phys. stat. sol. (b) 172, 337 (1992)

Subject classification: 65.50 and 64.60; S10

Department of Physics and Center for Micro Engineered Ceramics, University of New Mexico, Albuquerque\(^{1)}\) (a) and Los Alamos National Laboratory\(^{2)}\) (b)

**Model for Anomalous Power Dependence of Microwave Heating in Ceramic Materials**

By

V. M. Kenkre (a), M. Kus\(^{3)}\) (a), and J. D. Katz (b)

A model is presented to explain the anomalous power dependence of microwave heating in ceramic materials. The anomaly considered consists of a saturation effect wherein the dependence of the heating rate on microwave power becomes sublinear. Our proposed explanation is based on the idea of spatial confinement of the absorbing charges in grain boundary regions. The mechanism of saturation, more generally, of a decrease in absorption efficiency, arises from a mismatch of two lengths, one of which measures the maximum distance an absorbing charge travels under the combined action of damping and of the oscillating microwave field, and the other measures the spatial extent of the confining region. Explicit calculations for a simple model are presented and the results are examined in the light of observations on ferric oxide.

Ein Modell zur Erklärung der anomalen Leistungsabhängigkeit der Mikrowellenaufheizung keramischer Materialien wird dargestellt. Die Anomalie besteht in einem Sättigungseffekt, wobei die Heizgeschwindigkeit sublinear von der Mikrowellenleistung abhängt. Unsere Erklärung basiert auf der räumlichen Beschränkung der absorbierenden Ladungen in den Korngrenzenbereichen. Die Sättigung oder, allgemeiner, die Abnahme der Absorption entsteht durch die Fehlanpassung zweier Längen; die eine mißt die maximale Entfernung, die eine Ladung unter der Wirkung von Dämpfung und oszillierendem Mikrowellenfeld zurücklegt, die andere mißt die Abmessungen des beschränkenden Gebietes. Explizite Rechnungen werden für ein einfaches Modell durchgeführt, die Ergebnisse werden anhand von Beobachtungen an Eisenoxyd überprüft.

**1. Introduction**

Reports by several investigators in the recent literature [1 to 3], that microwave heating can lower the sintering temperature in a number of materials by several hundreds of degrees and shorten the sintering time by several hours, have led to a lot of intense activity [4 to 9] in the study of the effects of microwave sintering on ceramics. It has been suggested that, in addition to increasing the heating efficiency by concentrating the heating process within the material rather than in the furnace in which the material is placed, microwaves might have basic consequences such as more efficient atomic diffusion within the material. An investigation of such basic features of the interaction of microwaves on ceramics has been undertaken, as a consequence, on both experimental [8] and theoretical [9] fronts. As a result of some of these efforts, a general picture [7, 9] has emerged. The underlying theoretical framework has been found to be successful in the explanation of a number of observations.

\(^{1)}\) Albuquerque, NM 87131, USA.

\(^{2)}\) Los Alamos, NM 87475, USA.

\(^{3)}\) On leave of absence from the Institute for Theoretical Physics, Polish Academy of Sciences, Warsaw, Poland.
including those concerning the phenomena of thermal runaway and dielectric response. Satisfactory description of dielectric loss observations at several different frequencies, as well as of thermal runaway measurements displaying a diversity of time–temperature curves, has been provided by the theory. The materials analyzed have included silica, zinc oxide, ferric oxide, alumina, and strontium titanate.

The theory is based on the idea of a nonlinear feedback whereby heating frees absorbing entities such as vacancies, bivacancies, or interstitials and the freeing of the absorbers results in enhanced absorption and consequently of enhanced heating. The resulting temperature–time equation is given by [7, 9]

$$\frac{dT}{dt} = [k_A + k_M f(T)] P - \sigma_1 T^4,$$

(1.1)

where $k_A$ and $k_M$ are related to the absorption coefficients of two kinds of species of absorbers present in the material and $f(T)$ denotes the fraction of the M-absorbers which are free, $P$ is the incident power, and $\sigma_1$ is proportional to the Stephan-Boltzmann constant. The peculiar runaway nature of the heating arises generally from the “switching” nature of $f(T)$, which, in its simplest form, may be written as

$$f(T) = \frac{e^{-A/k_BT}}{1 + e^{-A/k_BT}},$$

(1.2)

$A$ being an energy barrier and $k_B$ the Boltzmann constant. Although the nonlinear nature of (1.1) has a structure which is rich enough to describe the diversity of time–temperature shapes encountered in the various materials addressed [7], it predicts a heating rate whose dependence on the microwave power $P$ is linear: the heating rate, equivalently the power absorbed, depends quadratically on the electric field $E$ of the microwave field, this behaviour being indicative of standard Joule heating. Such a linear dependence of the heating rate on the incident microwave power is observed in some materials such as silica [1, 5]. However, a saturation effect, more generally a sublinear dependence of the rate on the microwave power, has been observed in other materials, typified by ferric oxide [5]. Neither (1.1), nor any other theoretical starting point in the literature known to us, is currently able to address this effect. The present paper is an effort in this direction.

The rest of the paper is set out as follows. In Section 2 we introduce the model. In Section 3 we provide the details of our calculations and obtain expressions for the dependence of the heating rate on the input microwave power. A discussion including a comparison to experiment constitutes Section 4.

2. The Basic Ideas behind the Model

The primary observational feature under investigation is the decrease in the efficiency of absorption with increase in microwave power, i.e. with increase in the applied electric field. Our model explanation is based on the following spatial confinement idea. When in a spatially unrestricted medium, charges absorb power from an applied oscillating electric field by following the field oscillations and dissipating the absorbed power through whatever damping agency is present. The instantaneous power absorbed is proportional to the square of the velocity of the charge, equivalently to the square of the applied electric field. The time average yields the average power absorbed. If, however, the charges are constrained to
move in a finite region of space, they may find themselves unable to follow the time
dependence of the applied field simply because they may reach the walls of the confining
region before the electric field has reversed its direction. If the encounter with the wall
results in velocity reversal, the charges will find themselves moving against the field. Complex
motion will ensue which will work against efficient absorption of power from the field. At
very small fields or in very large regions, this effect will not occur. However, for a given
extent of the confining region, if the input power is increased, the electric field will eventually
reach a magnitude consonant with a traversal distance which is larger than the confining
length. The consequence will be a sublinear dependence of the absorbed power on the
incident power.

We will denote the confining length by $L$. If the maximum magnitude of the applied
field and its frequency are $E$ and $\omega$, respectively, the maximum displacement of a
charge $q$ of mass $m$ in an unconfined region, calculated in the limit of large damping, is
$(2qE/m\gamma\omega)$, where $m\gamma$ is the damping rate. We will call this distance the traversal
distance $L_E$,

$$L_E = \frac{2qE}{m\gamma\omega}.$$  \hspace{1cm} (2.1)

It is clear that the crucial parameter is $L_E/L$, the ratio of the traversal distance to the
confining length. When it is small, the dependence of the heating rate on the input power
(which is proportional to $E^2$) is linear. When it is large, the dependence will turn out to be
sublinear.

What might the charged particles and the confining regions of our model correspond to?
In many ceramics, particularly those which are clearly ionic in character, atomic diffusion
is thought of as occurring in two parts: diffusion of the cations through the bulk, and the
diffusion of larger anions primarily through the grain boundary regions. If we focus attention
on the anions, and assume for simplicity that the grain boundary is made up of a number
of segments along which the anion motion takes place, we have one possible realization
of our model. In such a situation, the linear segments forming the boundary provide the
spatially constrained regions, the confining length $L$ being the length of a typical segment.
The anions move less easily from one segment to another than on a given segment. A
representative value of the confining length $L$ is a micron or a tenth of a micron. A different
realization of our model is provided by the space-charge accumulation region across a
grain boundary. It is well known [10, 11] that electrostatic interactions lead to the
accumulation of space charges on the two sides of a grain boundary and that these charges
give rise to potentials which are felt by charged interstitials or vacancies. The confining
length in this case can be much smaller, viz. of the order 200 nm. In both these realizations,
the nature of charge motion in the confined region will be diffusive and complex in general
but, for simplicity, will be assumed to be free except for damping and the action of the
electric field. The equation of motion is

$$\frac{dv}{dt} + \gamma v = e \sin \omega t,$$  \hspace{1cm} (2.2)

where $v$ is the velocity of the charge, and $e = qE/m$. In the next section we examine the
solutions of (2.2) with boundary conditions imposed by spatial constraints.
3. Dependence of the Heating Rate on the Input Power

On introducing the phase lag angle $\varphi$ through

$$\tan \varphi = \frac{\omega}{\gamma},$$

the solution of (2.1) is written in a straightforward manner as

$$v(t) = e^{-\gamma t} \left[ v(0) + \frac{\omega \epsilon}{\gamma^2 + \omega^2} \right] + \frac{\epsilon}{\sqrt{\gamma^2 + \omega^2}} \sin(\omega t - \varphi).$$

In an unconfined region, after the transients have died down, the charge will follow the time dependence of the applied electric field with phase lag $\varphi$, and the instantaneous power absorbed will be given by $m(\gamma^2 + \omega^2)^{-1/2} \epsilon^2 \sin(\omega t - \varphi) \sin(\omega t)$. The heating rate $R$, which is proportional to the time average of this quantity, is thus given by

$$R = \frac{1}{2} \frac{q^2}{m} \frac{\gamma}{\gamma^2 + \omega^2} E^2.$$

Since the incident power is proportional to $E^2$, the rate as given in (3.3) depends linearly on the incident power.

Let us now consider the case of a spatially restricted region. The charge is accelerated but hits a confining wall before the oscillating electric field has reversed sign. The charge bounces back, with its velocity reversed in sign. If the collision with the wall is elastic, the magnitude of the velocity is unchanged. The velocity now decreases in magnitude because the electric field acts against the motion. The charge may return to the wall one or more times before the electric field reverses and forces it to move towards the other restraining wall. Fig. 1 shows a situation when the charge hits the wall twice, travels towards the wall a

![Diagram](image-url)

**Fig. 1.** Time evolution of the velocity in a confined region in the general case of finite damping. Units are arbitrary. For the case shown, the charge travels towards the right confining wall thrice, but hits it twice, before returning to the other wall under the slaving action of the electric field.
third time but reverses direction before meeting the wall, and starts to move back towards the other wall. For our further calculations, we will consider the simplified case of very large damping, which is known to be valid in the area of microwave interactions with ceramics.

Under the assumption of very large damping, the solution given in (3.2) simplifies significantly: we may neglect the initial condition term because it decays very quickly, eliminate the phase lag angle in light of (3.1), and neglect \( \omega \) in comparison to \( \gamma \) in the last term of (3.2). The result is that the velocity is completely slaved by the electric field

\[
v(t) = \frac{qE}{m\gamma} \sin \omega t .
\]  

(3.4)

Equation (3.4) could be written down also by realizing that the assumption of infinite damping is equivalent to the complete neglect of the derivative term in (2.2).

Let us assume that the charge is at rest at one of the walls initially and that it begins to travel towards the other wall under the action of the electric field \( E \sin \omega t \). As discussed in Section 2, we must consider two different situations according to the relative value of the ratio \( L/L_E \), where \( L_E \) is given by (2.1).

If the distance which the charge travels in one half-period of the electric field, i.e., before the field changes direction, is smaller than the confining length \( L \), the charge does not feel the existence of the confining wall, and the entire motion is governed by (3.4). The energy gain of the charge during a half-period of the applied field is calculated as

\[
\Delta \mathcal{E} = \int_{0}^{\pi/\omega} dt \, v(t) \, qE \sin \omega t = \frac{1}{2} \frac{q^2}{m} \frac{\pi}{\gamma \omega} E^2 ; \quad \frac{L}{L_E} \geq 1 .
\]  

(3.5)

Dividing the energy gain by the time \( \pi/\omega \) during which it occurs, we obtain the average heating rate as

\[
R = \frac{1}{2} \frac{q^2}{m\gamma} E^2 ; \quad \frac{L}{L_E} \geq 1
\]  

(3.6)

which is, of course, the large damping limit of (3.3).

Equation (3.6) is, however, not valid if the charge reaches the confining wall before the direction of the field changes. If the encounter with the wall reverses the velocity of the charge, it finds itself moving against the field. High damping forces the charge to follow the field very quickly, i.e. to stop and reverse its direction again and again after bouncing back from the confining wall. For infinite damping, the charge stops immediately after hitting the wall, and stays at the wall until the field reverses direction, allowing further motion (now towards the other wall) under the slaving action of the field. This case is shown in Fig. 2a. The time for the first encounter with the wall, \( t_0 \), is calculated by inverting the relation

\[
L = \int_{0}^{t_0} dt \, v(t) = \frac{qE}{m\gamma \omega} \left( 1 - \cos \omega t_0 \right) .
\]  

(3.7)
Fig. 2. Time evolution of a) the velocity and b) the instantaneous power absorbed, for a charge in a confined region in the special case of infinite damping. Units are arbitrary. The reduction in the average heating rate is clear from b).

The result is

$$t_0 = \frac{1}{\omega} \cos^{-1} \left(1 - \frac{2L}{L_E}\right),$$

where we have used (2.1). The counterpart of (3.5) for the energy gain in one half-period now involves an integration not over a time interval of $\pi/\omega$, but only over $t_0$,

$$\Delta\varepsilon' = \frac{1}{2} \frac{q^2}{m} \frac{E^2}{\gamma \omega} \left[ \cos^{-1} \left(1 - \frac{2L}{L_E}\right) - 2 \left(1 - \frac{2L}{L_E}\right) \sqrt{\left(1 - \frac{L}{L_E}\right) \frac{L}{L_E}} \right]; \frac{L}{L_E} \leq 1. \tag{3.9}$$

The average heating rate, on the other hand, is obtained by dividing the right-hand side of (3.9) by the entire half-period $\pi/\omega$. Since the charge remains at rest for a time interval equal to the difference of $\pi/\omega$ and $t_0$, the heating rate is reduced in comparison with (3.6),

$$R = \frac{1}{2} \frac{q^2}{m \gamma \pi} E^2 \left[ \cos^{-1} \left(1 - \frac{2L}{L_E}\right) - 2 \left(1 - \frac{2L}{L_E}\right) \sqrt{\left(1 - \frac{L}{L_E}\right) \frac{L}{L_E}} \right]; \frac{L}{L_E} \leq 1. \tag{3.10}$$
The rate depends on the electric field not only through the $E^2$ term as in (3.6) but also through the dependence of $L_E$ on the electric field as given by (2.1). The overall effect is to lower the heating efficiency, as discussed in Section 2. Fig. 2b makes this clear.

Equation (3.6) and (3.10) for the heating rate together constitute the central result of this paper. For a given amount of incident power, the heating rate is independent of the confining length $L$ if the latter exceeds $L_E$, but decreases with $L$ if it is smaller than $L_E$. This is shown in Fig. 3a. For a given confining length, the heating rate is linear in the input power if the latter is sufficiently small but becomes sublinear for larger values. This is shown in Fig. 3b. It is difficult to arrive at the value of the damping constant $\gamma$. However, typical estimates show that confining lengths of a tenth of a micron could correspond to behavior observed in materials such as ferric oxide.

We mention in passing, that in the realistic case of finite damping, the energy gained or lost during the repeated bouncing against a wall as depicted in Fig. 1, has little influence on the value of the heating rate. Gains and losses of energy during the bouncing cancel each other exactly for the case of a constant driving force as in the familiar problem of a particle falling under the influence of gravity. For a slowly varying field the cancellation is approximate but results in a negligible net loss of energy.

Fig. 3. a) Heating rate as a function of $1/L$, where $L$ is a measure of the spatial extent of the confining region, showing standard behavior (lack of dependence of the rate on $L$) when $L$ exceeds a characteristic value, and novel predicted behavior (decrease of the rate with decrease of $L$) when $L$ is less than that value. The characteristic value of $L$ is seen to increase with the microwave power, i.e. with the applied electric field. Here and in Fig. 3b, units are arbitrary and various combinations of quantities appearing in the heating rate expressions ((3.6) and (3.10)) have been put equal to unity for simplicity. b) Heating rate as a function of the incident microwave power, for several values of the confining length $L$. The limit of infinite $L$ is represented by the standard linear dependence and is approached by all finite-$L$ curves for appropriately small values of the incident power.
4. Comparison to Experiment and Discussion

Equation (1.1), which has provided a successful description of the temperature–time evolution in a large number of materials, predicts the following relation between the input microwave power \( P \) and \( T_s \), the value at which the temperature saturates as a result of a trade-off between the heat input and heat losses,

\[
P = \frac{\sigma_s T_s^4}{k_A + k_M f(T_s)}.
\]  

(4.1)

An inspection of the heating curves for some materials such as ferric oxide shows that (4.1) is obeyed rather poorly. The theory developed in the present paper replaces (4.1) by

\[
g(P) = \frac{\sigma_s T_s^4}{k_A + k_M f(T_s)}.
\]  

(4.2)

where the function \( g(P) \) is given by

\[
g(P) = P; \quad \frac{L}{L_E} \geq 1,
\]

\[
g(P) = \frac{P}{\pi} \left[ \cos^{-1}\left(1 - \frac{2L}{L_E}\right) - 2 \left(1 - \frac{2L}{L_E}\right) \sqrt{\left(1 - \frac{L}{L_E}\right) \frac{L}{L_E}} \right]; \quad \frac{L}{L_E} \leq 1.
\]  

(4.3)

In (4.3), \( L_E \) is a function of \( P \) in keeping with (2.1).

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Fig. 4. Comparison of the result for no spatial confinement (represented by (4.1)) and that for spatial confinement (represented by (4.2)) to data deduced from experiments on ferric oxide by McGill et al. [5]. Shown is the dependence of the saturation temperature on the incident microwave power. Values of the saturation temperature were estimated from the time–temperature curves reported in [5]. Error bars shown are the result of the estimating procedure.
In Fig. 4 we plot the saturation temperature $T_s$ as a function of input microwave power with and without the effect described in the present paper. The dashed line represents the inversion of (4.1) while the solid represents the inversion of (4.2) with (4.3). While it is difficult to deduce the saturation temperatures from the reported temperature-time curves for ferric oxide unambiguously, we have estimated them by inspection. The estimated values, along with a probable error of $\pm 50$ K in the saturation temperature, are depicted in Fig. 4. We see that the solid line, which represents the theory developed in the present paper, provides a much better description of the data than the dashed line does. The value of the confining length which corresponds to the solid curve is of the order of a tenth of a micron, and thus appears to be compatible with the length of a grain boundary segment. However, our estimating procedure is rather rough and there are too many uncertainties in the values of the quantities involved. That the observed increase of the saturation temperature (with an increase in the microwave power) is more slow than that predicted by the standard linear formula (4.1) is certain. That the data lie closer to the prediction of the theory presented in the present paper than to the prediction of the standard formula is also certain. However, the values of the quantities involved are not known with certainty. At the present stage of the investigation, we can only suggest that the spatial confinement mechanism we have described could be a source of the observed saturation of the heating rate in ferric oxide. A more definite conclusion must await further quantitative studies.

We mention in passing an allied consequence of our analysis. Our results (3.6), (3.10) show that the heating rate displays interesting behavior not only in its field dependence but also in its frequency dependence. In light of (2.1) we may write

$$R = R(\omega); \quad \omega \geq \omega_1,$$

$$R = R(\omega) \frac{1}{\pi} \left[ \cos^{-1} \left( \frac{1}{\omega_1} - 2 \left( \frac{1}{\omega_1} - \frac{2\omega}{\omega_1} \right) \sqrt{\left( \frac{1}{\omega_1} - \frac{\omega}{\omega_1} \right) \frac{\omega}{\omega_1}} \right) \right]; \quad \omega \leq \omega_1.$$  

(4.4)

We see that the heating rate is independent of frequency and has the value $R(\infty) = q^2 E^2/2mr_f$ at frequencies higher than the critical frequency $\omega_1$ (this behavior being characteristic of the infinite damping limit considered), but that it tends to zero when the frequency drops below $\omega_1$. The critical frequency is given by $\omega_1 = 2qE/Lm_f$. Fig. 5 depicts the frequency variation of the heating rate given by (4.4) along with the corresponding variation when the confinement is produced not by walls spaced $L$ apart but by a harmonic potential of frequency $\omega_0$. A smooth variation of the heating rate is obtained in this latter case,

$$R = R(\omega) \frac{\omega^2}{\omega^2 + \Gamma^2},$$

(4.5)

the characteristic frequency $\Gamma$ being identical to the Debye relaxation frequency $\omega_0^2/\gamma$. We note the curious fact that, in the case of the harmonic oscillator confinement, the sublinear dependence of the heating rate on the electric field does not occur. The reason is that the confining length in this case is itself (accidentally) proportional to the electric field magnitude. The ratio of the confining length to the traversal distance $L_E$ turns out, therefore, to be independent of the field. No interesting dependence of the absorption on the field appears.

Scarcity of available data on frequency variation of observables in ceramic materials in the microwave range has prevented us from comparing our result (4.4) with ex-
Fig. 5. Frequency dependence of the heating rate normalized to its value at infinite frequency in the case of confinement between walls as well as in the case of confinement by a harmonic oscillator potential. The abscissa is the ratio of the frequency to a characteristic frequency. The characteristic frequency is $\omega_1 = 2qF/Lm_r$ in the wall case and the Debye frequency $\Gamma = \omega_D/\gamma$ in the harmonic case.

experiment. We hope, however, that the relevant observations will be made in the future. They could serve as an additional check on the spatial confinement idea presented in this paper.

The model we have presented is highly simplified and the calculation is nearly trivial. There are a number of important issues we have not touched upon which could complicate the analysis of the model. However, our aim has been to arrive at the essence of anomalous power dependence of microwave heating in as simple a manner as possible. The following are some of the questions that we are in the process of analyzing. The assumption that the moving charges are completely confined to segments of the grain boundary obviously entails an oversimplification. One of the ways to avoid this assumption is to allow for a leakage of the charges from segment to segment at a rate which could depend on a number of factors including the angle between the segments, and eventually make the model continuous along the boundary. Another assumption we have made is that the nature of the motion in the confining region is free. This is consonant with the result \cite{7, 9} that the absorption by freely moving charges is considerably greater than that by bound charges. Nevertheless, it is of interest to work out the confinement features under the assumption that the motion is diffusive rather than free. We are in the process of carrying out such an analysis. We are also conducting a detailed investigation of the applicability of what we called in Section 2 the second realization of the spatial confinement model, viz. of the idea that the confining region is the space-charge accumulation region across the grain boundary.

**Acknowledgements**

It is pleasure for us to thank D. H. Dunlap and D. Sheltraw for helpful conversations.
References


(Received January 2, 1992)