Thermal Runaway in Ceramics Arising from the Temperature Dependence of the Thermal Conductivity

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Thermal runaway during microwave heating in ceramic materials has been explained recently in terms of a microscopic model involving a nonlinear feedback between microwave absorption and heat dissipation. We study here an alternate model of thermal runaway which is based on the temperature dependence of the thermal conductivity of the material being heated. If the thermal conductivity decreases as the temperature increases, a decrease could occur in the removal of heat from the central regions of the sample which, unlike the surface, cannot cool off quickly through processes such as radiation. The result would be an enhanced heating of the central regions and a further decrease of the thermal conductivity thus providing a feeback loop. A quantitative analysis of these concepts is provided.

1. Introduction

There has been considerable study in recent years of the interaction of microwave radiation with ceramic materials [1—12]. This interest has been driven by interest in the possibility of sintering ceramic powders through the uniform heating which occurs with microwave irradiation. An interesting but undesirable feature often associated with microwave heating is the phenomenon of "thermal runaway", wherein a sample subjected to a constant source of microwave power will be observed to exhibit a slow rise in temperature for a considerable period of time, after which the temperature will suddenly be observed to increase at a more rapid rate, ultimately destroying the sample.

A recent theory of thermal runaway constructed by Kenkre et al. [6—12] is based on the possibility that the material contains a species of absorber (such as vacancies, bivacancies, or interstitials), which can be in one of two regions of states: a free region which can absorb strongly from the microwave field, and a bound region in which the absorption is negligible. Microwave heating is shown in that theory to transfer absorbers from the bound region to the free region. This results in heightened absorption, enhanced heating, and consequently further transfer of absorbers to the free region, thereby repeating the cycle. That theory has been quite successful quantitatively, particularly in

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its ability to model the dynamics of the runaway process. It is, however, a dynamical theory that studies the time dependence of the heating process "in the bulk" and is, in its present form, incapable of addressing the spatial dependence of the runaway phenomena. As such, it is unable to predict a prominent feature observed in many samples which have undergone thermal runaway, namely, the way in which the destruction of the sample appears to be initiated in the interior regions.

In this paper, therefore, we investigate an alternate mechanism which may contribute to the phenomenon of thermal runaway, and in the process present an analysis that allows investigation of the spatial dependence of the thermal profile in the sample. Specifically, we address the question as to whether thermal runaway could result solely from a decrease in the thermal conductivity of the sample with increasing temperature. Physically, we envision that microwave heating of the sample produces a continuous and uniform source of heating throughout. If the sample is to reach a steady-state temperature distribution, the net heat per unit time into the sample must equal the flux of heat out through its surface. Heat created in the interior must obviously migrate to the surface before it can escape. Because of the uniform heating which occurs during microwave absorption, however, the temperature in the interior is elevated with respect to that of the surface. If the thermal conductivity decreases with increasing temperature, then it follows that the thermal conductivity may be lower in the interior than near the surface. This would decrease even further the ability of the internally generated heat to leave the interior. Such a mechanism would then serve to further increase the internal temperature and, in turn, lead to a further decrease in the thermal conductivity. This is the feedback mechanism which we investigate as a possible source of thermal runaway. Our investigation reveals that this mechanism alone is probably *not* the major cause of thermal runaway observed in most ceramics. Nonetheless, it could, when combined with the mechanism proposed by Kenkre et al. [6−12], be a significant factor in determining the critical parameters associated with the runaway process.

2. Formulation of the Model

Consider a spherical ceramic sample of radius R exposed to a continuous source of microwave radiation. The skin depth at the radiated frequency is assumed to be large compared to the size of the sample. As a result, power is uniformly absorbed from the radiation field and converted into heat in the interior. The temperature $T(r,t)$ at a distance r from the center of the sample evolves in time due to the continuous absorption and diffusion of heat throughout. Its evolution is given by the radial heat equation

$$\frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} r^2 k \frac{\partial T}{\partial r} + f - \sigma T^4 \, \delta(r - R) \tag{1}$$

in which

$$f = \frac{P}{\varrho C_v} \frac{\varepsilon''}{\varepsilon_0} \tag{2}$$

represents the uniform heating rate associated with the microwave field, and

$$k = \frac{\varkappa}{\varrho C_v} . \tag{3}$$

In these expressions P is the power density of the radiation field, and ε'' is the absorption coefficient, ϱ the mass density, C_v the specific heat, and $\varkappa$ the thermal conductivity of the ceramic. The last term on the right-hand-side of (1) describes the heat lost through the surface due to radiation. In general, we expect a variation with temperature (and therefore position) of both the heating rate (due, e.g., to changes in the absorption coefficient) and the thermal conductivity. In what follows, however, we are interested in investigating the possibility that thermal runaway could result primarily from the variation of the parameter $k = k(T) = k[T(r)]$ associated with the thermal conductivity, and so ignore any temperature or spatial variation of the absorbed power f.

If a steady state temperature distribution is eventually reached, it will obey the equation obtained by setting the left-hand side of (1) equal to zero. To obtain this steady-state distribution it suffices to solve the homogeneous steady-state equation

$$\frac{\mathrm{d}}{\mathrm{d}r} r^2 k \frac{\mathrm{d}T}{\mathrm{d}r} + fr^2 = 0 \tag{4}$$

along with the boundary condition

$$J_Q(R) = \lim_{r \to R} \left(-k \frac{\mathrm{d}T}{\mathrm{d}r} \right) = \sigma T^4(R) , \tag{5}$$

which accounts for heat loss at the surface. We also impose regularity of the temperature field at $r = 0$. Specifically, the lack of any concentrated heat sources or sinks at $r = 0$ means that the heat flux out of the origin must vanish; hence $J_Q(0) = 0$.

On integrating Eq. (4) and applying the boundary condition at $r = 0$ we find that

$$J_Q(r) = -k \frac{\mathrm{d}T}{\mathrm{d}r} = \frac{fr}{3} , \tag{6}$$

from which the surface temperature $T_R = (fR/3\sigma)^{1/4}$ is obtained by applying boundary condition (5).

We now introduce an invertible function $g(T)$ related to the thermal conductivity through the relation

$$k(T) = \frac{\mathrm{d}g}{\mathrm{d}T} . \tag{7}$$

It is convenient to specify $g(T)$ explicitly as

$$g(T) = -\int_T^{\theta_0} k(T') \, \mathrm{d}T' \tag{8}$$

where θ_0 is a (presumably large) reference temperature beyond which the properties of the material are adversely and irreversibly altered, leading to damage of the sample. The assumption that $g(T)$ is invertible simply means that there exists a function $\theta(g)$ such that $T = \theta[g(T)]$. Insofar as $k(T)$ is a known function for any given material, the construction of suitable functions $g(T)$ and $\theta(g)$ imposes no difficulty. We observe that g, like k, defines an implicit function of r, i.e., $g = g[T(r)]$. The radial derivative of $g(r)$ can therefore be written

$$\frac{\mathrm{d}g}{\mathrm{d}r} = \frac{\mathrm{d}g}{\mathrm{d}T} \frac{\mathrm{d}T}{\mathrm{d}r} = k \frac{\mathrm{d}T}{\mathrm{d}r} \tag{9}$$

which allows us to rewrite Eq. (6)

$$\frac{dg}{dr} = -\frac{fr}{3}$$ (10)

in terms of the function $g(r)$. Solution of this equation gives the function

$$g(r) = g_R + \frac{f}{6}\left(R^2 - r^2\right),$$ (11)

where $g_R = g(T_R)$. This gives the steady-state temperature distribution

$$T(r) = \theta[g_R + \frac{f}{6}\left(R^2 - r^2\right)].$$ (12)

3. Conditions for Thermal Runaway and Discussion

In applying the analysis presented above to the phenomenon of thermal runaway, we argue that the characteristic signature of thermal runaway is a steady-state temperature distribution which at some points exceeds the characteristic temperature θ_0 at which the material is destroyed. At sufficiently low heating rates, the temperature at the surface will be less than this characteristic temperature and so, from (8), the function $g(T)$ will be negative when evaluated at the surface temperature $T_R = (fR/3\sigma)^{1/4} < \theta_0$. As we move in from the surface, the temperature increases and the argument of the decreasing function $\theta[g(r)]$ has the possibility of vanishing at some finite critical radius

$$r_{\rm c} = \sqrt{R^2 - \frac{6\,|g_R|}{f}},$$ (13)

provided that the second term in the radical is smaller in magnitude than the first. According to (8) such a vanishing of $g(r)$ corresponds to a point inside of which the temperature exceeds the critical temperature θ_0. Thus, we argue, thermal runaway occurs whenever

$$\sqrt{\frac{fR^2}{6\,|g_R|}} > 1.$$ (14)

In what follows we consider two mathematical forms for the thermal conductivity which illustrate the way in which thermal runaway can occur. The first example describes a hypothetical system in which the thermal conductivity decreases exponentially with temperature. With such a rapidly decreasing form, thermal runaway can occur no matter how large the critical temperature θ_0, provided that the sample size or microwave power is sufficiently large. It is worth noting that the incorporation of anharmonic effects in the Debye model leads to the prediction of an exponential decrease of κ with temperature over an *intermediate* temperature regime $\theta_D > T > 0$, where θ_D is the Debye temperature [13, 14]. While this provides some justification for consideration of this exponential model, at the elevated temperatures at which runaway and destruction of the sample occurs (for which $T > \theta_D$) the same model predicts a power law decrease with temperature of the form $k \sim T^{-1}$, and observations on many crystalline materials are

consistent with a high temperature decrease of the form[2]) $k \sim T^{-s}$, with $2 \geq s \geq 1$. Thus, in the second model discussed below we consider the situation in which the thermal conductivity decreases according to a power law. We find that for $s > 1$ thermal runaway can always occur for sufficiently high heating rates, as with the exponential form. However, the limiting case $s = 1$ associated with the commonly encountered high-temperature form $k(T) \sim T^{-1}$, has a qualitatively different behavior, in that thermal runaway occurs only for finite values of the critical temperature θ_0.

3.1 Exponentially decreasing thermal conductivity

We consider a hypothetical material in which the thermal conductivity is an exponentially decreasing function of the temperature, with $k(T)$ having the form

$$k(T) = k_0 \, e^{-T/T_0} \, . \tag{15}$$

We assume, for convenience, an infinite critical temperature $\theta_0 \to \infty$, so that thermal runaway in this system is characterized by an interior region which is at infinite temperature out to the critical radius. The function $g(T)$ corresponding to this thermal conductivity is given by the expression

$$g(T) = -k_0 T_0 \, e^{-T/T_0} = -T_0 k(T) \, . \tag{16}$$

which is easily inverted to yield

$$\theta(g) = T_0 \ln \left[\frac{k_0 T_0}{-g} \right] \, . \tag{17}$$

Using this result with Eq. (12) gives us the steady-state temperature distribution

$$T(r) = \theta \left[g_R + \frac{f}{6}(R^2 - r^2) \right] = T_0 \ln \frac{6 k_0 T_0}{6 \, |g_R| - f(R^2 - r^2)} \tag{18}$$

inside the sample, where

$$g_R = g[T(R)] = -k_0 T_0 \, \exp \left[-(fR/3\sigma T_0^4)^{1/4} \right] \tag{19}$$

is negative definite. According to this expression, thermal runaway will occur at all points inside a radius

$$r_c = \sqrt{R^2 - \frac{6 k_0 T_0 \, \exp \left[-(fR/3\sigma T_0^4)^{1/4} \right]}{f}} \tag{20}$$

whenever

$$fR^2 > 6 k_0 T_0 \, \exp \left[-(fR/3\sigma T_0^4)^{1/4} \right] \, . \tag{21}$$

This implicitly defines critical values for each of the parameters appearing in this expression when the remaining ones are held fixed. For example if we keep all quantities fixed except the power absorbed, the critical value f_c above which thermal runaway occurs is given as the root of the equation $f_c = \gamma \exp \left(-\beta f_c^{1/4} \right)$, where $\gamma = 6 k_0 T_0/R^2$, and

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[2]) In the Debye model the thermal conductivity in the intermediate regime has the form $f(T) \exp \left(\theta_D/T \right)$, where $f(T)$ is a polynomial function of temperature. See, e.g., the discussion in [14].
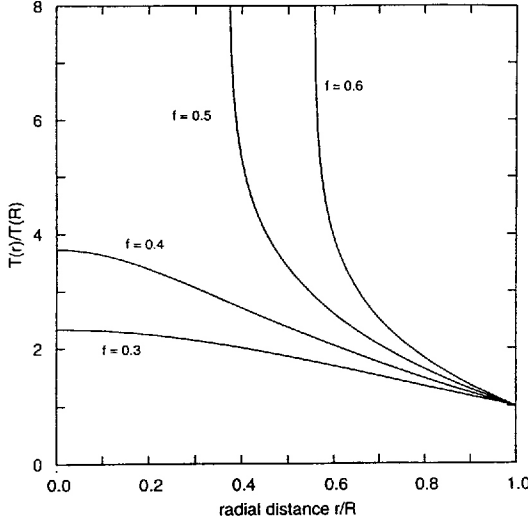
Fig. 1. Plot of the normalized steady-state thermal distribution $T(r)/T(R)$ as a function of normalized radial distance r/R for a ceramic with exponentially decreasing thermal conductivity. In these plots all parameters are kept fixed except the absorbed power f which is increased from values below the critical value f_c required for thermal runaway, to values above f_c

$\beta = (R/3\sigma T_0^4)^{1/4}$. Alternatively, as the prefactor k_0 of the thermal conductivity is varied, runaway will occur whenever k_0 drops below the value $k_c = (fR^2/6T_0)\exp[(fR/3\sigma T_0^4)^{1/4}]$.

We plot in Fig. 1 the normalized temperature distribution

$$T(r)/T(R) = -\frac{1}{\beta f^{1/4}}\ln\left[e^{-\beta f^{1/4}} - \frac{f}{\gamma}\left[1 - \left(\frac{r}{R}\right)^2\right]\right] \tag{22}$$

for four values of the absorbed power f showing the existence of thermal runaway for $f > f_c$ and its absence for $f < f_c$. We use arbitrary units and take $\beta = \gamma = 1$. The value of f_c for this situation is 0.4424.

3.2 Power law decrease of the thermal conductivity

We now consider the situation in which the thermal conductivity decreases with temperature as a power law,

$$k(T) = AT^{-s} = AT^{-\lambda-1}. \tag{23}$$

We first consider the situation with $s > 1$, and $\lambda = s - 1$. We can then write, again assuming an infinite critical temperature,

$$g(T) = -\frac{AT^{1-s}}{s-1} = -\frac{AT^{-\lambda}}{\lambda}. \tag{24}$$

Inverting this to find T as a function of g yields

$$\theta(g) = \left[\frac{A}{\lambda|g|}\right]^{1/\lambda}, \tag{25}$$

which implies the spatial profile

$$T(r) = \theta\left[g_R + \frac{f}{6}\left(R^2 - r^2\right)\right] = \left[\frac{6A/\lambda}{6\,|g_R| - f(R^2 - r^2)}\right]^{1/\lambda}. \tag{26}$$

Using the fact that $g_R = -(A/\lambda)T_R^{-\lambda}$ this becomes

$$T(r) = T_R \left[\frac{1}{1 - f\lambda T_R^\lambda (R^2 - r^2)/6A} \right]^{1/\lambda}. \tag{27}$$

From this expression it is clear that thermal runaway can again occur for $\lambda > 0$ or $s > 1$ for large enough samples or high enough power. When thermal runaway does occur, the critical radius r_c is given by the expression

$$r_c = \sqrt{R^2 - \frac{6A}{\lambda f T_R^\lambda}} = \sqrt{R^2 - \frac{6A}{\lambda f} \left(\frac{3\sigma}{fR} \right)^{\lambda/4}}. \tag{28}$$

which gives a positive radius whenever

$$\lambda f R^2 > 6A \left(\frac{3\sigma}{fR} \right)^{\lambda/4}. \tag{29}$$

This leads to the following critical values beyond which thermal runaway will occur:

$$f > f_c = [(6A/\lambda)^4 (3\sigma)^\lambda R^{-(8+\lambda)}]^{1/(4+\lambda)} \tag{30}$$

$$A < A_c = \frac{\lambda}{6} [f^{4+\lambda} R^{8+\lambda} (3\sigma)^{-\lambda}]^{1/4} \tag{31}$$

$$R > R_c = [(6A/\lambda)^4 f^{-(4+\lambda)} (3\sigma)^\lambda]^{1/(8+\lambda)} \tag{32}$$

The analysis presented above does not apply to the limiting case $s \to 1$ or $\lambda \to 0$, as can be seen from the fact that the critical parameters just derived all approach zero or infinity in this limit. Thus, we must study this case separately. To proceed, we assume that

$$k(T) = \frac{B}{T}, \tag{33}$$

and take a finite critical temperature θ_0, so that

$$g(T) = B \ln (T/\theta_0). \tag{34}$$

Inverting this to find T as a function of g yields

$$\theta(g) = \theta_0 \exp (g/B). \tag{35}$$

The spatial profile of the temperature distribution is then a Gaussian

$$T(r) = \theta \left[g_R + \frac{f}{6} (R^2 - r^2) \right] = \theta_0 e^{g_R/B} \exp \left[\frac{f}{6B} (R^2 - r^2) \right], \tag{36}$$

which can also be written

$$T(r) = T_R \exp \left[\frac{f}{6B} (R^2 - r^2) \right]. \tag{37}$$

We see that for this situation the temperature distribution remains finite everywhere inside the sample, so that thermal runaway in the strong sense observed earlier does not occur. In Fig. 2 we plot the normalized temperature profile for four different power laws
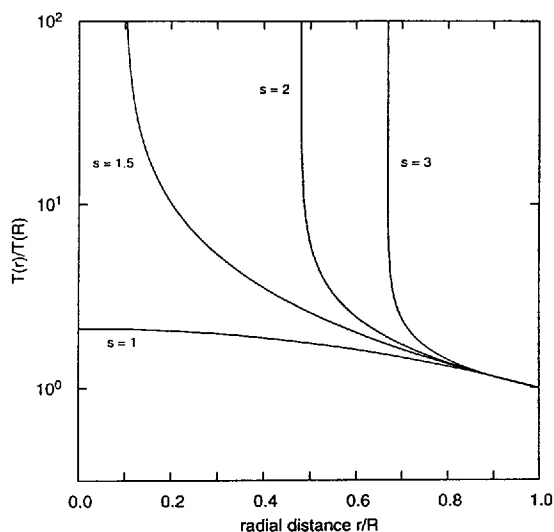
Fig. 2. Plot of the normalized steady-state thermal distribution $T(r)/T(R)$ as a function of normalized radial distance r/R for ceramic materials with power law decrease of the thermal conductivity, with powers s as labeled In all curves we have taken $f = 0.75$

for one fixed value of the power $f = 0.75$. In these plots we have taken $\beta = (R/3\sigma)^{1/4}/6A = 1$ for the power law curves with $s > 1$, and taken $B/R^2 = 1$ for the curve corresponding to $s = 1$, which for this value of f is the only one shown which does not achieve an infinite temperature at the center of the sample.

Even for $s = 1$, it is obviously possible to make the temperature in the sample exceed any finite but large value θ_0. Indeed, for any $\theta_0 > 0$ we find a critical radius

$$r_{\rm c} = \sqrt{R^2 - \frac{6\gamma \ln \left(fR/3\theta_0^4\sigma\right)}{4f}} \tag{38}$$

inside of which the temperature exceeds θ_0 whenever

$$4fR^2 > 6\gamma \ln \left(fR/3\theta_0^4\sigma\right). \tag{39}$$

Thus, the physically interesting case of a thermal conductivity which falls off at high temperatures with the inverse first power of T represents the most rapidly decaying falloff that can occur before thermal runaway of the strong type (i.e., thermal runaway with an infinite critical temperature) becomes possible. It may be argued that the dramatic effects associated with thermal runaway imply a critical temperature which is, effectively, infinite. If so, then our results can be taken as a "proof" that thermal runaway cannot occur in most materials as a result of a decrease with temperature of thermal conductivity *alone*. This finding, of course, lends support to the earlier analyses of Kenkre *et al.* who attributed the runaway phenomena to an effective temperature dependence of the absorption coefficient [6–12]. A few points are worth emphasizing, however. First, the dynamical analyses previously performed were insensitive to the boundary conditions of the sample. It seems plausible, in view of the analysis presented here, that an absorption coefficient that even mildly increases with temperature could push a system with $k(T) \sim T^{-1}$ into the regime where thermal runaway in the strong sense can occur. If so, then it would be expected that the critical parameters (of power, sample size, etc.) delineating the regimes where runaway does and does not occur would be

sensitive to the parameters which determine the variation of the thermal conductivity with temperature. In addition, the argument that this mechanism cannot, by itself, lead to thermal runaway, presumes that the functional *form* of the conductivity remains the same at extremely elevated temperatrues. This ignores the possibility that with increased degradation of the sample and the possible emergence of other chemical phases, changes in the thermal conductivity could indeed arise which would lead to a stronger dependence of the thermal conductivity with temperature than would otherwise be expected from conventional theories.

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