Computational Study of Microwave Processing of Zirconia: Effects of Frequency and Temperature-Dependent Material Parameters

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Abstract – The ultimate goal of the reported study is to predict the heating behavior of materials in a microwave field, in order to optimize the microwave power consumption, uniformity of heating, and heating rate. This study focused on modeling *only the microwave portion* of a high temperature microwave assist technology kiln, used for laboratory experiments at Ceralink Inc. An accurate and adequate computer model of this furnace was developed for precise simulations of microwave heating of cylindrical samples of zirconia. Detailed data on temperature-dependent electromagnetic and thermal material parameters (obtained both experimentally and from literature) of zirconia and alumina-based insulation is presented as a key element of the computer model. It is shown that these characteristics are responsible for the efficiency of microwave heating in the considered furnace, the level of non-uniformity of the temperature field, and occurrence of thermal runaway. It is demonstrated that, at initial temperatures, the system is highly resonant, so energy efficiency of microwave processing strongly depends on frequency. Computed time-temperature histories show that if the furnace operates at a resonance closest to 2.45 GHz, the rate of heating can be accelerated by up to 15-25 times and can lead to a temperature field with a hot spot strongly localized in the interior of the zirconia sample.

Introduction

In recent years, the increase in energy costs have lead more companies to explore microwave processing of materials as a fast, efficient method for heating processes. This in turn has lead to the need for powerful, high temperature microwave-modeling capabilities. The growing use of computers and computational techniques has stimulated a rising interest in computer modeling of processes and systems of microwave power engineering [1]. However, based on the literature, the significant benefits provided by the modern advanced numerical techniques of modeling and optimization have been demonstrated primarily in food related applications [2, 3]. In the field of materials science, the known attempts of multiphysics modeling on a macroscopic level [4-8] appear to be based on severe idealizations and do not account for the full picture of microwave interactions with materials as a complex multiphysics phenomenon.

This paper presents an innovative advanced electromagnetic-thermal model for simulating the process of high temperature heating of zirconia samples in a microwave furnace. The model is based on the laboratory 2.45 GHz system employed by Ceralink, Inc. for microwave thermal processing of a variety of ceramic and composite materials. Ceralink would use this model as a process development tool, in order to predict how materials will behavior and optimize the process parameters before an actual experiment is conducted. This could help to decrease process development time, especially for material of different composition or size/configurations.

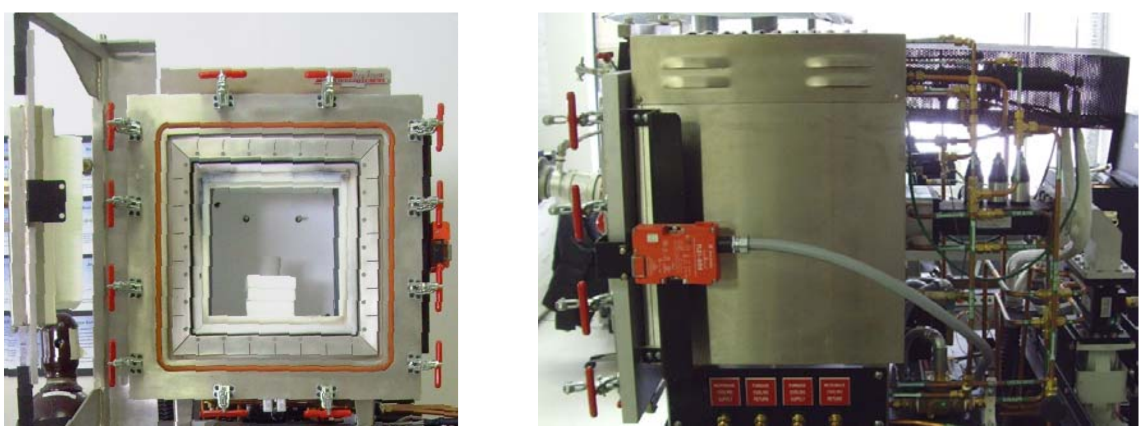
There is no method for in-situ monitoring of electromagnetic and thermal processes in the furnace in order to understand interaction of microwaves and materials at high temperature (up to

1600 °C and above). These circumstances provide strong motivation for development of adequate and accurate computer models of the microwave systems that would allow one to study major characteristics of high temperature processing prior to (or in parallel with) the experiments and thus reduce the resources required for purely experimental development of new processes.

Here we report some results in computer simulation of electromagnetic and thermal processes associated with microwave heating of cylindrical zirconia samples in the considered furnace. The model accurately takes into account internal temperature evolution of material parameters that follows the intrinsically non-uniform microwave heating. The presented results show that the heating rates and temperature patterns strongly depend on whether the system is fed by a magnetron operating at a resonance nearest to 2.45 GHz. In any case, a slightly increasing temperature characteristic of zirconia's loss factor causes the development of thermal runaway, and the time of this occurrence is predicted by the model.

Microwave System

The considered microwave furnace (Fig. 1) consists of a cubic metal cavity with edge length 406 mm. The cavity is filled with insulation material (Al-25/1700, by Zircar Ceramics, Inc.) with a nominal composition of 80 wt% alumina (Al_2O_3) and 20 wt% silica (SiO_2) – the 51 mm layers of this material are adjacent to all walls of the cavity. The sample of the processed material (zirconia with 3 mol% yttria) is centrally located on the top of a cubic block (made also of the insulation material) with edge length 152 mm centrally placed on the bottom insulation layer. The system is fed by a WR340 rectangular waveguide which connects the cavity with a microwave generator. We report here on a series of computational experiments considering a cylindrical zirconia compact with the diameter *D* and height *H* both varying from 38 to 62 mm.



(a) (b)

Fig. 1. Ceralink's microwave furnace for high temperature processing of materials: front view with an open door, demonstrating the Y-Z plane of the model (a) and side views showing the waveguide feed on the back of the furnace, shown to the right, demonstrating the X-Z plane of the model (b).

Computer Model

The microwave furnace in question has been represented in a 3D fully parameterized model developed using the 3D conformal FDTD software package *QuickWave-3D* (*QW-3D*) [9]. The mesh discretizing the scenario has been optimized in the framework of a special sensitivity analysis and set in accordance with the maximum values of dielectric constant of both materials in the entire temperature range. Depending on the size of the zirconia cylinder, the model consists of 730,000 to 806,000 cells, with the minimum cell size 2.9 mm.

When solving for temperature in the framework of an iterative procedure implementing an FDTD solution of the coupled problem [2] and using the *QW-3D*'s *Basic Heating Module* (*BHM*) [9], on the first iteration, a converged electromagnetic solution is supposed to be reached after 650 periods (nearly 55,000 FDTD time steps); at each subsequent iteration, 300 periods (about 25,500 steps) are involved. Heating pattern construction is carried out during 3 periods (nearly 250 steps).

Computations were performed using an OMP version of the multithread implementation of the *QW-3D* simulator. It was run on a Dell T4700 workstation with 16 GB of RAM and two quad-core Intel Xeon 3.2 GHz processors. Computations were accelerated by an Acceleware card A30 (NVIDIA Quadro FX 5600) implementing GPU technology in the form of an integrated FDTD hardware accelerator. On this machine, depending on the size of the model, the first and all subsequent iterations of the coupled solution take 13-14 and 6-6.5 min, respectively.

In the framework of electromagnetic-thermal iterations, computation of temperature was performed with a variable heating time step to accommodate highly non-linear profiles of time-temperature history of the considered process. The results presented below are obtained for adiabatic thermal boundary conditions on all air-dielectric interfaces.

Material Parameters

Prior to modeling, the thermal parameters of the insulation and the processed zirconia were carefully specified (using both our own measurements and data from literature) as temperature-dependent characteristics in a wide temperature range. Density ρ of the ceramic insulation material at room temperature was provided by the manufacturer. Since thermal expansion causes a decrease in density as a material heats and expands and due to fixed dimensions of the insulation used in the model, its mass slightly decreases as the temperature increases. The density as a function of temperature was calculated in accordance with the law of thermal expansion.

Preparation of zirconia for sintering involves pressing powder in compacts, and these compacts often range from 30-60% of the theoretical density. During sintering, the compact shrinks, and density increases, eventually approaching the theoretical density which, for 3 % yttria zirconia, is ~6 g/cm^3. The shrinkage in zirconia typically occurs above 1100°C, as the particles begin to sinter. Below 1100°C, the density decreases approximately linearly due to thermal expansion. Using a thermal expansion of 10.3 x 10^{-6} /°C and an initial density of 2.85 g/cc (based upon measured typical green densities of zirconia), the values of ρ were calculated up to 1100°C using the same technique as for the insulation. This approach is not valid above that temperature, as the zirconia begins to densify. It has therefore been decided to limit the computational study in this work to the temperatures below 1100°C.

When determining specific heat *c*, a rule of mixtures was applied to determine the resultant heat capacity of the ceramic insulation, based on the calculated phase content of 37 mol % mullite ($3Al_2O_3{\cdot}2SiO_2$) and 63 mol % alumina (Al_2O_3); the original data were taken from [10-13]. An approximation for *c* of zirconia was obtained by combining data from a variety of

sources and matching them with those which agree with c of 3 % yttria stabilized zirconia (YSZ) obtained in [14].
Thermal conductivity k of the insulation was provided by the manufacturer at 400, 800, 1100, and 1650°C and determined by interpolation for other temperatures. For zirconia, thermalconductivity was calculated (using data for 3 % YSZ zirconia [14] and the green property data measured on alumina, tin oxide, and zinc oxide) as $k = \alpha\rho c$, where α is the thermal diffusivity.

Dielectric properties of the insulation and the zirconia sample (dielectric constant ε' and the loss factor ε'') were measured using the TM_{010} cavity perturbation method based on the principles outlined in [15]. Measurements were taken at approximately 50°C intervals from room temperature to 1400°C.

In term of composition, zirconia with 3 mol % yttria is a mixture of monoclinic and stabilized tetragonal phases. A phase change taking place in the zirconia between 1100 and 1200 °C is associated with transformation of all of the monoclinic ZrO_2 to tetragonal ZrO_2. This causes a non-monotonic behavior of the measured complex permittivity in this temperature interval and requires a high precision in measurement. On the other hand, it is known from corresponding experiments that at the temperatures near the phase change and above, zirconia undergoes a notable densification (shrinkage), so application of an electromagnetic-thermal model in the conditions of changing geometry may be inadequate. This gives us another motivation for limiting this computational study to temperatures below 1100°C.

The resulting sets of electromagnetic and thermal material parameters of the zirconia compact and the alumina-based insulations are given in Tables 1 and 2 (where the values of electric conductivity σ appear as calculated from corresponding data for ε'' at 2.45 GHz).

Table 1. Electromagnetic (at 2.45 GHz) and Thermal Parameters of Zirconia

Temp. °C	ε'	σ S/m	c J/(g*C)	ρ g/cm^3	k W/(cm*C)
25	6.69	0.0258	0.217	2.848	0.00198
69	5.86	0.0045	0.324	2.844	0.00290
100	5.78	0.0033	0.363	2.841	0.00320
139	5.75	0.0029	0.398	2.838	0.00344
181	5.77	0.0036	0.426	2.834	0.00362
228	5.82	0.0043	0.450	2.830	0.00373
276	5.90	0.0050	0.470	2.826	0.00381
324	5.98	0.0058	0.487	2.821	0.00385
371	6.08	0.0078	0.501	2.817	0.00381
420	6.18	0.0121	0.514	2.813	0.00391
471	6.32	0.0185	0.526	2.809	0.00399
523	6.47	0.0288	0.537	2.804	0.00407
574	6.60	0.0442	0.547	2.800	0.00414
636	6.77	0.0664	0.558	2.794	0.00405
698	6.97	0.0975	0.568	2.789	0.00412
752	7.22	0.1416	0.575	2.785	0.00417
809	7.53	0.2003	0.583	2.780	0.00421
865	7.93	0.2786	0.590	2.775	0.00426
921	8.53	0.4083	0.597	2.770	0.00430
973	9.44	0.5942	0.603	2.766	0.00433
1019	10.46	0.8220	0.607	2.762	0.00436
1065	12.46	1.2190	0.612	2.758	0.00439
1100	14.77	1.6661	0.615	2.755	0.00441

Table 2. Electromagnetic (at 2.45 GHz) and Thermal Parameters of Zircar

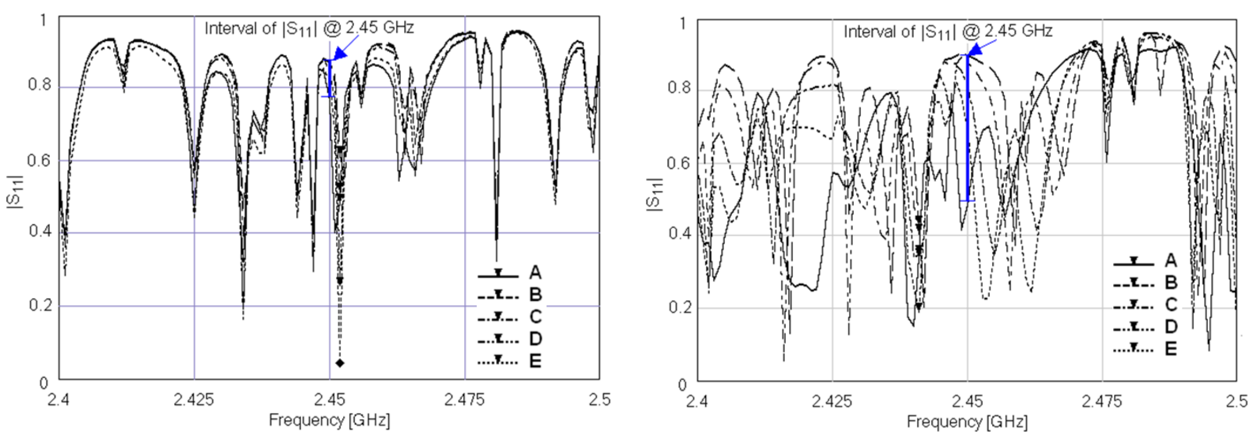
Temp. °C	ε'	σ S/m	c J/(g*C)	ρ g/cm^3	k W/(cm*C)
25	1.520	0.00005	0.764	0.4400	0.000631
100	1.520	0.00007	0.950	0.4392	0.000725
200	1.517	0.00015	1.042	0.4382	0.00085
300	1.513	0.00035	1.097	0.4371	0.000975
400	1.523	0.00062	1.135	0.4361	0.0011
500	1.540	0.00081	1.165	0.4350	0.001225
600	1.563	0.00091	1.190	0.4340	0.00135
700	1.573	0.00113	1.210	0.4329	0.001475
809	1.584	0.00131	1.230	0.4318	0.0016
900	1.593	0.00159	1.244	0.4309	0.0018
1000	1.600	0.00234	1.258	0.4299	0.0020
1100	1.608	0.00315	1.271	0.4288	0.0022

Table 3. Temperatures of Materials in Computation of Reflections in Fig. 2

Test	Temperature in zirconia (°C)	Temperature in insulation (°C)
A	25	25
B	140	25
C	250	25
D	370	25
E	420	100

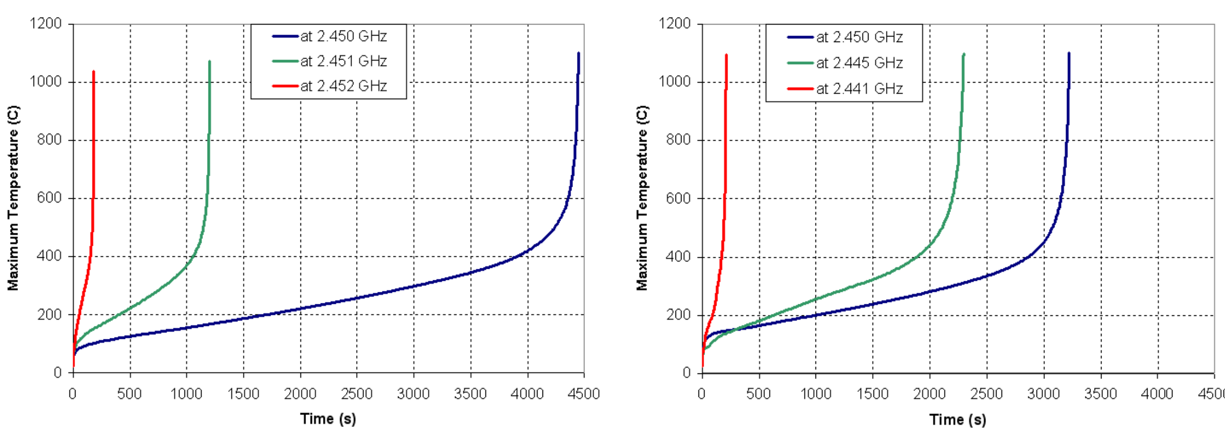
Computational Results

The first computational test (performed with arbitrary constant heating time step sufficiently small to follow the non-linear variation of the loss factor of zirconia shown in Table 1) in determination of temperature fields of the process carried out at 2.45 GHz confirms the known experimental fact that the zirconia sample is heated notably more quickly than the insulation. Although in reality the materials are heating non-uniformly, we initially assume homogeneous distribution of material parameters (in accordance with the temperature values in Table 3) in order to approximate the frequency characteristics of the reflection coefficient $|S_{11}|$ at the initial stage of microwave heating process.



(a) (b)

Fig. 2. Evolution of frequency characteristics of the reflection coefficient in the course of microwave heating: materials temperatures are assumed to vary in accordance with Table 3; the zirconia sample of $D = H = 38$ mm (the resonance at 2.452 GHz) (a) and $D = H = 62$ mm (the resonance at 2.441 GHz) (b).



(a) (b)

Fig. 3. Time histories of maximum temperature for different magnetron frequencies for the zirconia sample of $D = H = 38$ mm (a) and 62 mm (b).

The resulting curves are shown in Fig. 2. They reveal a highly resonant profile of the microwave system, and this looks fairly natural given the low values of the loss factors of both materials and the small size of the zirconia compact. If the furnace with a small sample is excited at 2.45 GHz, the level of reflections appears to be high and almost not changing in the course of heating. This indicates that energy coupling of the process is low (for $D = H = 38$ mm, its average value $\hat{C}$ in the considered temperature range is ~30%), so one cannot expect quick heating of the sample of that size. When a larger cylinder is processed, the coupling varies with temperature (as marked by a blue line in Fig. 2(b)), but remains low ($\hat{C} \sim 40\%$ for $D = H = 62$ mm).

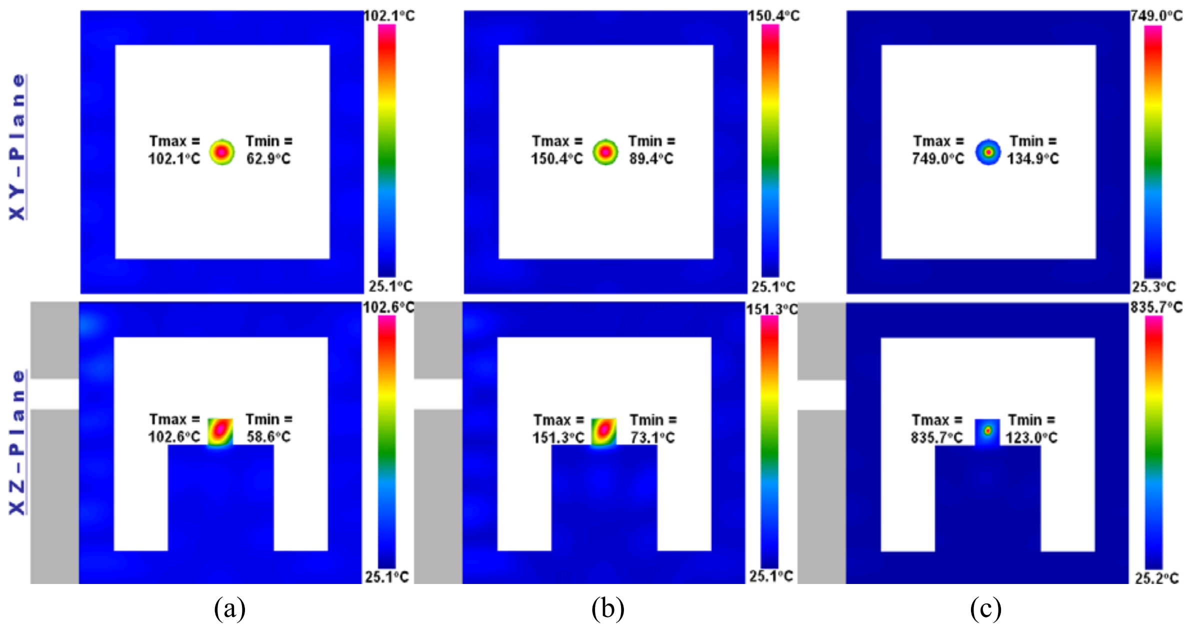


Fig. 4. Temperature field in the system after 180 s of microwave processing at the resonant (2.452 GHz) (c) and non-resonant (2.450 and 2.451 GHz) (a), (b) frequencies; zirconia sample: $D = H = 38$ mm. The XZ-Plane of the model includes the waveguide transition into the furnace at the left of (a), (b), and (c).

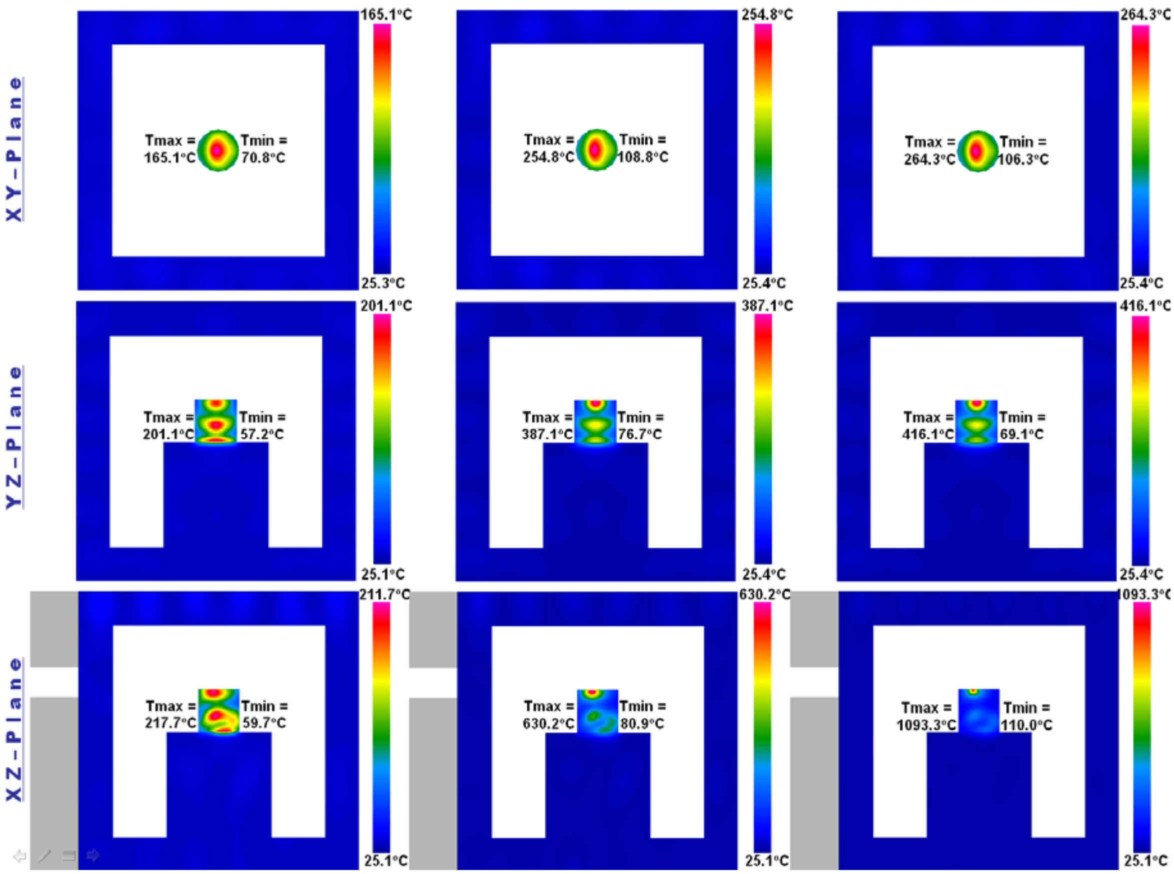
However, if the magnetron is tuned to the closest deepest resonance (located at 2.452 GHz for $D = H = 38$ mm and at 2.441 GHz for $D = H = 62$ mm), the situation dramatically changes: energy coupling become substantially higher for all temperatures in the range – 81 and 88 %, respectively. These results allow us:

(a) to conclude that, from an energy efficiency viewpoint, the performance of the considered microwave system strongly depends on the operational regime of the magnetron (and, thus, in general, may be fairly unstable), and
(b) to detect the resonances closest to 2.45 GHz as the potential operational frequencies of the furnace.

Next step of our computational study is dedicated to mimicking the process of heating of zirconia by 1 kW microwave power in the entire temperature range both at the resonant and non-resonant frequencies. The simulated time characteristics of maximum temperature in the zirconia cylinder are shown in Fig. 3. These curves were generated in the framework of an iterative solution of the electromagnetic-thermal coupled problem using

(*i*) from 59 (at 2.452 GHz) to 465 (at 2.450 GHz) iterations with the heating time step varying from 10 to 0.25 s (for the smaller cylinder $D = H = 38$ mm), and
(ii) from 68 (at 2.441 GHz) to 325 (at 2.450 GHz) iterations with the heating time step varying from 20 to 0.1 s (for the larger cylinder $D = H = 62$ mm).

It is important to emphasize that computation of the temperature field after each iteration is performed with a rigorous upgrade of material parameters that is performed in each FDTD cell (inside the insulator and zirconia) in accordance with the detailed temperature-dependent data given in Tables 2 and 3.



(a) (b) (c)

Fig. 5. Temperature field in the system at the resonant frequency (2.441 GHz) after 100 s (a), 200 s(b), and 208.6 s (c) of microwave processing; zirconia sample: $D = H = 62$ mm.

The computed characteristics motivate two major observations. First, it becomes evident that the frequency at which the considered microwave system is excited is critical for the process as the position on or off the closest resonance results in a difference in the heating rate from nearly 15 (larger cylinder) to 25 times. Second, the minor temperature increase in the loss factor of zirconia, occurring near 400^{o}C, results in quick development of thermal runaway, and it takes place regardless of the frequency of excitation: the zirconia samples of smaller and larger size are heated from 400 to 1100°C at all considered frequencies for ~15% of the total heating time.

Finally, the patterns in Figs. 4 and 5 reveal details of the heating process in the considered furnace in term of volumetric temperature distribution. It is seen that microwave heating at a high heating rate produces strongly non-uniform temperature fields and the detected thermal runaway occurs at one sharply localized hot spot – the resulting ratio of maximum/ minimum temperatures in the zirconia cylinder may reach 8.5 to 10. On the other hand, a slow heating obtained by keeping the exciting frequency off of the resonant frequency results in a much higher level of uniformity of temperature distribution.

While all generated computational results appear to be consistent with the experiments performed in the considered furnace with the zirconia samples, the details of the present study show the capability of the developed model to help with development of new microwave-assisted processes by predicting occurrence of thermal runaway and 3D temperature distributions.

Conclusion

An advanced innovative approach to multiphysics modeling of microwave high temperature thermal processing of materials has been reported. Computational results obtained for the process of microwave heating by 1 kW power of cylindrical zirconia compacts reveal practicality of the developed modeling technique and emphasizes a critical importance of availability of detailed data on temperature-dependent material parameters. Future development of the model includes incorporation of convective thermal boundary condition on the air-dielectric interfaces.

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