

# Microwave frequency material properties of PBS 9501 and PBX 9501 and small scale heating experiments

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**Abstract.** This work reports the microwave frequency dielectric properties of PBX 9501 and one of its representative mocks, PBS 9501, within 1-20 GHz. From these measurements it is shown that the binder system has a strong influence on microwave heating of such compositions resulting in significant temperature gradients within the individual HMX or sugar crystals at high microwave heating rates. Using the measured dielectric properties, COMSOL 4.3 Multiphysics was used to simulate and optimize a microwave applicator with a high electric field to input power ratio. The simulated applicator design indicated subsecond heating to decomposition for PBX 9501 and was validated with small scale experiments on both PBS 9501 and PBX 9501. At approximately 2.45 GHz and 100 W applied power, PBS 9501 decomposition was observed shortly (< 34 ms) after a measured surface temperature of 70 °C (binder system melts). Finally, rapid heating of PBX 9501 was also shown in the optimized cavity.

## 1. Introduction

Neat explosives, such as HMX, RDX, etc. are regarded as low dielectric loss materials and therefore poor absorbers of microwave energy [1-3]. Consequently, microwave irradiation of plastic bonded and neat explosives has typically been studied with the inclusion of high dielectric loss (absorptive) particles like silicon carbide or carbon nanotubes in order to lead to effective heating and ignition [3-6]. As a result, there is little work, to the author's knowledge, exploring solely the influence of the binder system on the possibility of achieving volumetric microwave heating. While the binder system is only a minor constituent (3-30 vol.%) in some cases it may be much more absorptive than the neat energetic crystals themselves and may lead to significant localized heating.

Therefore, the objective of this brief study was to measure the relative complex permittivities of both PBX 9501 and its mock PBS 9501 and examine the role binder system has on such compositions when subjected to microwave irradiation, both theoretically and experimentally.

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## 2. Experimental

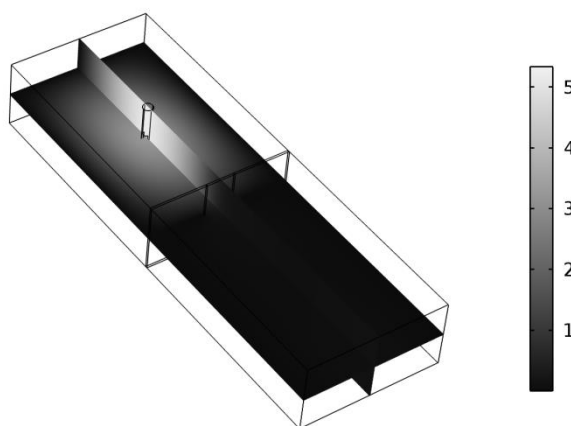
### 2.1. Complex permittivity measurements

**2.1.1. Materials and preparation.** PBX 9501 (HMX/Estane/BDNP-F/A – 95/2.5/2.5 wt. %) received from Los Alamos National Laboratory was used as received. PBS 9501 (Sugar/Estane/BDNP-F/A – 95/2.5/2.5 wt. %) was prepared following the procedure of Lui et al. [7]. The initial particle size for PBS 9501 is such that it closely matches that used for PBX 9501. Materials were pressed into pellets with ODs of 6.35 mm and 12.7 mm and heights of 10.16 mm and 5.08 mm, respectively. A Carver 12 Ton hydraulic assembly was used to press pellets to high density, typically above 95 % of the theoretical maximum density (TMD).

**2.1.2. Apparatus and permittivity extraction.** Prepared samples were measured in a Damaskos Model 400 Circular Cavity with a diameter of 10.16 cm and inserts with thicknesses of 10.16 mm and 5.08 mm. An Agilent 8364B or 8362B Vector Network Analyzer was used in conjunction with CAVITY<sup>TM</sup> software from Damaskos to calculate the complex permittivity by measuring the change in resonant frequency and quality factor between the unloaded and loaded cavity between 1 and 20 GHz. Interested readers may refer to [8] for an explanation of an analytic model used to calculate the permittivity from the shift in frequency and quality factor. Finally, the Maxwell-Garnett mixing formula was used to correct for density where  $\epsilon_{\text{eff}}$  was the effective permittivity of the mixture,  $\epsilon_e$  is the spatially averaged permittivity of the composition,  $\epsilon_i$  is the permittivity of the inclusion (in this case air,  $\epsilon_i = 1$ ), and  $f$  is the volume fraction of the inclusion [8].

### 2.2. Experimental Microwave Cavity

**2.2.1. Cavity Geometry.** A microwave applicator with a high electric field to input power ratio was built based on an optimized TE<sub>10n</sub> cavity design (figure 1) using Gerling waveguide components. Full details can be found in reference [9]. In brief, the applicator consists of a three half wavelength ( $n = 3$ ) WR-284 cavity with a numerically (COMSOL 4.3 Multiphysics) optimized inductive iris for enhanced coupling. Within the cavity a 3 mm ID by 5 mm OD fused quartz tube serves as the sample holder. A 3 mm OD unit aspect ratio cylinder of the material of interest at 50-60 % of TMD was hand pressed within the sample holder. The cavity was operated at 2.4391 GHz (Agilent N9310A signal generator) and 100 W (Empower 1189 high power amplifier). The temperature of the surface of the pellet was monitored using a FLIR model A325sc camera (30 fps). An emissivity of 0.95 was assumed based on previous work [10].



**Figure 1.** Normalized electric field distribution in an inductive iris coupled TE<sub>10n</sub> cavity. The inductive iris aperture is 0.2 times the width of the cavity and a 3 mm OD unit aspect ratio PBX 9501 cylinder is held by a 5 mm OD fused quartz tube.

### 3. Results and Discussion

#### 3.1. Permittivity measurements and analytic heating model

**3.1.1. Measurements.** The average relative complex permittivity, in the range of 1-20 GHz, is found to be  $3.58-0.039i$  for PBX 9501 and for PBS 9501,  $4.02-0.068i$ . In contrast the average relative complex permittivity, in the same frequency range, of HMX is  $3.50-0.003i$  [9]. From these measurements it is clear that addition of a small volume fraction,  $\sim 7\%$ , of the binder system Estane-BDNP-F/A to HMX will dramatically increase the per unit volume microwave absorption for a given spatially averaged electric field value. This increase in microwave absorption is so significant (1200 %) that microwave absorption in the HMX particles can practically be neglected. Heating of the individual particles within the PBX 9501 sample is therefore dominated by binder absorption and particle thermal conductivity and size.

**3.1.2. Heating Model.** Assuming uniform heating in the binder, a time dependent temperature boundary condition for the crystals can be assumed. Given the particle size distribution of HMX crystals in pressed PBX 9501 (figure 2 [11]) the temperature difference between  $r = r_{\max}$  and  $r = 0$  in the individual binder bounded particles can be calculated to a reasonable approximation with the following analytical expression [12]:

$$\Delta T = - \left( mt \left( \left( 1 + \frac{R^2 - 1}{6F} \right) - \frac{2}{\pi^3 F} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^3} e^{-n^2 \pi^2 F \frac{\sin n\pi R}{R}} \right) + T \right), \quad (1)$$

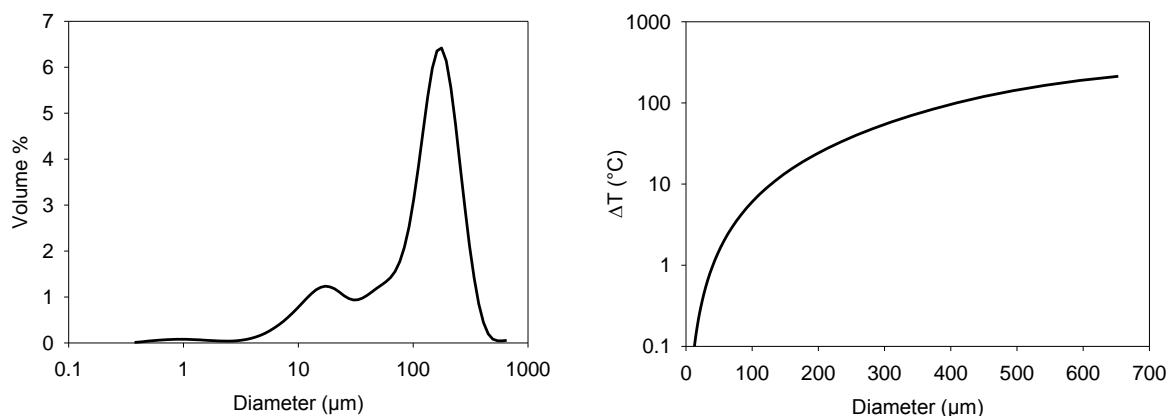
where

$$F = \frac{kt}{\rho C_p R^2}, \quad (2)$$

and

$$m = \frac{5.56 \times 10^{-11} \epsilon'' f E_{RMS}^2}{\rho C_p} \quad (3)$$

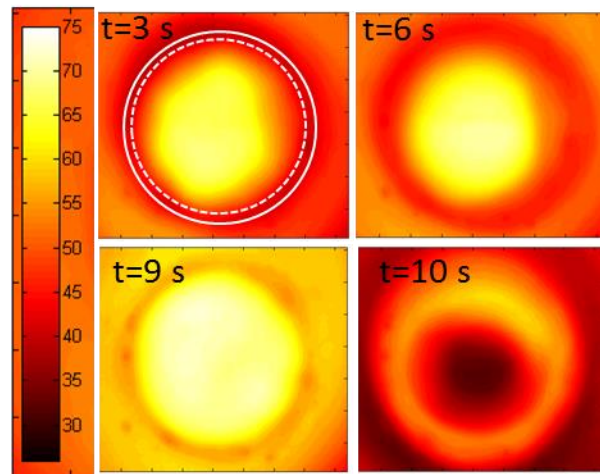
with  $R = r_{\max}$ ,  $k$ -thermal conductivity,  $\rho$ -density,  $C_p$ -heat capacity,  $\epsilon''$ -relative imaginary permittivity,  $E_{RMS}$ -root mean square electric field, and  $f$ -frequency. Calculations using the simple analytic model above show that due to significant preferential heating of the absorptive binder and the effects of particle size, increasingly localized heating is expected for microwave power absorption above about  $4.7 \text{ GW/m}^3$ . For microwave power absorption of  $4.7 \text{ GW/m}^3$  for 128 ms and the particle size distribution shown in figure 2 below for PBX 9501, only HMX particles below a diameter of approximately  $50 \mu\text{m}$  can be considered approximately, uniformly heated to  $1^\circ\text{C}$  of the binder temperature.



**Figure 2.** (left) Size distribution of HMX crystals in pressed PBX 9501 taken from reference [11]. (right) Temperature difference between  $r=r_{\max}$  and  $r=0$  for individual binder bound HMX particles.

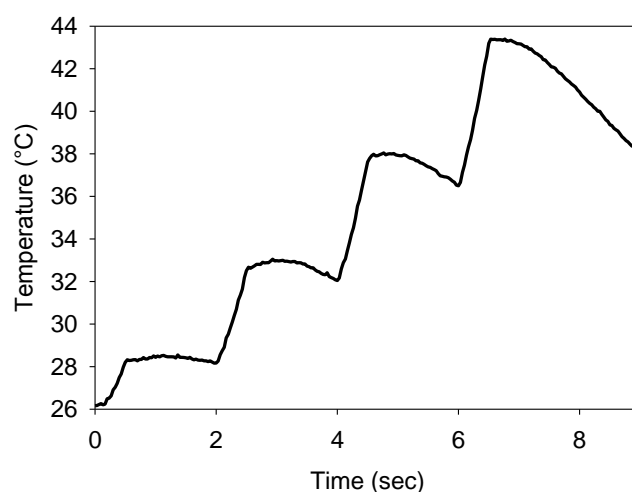
### 3.2. Applied cavity heating

**3.2.1. Microwave heating of PBS 9501.** To validate the cavity design and prediction we first considered heating experiments of the sugar mock, PBS 9501. The sample was subject to pulsed applied power with varying pulse widths (0.25 s, 0.5 s, 0.75 s, 1.0 s). In all experiments rapid heating occurred but non-uniform temperatures within the sample were observed. For brevity, results are only shown for a pulse width of 1 s. As indicated by the surface temperature distributions (figure 3), non-uniform heating was observed due to electric field variations and energy localization. Decomposition of the sample was observed less than 34 ms after a measured surface temperature of greater than 70 °C was achieved. This is near the temperature at which the binder system is expected to begin melting [13] and exhibit increased susceptibility to microwave heating. While these experiments clearly show the feasibility of rapid heating of the sample to the point of decomposition or ignition, they also demonstrate the requirement for temperature measurements with improved temporal resolution [3].



**Figure 3.** Surface temperature for a 3 mm OD, 50 % TMD PBS 9501 sample subject to 100 W input power in the WR-284 applicator operating at 2.4391 GHz. Note: Color mapping corresponds to temperature in °C as indicated by the scale on the left. White lines on first frame indicate approximate location of the sample holder outer and inner edges.

**3.2.2. Microwave heating of PBX 9501.** A 3 mm OD unit aspect ratio cylinder of PBX 9501 was also subjected to microwave heating in the WR-284 applicator. The surface temperature as a function of pulse duration is shown in figure 4. A pulse width of 0.5 s was used with a total duration of 2 s to avoid the decomposition observed in the PBS 9501 experiment. This demonstrates the same trend observed for PBS 9501 – rapid heating of the binder system and HMX particles during the pulse (0.5 s) – followed by conductive heat transfer to the sample holder (1.5 s) and shows the feasibility of microwave heating low dielectric loss plastic bonded explosives in reasonably short time scales.



**Figure 4.** Maximum temperature observed on the surface of a 3 mm OD, 60 % TMD PBX 9501 sample exposed to 0.5 s pulse width, 2.4391 GHz electromagnetic energy at 100 W input power.

#### 4. Conclusions

It is observed from complex permittivity measurements of the plastic bonded explosive PBX 9501 and one of its representative mocks, PBS 9501 that in contrast to neat HMX these composite materials are an order of magnitude more susceptible to microwave heating. This is directly attributed to the binder system Estane-BDNP-F/A. Microwave irradiation of plastic bonded explosives with an Estane-BDNP-F/A binder system, or a binder system with comparable dielectric loss, will result in preferential energy deposition within the binder, which can lead to highly inhomogeneous heating at high heating rates. It was shown for the sample size considered in this study that when an applied power of greater than 100 W is considered less than 20 vol.% of the HMX crystals in the composite will be within 1 °C of the binder temperature. Experimentally, when such samples were exposed to approximately 100 W applied power (2.4391 GHz) somewhat non-uniform surface temperatures were observed. Full decomposition of the sugar mock, PBS 9501 was observed in less than 34 ms after a measured surface temperature of about 70 °C was observed, which was after 9 s of microwave heating. The rapid onset of decomposition occurring in less than 34 ms indicates that the microwave absorption of the binder system Estane-BDNP-F/A is significantly increased at temperatures greater than 70 °C.

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