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ABSTRACT

This paper describes a new approach, based on the Mie theory, to measure the dielectric properties of lossless materials at temperatures greater than 1500 °C. For the reliable operation of microwave transmitting materials in harsh environments, it is crucial to correctly characterize the permittivity under various temperature conditions. Heating and measurement systems using a propane torch and a single horn antenna were designed to estimate such dielectric properties. The reflection spectrum of Al_2O_3 ceramics at room temperature was determined using the Mie theory to derive the permittivity and validate the approach. High-temperature dielectric constants are derived from simulated values, which are reliable and have only a slight slope as a function of temperature. These results indicate that the permittivity measurement technique can provide powerful information for the optimal design and accurate evaluation of the dielectric properties of various lossless materials at high temperatures.

I. INTRODUCTION

Microwave transmitting materials have been of interest in a variety of applications, including mobile communications and aerospace vehicles.[1,2] When such materials are used in aerospace applications, the surface temperature of the material is significantly elevated due to friction from the air, which changes the dielectric properties of the material. Therefore, there have been increased demands for an accurate technique to measure the high-temperature permittivity of materials at microwave frequencies, with many potential techniques being introduced over the past several decades. Some of these well-known methods have been proposed to characterize permittivity in the microwave frequency using waveguides or coaxial transmission lines, such as the free-space method and resonance methods. One important method to measure the permittivity or permeability of materials is the waveguide transmission line method.[3–5] This approach uses the coefficient of electromagnetic power transmitted to or reflected by a specimen placed in a waveguide when an electromagnetic field is applied. However, this approach has inaccuracies because of the discontinuous air gaps that appear between the sample and the waveguide. In addition, differences in the thermal expansion coefficients between the sample and the waveguide cause imprecision in the measured values at higher temperatures.[6]

Another method, the free-space technique, is capable of performing measurement at high frequencies.[4,7,8] The specimens are placed inside two horn antennas and a Gaussian beam is used to focus the electromagnetic energy onto the specimen. The dielectric properties are then evaluated using the scattering, transmission, and reflection properties at a specific frequency of the focused beam. This method can measure the electromagnetic properties at a high temperature because there is no need for direct contact with the samples.[9] However, this method is less accurate than the cavity system method since there are diffraction effects and multiple reflections at the samples.[10] The resonant method, widely known as the Hakki-Coleman method, is useful for low-loss dielectric materials in cavities and open resonators.[11–13] In this approach, if pores in the material or a complex are formed, the resonance is complicated by the incident microwaves, making it difficult to precisely extract the dielectric properties.[14]

Notably, these widely used permittivity measurements vary in accuracy depending on the type and dimensions of the specimen, while high temperatures cause reliability problems in the measured values. Especially, when dielectric properties are measured at high temperatures above 1500 °C, a new heating source must be utilized due to the limitations of conventional heating methods, which uses a ramp heating apparatus or a furnace. Although an acetylene or plasma torch system can increase the sample temperature above 1500 °C, the heating zone is too small relative to the size of the sample, causing a temperature gradient in the lateral direction. This temperature gradient scatters the microwave beam in various directions, which degrades the accuracy of the extracted permittivity. Therefore, it is necessary to develop a design that reliably determines the dielectric properties of materials in harsh environments, even under high frequencies and at high temperature (above 1500 °C) conditions.

This work considers measurements for the permittivity of lossless dielectric materials at temperatures above 1500 °C using the Mie theory. Mie scattering is a phenomenon applied to particles that are approximately the same size as the incident wavelength. However, the properties of Mie scattering strongly depend on the shape and size of the particle. This phenomenon can be understood as a combination of multiple electromagnetic resonance modes, also known as the multipole expansion.[15,16] Controlling these electric and magnetic modes enables many novel functionalities, such as artificial magnetism with nonmagnetic materials or directional scattering. Many studies have actively utilized this controllability. It was experimentally revealed that Mie scattering can also be utilized in the microwave regime.[17] To give a more intuitive description of the Mie theory, an analysis based on a Fabry–Perot resonator was described in the literature.[18]

In this study, we design a Mie resonator whose backward-scattering cross section peak is in the Ku-band frequency. This peak position changes with the dielectric constant of a specimen due to temperature changes. Since the specimen is relatively small, one can minimize the temperature gradient inside the resonator through heating, which can simplify the experimental conditions. The dielectric constant is deduced from this peak position in the measured reflection pattern. The reflection patterns from Al_2O_3 ceramics in the Ku-band are obtained using a free-space antenna system. The hardware for the conventional resonance and the free-space method have obvious limitations when attempting to obtain the permittivity at temperatures above 1500 °C. Therefore, the heating and the refractory space are changed by employing only a single horn antenna system, and the entire region of the specimen is heated to high temperatures using a propane torch. After analyzing the reflection spectra, the dielectric properties of the Al_2O_3 ceramics are estimated as functions of temperature.

II. EXPERIMENTAL PROCEDURE

A. Specimen design

The Mie resonance frequency is strongly dependent on the shape and dimensions of the target samples. This study considers measurements for the permittivity of Al_2O_3 specimens in the Ku-band frequency regime. The size of the specimen was varied from $4 \times 4 \times 40\,\text{mm}^3$ to $12 \times 12 \times 40\,\text{mm}^3$ to determine the proper specimen size that exhibits a backward-scattering cross section peak within this frequency range. Furthermore, thin Al_2O_3 pillars sized at $3 \times 3 \times 50\,\text{mm}^3$ were attached on both sides of the specimen to suspend it in air. Since the dimensions of the side pillar structures are much smaller than that of the main specimen, their effects on the cross section are neglected. In this configuration, the entire main specimen area can be heated using the flame from a torch. The polarization direction of the microwave was set to be along the longest direction of the sample.

A finite-difference time-domain (FDTD) analysis was performed to optimize the shapes and sizes of the specimens and to calculate their scattering cross section over the Ku-band frequency regime. If the dimensions of the specimen are too small, it is not possible to reliably measure its cross section. On the other hand, if they are too large, it will be difficult to define an apparent peak because of the multiple higher-order modes that would be present. When calculating the scattering cross section, the experimental setup conditions are taken into account such as the antenna size and the antenna-sample configuration so that the exact scattering cross section of the light on the antenna area can be calculated from the far-field simulation results. Once the sample dimensions are determined, a database that relates the peak frequency to the permittivity is built from simulations of various permittivities. After the measurements, the dielectric constant is retrieved by comparing the measured peak frequency in the reflection spectrum with the data from the database.

B. Measurement and retrieval of permittivity at high temperature

Prior to using the Mie theory to determine the dielectric constant, the dielectric properties of the measuring system are determined using the waveguide through-reflection-line (TRL) approach and the free-space gate-reflection-line (GRL) method to minimize any errors. Firstly, we use the through connection, reflection, and line-matching in the waveguide system to obtain the permittivity values of air and the reference materials. After calibration using the TRL method, the transmitter and receiver horn antennas are employed for calibration using the GRL technique. After the GRL calibration, one horn antenna is removed to construct the high-temperature torch and refractory system.

After the calibration procedure, we use the single antenna-propane torch system to estimate the high-temperature permittivity of the lossless ceramic samples. A schematic illustration of the measurement configuration is shown in Fig. 1. A planar Al_2O_3 sample with dimensions $8.5 \times 8.5 \times 40\,\text{mm}^3$ is surrounded by refractory blocks (SK-38, Hanse refractories, Korea) for thermal insulation and is located in front of a spot-focusing Ku-band (12.4–18.0 GHz) horn antenna with a focal distance of 30.5 cm. A high-temperature propane torch (Nortel Red Rocket, Wale Apparatus, USA) is placed in front of the target specimen. The horn antenna is covered with cerawool to protect it from the heating process. The reflection spectrum is received by the horn antenna, which is aligned with the incident beam and the Al_2O_3 specimen. The reflection parameters are

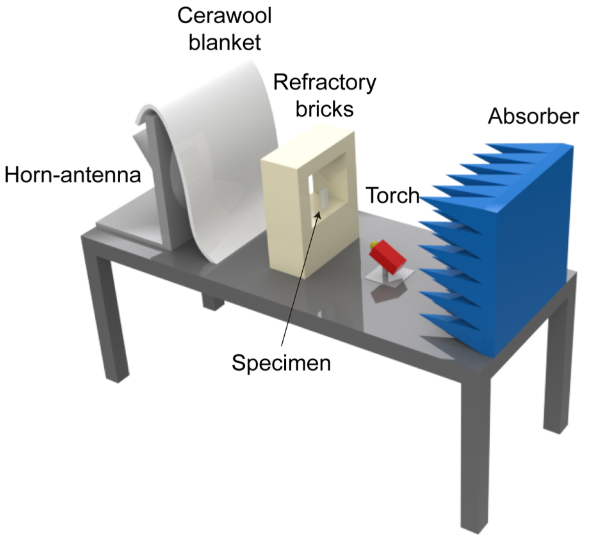

FIG. 1. Scheme of permittivity test apparatus using a single horn antenna-propane torch system at high temperature.

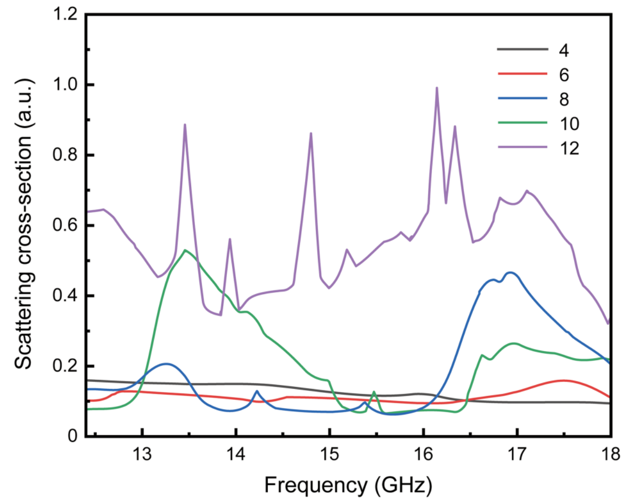

FIG. 2. Change of reflection peak position of Al_2O_3 specimen for a variety of sample sizes in the Ku-band frequency regime. Height of all samples is fixed at 40 mm.

obtained from a vector network analyzer (HP 8510C, Agilent, USA). To study the relationship between the temperature and permittivity, the temperature is varied using a mass flow controller (M3030 V, Line Tech, Korea) for both the liquefied petroleum gas (LPG) and oxygen flow rates. The LPG to oxygen ratio in the flow is fixed at 3. The specimen temperature is measured using a two-color pyrometer (Pyrosoft DSR 10NV, DIA Infrared, Germany). After evaluating the dielectric properties at room temperature, the dielectric constant of the sintered Al_2O_3 sample is measured from room temperature to 1550 °C within the frequency range of 12.4–17.5 GHz.

III. RESULTS

Before measuring the permittivity, the Mie resonance method is applied to estimate it using several assumptions. First, it is assumed that there is no temperature gradient inside the specimen during the heating because the sample size is smaller than the heating area of the torch and the size is sufficiently thin. Through this assumption, the complexity of the simulation is simplified and is feasible by reducing the number of variables that are considered. Second, the sample is assumed to be a lossless dielectric material. Since the Mie resonance method uses the singular information of the reflection peak position to derive the permittivity, it is impossible to obtain both the real and imaginary components of the permittivity simultaneously. Finally, the thermal expansion of the Al_2O_3 sample is not considered in the calculations. Based on these assumptions, the dielectric constants of the Al_2O_3 ceramics are measured from the Mie resonance.

It is essential to define the size of the target sample for which the Mie resonance occurs to ensure that the position of an intense reflection peak is within the Ku-band frequency region. As shown in Fig. 2, we predict the reflection peak position in the Ku-band region by varying the dimensions of the Al_2O_3 samples from $4 \times 4 \times 40\ mm^3$ to $12 \times 12 \times 40\ mm^3$. The height for all samples is fixed at 40 mm. It is noted that the Al_2O_3 at $8 \times 8 \times 40\ mm^3$ has obvious reflection peaks around 16.9 GHz. The room-temperature permittivity is used to design the sample size so that a clear peak occurs in the higher frequency regime of the Ku-band. Since the permittivity of Al_2O_3 ceramics generally increases with temperature, simulations with a higher permittivity than at room temperature are performed, and the results are arranged in a database to show the relationship between the permittivity and the peak position of the scattering cross section. When the specimens are large, as in case of the $12 \times 12 \times 40\ mm^3$ sample, the higher-order modes enter into the Ku-band region. Although these modes have the advantage of a high reflectance in the frequency domain, it is difficult to determine the dielectric constant from these because there are multiple modes that interact with each other. In this study, because it is preferable to use only the strongest peak, such large samples are not preferred. Moreover, as it is assumed that there is no thermal gradient on the surface of samples during heating, the dimensions of samples should be minimized to ensure that this assumption is valid. When the sample size decreases to $4 \times 4 \times 40\ mm^3$, there is no obvious resonance peak in the reflection spectrum of the Ku-band frequency range. Therefore, the sample dimensions are fixed at $8.5 \times 8.5 \times 40\ mm^3$. The measured reflection spectrum for the Al_2O_3 sample of $8.5 \times 8.5 \times 40\ mm^3$ at room temperature is given in Fig. 3. Figure 3(a) shows that the peak obtained from a single antenna system requires a postprocessing to clearly define its position due to the noise.

Firstly, a Savitzky-Golay filter is applied to smooth the experimental reflection spectrum. However, since it is difficult to distinctly define the peak position simply by applying this filter, a 3rd order polynomial curve fitting is also performed over the frequency range where reflectance has more than 70% of the maximum value. The postprocessed values clearly indicate the peak position from the experimental results. The measured and simulated reflection spectra of the Al_2O_3 samples in the Ku-band frequency region at room temperature are shown in Fig. 3(b), where an obvious peak appears from 12.4 to 17.5 GHz with a peak at 15.7 GHz. The simulated pattern for the $8.5 \times 8.5 \times 40\,mm^3$ Al_2O_3 sample corresponds with the dielectric constant of 9.77, which is well-matched with the results of other works.[19–21] Therefore, it is confirmed that the method to derive the permittivity of ceramics using the Mie theory with a single antenna is validated at room temperature.

It is important to measure the changes in the dielectric characteristics at various temperatures since ceramics with low permittivities are often employed in harsh environments. We heated the specimen to 1550 °C by increasing the gas flow rate of the heat source. No obvious deformation of the Al_2O_3 sample is observed during the experiment. Figure 4 shows shifts in the reflection spectra of the Al_2O_3 ceramic sample as a function of temperature. As can be seen from Fig. 4(a), the reflection spectrum shifts to lower frequencies with higher temperatures. The maximum peak position changes from 15.63 to 14.35 GHz from room temperature to 1550 °C [Fig. 4(b)]. In addition, the full width at half maximum (FWHM) of the resonance bandwidth gradually increases, which is in agreement with the results for the Fabry–Perot resonance.[22,23] It is well-known from the Debye model that the permittivity changes as a function of frequency and

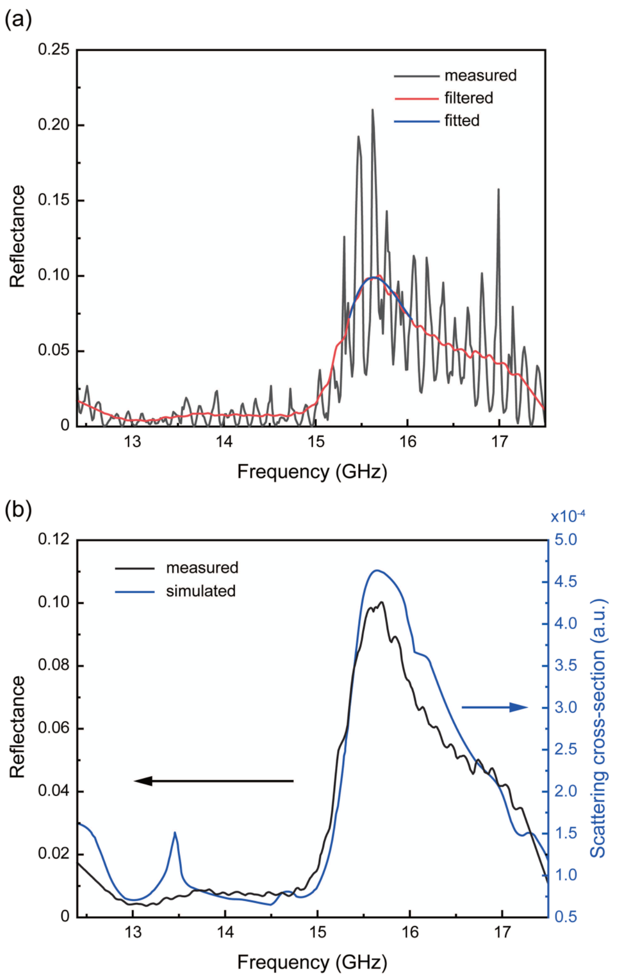

FIG. 3. (a) Measured Al_2O_3 reflection spectrum in the Ku-band region before and after using the Savitzky-Golay filter for smoothing. The blue line indicates the reflection spectrum of the Al_2O_3 specimen after 3rd order polynomial curve fitting. (b) Measured and simulated Al_2O_3 reflection spectrum at room temperature in the Ku-band region.

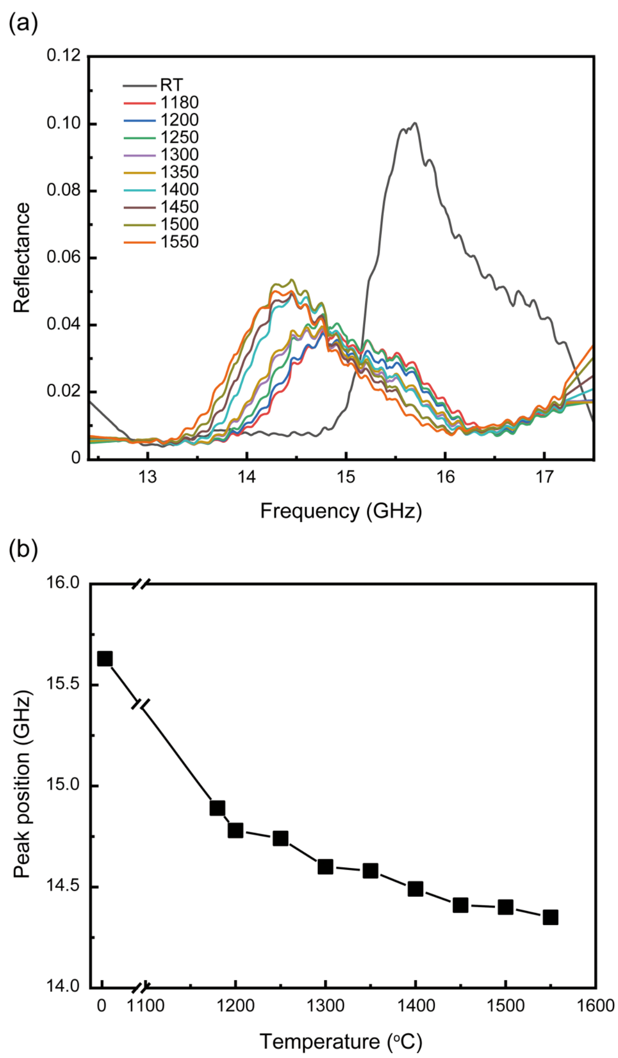

FIG. 4. (a) Change of measured reflection spectrum and (b) maximum peak position of Al_2O_3 specimens after using Savitzky-Golay filtering with an increase of temperature up to 1550 °C.

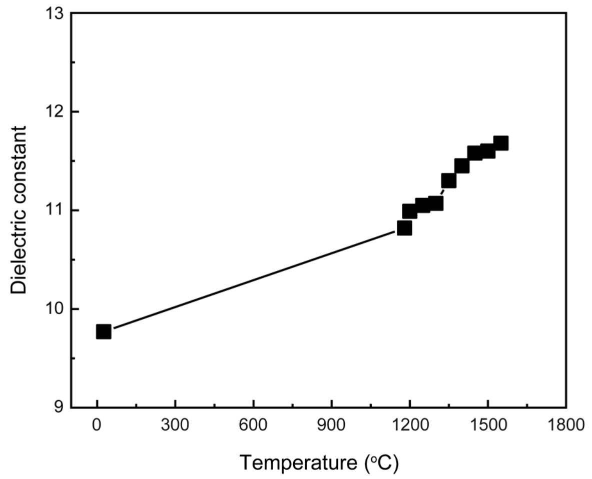

FIG. 5. Measured dielectric constant of Al_2O_3 ceramics with an increase of temperature to 1550 °C in the Ku-band frequency region.

temperature.[24–27] The real permittivity can be expressed by[28]

$$\varepsilon' = \varepsilon_\infty + \frac{\varepsilon_S - \varepsilon_\infty}{1 + \varpi^2 \tau(T)^2}, \tag{1}$$

where $\tau(T)$ is the relaxation time, ϖ is the angular frequency, ε_∞ is the high-frequency permittivity, and ε_S is the static permittivity. The relaxation time strongly depends on the sample temperature, which is described by

$$\tau(T) = \frac{1}{2\nu} e^{U/kT}, \tag{2}$$

where U is the potential barrier, ν is the vibration frequency, k is Boltzmann's constant, and T is the absolute temperature, respectively. With an increasing temperature, the thermal energy stimulates the migration of carriers for a faster response. Therefore, the relaxation time decreases at higher temperatures, thereby increasing the permittivity of the Al_2O_3 samples.[28]

Figure 5 shows the variations in the dielectric constant for the $8.5 \times 8.5 \times 40\ \mathrm{mm}^3$ Al_2O_3 sample in the Ku-band region from room temperature to 1550 °C in air. The dielectric constant varied from 9.77 at room temperature to 11.68 at 1550 °C. It is clearly seen that the measured dielectric constant of polycrystalline ceramics gradually increases with rising temperatures.[29,30] The rate of increase for the Al_2O_3 sample relative to the temperature is 0.0013/°C, which is a smaller slope compared to previous results for the permittivity as reported by Park *et al.* due to different grades and density of used specimens,[6] whereas the trends in the temperature dependence of the permittivity fit well with previous results.

IV. CONCLUSIONS

In this study, we used the Mie theory to estimate the dielectric properties of Al_2O_3 ceramics as the temperature increases up to 1550 °C. We have successfully designed a new heating and measurement system to determine the dielectric properties at high temperatures using a single horn antenna and a propane torch. The dimensions of the specimen that are needed to obtain a reflection peak in the Ku-band region were determined through simulations. The dielectric constant of the Al_2O_3 sample was also measured and was found to be consistent with the reference values. To investigate the dielectric properties of Al_2O_3 ceramics at high temperatures, the sample was heated to 1550 °C using a propane torch. The measured high-temperature permittivities were reliable, showing a slight slope with an increasing temperature. Based on these results, we demonstrated that this dielectric constant measurement technique enables the determination of information to aid in the optimal design of ceramics and can be used to accurately evaluate dielectric properties at high temperatures.

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REFERENCES

[1]I. M. Reaney and D. Iddles, J. Am. Ceram. Soc. **89**, 2063 (2006).
[2]N. Khatavkar and K. Balasubramanian, RSC Adv. **6**, 6709 (2016).
[3]C. Larsson, D. Sjoberg, and L. Elmkvist, IEEE Trans. Instrum. Meas. **60**, 2872 (2011).
[4]J. Wu and L. Kong, Appl. Phys. Lett. **84**, 4956 (2004).
[5]K. I. Murata, A. Hanawa, and R. Nozaki, J. Appl. Phys. **98**, 084107 (2005).
[6]M. K. Park, H. N. Kim, S. S. Baek, E. S. Kang, Y. K. Baek, and D. K. Kim, Solid State Phenom. **124-126**, 743 (2007).
[7]D. K. Ghodgaonkar, V. V. Varadan, and V. K. Varadan, IEEE Trans. Instrum. Meas. **38**, 789 (1989).
[8]D. K. Ghodgaonkar, V. V. Varadan, and V. K. Varadan, IEEE Trans. Instrum. Meas. **39**, 387 (1990).
[9]H. L. Bassett, Rev. Sci. Instrum. **42**, 200 (1971).
[10]A. L. Cullen, Radio Sci. **22**, 1168 (1987).
[11]J. Dhanya, A. V. Basiluddeen, and R. Ratheesh, Scr. Mater. **132**, 1 (2017).
[12]P. P. Ma, X. Q. Liu, F. Q. Zhang, J. J. Xing, and X. M. Chen, J. Am. Ceram. Soc. **98**, 2534 (2015).
[13]A. N. Unnimaya, E. K. Suresh, and R. Ratheesh, Mater. Res. Bull. **88**, 174 (2017).
[14]M. T. Sebastian, R. Ubic, and H. Jantunen, Int. Mater. Rev. **60**, 392 (2015).
[15]C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, 1983).
[16]A. I. Kuznetsov, A. E. Miroshnichenko, M. L. Brongersma, Y. S. Kivshar, and B. Luk'yanchuk, Science **354**, aag2472 (2016).
[17]J. M. Geffrin, B. García-Cámara, R. Gómez-Medina, P. Albella, L. S. Froufe-Pérez, C. Eyraud, A. Litman, R. Vaillon, F. González, M. Nieto-Vesperinas, J. J. Sáenz, and F. Moreno, Nat. Commun. **3**, 1171 (2012).
[18]P. E. Landreman, H. Chalabi, J. Park, and M. L. Brongersma, Opt. Express **24**, 29760 (2016).
[19]R. Vila, M. González, J. Mollá, and A. Ibarra, J. Nucl. Mater. **253**, 141 (1998).
[20]M. Habib Ullah and M. T. Islam, Chalcogenide Lett. **9**, 61 (2012).
[21]W. B. Westphal and J. Iglesias, *Dielectric Measurements on High-Temperature Materials* (Cambridge Laboratory for Insulation Research, 1970).
[22]H. Lu, B. Sadani, G. Ulliac, C. Guyot, N. Courjal, M. Collet, F. I. Baida, and M.-P. Bernal, Opt. Express **21**, 16311 (2013).

[23]Y. Zhang, E. Li, J. Zhang, C. Yu, H. Zheng, and G. Guo, Rev. Sci. Instrum. **89**, 024701 (2018).
[24]S. Shao, H. Luo, L. Deng, J. He, and S. Huang, AIP Adv. **8**, 075127 (2018).
[25]Y. Q. Tan, J. L. Zhang, W. T. Hao, G. Chen, W. B. Su, and C. L. Wang, Mater. Chem. Phys. **124**, 1100 (2010).
[26]C. Ronne, L. Thrane, P.-O. Astrand, A. Wallqvist, K. V. Mikkelsen, and S. R. Keiding, J. Chem. Phys **107**, 5319 (1997).
[27]Z. Haijun, L. Zhichao, M. Chengliang, Y. Xi, Z. Liangying, and W. Mingzhong, Mater. Sci. Eng. B **96**, 289 (2002).
[28]Y. Wang, F. Luo, P. Wei, W. Zhou, and D. Zhu, J. Electron. Mater. **44**, 2353 (2015).
[29]W. W. Ho, Mat. Res. Soc. Symp. Proc. **124**, 137–148 (1988).
[30]E. Li, Z. Nie, G. Guo, Q. Zhang, Z. Li, and F. He, Prog. Electromagn. Res. **92**, 103 (2009).