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MATHEMATICAL MODEL OF THERMAL SPIKES IN MICROWAVE HEATING OF OXIDE CERAMIC FIBERS

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ABSTRACT

Experiments on microwave sintering of ceramic fibers in a single-mode cavity have revealed the presence of thermal spikes and "hot spots" which sometimes travel along the fiber and eventually disappear. They are triggered by relatively small increases in microwave power, and thus have obvious implications for the development of practical microwave-based fiber processing systems. These hot spots are conjectured to originate at slight irregularities in the tow morphology, and propagate as the result of solid phase transitions which take place at elevated temperatures and reduce the dielectric loss coefficient ϵ''.

An elementary mathematical model of the heat transfer process was developed which reproduces the essential features of the observed phenomena, thus lending support to our conjecture. This model is based on the assumption of one-dimensional heat conduction along the axis of the fiber tow, and radiation losses at the surface.

INTRODUCTION

Experiments on the microwave heating of alumina/silica fiber tows in a single-mode microwave cavity at 2.45GHz have revealed "hot spots" which brighten rapidly, persist for a few seconds, and rapidly extinguish [1]. Some hot spots propagate along the fiber in one direction. Since these hot spots are triggered by relatively small (c.a. 10%) increases in power, they have obvious implications for the development of practical microwave processing systems.

As reported elsewhere [1], it is conjectured that these hot spots are partially the result of a partial phase transformation of the tow material into mullite, coupled with the known temperature dependence [2] of $\epsilon''$of alumina/silica ceramics. Other contributing processes are thought to be burnoff of the carbon coating which is applied prior to processing to aid microwave coupling, and the decrease of radiative emissivity with increasing temperature of the tow. In an attempt to understand the relative importance of these various factors and their interdependence, an elementary mathematical model of the microwave sintering process has been developed. The predictions of this model are in qualititative agreement with experimental data as will be shown.

MATHEMATICAL MODEL

The mathematical model of the fiber sintering experiments is based on the following principal assumptions:

- the fiber tow is heated volumetrically and uniformly by the microwave field;
- the tow loses heat by conduction along its length and by radiation and convection to the environment (the microwave cavity);
- radial temperature gradients in the fiber tow are negligible;
- when the temperature at any point along the tow exceeds the phase transformation temperature ($1200°C$) a phase transition begins at that point which lowers the local value of the dielectric loss coefficient ϵ''.

Thus the heat conduction equation describing the temperature field is:

$$\frac{\partial}{\partial x}(k\frac{\partial T}{\partial x}) + \dot{q}(x,T) - (P/A)\{\epsilon\sigma[T(x,t)^4 - T_\infty^4] + h[T(x,t) - T_\infty]\} = \rho c_p \frac{\partial T}{\partial t}. \quad (1)$$

Here $T(x,t)$ represents the tow temperature at location x and time t, k is the thermal conductivity, x is the distance coordinate along the length, $\dot{q}$ is the volumetric heat generation rate, ϵ is the emissivity of the tow, σ is the Stefan-Boltzmann constant, ρ is density, and c_p is the specific heat capacity at constant pressure. The ratio P/A is the perimeter of the tow divided by its area, and T_∞ is the environment temperature. Since the microwave cavity is cooled by a water jacket, the cavity walls remain at ambient temperature.

The tow temperature at each end is set equal to the temperature of the environment at all times:

$$T(0,t) = T(L,t) = T_\infty, \quad (2)$$

and the initial temperature is also taken to be the environment temperature:

$$T(x,0) = T_\infty. \quad (3)$$

The local heat generation rate $\dot{q}(x,t)$ is determined from the local electric field strength E_f through the relation

$$\dot{q}(x,T) = 2\pi f \epsilon''(T) \mid E_f \mid^2, \quad (4)$$

and $\epsilon''(T)$ is calculated from a polynomial or exponential fit to literature data.

Eq.(1) is solved by a fully-implicit finite-difference scheme using 400 mesh intervals over the length of the tow. The time step size is adjusted during the solution as described below.

A typical experiment produces a time-temperature trace like that shown in Fig. 1 (the measured power applied to the cavity in this experiment is shown in Fig. 3). During a simulation, the electric field strength is set equal to a value determined from the measured

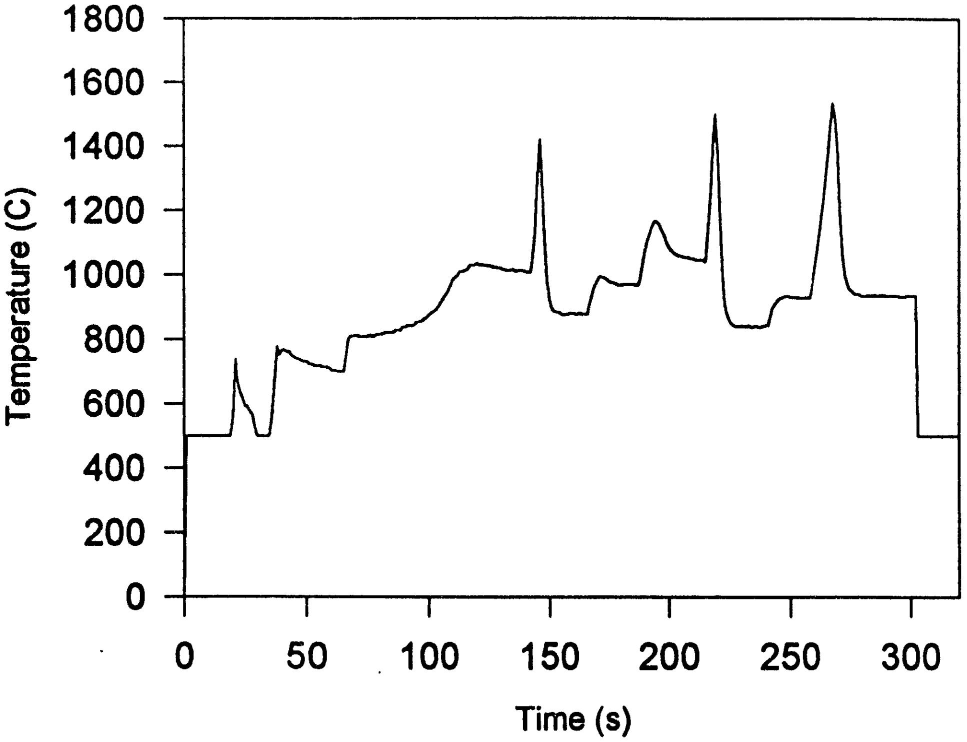

Figure 1: Temperature spikes observed during a typical experiment. The power input to the cavity is shown in Fig. 3

power data during an actual experiment, and the solution of the electromagnetic field equations for the TE_{103} resonant cavity. After each increase in field strength, the time step is shortened to 0.01 seconds until the temperature rise is nearly complete, and then it is set back to the default value of 1 second.

In order to induce a localized thermal "spike", the dielectric loss coefficient over a small interval about an arbitrary point is made larger by a small amount (ca 10 - 20%) at each time:

$$\epsilon''(x,T) \rightarrow (1+\Delta)\epsilon''_0(x,T), \quad x \in [L/2-\delta, L/2+\delta]. \tag{5}$$

To simulate the effect of a phase change on ϵ'', it is assumed that the extent of new phase present at time t is related to the accumulated time that the local temperature has exceeded the phase transition temperature T_p. Thus a quantity $s(x)$ is computed at each point, where $s(x)$ is defined as

$$s(x) = S_f \int_0^t [T(x,t') - T_p]dt, \tag{6}$$

where T_p is the phase-change temperature, and S_f is a scale factor. The loss coefficient $\epsilon''$is then adjusted downward by $\epsilon'' = F\epsilon''(T)$, where

$$F = f_1 + (1-f_1)e^{-s(x)}, \tag{7}$$

where f_1 is a positive constant < 1. The scale factor S_f and the constant f_1 are chosen so as to produce the best agreement with experimental data. It is known from $X-$ray diffraction analysis that the fraction of mullite formed in a typical experiment is about 40%.

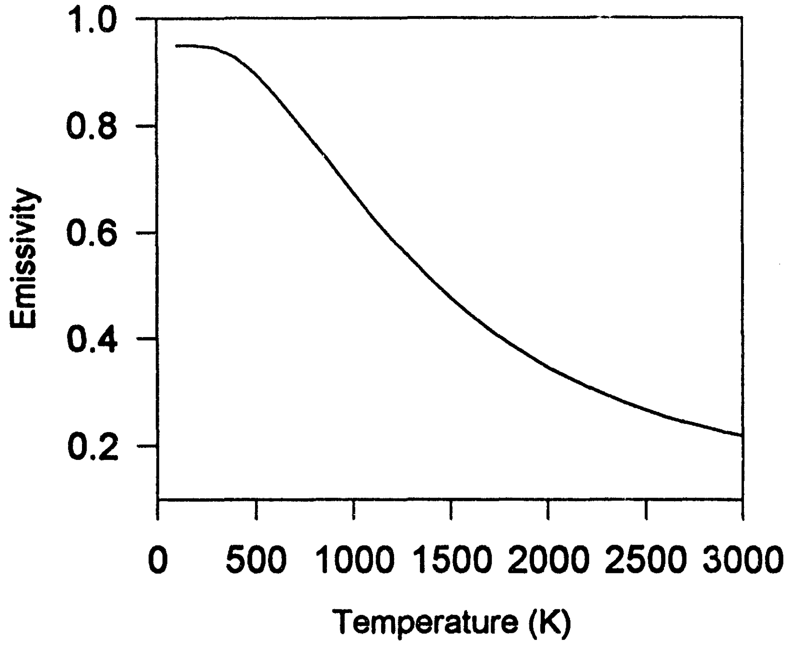

Figure 2: Temperature behavior of total hemispherical emmisivity of fiber tow

As described elsewhere [1], the fiber tows were dip-coated with carbon to provide hybrid preheating from room temperature to approximately $600°C$. Visual inspection reveals that the carbon is completely burned off by the time the samples reach approximately $1000°C$. To simulate this effect, a small uniform increment in ϵ'' was added and then decreased in magnitude according to a scheme similar to that given by Eqs. (6-7), but with different constants, assuming that the carbon begins to burn off at c.a. $600°C$.

Temperature dependence of thermal conductivity k and specific heat capacity c_p was included as polynomial fits to data from Ref. [3]. The spectral hemispherical emissivity of the fiber tow was also taken from Ref. [3] and integrated over a blackbody spectrum at various temperatures to yield the emissivity function $\epsilon(T)$ shown in Fig. [2]. The rapid decrease in emissivity with temperature is an important factor in producing the large thermal spikes seen in the experiments, since thermal radiation is the dominant heat loss mechanism.

A number of the parameters used in the simulation are not known and must be chosen by trial and error to produce the best match to experimental data. These parameters include the scale factors in the phase change and carbon burnoff models, the enhancement of the dielectric loss produced by the carbon coating, and the precise temperature dependence of the dielectric coefficient. The dielectric loss data for alumina silicate given by Tinga[2] were used as a starting point in the simulations. However, small adjustments were made to this data to improve agreement between model simulations and experimental data, particularly at temperatures greater than $1100°C$, where no data is available.

RESULTS

A typical simulation result is shown in Fig. 3, in which the average temperature over a small region near the center of the fiber, corresponding to the field of view of the optical

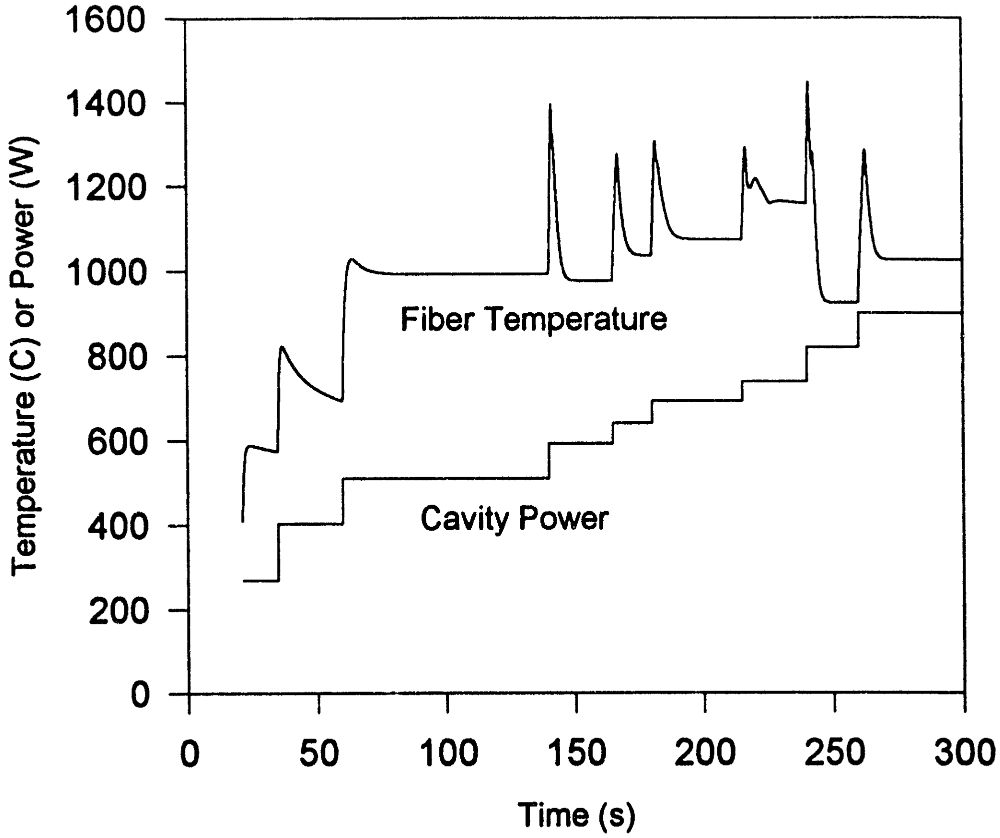

Figure 3: Result of simulation run for temperature of fiber tow subjected to the magnetron power history shown. Experimental data for comparison are shown in Fig. 1

fiber sensor, is plotted *vs.* time. This result is to be compared to the experimental curve of Fig. 1. To develop these results, we first adjust the carbon burnoff parameters to yield good agreement for the portion of the curve up to 140 seconds since visual observation reveals that the carbon has been completely removed after the first spike. The parameters describing the phase change effect on the dielectric loss are then adjusted to yield temperature spikes in qualitative agreement with the experimental data.

It is apparent that the simulation is in good qualititative agreement with the experimental data. Given the degree of uncertainty in the input parameters, this is all we had hoped for. It is quite clear from our numerical experiments that there is no unique set of parameters that would yield a good result. However we believe that our model does include all important physical processes.

Visual observation of the fiber tow during a sintering run reveals that some hot spots develop and migrate along the tow to the cavity exit. To attempt to understand this phenomenon, we induced a local hot spot in the simulation by arbitrarily increasing the dielectric loss over a very small zone as in Eq. (5). This always produced a hot spot which splits as the local temperature exceeds the phase change temperature, thus *reducing* the local dielectric loss coefficient below that of the surrounding tow material. This behavior is illustrated in Fig. 4, in which all parameters are the same as those in Fig. 3 except that $\epsilon''$was increased by 10% for $0.48 \leq x/l \leq 0.52$. Our simulations always produce two hot spots which migrate in both directions, while experimental observations usually show only a single moving hot spot. The reason for this discrepancy is still undergoing investigation.

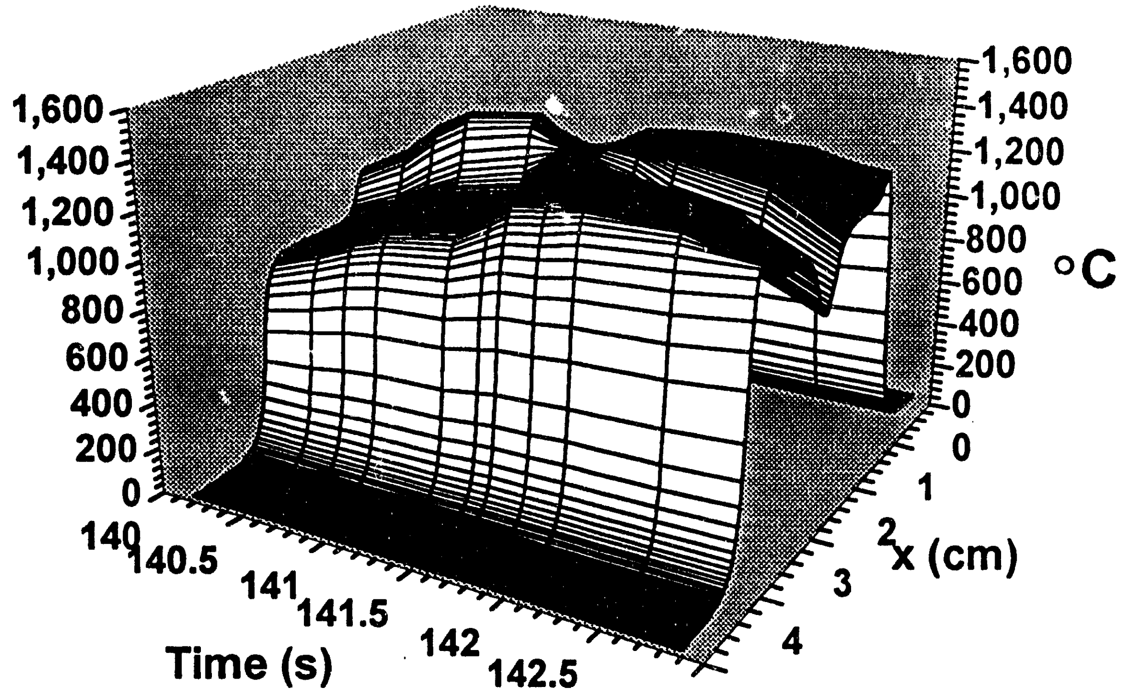

Figure 4: Simulated "splitting" of thermal spike induced by a 10% local perturbation in ϵ''. This is the spike beginning at 140.0 s in Fig. 3.

CONCLUSIONS

Thermal spikes in microwave heating of ceramic oxide fibers are apparently caused by reduction in dielectric loss as temperatures exceed a phase transition temperature which leads to a new phase with smaller dielectric loss ϵ''. A relatively simple heat transfer model which ignores coupling between the microwave field and power absorbed by the fiber leads to good qualitative agreement with experimental observations and measurements. Quantitative agreement must await better dielectric property data and a fully coupled analytical model, both of which are the subject of current research.

References

1. Gerald J. Vogt, Wesley P. Unruh, and J. R. Thomas, Jr., "Experimental Observations of Thermal Spikes in Microwave Processing of Ceramic Oxide Fibers" (This Proceedings)

2. Weiguo Xi and W. R. Tinga, in *Microwave Processing of Materials III*, edited by R. L. Beatty, W. H. Sutton, and M. F. Iskander (Mater. Res. Soc. Proc. **269**, Pittsburgh, PA, 1992) pp. 569-577.

3. "Properties of Nextel^{TM} 312, 440, & 480 Ceramic Fibers", 3M Ceramic Materials Department, St. Paul, MN (1989).

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