Modeling of Microwave Heating of Ceramic Materials with Respect to Changes in Sample Size

Andreas Rosin and MonikaWillert-Porada

Chair of Materials Processing, University of Bayreuth, Bayreuth, Germany

Abstract – An approach for consecutive electromagnetic simulations by FDTD and thermal simulation by FEM is developed with respect to change in sample volume during microwave sintering. Volume-related heat dissipation by microwaves is calculated as a step function of temperature and material porosity with help of FDTD simulations. The material of choice for this study is fully yttria-stabilized zirconia (FYSZ). A cylindrical green body with 50% porosity is virtually sintered to >99% theoretical density, i.e. the sample shrinks by 50%. The transfer of dissipated heat data from microwave modeling to thermal modeling follows a pragmatic volume-by-volume method for faster results but at the expense of local accuracy. The thermal model includes radiative and convective heat transfer to environment. A feedback loop ensures constant heating rates in order to achieve comparable results on different sintering setups.

Introduction

Microwave heating and sintering at lab scale and prototype scale is typically performed at 2.45 GHz with λ_0 = 122 mm in free space. In this field of application the size of heated objects is often in the same size range as the microwave wavelength. From investigations on optical scattering it is known that the absorption of electromagnetic waves is prone to resonances in this so-called Mie range [1]. Absorption and scattering of electromagnetic waves for single, isolated spherical objects can be calculated by the Mie theory [2]. A useful C code is provided in [2], which is extended and used here to calculate the absorption at 2.45 GHz of an isolated FYSZ sphere in air for radius R = 0.5 to 200 mm and temperature T = 20°C to 1200°C (see Fig. 1) [3]. The permittivity of dense FYSZ is taken from [4]. This behavior is the reason to consider microwave absorption to be a function of permittivity and sample volume as described by [5]. The permittivity in turn is depending on the material composition, microwave frequency, porosity and temperature.

However, Mie calculations cannot be transferred to microwave processing in closed resonators, because they provide their own resonance conditions. The resonator geometry is a key parameter for designing microwave processes, see e.g. [6]. For the present investigation a pre-densified green body of FYSZ will be virtually microwave heated to >1400 °C, and thus change its volume and porosity. The influence of decreasing sample volume is implemented by stepwise volume reduction. Its thermal behavior is evaluated by coupling of microwave and thermal simulation.

Technique

In recent years electromagnetic simulation has been extended to multiphysics simulation of microwave processes in order to couple electromagnetic interactions with material changes in order to evaluate e.g. thermal [7], radiative [8] or fluid flow interactions with microwave heating [9]. This requires either real multiphysics software packages, which combine all physical models into one tool, or coupling results from two or more specialized software tools externally [10]. For microwave modeling the FDTD-based software QuickWave-3D

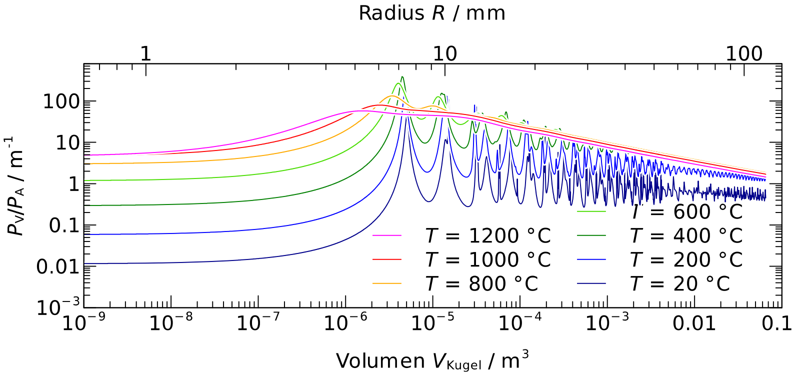


Fig. 1: Volume specific absorption of a single FYSZ sphere as a function of radius and temperature.

(v.7.5c) is used. For thermal simulation the FEM-based COMSOL Multiphysics (v.3.5a) is chosen. A cubic microwave cavity is selected with a dominating TE_{031} mode (length 200 mm, width 190 mm and height 162 mm). A cylindrical FYSZ sample is placed exactly in the center, levitating in air (see Fig. 2a). A standard WR340 waveguide connects the applicator to the microwave source. The frequency f of the incident wave is 2.45 GHz, and the amplitude is 20, i.e. the time-averaged microwave power is 200 W. The walls of the applicator and waveguide are considered to be loss-free conductors and to be tempered to ambient temperature T_∞. The initial diameter d_0 and height h_0 of the cylindrical sample is 48 mm, initial Mie coefficient is $x_0 = 1.24$ and initial porosity is 50%. After reaching temperatures above 1400 °C the final diameter (and height) is shrunken to 38.1 mm, and Mie coefficient is decreased to 0.98.

Two sintering setups are considered: (1) direct microwave heating from room temperature to 1430 °C ($T_\infty = 25$ °C), and (2) microwave hybrid heating from 800 °C to 1430 °C ($T_\infty = 800$ °C). In the latter case the complete microwave applicator is considered to be heated to 800 °C by conventional heating means and is kept at this level while microwave heating brings the sample to the final temperature. The sintering behavior of YSZ is taken from [11] by extracting the relation of theoretical density to temperature curve. Until 800 °C no volume shrinkage is observed. From 800 °C to 1400 °C the volume of the sample is modified in 13 steps from $d_0 = 48$ mm to $d_n = 38.1$ mm in temperature step widths of $\Delta T = 25 \ldots 50$ K ($\Delta d = 0.1 \ldots 1.5$ mm). For each step in sample size, a separate microwave (and thermal) simulation is performed. The permittivity of the FYSZ sample is defined, as mentioned above, as a function of temperature and porosity. The influence of the voids fraction φ on permittivity is calculated from the generalized mixing rule described in [12] with $\varepsilon_{air} = 1$ and the coefficient $\nu = 2.3$, which was determined experimentally [3]. The results for permittivity calculation are presented in Fig. 2.

$$\frac{\epsilon_{\mathrm{mix}}(T) - \epsilon_{\mathrm{air}}}{\epsilon_{\mathrm{mix}}(T) + \epsilon_{\mathrm{air}} + \nu\left[\epsilon_{\mathrm{mix}}(T) - \epsilon_{\mathrm{air}}\right]} = \varphi(T)\frac{\epsilon_{\mathrm{YSZ}}(T) - \epsilon_{\mathrm{air}}}{\epsilon_{\mathrm{YSZ}}(T) + \epsilon_{\mathrm{air}} + \nu\left[\epsilon_{\mathrm{mix}}(T) - \epsilon_{\mathrm{air}}\right]}$$

The mesh size s_{mesh} for FDTD modeling depends on wavelength λ_0 and relative permittivity ε_r (and permeability μ_r) of the material and follows the rule [13]:

$$s_{\mathrm{mesh}} = \frac{\lambda_0}{10\sqrt{\epsilon_{\mathrm{mix}}(T)}}$$

Accordingly, the mesh size for FYSZ is initially 3.7 mm and finally 2.2 mm at 1400 °C. The air-filled area has a mesh size of 10 mm. The number of cells increases from 700,000 to

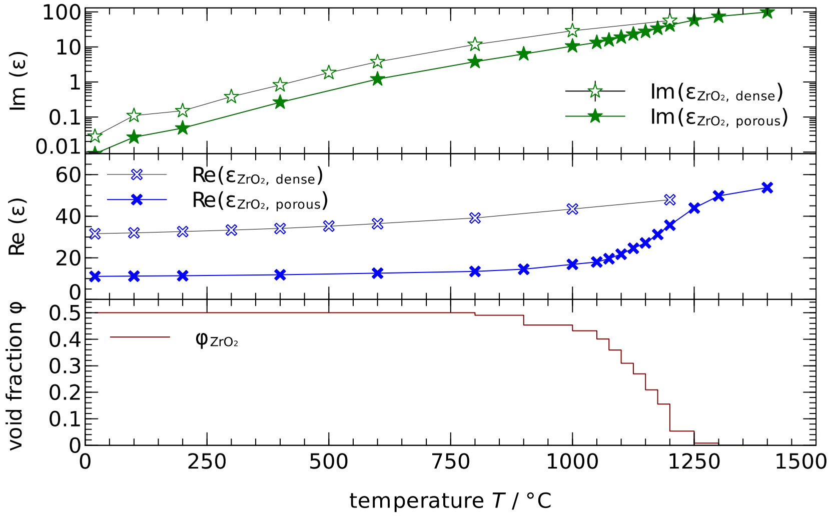
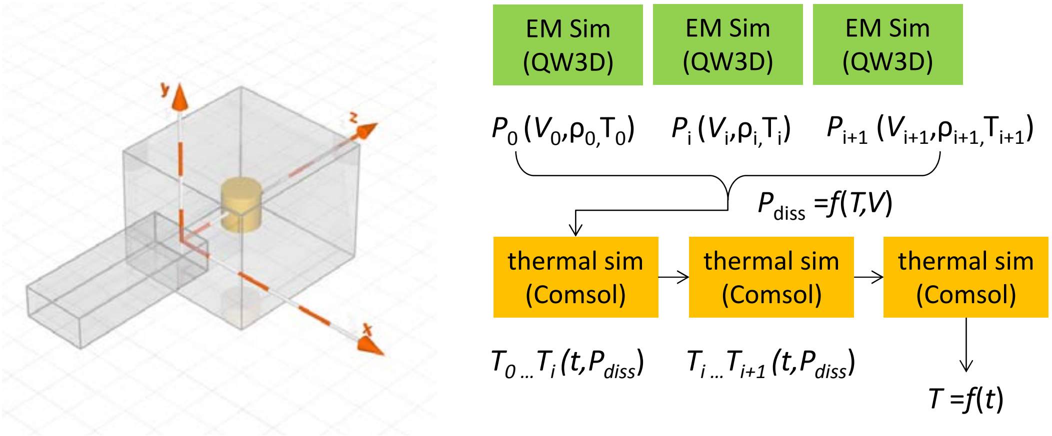


Fig. 2: Temperature dependent permittivity of dense FYSZ and as a function of the densification during sintering.

more than 2 million, and thus demand for RAM increases from 60 to 200 MB. The FDTD simulations are carried out on a quad-core processor with 3 GHZ and 8 GB RAM without use of multithreading. After passing a transient initial phase in FDTD calculation of 50,000 to 60,000 iterations, a time-averaging envelope phase is started to reach stationary results after at least 25,000 additional iterations. The simulation is stopped arbitrarily after passing this mark. The initialization phase takes about 0.5 to 2 hours; the enveloping phase takes about 4 to 22 hours depending on model size.

The coupling between microwave and thermal simulation is done consecutively, i.e. first microwave simulations are completed and a temperature-related function for the absorbed microwave power is generated (see Fig. 2b). This approach differs from other approaches as reported by others [10] where a serial cycling between microwave and thermal model is realized in a semi-closed loop with appropriate selection of time steps and a reliable data transfer. According to the different meshing methods of FDTD and FEM, point-to-point transfer of Joule heat data necessitates additional interpolation routines [14]. In the present work the Joule heat information is instead transferred by a volume-to-volume approach. The FYSZ sample is divided into adequate areas like outer shell and inner core, representing 65% of total volume (for more complex arrangements the number of sub-volumes will be increased). Microwave dissipation is extracted for these two sub-volumes as volume-specific, temperature dependent step-functions.

The thermal model in COMSOL Multiphysics® is using the “general heat transfer” module, which includes thermal radiation. According to rotational symmetry of the cylindrical sample and the constant ambient temperature of the surrounding applicator the thermal model is reduced to 2D, which reduces the cell number to 3150 elements and accelerates the thermal simulation while maintaining a sufficient level of accuracy. The mesh size is coupled to volume shrinkage of the FYSZ sample. For each size/temperature range a separate thermal simulation is performed, where the results of the previous step are used as initial conditions. Temperature dependent values for heat conductivity and capacity are found in [15, 16]. The effective heat conductivity of the porous FYSZ is calculated by simplified Maxwell’s equations [15]. The total emissivity is assumed to be constant with temperature [17]. The convective heat loss to the surrounding gas phase is calculated by Nusselt, Rayleigh and Prandtl numbers for natural convection [18]. The temperature of the surrounding gas



(a) (b)

Fig 3. Geometrical model of microwave cavity and cylindrical sample levitating at central position (a); procedure of serial coupling of microwave and thermal simulation in the present approach (b).

phase is equal to the ambient applicator temperature. Properties for air are taken from the software's material database.

$$\mathrm{Nu} = \left(0.825 + 0.387\left(\mathrm{Ra}\left[1+\left(\frac{0.559}{\mathrm{Pr}}\right)^{\frac{9}{16}}\right]^{-\frac{16}{9}}\right)^{\frac{1}{6}}\right)^{2} + 0.435\frac{h}{d}$$

A feedback control loop, which is designed as a PID algorithm, controls the total amount of microwave heating power needed to keep track of the defined heating rates of 5 K/min and 20 K/min. Reference point is the center of the sample. The constants K_P, K_I and K_D are determined by the Ziegler-Nichols method [20] with some individual adjustments.

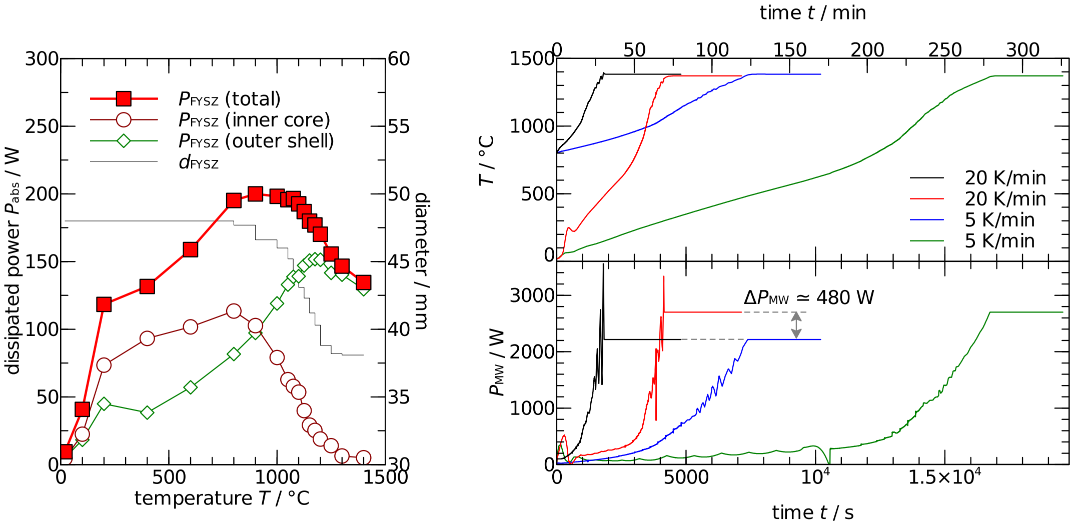
$$P_{\mathrm{MW}} = K_P \cdot e(t) + K_I \cdot \int_0^t e(t)\,dt + K_D \frac{d}{dt} e(t)$$

Results

Fig. 4a shows the calculated microwave absorption of the sample as a function of sample temperature and diameter. At temperatures below 200 °C microwave absorption of FYSZ is quite low due to the low permittivity of FYSZ. The maximum microwave dissipation occurs at 800 to 1100 °C (d = 48 to 43 mm), reaching almost 100% microwave absorption. After passing this maximum range, microwave efficiency decreases to about 65% at 1400 °C. The ratio of absorbed power in the inner core and outer shell changes significantly with increasing temperature, changing from volumetric to surface heating above 1000 °C. The result of thermal simulation is demonstrated in Fig. 4b, showing the average temperature of the FYSZ samples and the required microwave power as function of time with respect to heating rate and microwave heating method. High heating rate and hybrid heating result in less energy consumption as expected. In the present case hybrid heating demands about 18% less heating energy and 20 K/min heating rate saves about 80% energy.

Conclusion

Although the presented volume-by-volume approach for coupling models fulfills the law of conservation of energy, it is not considered to highly resolve heat distribution inside the



(a) (b)

Fig. 4. Extracted total microwave absorption of the sample as function of temperature and diameter (a); temperature-time and power-time curves for direct microwave heating (T_0 = 25 °C) and hybrid microwave heating (T_0 = 800 °C) of a cylindrical FYSZ sample (d_i = 48 mm) with heating rates of 5 and 20 K/min (b).

sample in comparison to other approaches [19]. In particular, thermal runaway or hot spot formation will be missed due to the "smoothing" effect of this approach. The confidence level of this volume-by-volume method is about 90%. However, its consecutive approach offers the possibility for early re-designing a microwave cavity in order to increase the microwave heating efficiency with respect to maximum power absorption at highest process temperature, resonator geometry and changes in object size. The presented method can be used for basic engineering of microwave processes, providing sturdy estimates on microwave heating.

References

[1] H. C. Van De Hulst, *Light Scattering by Small Particles,* Dover Publications, Mineola, 1982.

[2] C. F. Bohren and D. R. Huffman, *Absorption and scattering of light by small particles*, Wiley, New York, 1983.

[3] A. Rosin, *Entwicklung einer simulationsgestützten Methodik zur Auslegung und Bewertung von Hochtemperatur-Mikrowellenprozessen*, Dissertation, University of Bayreuth, in preparation.

[4] J. Batt, J.G.P. Binner, T.E. Cross, N.R. Greenacre, M.G. Hamlyn, R.M. Hutcheon, and W.H. Sutton, A parallel measurement programme in high temperature dielectric property measurements: an update, In: *Microwaves: Theory and Application in Materials Processing II,* vol. 36, D.E. Clark, W.R. Tinga & J.R. Laia, Eds., American Ceramic Society, 1993, pp. 243-250.

[5] M. Rother, *Über das Konkurrenzverhalten von Dielektrika bei der Mikrowellenerwärmung,* KIT Scientific Publishing, Karlsruhe, 2010.

[6] T.V.C.T. Chan and H.C.Reader, *Understanding Microwave Heating Cavities*, Artech House, Boston, 2000.

[7] L. Ma, D.-L. Paul, N. Pothecary, C. Railton, J. Bows, L. Barratt, J. Mullin, and D. Simons, Experimental validation of a combined electromagnetic and thermal FDTD model of a microwave heating process, *IEEE Trans. Microwave Theory and Tech.*, vol. 43, pp. 2565-2572, 1995.

[8] S.M. Allan, M.L. Fall, E.M. Kiley, P. Kopyt, H.S. Shulman, and V.V. Yakovlev, Modeling of hybrid (heat radiation and microwave) high temperature processing of limestone, In: *IEEE MTT-S Intern. Microwave Symp. Dig. (Montreal, Canada, June 2012)*, 978-1-4673-1088-9/12.

[9] C.M. Sabliov, D.A. Salvi, and D. Boldor, High frequency electromagnetism, heat transfer and fluid flow coupling in ANSYS multiphysics, *J. Microwave Power and Electromag. Energy*, vol. 41, pp. 5-17, 2007.

[10] P. Kopyt and M. Celuch, Coupled electromagnetic-thermodynamic simulations of microwave heating problems using the FDTD algorithm, *J. Microwave Power and Electromag. Energy*, vol. 41, pp. 18-29, 2007.

[11] D.-J. Chen and M.J. Mayo, Rapid rate sintering of nanocrystalline ZrO_2-3 mol% Y_2O_3, *J. Amer. Ceramic Soc.*, vol. 79, pp. 906-912, 1996.

[12] A. Sihvola, Self-consistency aspects of dielectric mixing theories, *IEEE Trans. Geoscience and Remote Sensing*, vol. 27, pp. 403-415, 1989.

[13] D.B. Davidson, *Computational Electromagnetics for RF and Microwave Engineering*, Cambridge University Press, 2011.

[14] S. Demjanenko, K. Nowak, R. Northrup, S. Bogachev, E.M. Kiley, D. Bouvard, S.L. Weekes, and V.V. Yakovlev, Interpolation algorithms for interfacing FDTD and FEM meshes in multiphysics modeling of microwave sintering, *Proc. 12th Seminar "Computer Modeling in Microwave Engng & Applications - Advances in Modeling of Microwave Sintering" (Grenoble, France, March 2010)*, pp. 62-64.

[15] K.W. Schlichting, N.P. Padture and P.G. Klemens, Thermal conductivity of dense and porous yttria-stabilized zirconia, *J. Materials Science*, vol. 36, pp. 3003-3010, 2001.

[16] X. Song, M. Xie, F. Zhou, G. Jia, X. Hao, and S. An, High-temperature thermal properties of yttria fully stabilized zirconia ceramics, *J. of Rare Earths*, vol. 29, pp. 155-159, 2011.

[17] A.V. Palagin, *On the Thermo-Physical Properties of Zircaloy-4 and ZrO_2 at High Temperatures: Part 2. Determination of Zircaloy and ZrO_2 Emissivities Using Experimental Data of Empty Rods Cool-Down Tests*, Wissenschaftliche Berichte, Forschungszentrum Karlsruhe, 6739, 57-78, 2002.

[18] V.D.I. Wärmeatlas, *Berechnungsblätter für den Wärmeübergang* Verein Deutscher Ingenieure, Springer, 2006.

[19] V.V. Yakovlev, S.M. Allan, M.L. Fall, and H.S. Shulman, Computational study of thermal runaway in microwave processing of zirconia, In: *Microwave and RF Power Applications*, J. Tao, Ed., Cépaduès Éditions, 2011, pp. 303-306.