

# The influence of microwave frequencies at high temperatures on structural properties of h-BN

H.A. Al-Jawhari<sup>\*</sup>, T. A. Baeraky and Y. H. Afandi

**Abstract**—The influence of microwave frequencies at high temperature on the structural properties of Boron Nitride (BN) powder had been investigated. Crystallite size, microstrain and the graphitization index (G.I.) were determined before and after the microwave measurements. The x-ray diffraction patterns showed a remarkable increase in both h(002) and h(004) reflections. This means that after the microwaves treatment the preferential orientation factor increased and the powder became more crystallized. This increase in the crystallinity was confirmed by measuring the graphitization index (G.I.), which was found to be decreasing from 1.5 to 1.24, and further established by the small red-shift of the 1363  $\text{cm}^{-1}$  h-BN band in the FTIR spectrum. The SEM micrograph further confirmed this conclusion by showing the grains more closely packed with less porosity.

**Index Terms**—Hexagonal Boron Nitride, microwaves, graphitization index, dielectric properties.

## I. INTRODUCTION

The remarkable physical properties of Boron Nitride are mostly governed by its atomic structure. In its hexagonal phase, BN layers are bonded by weak Van der Waals forces, which enable the layers to slide easily against each other. As a result, h-BN can be used as a release agent or solid lubricant and can be easily machined to produce complex shapes. Another advantage of h-BN is its significant stability even in the presence of moisture, a fact which makes it a promising support for catalysts working under high temperature in presence of humidity [1]. It shows as well a poor wettability by many glass and metal melts. In addition, h-BN is a good candidate for application in power electronics since it has the ability to be doped whether p or n type along with the capability to respect the substrate surface [2]. Other important properties of BN are its high-temperature resistance,

thermal shock resistance, high-thermal conductivity, non-toxicity and environmental safety.

The effect of microwaves on the electrical conductivity of BN had been studied previously [3]. The results showed that Boron Nitride is transparent for microwave radiations up to 1000 °C. However, above that temperature BN absorbed some microwave frequencies and therefore its electrical resistivity decreased. In this work, we investigate the influence of microwave frequencies at high temperature on the structural properties of this nitride ceramic.

## II. SAMPLE CHARACTERIZATIONS

In this study we used a commercial white powder Boron Nitride of density 2.25  $\text{g cm}^{-3}$  supplied by (Goodfellow). Cavity perturbation technique was used to measure the complex permittivity of BN powder in microwave frequencies, 615 MHz, 1412 MHz, 2214 MHz, 3017 MHz, and 3820 MHz at temperature ranges from 25°C to 1700°C. In order to study the effect of these microwave measurements on the structural properties of BN, the powders were characterized by XRD, FTIR and SEM techniques.

XRD patterns were recorded on an Xpert x-ray powder diffractometer ( Philips PW1398) using  $\text{CuK}\alpha$  radiation ( $V=40$  KV,  $I=30$  mA); with a step size of ( $2\theta = 0.05^\circ$ ). The XRD spectrum of Si crystal was used as a standard to calibrate the scanning angles.

Fourier transformation infrared spectroscopy (FTIR) measurements were carried out using Perkin Elmer Spectrum 100 spectrometer with the wave number resolution of ( $4 \text{ cm}^{-1}$ ) and 50 scans.

The morphology of the powder was analyzed by the scanning electron microscope (SEM). Samples were mounted on metal stubs with double sided tape, covered with gold under vacuum and photographed by a JEOL/EO JSM-6360 (30 KV) unit.

## III. RESULTS AND DISCUSSIONS

### A. Dielectric property

The temperature and frequency dependence of the two parts of the complex permittivity  $\epsilon'$  and  $\epsilon''$ , was studied earlier [3]. The obtained results are presented in Fig.1. This figure shows

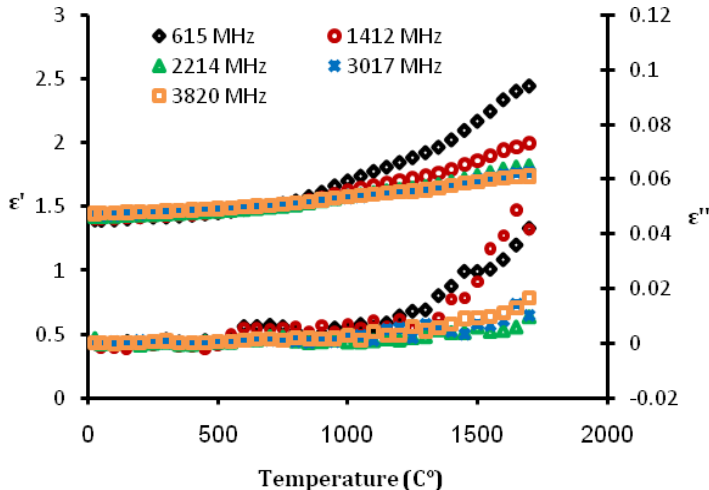
Manuscript received January 8, 2011.

H. Al-Jawhari, Department of Physics, King Abdulaziz University, Jeddah, Saudi Arabia (\*Corresponding author: e-mail: haljawhari@kau.edu.sa, Phone: +966 2 640 0000 Ext.: 26904.)

T. Baeraky, Department of Physics, King Abdulaziz University, Jeddah, Saudi Arabia (e-mail: tbaerky@kau.edu.sa)

Y. H. Afandi, Department of Physics, King Abdulaziz University, Jeddah, Saudi Arabia (e-mail: yafandi@hotmail.com).

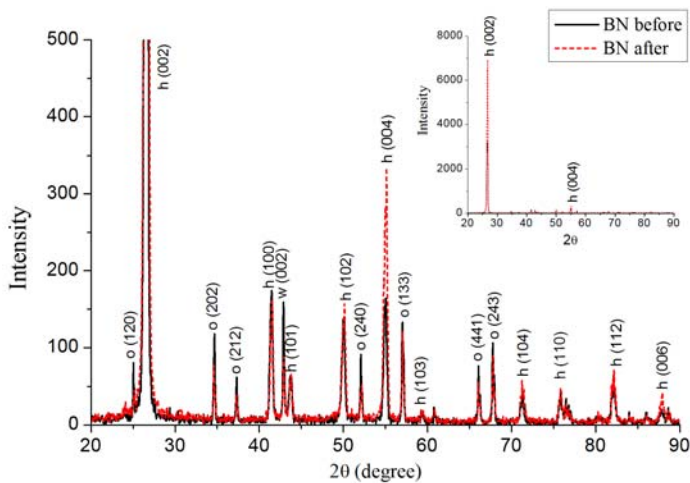
that the real and imaginary parts  $\epsilon'$  and  $\epsilon''$  are varied, respectively, from 1.5 to 2.5 and from zero to 0.05 during the temperature range of 1000 to 1800°C. These notable variations occurred at the frequencies 615 MHz and 1412 MHz. The effect of higher frequencies on  $\epsilon'$  and  $\epsilon''$  was negligible.



**Fig. 1.** The temperature and frequency dependence of the complex permittivity  $\epsilon'$  and  $\epsilon''$ , of BN.

### B. Fine structure

XRD reflections of BN powder before and after microwave measurements are shown in Fig.2. The crystallite size measured in X-ray diffraction is concerned with the space region that reflects the radiation in a coherent manner which is not identical with the grain size as measured by SEM.



**Fig. 2.** The XRD pattern of BN powder before and after microwave measurements were performed.

XRD reflections show that the main crystalline structure of our powder is the hexagonal phase [4], with some combination of orthorhombic [5,6] and wurtzite [7,8]. The cell parameters  $a$  and  $c$  of h-BN were calculated using Cohen's method and their values are shown in Table 1.

TABLE 1

Experimental parameters derived from x-ray diffraction for the h-BN phase.

Parameter	Before microwave measurements	After microwave measurements
$a$ (Å)	2.511	2.507
$c$ (Å)	6.672	6.666
G.I.	1.56	1.24
$D$ (Å)	239.13	301.37
$\epsilon$	-0.00047	0.001455
$La_{100}$ (Å)	495.565	460.687
$Lc_{002}$ (Å)	220.646	250.143
$La/Lc$	2.246	1.842

After the microwaves treatment intensities of all h-BN Bragg peaks, except for (100), increased indicating a little enlargement in the crystallite size. The effect was more noticeable at (002) and (004) reflections since the tendency of the particles to arrange themselves with axis perpendicular to the free surface of the powder is more pronounced if the particle size is large [9]. On the other hand, both lattice constants  $a$  and  $c$  of h-BN found to be decreased by a 0.16% and 0.08% respectively.

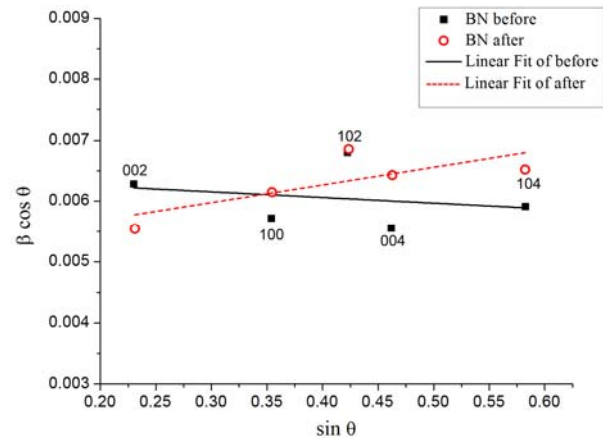
Crystallite size and microstrain were determined before and after the microwave treatment from the broadening of XRD peaks using Hall-Williamson formula

$$\beta \cos \theta = \frac{\lambda k}{D} + 2\epsilon \sin \theta$$

where  $\beta$  is the full width at half maximum (FWHM),  $\lambda$  is the wavelength of  $CuK\alpha$ ,  $k$  is a shape factor ( $\approx 1$ ),  $D$  is the crystallite size and  $\epsilon$  is the microstrain. The FWHM were measured after removing the instrumental broadening effect  $b$  according to Gauss approximation

$$\beta = B^2 - b^2$$

Where  $B$  is the experimental broadening. The corresponding results are presented in Fig.3 and Table 1.



**Fig.3.** Hall-Williamson plots before and after the microwave measurements

The observable microstrain (0.001455) after the treatment indicates an increase in the material stiffness.

To specify how flaky the crystallites of h-BN are, we calculated the aspect ratio  $L_d/L_c$  (average diameter / average thickness) based on the well known Sherrer method.

In this case, we have used the values  $k=1.84$  for obtaining the size parameter  $L_{a100}$  and  $k=0.9$  for obtaining the size parameter  $L_{c002}$ . Those values were proposed by Warren [10] and recently used by Balint and Petrescu [9] when Sherrer relationship was applied to hexagonal structures perfectly ordered in their basal plane but randomly stacked in the perpendicular direction. From Table 1, it seems that the crystallites became less flaky after the treatment.

### C. The degree of crystallization

The degree of crystallization of the h-BN phase was evaluated in terms of the "graphitization index" (G.I.) as

$$G.I. = \frac{Area[(100) + (101)]}{Area[(102)]}$$

Based on this definition, a higher value of G.I. would mean less three-dimensional ordering in h-BN and vice versa. According to [9] powders with G.I. value 1.6 are considered to have the completely graphitized (crystallized) h-BN and a value of about 50 is considered to be the upper limit for the least three dimensionally ordered h-BN. The data in Table 1 shows a reduction in the value of G.I after microwave treatment which means the powder became more graphitized.

Fig.4 shows the FTIR spectrum of the sample before and after microwave and heating effects. The strong band at  $775\text{cm}^{-1}$  refers to h-BN out-of-plane bending vibration, while the one at  $1363\text{cm}^{-1}$  stands for h-BN in-plane B-N bond stretch. It obvious that h-BN is the dominant phase in the sample. The wide broadening of these peaks indicates overlapping with other BN phases; mainly orthorhombic and wurtzite [8].

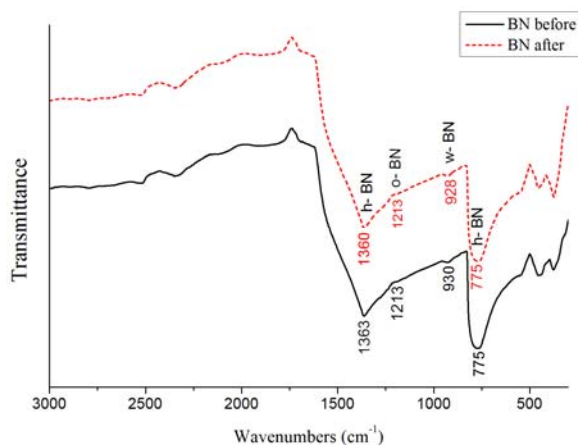


Fig.4. FTIR spectra of BN powder before and after the microwave measurements

After microwave measurements were done, the only observed change is a small red-shift from  $1363\text{cm}^{-1}$  to  $1360\text{cm}^{-1}$  which indicates a small improvement in the

crystallinity of the sample [11] which confirms the previous results.

### D. Microstructure

The SEM micrographs of the polished surfaces of BN powders before and after the microwave measurements are shown in Fig.5 (a) & (b) respectively. The SEM images indicate that the BN grains in both cases have the pin-leaf-like shape with an average grain size ranges from 3–15  $\mu\text{m}$  in diameter. However, as obviously seen from Fig. 5 (b), after the powder had been treated by microwaves at high temperatures it became more crystallized with less porosity. This decrease in porosity and the increase in the percentage of crystallinity of h-BN have a direct effect on increasing the dielectric constant.

Similar dependence of dielectric constant on the crystallite size and porosity has been reported on other materials such as diamond,  $\text{TiO}_2$  and  $\text{Bi}_2\text{O}_3$  [12-14].

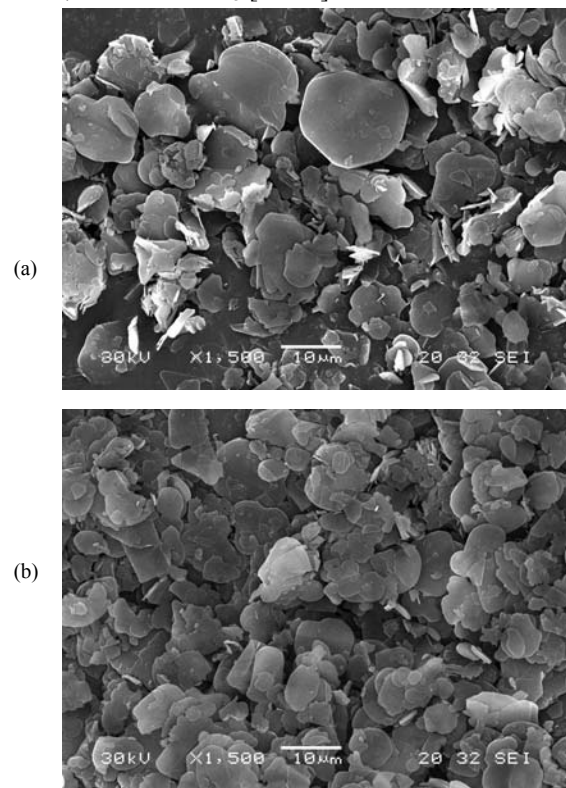


Fig. 5. The SEM micrographs of BN powder; (a) before and (b) after the microwave measurements.

## IV. CONCLUSION

Our results indicate that the regular treatment of microwaves range from 615 MHz to 1412 MHz along with heating up to  $1800^\circ\text{C}$ , does not affect the phase of h-BN. However, it causes the structure of this ceramic to be more crystalline with less porosity; this in turn affects its dielectric

characteristics and allows it to absorb some microwave frequencies at temperatures over 1000°C.

#### ACKNOWLEDGMENT

Authors would like to thank Dr. S. Al-Heniti for his technical support during XRD measurements.

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