Computational Study of Thermal Runaway in Microwave Processing of Zirconia

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Abstract—**Up to now, reported attempts of multiphysics modeling of high-temperature microwave processing have been limited to fairly idealized models. In this contribution, an original accurate computer model enabling virtual experimentation in microwave processing of zicronia is presented. The model, implementing fully coupled FDTD electromagnetic and thermal algorithms, uses detailed data on temperature-dependent electromagnetic and thermal material parameters of zirconia and alumina-based insulation in the temperature range from 25 to 1100°C. Computational results including characteristics of energy coupling and 3D temperature fields are analyzed in the context of conditions for occurrence of thermal runaway. The rate of microwave heating is found to be dependent on the frequency of processing: operation at a resonance closest to 2.45 GHz results in substantial acceleration of heating and generation of a particularly strong hot spot. It is demonstrated that thermal runaway is caused by an increasing loss factor and develops independently of energy coupling.**

Keywords – energy coupling, hot spot, loss factor, multiphysics modeling, thermal runaway.

I. INTRODUCTION

In recent years, the increase in energy costs has resulted in exploring microwave processing of materials as a fast and efficient method for heating processes. At the same time, the growing use of computers and advancements in computational techniques have stimulated a rising interest in computer modeling of microwave heating processes and systems. However, the significant benefits provided by the modern techniques of modeling and optimization have been demonstrated primarily in food related applications [1, 2]. In the field of materials science, the known attempts of multiphysics modeling on a macroscopic level [3-7] are 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 electromagnetic-thermal model for simulating the process of high temperature heating of materials in a microwave furnace. The model is based on the 2.45 GHz system employed by Ceralink, Inc. for microwave processing of a variety of ceramic and composite materials. The model is built as a process development tool in order to predict how materials will behave and attempt to optimize the process parameters. Indeed, the absence of a method for in-situ monitoring of electromagnetic and thermal processes in the furnace provides a 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 intrinsiccally 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.

II. 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 waveguide connecting the cavity with a generator. We report here on a series of computational experiments with a cylindrical zirconia compact with the diameter D and height H equal to 38 and 62 mm.

III. COMPUTER MODEL

The microwave furnace in question has been represented in a 3D fully parameterized model (Fig. 2) developed using the 3D conformal FDTD software package *QuickWave-3D* (*QW-3D*) [8]. 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 [1] and using the *QW-3D*'s *Basic Heating Module* (*BHM*) [8], on the first iteration, a converged electromagnetic

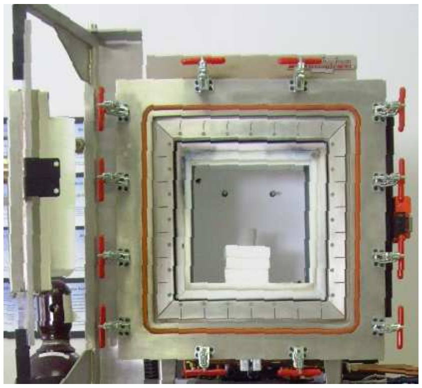


Figure 1. Ceralink's microwave furnace for high temperature processing of materials: front view with an open door.

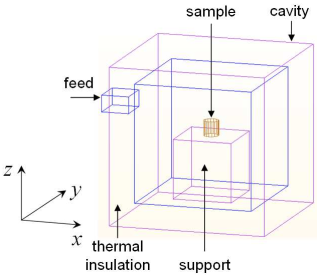


Figure 2. 3D view of the model of the furnace in Fig. 1.

solution was supposed to be reached after 650 periods (~55,000 FDTD time steps); at each subsequent iteration, 300 periods were involved. Heating pattern construction was carried out during 3 periods.

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 two quad-core Intel Xeon 3.2 GHz processors. Computations were accelerated by a GPU Acceleware card A30 (NVIDIA Quadro FX 5600). 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 course of electromagnetic-thermal iterations, computation of temperature was performed with a variable heating time step to take care of non-linear profiles of time-temperature history of the considered process.

IV. MATERIAL PARAMETERS

Prior to modeling, the dielectric and 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. The process of generation of these characteristics is described in [9]. Here we emphasize that since, during sintering, the zirconia compact shrinks and the shrinkage typically occurs above 1100°C, the employed technique of determination of density is not valid above that temperature. Moreover, application of the developed electromagnetic-thermal model in the conditions of changing geometry may be inadequate. It has therefore been decided to limit the computational study to the temperatures below 1100°C.

TABLE I. 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 II. ELECTROMAGNETIC (AT 2.45 GHZ) AND THERMAL PARAMETERS OF ALUMINA INSULATION

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 III. TEMPERATURES OF MATERIALS IN COMPUTATION OF REFLECTIONS IN FIG. 3

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

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.

V. COMPUTATIONAL RESULTS

The first computational test (performed with a constant heating time step sufficiently small to follow the non-linear variation of the loss factor of zirconia 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 more quickly than the insulation. Although in reality the materials are heated non-uniformly, we initially assume homogeneous distribution of material parameters (according to the temperatures in Table 3) in order to approxi-

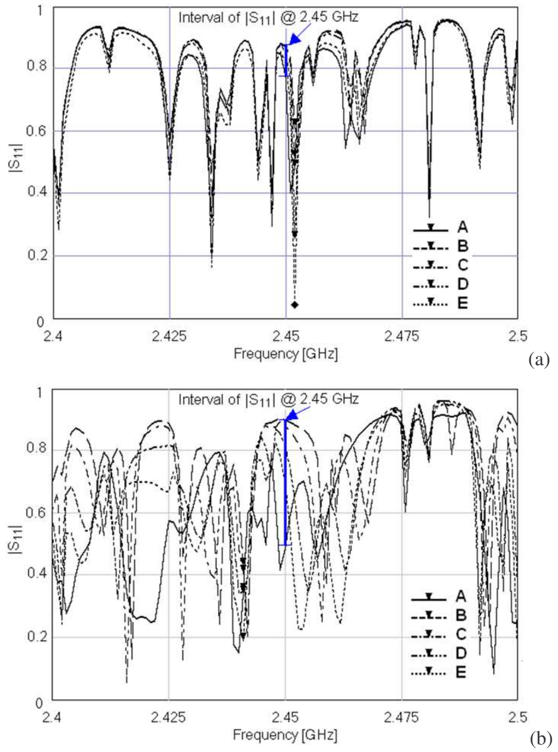


Figure 3. Evolution of frequency characteristics of the reflection coefficient in the course of microwave heating: materials temperatures are assumed to vary in line with Table 3; the zirconia sample of $D = H = 38$ mm (the resonance at 2.452 GHz) (a) and $D = H = 62$ mm (the resonance at 2.441 GHz)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

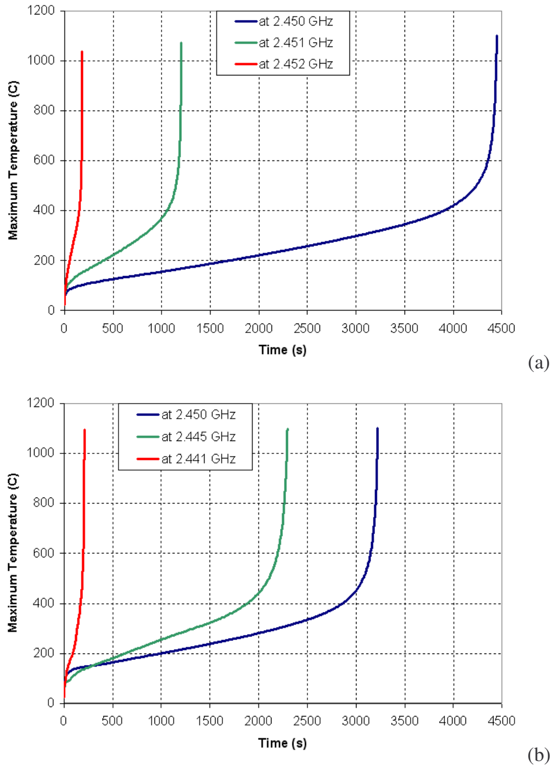


Figure 4. Time histories of maximum temperature for different magnetron frequencies for the zirconia sample of $D = H = 38$ mm (a) and 62 mm (b).

mate the frequency characteristics of the reflection coefficient $|S_{11}|$ at the initial stage of microwave heating process.

The resulting curves are shown in Fig. 3. 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 is 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 in the considered temperature range is ~30%, so one cannot expect quick heating of the sample. When a larger cylinder ($D = H = 62$ mm) is processed, the coupling varies with temperature (as marked by a blue line), but remains low (~40%). However, if the magnetron is tuned to the closest deepest resonance (at 2.452 GHz for $D = H = 38$ mm and at 2.441 GHz for 62 mm), the situation radically changes: energy coupling becomes much higher – 81 and 88 %, respectively.

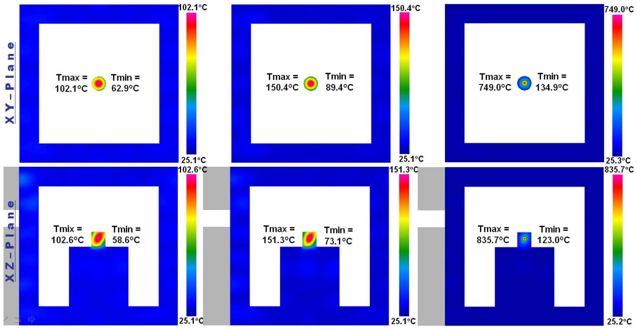
These results allow us to conclude that, in terms of energy efficiency, the performance of the considered microwave system strongly depends on the operational regime of the magnetron (and, thus, in general, may be fairly unstable) and detect the resonances closest to 2.45 GHz as the potential operational frequencies of the furnace.

Next step of our 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. 4. These curves were generated in the framework of an iterative solution of the electromagnetic-thermal coupled problem using:

- 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);
- 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).

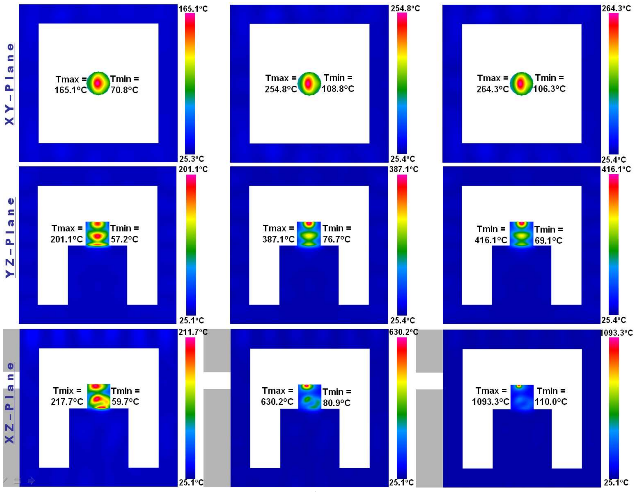
It is worth noting that computation of the temperature field after each iteration is performed with a rigorous upgrade of material parameters that is performed in each FDTD cell (inside the insulator and zirconia) in accordance with the detailed temperature-dependent data given in Tables 2 and 3.

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°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.



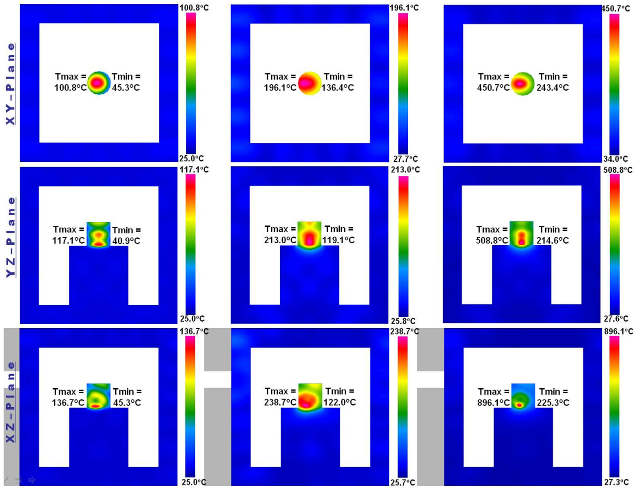
(a) (b) (c)

Figure 5. 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.



(a) (b) (c)

Figure 6. 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.



(a) (b) (c)

Figure 7. 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 patterns in Figs. 5-7 reveal more details of the heating process in the considered furnace. 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 max/min temperatures in the zirconia cylinder may reach 8.5 to 10. On the other hand, a slow heating by keeping the exciting frequency off of the resonance results in a much higher level of uniformity of temperature distribution.

While all generated computational results appear to be consistent with the experiments performed with the considered 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.

VI. 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 will include incorporation of convective thermal boundary condition on the air-dielectric interfaces.

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