemical marker analyses was taken about 3 mm away from the corner. This resulted in a lower calculated value for *M-1* yield compared with the measured value.

Generally, the results obtained by temperature sensors, intrinsic chemical marker, and marker kinetic calculation agreed with each other very well.

Time-temperature profile for macaroni and cheese

The results obtained by temperature sensors and color measurements of macaroni and cheese agreed with those expected, based on experience from work with whey protein gels. Thirty minutes were required for all 3 measured locations in food to reach the target temperature of 121 °C (Figure 8). The F_0 values of 10.9, 29.1, and 14.0 min with cook values of 85, 98, and 94 min were obtained for the core, corner, and middle, respectively, using RF heating (Table 4). Using a conventional retort, it took 85 min for the core of the food to reach 121 °C, resulting in a somewhat lower F_0 value of 7.3 min and much greater cook value of 154 min.

Color measurements for macaroni and cheese

Color measurements for both RF heating and conventional re-

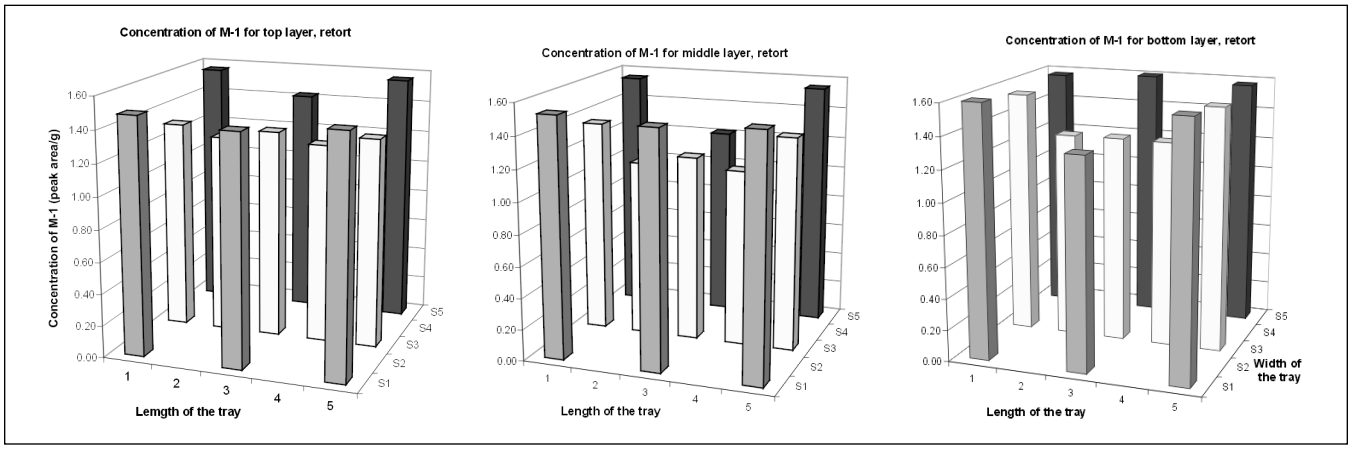

Figure 6—Concentration of *M-1* yields at different locations in 3 layers of the model food after retort treatment

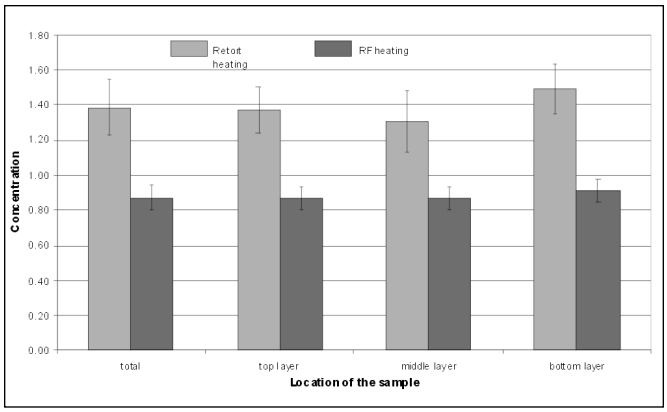

Figure 7—Average *M-1* yields for 3 layers and overall average *M-1* yield for the whole block of model foods thermally processed by radio frequency and conventional retort methods.

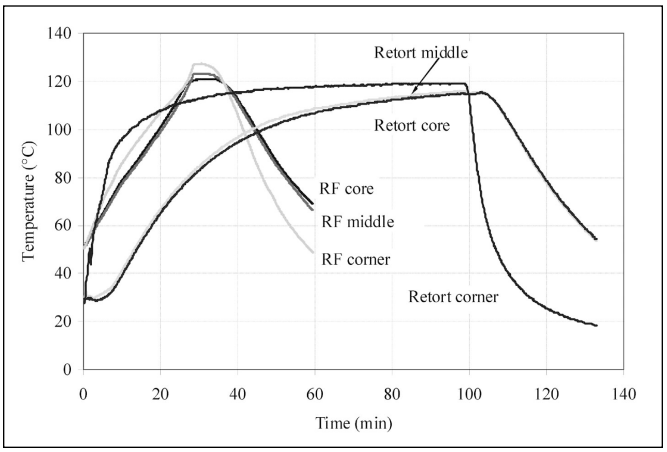

Figure 8—Time–temperature profile for macaroni and cheese thermally processed by radio frequency and conventional retort methods.

tort products are shown in Table 5. The results are expressed in L*, a*, and b* values. The L* value of the surface of the product heated by RF was 58.7, not significantly different from the value of 61.5 for the control ($P > 0.05$), and much higher than the value of 48.5 for the surface of product heated by conventional retort ($P < 0.05$). The L* values for cores of products heated by RF and conventional retort were 55.8 and 53.4, respectively, not significantly different ($P > 0.05$). Similarly, a* and b* values of RF-treated sample were much closer to that of the control than the retorted counterparts. The color change of the product was in general agreement of measured *M-1* yields and calculated cook value for the 2 processes. The retorted product gave a burned flavor of the macaroni and cheese, whereas RF-treated product was not much different from that of the control that had not been thermally processed.

Conclusions

THE YIELD OF *M-1* IN RF HEATING WAS RELATIVELY EVEN. THE COOK value was half of that of the conventional retort process while delivering an F_0 that was nearly identical to that of the retort. The results indicated that satisfactory temperature uniformity, low cook value, and lethality F_0 = 10 min were achieved within a short process time (30 min). This 30-min process time was, to a large degree, limited by the maximum RF power of the equipment. We expect to further reduce this process time with a larger RF power source. On the basis of the study, experiments were conducted on real food such as macaroni and cheese and similar result was obtained. Compared with the conventional method, the RF process produced better quality products with shorter time and less energy. Radio frequency heating will probably serve as an improved means of producing higher-quality, shelf-stable foods for civilian and military use.

Table 3—Comparison of predicted and measured *M-1* yield after retort and RF treatments

Method	Location	Calculated (peak area/g)	Measured (peak area/g)
RF	Core	0.78	0.79
	Middle	0.81	0.84
	Corner	0.60	0.84
Retort	Core	1.15	1.18
	Middle	1.19	1.12
	Bottom	1.31	1.31

Table 4—Sterilizing values (F_0) and cook values for macaroni and cheese

Method	Location	F_0 (min)	C_{100} (min)
RF	Core	10.9	85
	Middle	14.0	84
	Corner	29.1	98
Retort	Core	7.3	154
	Middle	9.1	166
	Bottom	33.2	277

Table 5—Color measurement for macaroni and cheese, average of triplicates

Method	Location	L*	a*	b*
Control		61.8	9.3	27.6
RF	Corner	58.7	8.0	25.0
	Core	55.8	7.7	24.5
Retort	Corner	48.5	7.4	19.7
	Core	53.4	5.6	23.6

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