PSFC/JA-10-46

Thermal Analysis of Graphite and Silicon Carbide with Millimeter-Wave Radiometry

P. P. Woskov and S. K. Sundaram*

* Pacific Northwest National Laboratory

.

November 2010

Plasma Science and Fusion Center
Massachusetts Institute of Technology
Cambridge MA 02139 USA

This work was supported by the U.S. Department of Energy. Reproduction, translation, publication, use and disposal, in whole or in part, by or for the United States government is permitted.

High Temperature Thermal Analysis of Graphite and Silicon Carbide with Millimeter-Wave Radiometry

Paul P. Woskov[1] and S. K. Sundaram[2]
[1] MIT Plasma Science and Fusion Center, 167 Albany Street, NW16-110, Cambridge, MA 02139, U.S.A.
[2] Pacific Northwest National Laboratory, 902 Battelle Boulevard, K6-24, Richland, WA 99352, U.S.A.

ABSTRACT

Millimeter-wave thermal analysis instrumentation is being developed for characterization of high temperature materials required for diverse fuel and structural needs in extreme high temperature reactor environments. A two-receiver 137 GHz system with orthogonal polarizations for anisotropic properties resolution has been implemented at MIT and is being tested with graphite and silicon carbide specimens at temperatures up to 1300 ºC. Real time measurement sensitivity to submillimeter surface displacement and simulated anisotropic surface emissivity is demonstrated.

INTRODUCTION

The development of Generation IV (Gen IV) very high temperature reactor (VHTR) technology depends on the development and characterization of high temperature materials that can reliably meet the diverse fuel and structural requirements in extreme VHTR environments. New thermal analysis tools are needed as well as the compilation of data bases on the characteristics of old and new materials in various previously untested high temperature situations. Thermal analytical tools that could be used in the laboratory and eventually applied *in situ* for reactor monitoring would be of particular value. Millimeter-wave (MMW) thermal analysis methods and technologies can meet these needs and address the short comings of present material diagnostic technology due to anisotropic and small sample size issues. MMW radiation refers to electromagnetic wavelengths in the 0.1 – 10 mm range that are shorter than microwaves but longer than infrared radiation. At these wavelengths propagation is possible through infrared/optically opaque pathways and materials, but with spatial resolution much finer than with microwaves. Past work has applied MMW techniques to nuclear waste glass melter measurements [1], water-ice freezing dynamics [2], and precision high-temperature superconductor resistivity measurements [3]. In this paper, we extend the technology to thermal analysis of graphite and silicon carbide materials in high temperature environments relevant to the Next Generation Nuclear Plant (NGNP).

EXPERIMENTAL

The amplitude, phase, and polarization of MMW thermal emission and probe beam reflection are used to provide information on the thermal characteristics of the materials under observation. The use of two orthogonally polarized receivers makes possible the study of anisotropic features. A schematic illustration of the laboratory implementation of the MMW thermal analysis hardware is shown in Figure 1. The MMW receivers are heterodyne receivers that operate at a frequency of 137 ± 2 GHz (λ = 2.19 mm). Receiver 2 is polarized perpendicular to Receiver 1. Receiver 1 views the electric field direction at the test specimen that is in the plane of Figure 1 and Receiver 2 views the electric field direction that is perpendicular to the plane of Figure 1. In addition to detecting the test specimen thermal emission heterodyne receivers also leak a local oscillator (LO) signal that is used as a coherent probe beam.

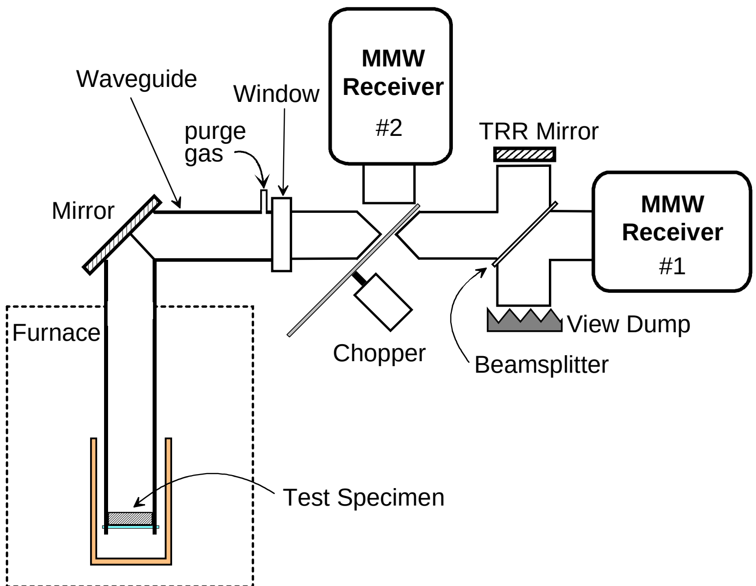

Figure 1. Schematic illustration of the MMW thermal analysis hardware setup.

Corrugated metallic waveguide (aluminum and brass) is used to propagate the MMW signals between the receivers and a miter mirror above the furnace where the waveguide material changes to smooth walled ceramic mullite to go inside the electric furnace (Deltech Model DT-31-RS-12). Test specimens were located inside the bottom of the 41 mm i. d. mullite waveguide. A Teflon window is used to seal the waveguide section on the furnace side for purge gas flow through the waveguide to displace the air and prevent uncontrolled test specimen oxidation at high temperatures. The chopper with a flat reflective blade serves two functions. It allows both receivers to view the test specimen and also modulates the signals for synchronized detection with lock-in amplifiers. When the chopper blade is not in the waveguide, the furnace signals pass to and from Receiver 1. When the chopper blade moves inside the waveguide at an angle of 45º to the waveguide axis, it reflects the furnace signals to and from Receiver 2. Typically the chopping frequency is about 100 Hz. When the receivers are not viewing the furnace, the background laboratory temperature reference is viewed. The measured thermal signals are difference signals between room temperature and sample temperature.

The other major hardware component in the setup is the beamsplitter in a four port crossed waveguide block in front of Receiver 1. A fused quartz beamsplitter was polished to a thickness of 0.90 mm to be approximately 50% reflective at 137 GHz. Half the thermal emission from the test specimen is reflected to the side port and can be partially returned to the sample by a thermal return reflection (TRR) mirror. By removing and replacing the TRR mirror on this port the thermal signal detected by Receiver 1 is modulated to a level that depends on the reflectivity of the test specimen, which in turn depends on the test specimen emissivity. The analytical formulas relating the TRR signal modulation to specimen reflectivity and emissivity have been previously derived [4]. The port opposite the TRR mirror is a signal dump and not used.

MEASUREMENTS

Displacement

The part of the waveguide that is inside the furnace is heated to high temperatures along with the test specimen. Consequently the waveguide will expand, changing the path length of the signals between the receivers and test specimen. This causes the reflected LO by the specimen to beat with itself constructively or destructively depending on the phase of the reflection as changed by the expanding path length. Since the wavelength is 2.19 mm at 137 GHz, a quarter wavelength expansion of 0.548 mm will cause the round trip signal to go through a half wavelength change or a complete cycle of minimum-maximum signal.

Figure 2 shows the LO reflection, labeled Video 1, as recorded by Receiver 1 when a graphite test specimen was heated to 1300 °C. The furnace temperature is shown by the upper thermocouple plot. The video signal consists of a number of fringes as the heated waveguide expands and then contracts as it is cooled down. The expansion of the test specimen is not significant since it is thin (2 cm) in comparison to the length of waveguide exposed to high temperature. The fringes are labeled 1 through 3 on the plot. A fourth fringe not completely formed at the maximum temperature is not labeled. When the temperature of the furnace is held constant at 520 and 1300°C, the video signal also does not change significantly confirming that this fringe pattern is due to thermal expansion.

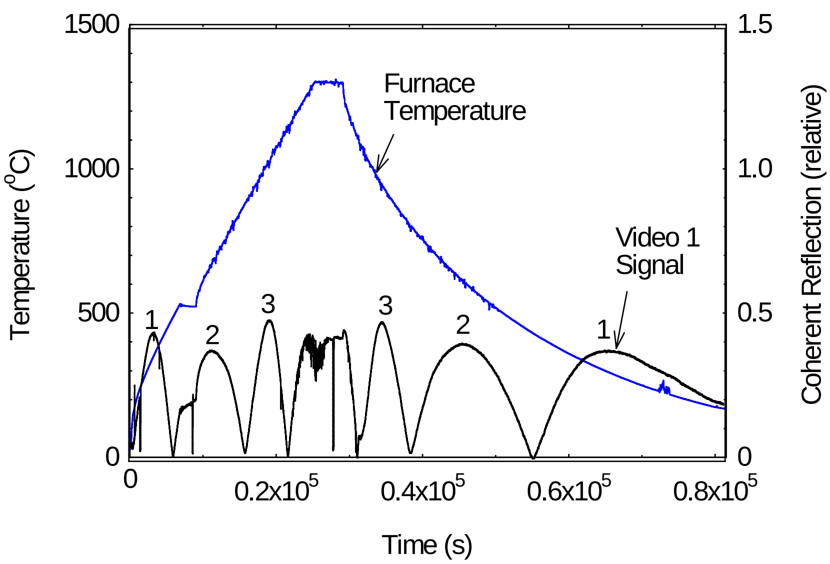

Figure 2. Expansion of the heated mullite waveguide is shown by interference fringes in the lower plot due to the reflected 137 GHz local oscillator signal beating with itself in the receiver.

The observed total fringe count of slightly over 3.5 on heat up to 1300°C corresponds to a waveguide expansion of approximately 2 mm. This is consistent with a thermal expansion coefficient of 5.4×10^{-6} /°C for mullite [5] and a 30 cm length exposed to high temperature as in the present setup. An important capability of MMW monitoring is demonstrated here to be able to observe surface swelling or erosion with sensitivity of the order of 50 μm (1/10 fringe at 137 GHz) at constant temperature. Higher MMW frequencies could be used to increase sensitivity. Even with changing temperature, having the calibration curve for the propagation path expansion, it should be possible to determine the contribution of the viewed material dimension change on the fringe pattern if it is a significant fraction of one fringe.

Graphite and Silicon Carbide Thermal Emission

Graphite and silicon carbide specimens 40 mm in diameter were acquired to fit inside the mullite waveguide. The graphite grade was GR001CC from Graphtek, LLC and the silicon carbide was molded by Ortech, Inc. These samples were polycrystalline and not expected to have anisotropic behavior in themselves, but as described further below anisotropy could be simulated by alignment. A comparison of Receiver 1 MMW thermal emission as calibrated at the window is shown in Figure 3 for a maximum flat top furnace temperature of 1300ºC for each sample. The MMW thermal signal is proportional to the product of emissivity and temperature (εT) and thus will be different at a given temperature if the emissivity is different.

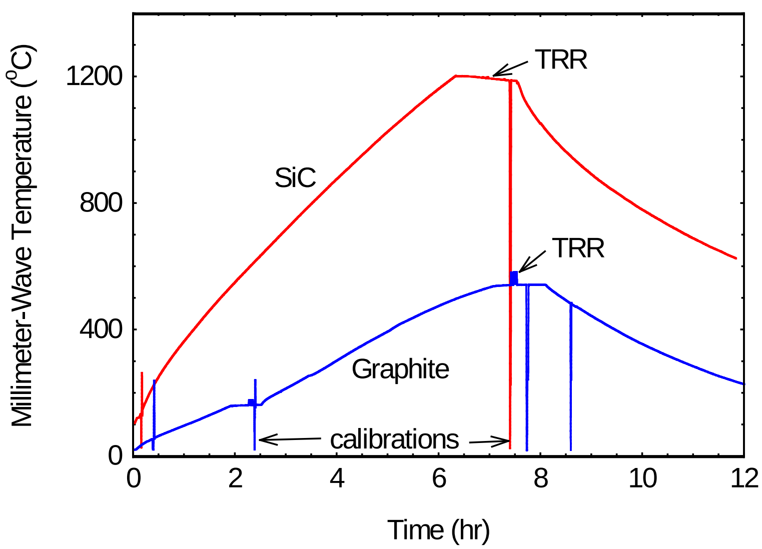

Figure 3. Comparison of the MMW thermal emission from graphite and silicon carbide for heating up to 1300 ºC showing the emissivity of SiC to be much higher than graphite .

It is readily evident that the 137 GHz emissivity of SiC is much higher than graphite. The TRR modulation provides another indication of emissivity that does not depend on an independent temperature measurement. It is larger for graphite and very small for SiC. Previous TRR measurements at 137 GHz have shown the emissivity of low resistivity SiC to be 0.76 [4]. Graphite with its high electrical conductivity would be expected to have emissivities < 0.20 in the MMW range, depending on surface condition. A surface roughness larger than 1/10 wavelength, as could be caused by surface corrosion would also increase surface emissivity.

Graphite Dual Polarization Measurement

The graphite sample was a 2 cm thick disk inserted into the end of the mullite waveguide and supported by a 5 cm long inner fitting mullite tube that kept the sample surface aligned orthogonal to the waveguide axis throughout the thermal cycling. The dual polarization data for a warm up to 520 ºC is shown in Figure 4. The MMW thermal emission observed in the two orthogonal polarizations is similar except for the time when a TRR measurement is done on Receiver 1. The ratio of the Receiver 1 to Receiver 2 signals is constant and approximately equal to 1.0 as expected for an isotropic sample.

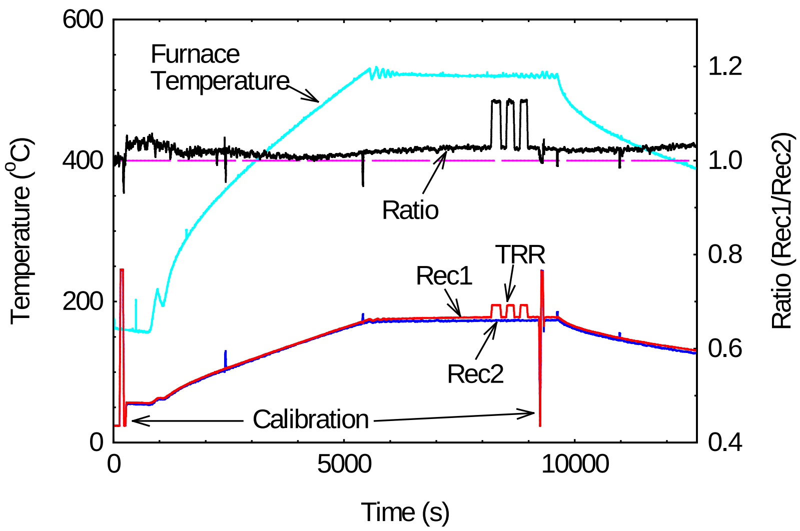

Figure 4. Data for a dual polarization measurement of graphite showing the furnace temperature, the two MMW receiver temperatures, and the ratio of the receiver temperatures, which is constant near 1.0 as expected for an isotropic sample.

Silicon Carbide Dual Polarization Measurement

The silicon carbide disk was only 1 cm thick and supported by one 3 mm diameter alumina rod behind the sample that was inserted across the center of the mullite waveguide diameter. There was no extended inner support to prevent wobble due to the disk diameter being slightly smaller than the waveguide i. d. The support rod was oriented along the electric field direction of the Receiver 2 view, maximizing the wobble polarization difference between the receiver views. The lack of sufficient support to prevent sample wobble was a setup error, but has provided data on the sensitivity of the instrumentation to detect anisotropic characteristics as shown in Figure 5. The furnace with a SiC sample was first heated to 520 ºC and then to 625 ºC followed by a long cool down. The MMW thermal signal ratio of the two receives is seen to change with warm up and then return on cool down. Since the sample was a polycrystalline material with no anisotropy to cause this behavior, it is attributed to the alignment change of the sample with waveguide thermal expansion. It is well known that the refection from an angled dielectric is sensitive to polarization and can be shown to be of the right magnitude to explain the observations. This result also shows promise for measuring any misalignment of critical structural materials in a reactor.

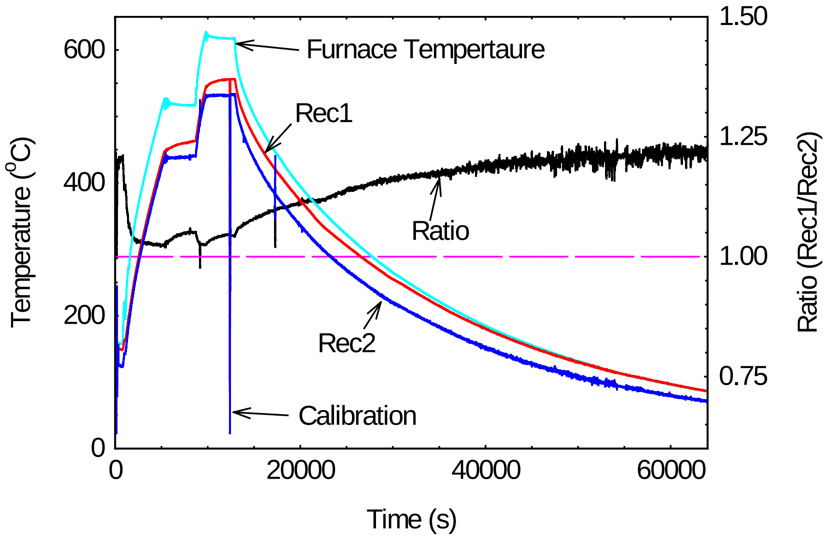

Figure 5. Dual polarization measurement of silicon carbide in a wobbly setup showing sensitivity to polarization as the sample tilts during the heating cycle.

CONCLUSIONS

The MMW range of the electromagnetic spectrum is well suited for materials science research relevant to the extreme environments of future reactors. MMW techniques can access extreme high temperature environments for real time data at engineering resolutions that avoid small size issues. The amplitude, phase, and polarization of MMW signals propagating through dielectrics and reflecting from material boundaries, discontinuities, hybrid/composites, and metallic surfaces can provide comprehensive information on the characteristics of the materials and interfaces. Our results show that real-time measurements would be possible to track material reactions, phase transformations, volumetric changes, and aging. In addition, the MMW sensor approach would be particularly applicable to simultaneous characterization of both bulk and surface transformation processes by simultaneous sensitivity to emissivity (phase transitions/corrosion) and displacement (creep) changes. Polarization sensitive measurements could unambiguously track anisotropic material characteristics of particular relevance to advance ceramic composite materials (e.g., SiC_f-SiC) where little data is now available.

REFERENCES

1. P. P. Woskov, S. K. Sundaram, G. Daniel, J. S. Machuzak, P. Thomas, "Millimeter-Wave Monitoring of Nuclear Waste Glass Melts – An Overview", *Environmental Issues and Waste Management Technologies VII*, (Ceramic Transactions, Vol. 132, 2002) pp. 189-201.
2. S. Sundaram, S. K. Sundaram and P. P. Woskov, *Journal of Infrared, Millimeter, and Terahertz Waves*, DOI 10.1007/s10762-008-9457-3, (2009).
3. P. P. Woskov, D. R. Cohn, S. C. Han, A. Gatesman, R. H. Giles, and J. Waldman, *Rev. Sci. Instrum*. **65**, 438-444, (1994).
4. P.P. Woskov and S. K. Sundaram, *J. Appl. Phys.* **92**, 6302-6310 (2002).
5. http://accuratus.com/mullite.html