

TRANSIENT TEMPERATURE BEHAVIOR OF A SPHERE HEATED BY MICROWAVES

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We have developed a model for microwave heating of a sphere in a rectangular resonant cavity. The model calculates transient temperature distributions within a sphere during the approach to steady state conditions or on the path to thermal runaway. The calculation takes advantage of spherical symmetry and divides the sample into concentric spherical shells. The temperature dependence of thermal and dielectric properties of the sample are taken into account and values of temperature, heat flux density, and microwave absorption in each shell are systematically updated as functions of time. The results of this transient model for alumina spheres agree with the predictions of a previously derived steady state model. Time dependent temperature profiles will be presented for various experimental conditions. An S-shaped microwave heating curve is obtained that is similar to the 1-1) model predictions of Kriegsmann.

INTRODUCTION

The theoretical background for our present work was developed and previously reported in two stages. The first stage dealt with total microwave power absorption by a homogeneous, lossy dielectric sphere in a resonant rectangular cavity [1]. The second stage extended those results and dealt with steady state conditions in lossy dielectric spheres [2] where thermal and dielectric properties were inhomogeneous overall, but could be treated as homogeneous in individual spherical shells.

The present work extends the previous studies to include the transient response of a microwave heated spherical sample. Modeling of transient temperature profiles in heated samples can be a powerful aid in processing of materials. Some important benefits that will be discussed here are (1) identification of parameters that determine preheating and processing times and prediction of those times for specified experimental conditions, (2) improved understanding of thermal runaway and how to avoid it, (3) control of temperature profiles to improve the quality of processed samples or to explore processing conditions that may produce new and useful materials. Significant progress has been reported by earlier workers in their treatments of cylindrical [3] and one-dimensional models [4], [5]. However, there clearly is much remaining to be investigated and understood.

The basic problem of interest in this new study is to find simultaneous time-dependent solutions of Maxwell's equations and a heat balance equation throughout the cavity and sphere. These equations may be strongly coupled in a non-linear

manner by a temperature dependent complex-valued dielectric constant. Just as in the model for the steady state [2], conditions are specified that enforce approximate spherical symmetry inside the sphere. Our theory takes advantage of this spherical symmetry and divides the sample into spherical shells. Each shell can have different thermal and dielectric properties. Parts of the problem can then be solved analytically while taking into account temperature dependence of thermal and dielectric properties of the sample, but other parts require application of numerical methods. A gray body model was assumed for these calculations. The solution yields values of temperature, heat flux density, and microwave absorption in each shell that are systematically updated as functions of time.

Some calculated results for alumina will be presented to illustrate benefits of such modeling mentioned earlier. Alumina was chosen since the temperature dependence of the thermal and dielectric properties of this material have been measured (see references 4 and 5 in [2]). These theoretical calculations include time-dependent temperature profiles during approach to steady state conditions or on a path to thermal runaway.

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Calculation of time-dependent temperature profiles in a sample requires simultaneous solution of a set of electromagnetic and thermal equations. The electromagnetic fields must satisfy Maxwell's equation throughout the cavity, including the interior of the sample. These equations imply that the electric field $E = E_0 e^{-i\omega t}$ satisfies,

$$\nabla^2 E + \frac{\omega^2}{\epsilon_r \epsilon_0 \mu_0} E = 0 \quad (1)$$

and

$$\epsilon_r = \epsilon_r' + i\epsilon_r'' \quad (2)$$

inside the sample. Thermal energy balance averaged over a cycle of electromagnetic field oscillation requires

$$\dot{T} = \frac{W}{C} - \frac{1}{C} \nabla \cdot \mathbf{q}. \quad (3)$$

where W is microwave power absorbed per unit volume:

$$W = \frac{1}{2} \omega \epsilon_0 \epsilon_r'' |E_0|^2. \quad (4)$$

The heat flux density \mathbf{q} satisfies

$$\mathbf{q} = -k \nabla T. \quad (5)$$

Also, $C = \rho c_p$, where C is heat capacity per unit volume, ρ is the density of the sample, and c_p is specific heat at constant pressure.

The model assumes radiative boundary conditions at the sample surface,:

$$q_s = \sigma \epsilon (T_s^4 - T_w^4). \quad (6)$$

Here q_s is the heat flux density normal to the surface, T_s and T_w are temperatures of the sample surface and of the cavity walls, respectively, ϵ is emissivity and σ is the Stefan-Boltzmann constant,

Equations (1) and (3) are directly coupled through the dielectric constant ϵ_r , which is temperature dependent. Of particular interest is ϵ_r'' which is strongly temperature dependent in cases presented in this paper. The set of electromagnetic and thermal equations are non-linear in the electric field and in the temperature. These equations can be solved in general only by using finite element methods.

Just as in the earlier treatment of the steady state [2], the complexity of our problem can be substantially reduced by confining attention to situations where the temperature varies only radially in the sample. In practice this condition can be imposed to good approximation by taking the sample to be a sphere that is properly positioned in a cavity with appropriately selected dimensions and mode of excitation. A specific example will be given later in this paper.

Under these circumstances, the sample can be treated as a collection of thin spherical shells. The temperature-dependent dielectric constant is regarded as a constant throughout each shell, but it can vary from one shell to another. Then for a specified distribution of shell dielectric constants, the electromagnetic fields can be treated analytically using scattering formalism, wherein any resonant mode of the cavity is resolved into transversely polarized plane waves that impinge on the sphere and are then scattered. Inside each spherical shell there are both incoming and outgoing spherical waves that are matched at shell boundaries. This scheme provides approximate values for the fields and for the power absorbed per unit volume in the system of shells.

The temperature-dependent quantities consisting of heat capacity per unit volume, thermal conductivity and power absorbed per unit volume are also treated as constant in each shell, but may vary from one shell to another. Temperature dependence of emissivity is also taken into account. One can linearize the temperature as a function of r in each shell and then analytically integrate Eq. (3) over the volume of any shell. Next one can use finite difference ratios to replace first derivatives with respect to time and radius and require that $T(r)$ and $q(r)$ be continuous at the shell boundaries. For a closely spaced set of shell radii

$\{r_i, i = 1, 2, \dots, N\}$ and time instants $\{t_\alpha, \alpha = 0, 1, 2, \dots, M\}$, one finds that the temperatures $T_i^\alpha \equiv T_i(t_\alpha)$ satisfy the following collations,

$$T_i^{\alpha+1} = T_i^\alpha + (t_{\alpha+1} - t_\alpha) \left\{ \frac{W_i^\alpha}{C_i^\alpha} - \frac{3}{C_i^\alpha} \frac{[r_i^2 q_i^\alpha - r_{i-1}^2 q_{i-1}^\alpha]}{r_i^3 - r_{i-1}^3} \right. \\ \left. - \frac{1}{4} \frac{q_i^\alpha - q_{i-1}^{\alpha-1} a_{i-1}^\alpha \kappa_i^\alpha}{\kappa_i^\alpha (t_\alpha - t_{\alpha-1}) (\kappa_i^\alpha)^2 t_\alpha - t_{\alpha-1}} \kappa_i^{\alpha-1} \right\} \\ \times \left[\frac{r_i^4 + r_{i-1}^3 (3r_{i-1} - 4r_i)}{r_i^3 - r_{i-1}^3} \right] \quad (7)$$

$$q_i^\alpha = -\kappa_i^\alpha \frac{T_{i+1}^\alpha - T_i^\alpha}{r_{i+1} - r_i} \quad (8)$$

For specified initial conditions one can calculate $\{W_i^{\alpha=0}, i = 1, 2, \dots, N\}$ and propagate the temperature profile and microwave absorption forward in time with the aid of Eqs. (6), (7), and (8), and results of the electromagnetic wave scattering theory. This formalism allows for the possibility that the normal mode amplitude for the cavity excitation and the temperature of the cavity walls may change with time.

When calculating temperature profiles using these finite difference equations, it is important to choose the mesh size for shell radii and time in a manner that meets certain stability conditions that are known from general theory of numerical analysis [6]. Calculated results based on these formulas will be discussed next.

DISCUSSION

This spherical shell model was previously applied to calculate the steady state behavior of alumina spheres heated in a single mode microwave cavity [2]. A TM 354 mode excited in a rectangular cavity with dimensions Of $L_x = 10.25$ cm, $L_y/L_x = 1.776$, and $L_z/L_x = 1.463$ was chosen to produce an essentially isotropic electric field intensity near the center of the cavity. For those experimental conditions, the TM 354 mode frequency was $f = 7.21$ GHz and the empty cavity wave number was $k = 1.51$ cm⁻¹. The present study uses these experimental conditions to further investigate the transient behavior of an alumina sphere as it approaches a steady state condition or experiences thermal runaway. Unless stated otherwise, the predictions presented in this paper were determined by dividing the sphere into 20 equally spaced spherical shells. The electric field strengths given in this paper are norms] mode characteristic amplitudes. Of course, the electric field varies throughout the cavity, including the interior of the sample.

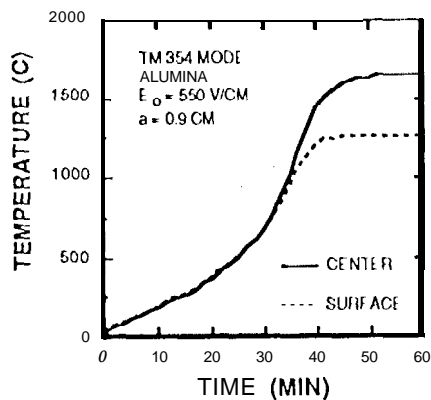


Figure 1. Transient response of a 0.9 cm radius alumina sphere.

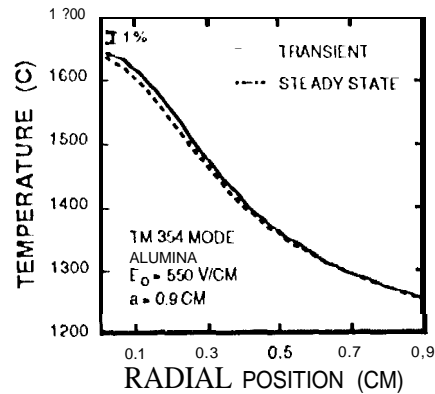


Figure 2. Comparison of calculated transient and steady state equilibrium profiles.

The transient response of an alumina sphere of radius 0.9 cm situated in the center of the cavity and heated with the TM 354 mode continuously excited at a field strength of 550 v/cm is shown in Fig. 1. The center and surface temperature of the sphere are shown as a function of time. For this experimental case, the center and surface temperatures remain essentially equal below ≈ 400 C. The sample surface temperature reaches a steady state condition before the center. An equilibrium temperature gradient between the center and surface is achieved after ≈ 50 minutes. The calculated resultant steady state temperature gradient within the sphere is shown in Fig. 2. The solid and dashed curves correspond to the present transient model and steady state model [2] respectively. For this comparison, the sphere was divided into 40 equally spaced spherical shells. The calculated internal gradients agree to better than 1% throughout the interior of the sphere. There is a temperature difference of ≈ 387 C between the center and surface of the sphere for these experimental conditions. Calculations using only 20 concentric spherical shells gave essentially the same results for the transient model shown in Fig. 2, however, the steady state model results were $\approx 0.7\%$ lower. This test demonstrates the faster convergence of the transient model over the steady state model.

The time required to reach steady state conditions is strongly dependent on the experimental conditions. This is illustrated in Fig. 3 where the transient behavior at the center of a small 0.1 cm radius sphere is shown for various electric field strengths. In this case, large field strengths are required to reach high equilibrium temperatures. A field strength of 4000 v/cm leads to a steady state condition at the center of the sphere in ≈ 10 minutes. As the field strength is gradually increased to 5000 v/cm, higher steady state temperatures are reached during the same 10 minute time interval. The time to achieve steady state increases as the electric field is increased further, until a critical field value (≈ 5200 v/cm) is reached that leads to thermal runaway. For this experimental case, thermal runaway occurs after 30

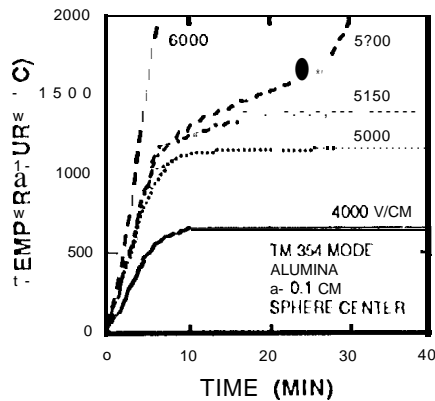


Figure 3. Transient response of a 0.9 cm radius alumina sphere.

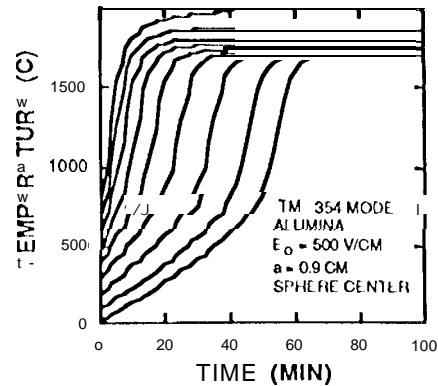


Figure 4. Effect of cavity wall temperature on approach to steady state or runaway.

minutes. Field strengths above this critical value produce the thermal runaway condition more quickly (i.e., ≤ 7 minutes for 6000 v/cm).

Figure 4 shows the transient heating behavior associated with various ambient cavity wall temperatures. These calculations are for the center temperature of a 0.9 cm radius sphere. The time required to reach steady state is the longest (70 minutes) for the cavity walls being held at room temperature throughout the heating process. As the ambient cavity wall temperature is increased, the equilibration time becomes significantly reduced (< 35 minutes for 400 C ambient wall temperature). Increasing the cavity wall temperature above 400 C leads to higher steady state temperatures since the walls are now providing significant sample heat input along with the microwaves. For these experimental parameters, wall temperatures ≥ 800 C lead to thermal runaway.

We have extensively explored the transient heating properties associated with thermal runaway in a microwave processed spherical sample. Figure 5 shows the predicted temperature profiles within a 0.875 cm radius sphere during the thermal runaway process. These curves correspond to a sphere heated with an electric field strength of 2000 v/cm. A 100 C temperature gradient between the sphere center and surface is generated after 83.5 seconds. At 96 seconds after starting the process the center of the sample has become 365 C hotter than the surface. For longer processing times, the surface temperature hardly changes (remaining near 600 C) while the sphere center temperature experiences thermal runaway. We have arbitrarily assumed that the sample will melt at 2100 C which corresponds to ≈ 100 seconds. These calculations suggest that it is difficult to anticipate thermal runaway by monitoring only the surface temperature of a sphere.

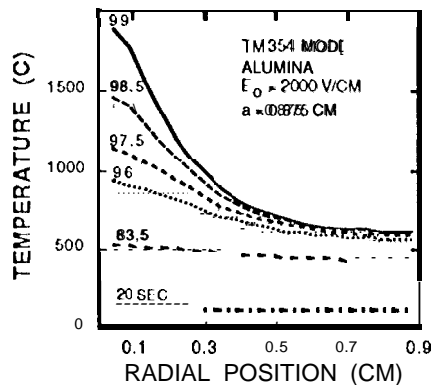


Figure 5. Temperature gradient within a sphere during thermal runaway.

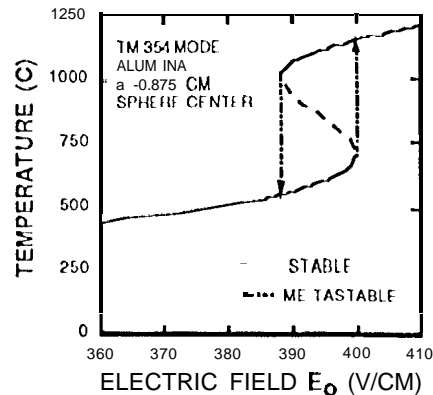


Figure 6. Internal S-shaped heating curve with thermal runaway without melting.

in the previous study of steady state behavior [2], parametric studies were performed primarily near the lowest absorption peak associated with electromagnetic resonance's within the sphere. Anomalous behavior similar to thermal runaway was predicted (see Fig. 3 in ref. [2]). The present transient model was used to explore, in more detail, the behavior of this predicted internal runaway phenomenon. The 2-D microwave modeling studies of Kriegsmann [4] have predicted an S-shaped heating curve that is a multivalued function of the microwave power. This new response curve implies that there are actually two branches to the heating curve; a lower temperature branch that can lead to thermal runaway and an upper branch that was not previously anticipated. This S-shaped heating curve prediction was tested within the context of the transient spherical shell model. Figure 6 shows the results of this analysis at the center of a 0.875 cm radius sphere. A sample of this size was previously studied using the steady state model (see Fig. 3 in ref [2.]). The earlier work predicted the lower branch of the heating curve, the thermal runaway jump (upward arrow) to the upper heating curve and the higher temperature behavior of the upper curve (see Fig. 4 of ref. [2]). This predicted thermal runaway phenomenon did not lead to sample melting because for this experimental case the enhanced microwave heating associated with the runaway becomes significantly reduced at higher temperatures. The results of the transient model agreed with the previous steady state predictions for these experimental parameters. The multivalued portion of the upper branch of this S-shaped heating curve was determined by initially heating the sample with a field strength of 410 v/cm that is greater than that associated with runaway (400 v/cm). The field strength was reduced to a new constant value when the temperature of the sample reached 1200 C which is higher than the upper branch in the region shown here. If there was no multivalued upper branch the system would cool back down to the lower branch value corresponding to the constant field strength. This procedure produced the predicted multivalued portion of the upper branch shown in Fig. 6.

The unstable branch of the S-shaped curve, in Fig. 6 defines the locus of points that separate regions attracted by the upper and lower branches. The unstable branch was determined using the following procedure. First, a field strength was chosen between the inflection point values associated with the upper and lower branches, i.e., between 388 - 400 v/cm. The sphere was again initially heated at 410 v/cm until its temperature reached a predetermined value between the inflection point temperatures associated with the upper and lower branches, i.e., between 700 - 1000 C. The field strength would then be lowered to the chosen value and the direction of the sphere temperature was determined. This procedure was repeated in temperature intervals of 20 C for the chosen field strength until the unstable temperature value was identified. The rest of the unstable curve was determined by repeating the procedure for a range of chosen field strengths.

The hysteresis behavior is one of the consequences of this S-shaped heating curve. This type of behavior is also illustrated in Fig. 6. During an initial heating phase, with the field strength slowly incremented up to 410 v/cm, the sample will follow the lower steady state branch until the inflection point temperature of 700 C is reached. At this time the sample will experience a thermal runaway effect (upward arrow) until it reached the upper branch at ≈ 1140 C. Now the sample will continue to following the upper branch as its temperature increased to the steady state temperature of 1205 C associated with the final field strength of 410 v/cm. A different processing curve will be attained if the field strength is now slowly incremented downward to zero. This time the sample follows the upper branch downward to the inflection point temperature at 1000 C where it abruptly decreases (downward arrow) to 535 C at the lower branch. Further reduction of the field strength causes the sample to move down the lower branch.

We have investigated the effect of the temperature gradient on the unstable intermediate branch of the heating curve. Any spatial gradient configuration can be analyzed with the transient model. A gradient in which the surface was hotter than the center by 200 C produced the same unstable curve as shown in Fig. 6. The hypothetical situation where the surface and center have equal temperatures was also investigated. For this case an inverted v gradient was used with the hottest region being half way between the center and the surface at a temperature 200 C above these extreme positions. For this gradient, the single case studied suggested that the unstable curve was approximately determined by the highest temperature in the starting inverted v -shaped profile.

To verify that the S-shaped heating curve shown in Fig. 6 was a general phenomenon, we analyzed the microwave heating curve for a different set of experimental conditions leading to thermal runaway. Figure 7 shows the S-shaped heating curve for a 0.825 cm radius sphere that illustrates a thermal runaway phenomenon leading to sample melting. This size sample was also previously studied using the steady state mode] (see Fig. 3 in ref [2.]). For this experimental case, the runaway effect that occurs upon heating will cause the sample to melt. The upper branch of the S-shaped curve is masked by the lower branch and can only be obtained by controlling the thermal runaway phenomena [5]. The simplest method for reaching the upper branch is to initially heat the sample with a field

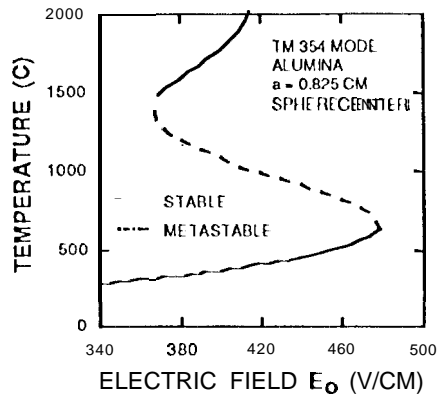


Figure 7. S-shaped heating curve with thermal runaway leading to melting.

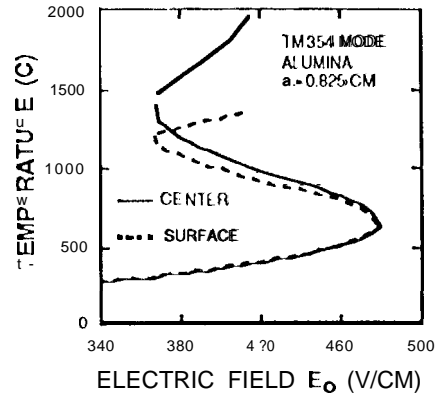


Figure 8. S-shaped heating curves for sphere center and surface.

strength larger than the critical field strength for runaway (480 v/cm corresponding to 620 C) and then at the appropriate time reduce the field strength sufficiently to cause the sample to equilibrate on the upper branch. The ability to access this upper branch would allow the processing of a material at high temperatures using lower power levels. This ability also would open up the temperature range for a given size sample which can be studied for possible new solid state phases [3].

The S-shaped heating curves shown in Figs. 6 and 7 are associated with the center temperature of the sphere. The variation of this heating curve on the position within the sphere is illustrated in Fig. 8 for both the sphere center and surface. The temperature difference between the center and surface gradually increases with temperature, becoming measurable (≥ 5 C) above 500 C. For temperatures on the upper branch of the heating curve the temperature difference can become very large (≥ 600 C at a center temperature of 1950 C).

In conclusion, we have developed a new transient model for the processing of a spherical sample in a rectangular cavity. This model, based on spherical symmetry, has elucidated the behavior of the large internal temperature gradients attained during the approach to steady state or thermal runaway. The ability to follow the time evolution of the sample during processing also provides a method for studying the features of the predicted multivalued microwave heating curve. We have investigated the behavior of this S-shaped curve in both the stable and unstable regions. Experimental access to the predicted high temperature branch of the heating curve should lead to improved methods for microwave sintering of ceramics. The present transient model can support the development of experimental techniques for reaching this branch.

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