Sintering of Ceramics with Microwave Radiation

V I Nefedov[1], P A Luchnikov[1], N A Trefilov[1] and K A Selenya[2]

[1]Moscow Technological University (MIREA), 119454, Vernadsky 78, Moscow, Russia

[2]Scientific Center of Applied Electrodynamics, 190103, Riga Avenue, 26, St. Petersburg, Russia

E-mail: xamdex@gmail.com

Abstract. The method of ceramics sintering in the EM-field of UHF band radiation (microwave heating) has been investigated. The paper presents analytical expressions to estimate temperature distribution in the material treated in normalized heating time and pulse duration with Bouguer and Fourier criteria applied. An installation for drying and sintering ceramic samples with microwave radiation (microwave-heating chamber of 30 dm^3 and a continuous wave magnetron of 3 kW) has been described.

1. Introduction

Microwave heating of the materials treated is carried out through the EM-wave energy absorption in the substance. Like radiation heating microwave heating is characterized by electromagnetic waves penetrating the material treated and providing its heating due to molecular friction of dielectric losses [1–4]. This method is highly distinguished from convective heating or heat-transferring ones [5-7]. Dielectrics, such as fluorine-polymers [8], have extremely small dielectric losses and are not actually heated as compared with organic semiconductors [9].

Microwave heating of materials has high efficiency in the technologies of microwave synthesis, continuous rubber vulcanization, ceramics and nano-ceramics sintering. The resulting effect of this leads to a considerable increase of chemical reaction rate as compared with the conventional ways of heating [10], thus providing a tenfold decrease of the technological process time [11].

2. Microwave heating model

The simplest microwave heating model is the plain-wave power absorption by semi-infinite dielectric lossy medium. In this case the volume density of heating power can be specified by the following:

$$D = \frac{1}{2}E_0^2 \exp(-2k_0 z\sqrt{\varepsilon\mu\sqrt{1+tg^2\delta}}\,\sin^2\frac{\delta}{2}),$$

where: E_0 – is the electric field intensity amplitude on the medium surface; z – is distance from the medium boundaries; k_0 – stands for the wave number for free space; ε,μ – are relative dielectric and magnetic permeabilities of the medium; $tg\delta$ – is the tangent of the medium dielectric losses' angle.

Fig. 1 represents the graphs of heating volume density P/P_{max} depending on the normalized coordinate z/z_0 at various $tg\delta$ values of the medium.

It is evident from the graph that microwave heating takes place unevenly in medium depth and the magnitude of the non-uniformity increases as the medium thickness and dielectric losses angle tangent increase. If the uniform temperature field in a stationary mode is required, one should determine the heating timeline taking into consideration the sample thickness. To a first approximation, the average complex relative dielectric permeability ε_m of the units threated under the sparse packing is determined by the Lichtenecker equation

$$\varepsilon_m = \varepsilon_{ou}^{\eta},$$

where: ε_{ou} – is the complex relative permeability of the treated units; η – is relative portion of the microwave-heating chamber volume taken up by the material being treated. When using sparse packing the average dielectric losses angle tangent decreases.
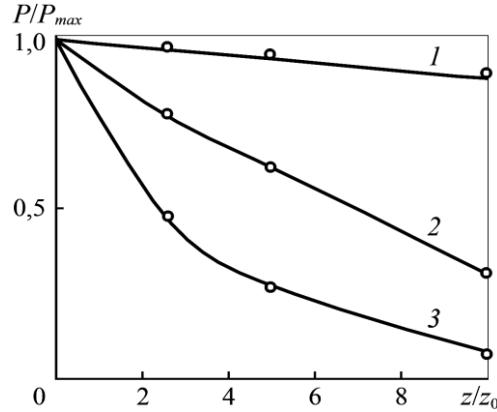
Figure 1. Heating relative volume density dependence P/P_{max} of z/z_0 normalized coordinate with medium $tg\delta$ values: 1 – 0,001; 2 – 0,1; 3 – 1,0.

The electromagnetic field power density carried by the wave inside the medium decreases as it travels away from the boundary and is described by the following ratio:

$$\frac{P}{S} = \frac{E_0^2}{2|W_c|}\exp(-2\alpha z)\cos\frac{\delta}{2},$$

where W_c – is the medium wave impedance.

The temperature distribution in the depth of the medium on microwave heating in the linear approximation satisfies the equation of thermal conductivity at radiation absorption according to the Beer–Lambert–Bouguerlaw which reads [12]:

$$\frac{\partial\vartheta}{\partial t} = a\frac{\partial^2\vartheta}{\partial z^2} + \Im\frac{2\alpha}{c\gamma}\exp(-2\alpha z),$$

with the second-type boundary conditions

$$\frac{\partial\vartheta}{\partial z}(0,t) = \frac{\partial\vartheta}{\partial z}(l,t) = 0$$

where: ϑ – is the excessive temperature; $\Im$ – stands for the irradiance within the medium; α – is the medium thermometric conductivity factor; c – is specific heat capacity of the medium; γ – is the specific weight of the medium; l – stands for the thickness of the heated layer.

$$\vartheta(z,t) = T^0(z,t) - T_0^0, \quad \Im = \frac{P}{S}.$$

The solution to the equation depends on the irradiance time function type. At microwave heating, rectangular functions are commonly used, and for them the solution to the time interval $t < 1$ and $t > 1$ equation has the following form [12]:

$$\vartheta(z, t\le 1) = t + \frac{2}{Fo[1-\exp(-Bu)]}\times\sum_{n=1}^{\infty}\frac{\left[1-(-1^n)\exp(-Bu)\right]}{\pi^2 n^2\left[1+\frac{\pi^2 n^2}{Bu^2}\right]}\left[1-\exp(-\pi^2 n^2 Fot)\right]\cos(\pi nz),$$

$$\vartheta(z, t\ge 1) = 1 + \frac{2}{Fo[1-\exp(-Bu)]}\times$$
$$\sum_{n=1}^{\infty}\frac{\left[1-(-1^n)\exp(-Bu)\right]}{\left[1+\frac{\pi^2 n^2}{Bu^2}\right]}\left\{\exp\left[-\pi^2 n^2 Fo(t-1)\right] - \exp(-\pi^2 n^2 Fot)\right\}\cos(\pi nz),$$

Here we use the normalized time t/t^* to the heating pulse duration t^* as well as Bouguer (Bu) and Fourier (Fo) criteria.

$$Bu = 2\alpha l, \quad F_0 = \frac{at}{l^2}.$$

Table 1 gives the calculated dependencies $\vartheta(z/l)$ for the time point $t = 1$, and Table 2 lists the surface heated temperature dependencies $\vartheta(z = 0, t)/\vartheta_{av}$ for different time points after heating completion in case of one-sided heating.

Table 1 - The temperature changes in the medium after microwave heating completion ($Fo = 10^{-2}$, $t = 1$).

	z/l	0	0,3	0,7	1,0
	10^{-2}	1,004	1,002	0,998	0,996
Bu	10^{-1}	1,043	1,02	0,9799	0,958
	$5*10^{-1}$	1,224	1,095	0,8967	0,8007
	∞	11,28	0,1725	<0,001	<0,001

Table 2 - Surface temperature change after microwave heating completion ($Fo = 10^{-2}$).

	$t/t*$	1	2	3	4	6	8	10
	10^{-2}	1,004	1,004	1,003	1,003	1,002	1,002	1,002
Bu	10^{-1}	1,043	1,037	1,032	1,029	1,024	1,019	1,016
	$5*10^{-1}$	1,224	1,188	1,165	1,147	1,119	1,097	1,079

t – Time, t* – Time of sample heating

The data given in Table 1 and Table 2 are obtained for a linear model with the values of Fourier and Bouguer criteria typical for ceramics. One should account for the real materials having non-linear thermophysical properties, thus the thermal conductivity factor can vary in magnitude during the heating process [13]. Moreover, the calculated dependencies in Table 1 and Table 2 do not take into account chemical transformations and palasticizers and moisture gasification.

In order to compare the microwave heating and the convection one the last line of Table 1 shows the temperature change in the medium when applying heat to the surface only with the heating time being the same.

It should be emphasized that from the electrodynamics standpoint, determining field structures in microwave heating chambers appears to be difficult for a variety of reasons [1–4]:

– the size of heating chambers is substantially more than the wavelength of the electromagnetic oscillation, which implies that numerous coherent eigenmodes with unknown amplitudes exist in the chamber volume;

– the spectrum of the MW-heating chamber eigenmodes depends on the way the material treated fills the chamber. The material characteristics (its geometric form, its layout inside the chamber, complex relative dielectric permeability, etc.) are mainly defined by accidental factors and may alter during the treatment.

3. Experimental research

When considering simplified models of the microwave-heating chambers it is reasonable to use approximate calculation methods.

So as to define fields in the closed-type chambers one usually applies the chamber model of a cavity resonator. Provided walls of the resonator have a simple form, coincide with the coordinate surface and have infinite conductivity, the electromagnetic field in the cavity at the moment of resonance is the solution of the Helmholtz equation, satisfies simple boundary conditions and is therefore determinable. Possible field types in the heating chamber represent the eigenfunctions of the corresponding electrodynamic problem and are known for the parallelepiped and cylinder chambers. Therefore, the full-field in heating chambers of the forms mentioned above can be presented as a series expansion by the eigenfunctions. The expansion coefficients depend on the type and location of the exciter in the heating chamber.

It is supposed that if even distribution of the microwave heating power is achieved in an empty chamber, it will remain when the chamber is filled with the material treated. Such analysis approach is an eigenfunction method.

A lab installation has been designed for drying and sintering ceramics with microwave radiation. The installation comprises the microwave heating chamber of 30 dm^3 volume, which is excited by a continuous wave magnetron with maximum output oscillation power of 3 kW.

The material treated was set on applicators — thin ceramic tube segments.. Chamber volume filling coefficient is equal to ~ 0.1…0.2. To control the technological process a witness sample disk made of the material treated was placed inside the chamber. A tungsten-rhenium thermocouple was embedded in the sample with its outputs screened from the microwave field exposure. A radio-wave phasing tester of the factor of the probing EM-wave transmitting through the thicknessof the witness sample was used as an additional control system for the material treatment process. For this the sample was placed in the gap of the dielectric wave-guide, created with the ceramic tubes embedded in the measuring radio-wave channel. The measurements were carried out at the microwave field wavelength of 3 cm. Microwave heating and sintering of small-sized ceramic units have been demonstrated to be promising.

Acknowledgments

4. References

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