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Microwave Dielectric Properties Measurements of Silicon Nitride at High Temperature

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ABSTRACT. The microwave dielectric properties of a powder ceramic, silicon nitride SiN will be measured using two cavity perturbation techniques in two different places: King Abdulaziz University Jeddah K.S.A and Nottingham University UK. The frequencies and the temperature ranges of these techniques, for the microwave dielectric properties are: 615 MHz, 1412 MHz, 2214 MHz, 3017 MHz, 3820 MHz and from 25 °C to 2000 °C respectively. Results of these measurements will determine the changes of the measured complex permittivity, ε, of SiN, at certain industrial microwave frequency ranges and at high temperature. The temperature and frequency-dependent electrical conductivity, σ, the activation energy, E_A, and the frequency exponent, n, of SiN will be calculated using the imaginary part, ε'', of the measured complex permittivity. Studying the changes of the measuring and the calculating electrical properties of SiN at microwave frequency ranges and at high temperature will help to understand the microwave material interaction. This understanding is very important to control the microwave processing of ceramics.

Introduction

Microwaves lie between radiowave frequencies (RF) and infrared frequencies in the electromagnetic radiation spectrum. Microwaves can be reflected, absorbed and transmitted by materials. Clark (1997) has shown that microwave energy is generated in the form of heat inside the material as a result of microwave absorption within the material, *i.e* what is called self-heating. Microwave heating has been successfully used for a wide range of applications such as drying, polymerization, food pasteurization and cooling, etc. Shawn *et al.*, (2003) and Xu., *et al.*, (2001) have shown that microwave processing of materials is a technology that can provide the ceramic material processor with a new, powerful, and significantly different tool to process materials that may not be amenable to conventional means of processing; or to improve the performance characteristics of existing materials. Several advantages over conventional heating techniques are realized because microwave energy is coupled directly into the inside of the object.

Ceramics are a class of materials broadly defined as inorganic and nonmetallic solids. They are divided into two groups: traditional ceramics (e.g. pottery, clay) and engineering ceramics such as: carbides (SiC), pure oxides (ZrO), and nitrides (SiN). Engineering ceramics are used and considered for high specific strength and high temperature applications due to their very high bond strengths. They display a wide range of properties which facilitate their use in many different product areas (Seal S. *et al.*, (2004); Zambov *et al.*, (2003); Alford *et al.*, (1998)). Microwave processing of ceramics such as: joining and sintering ceramics provide both high energy and time saving; Shulman *et al.*, (2003); Plucknett & Wilkinson (1995); Fernie (2001); Birnbon & Carmel (1999).

Unfortunately, there are two major problems facing the microwave processing of ceramics, inverse profile and thermal runaway. Inverse profile arises when the material inside is much hotter than the surrounding while thermal runaway occurs when one section of the material absorb the microwave energy preferentially over the bulk material. Lewis (1992) has shown that insufficient understanding of these problems and inadequate equipment in the form of applicators and generators of microwave energy are the reason for the failure of the microwave technology to more effectively penetrate the industrial market. It has been shown (Coccioli *et al.*, (1999); Meng *et al.*, (1994)) that the dielectric properties or the complex permittivity of any material plays an important role in microwave technology. Studying of dielectric properties of materials at different frequencies and different temperatures can be helpful in understanding the microwaves materials interaction. These properties can be determined by using the technique that is based on the cavity perturbation theory (Baeraky (2002)).

Silicon nitride has the best overall combination of properties and thus is the leading candidate for use as high-temperature structural ceramic. It has been shown (Vargheese & Rao (2001)) that silicon nitride is an important material in microelectronics due to its high resistivity, higher dielectric properties compared to silicon dioxide, mechanical strength, and chemical inertness. In the current work the dielectric properties of SiN measured by using the two new cavity perturbation techniques built in two different places: King Abdulaziz University Jeddah K.S.A and Nottingham University UK. Comparing the results of these techniques and studying the microwave interaction with silicon nitride material at high temperature and different frequencies are the aim of this work.

Materials and Methods

Full computerized systems in two different places: King Abdulaziz University Jeddah K.S.A and Nottingham University UK based on the cavity perturbation technique have been used to determine the dielectric properties of ceramic materials. The functions of these systems are described in detail (Baeraky (2002)). The system shown in Fig. 1 is developed in King Abdulaziz University Jeddah K.S.A to measure the dielectric properties of materials with conventional furnace in the temperature range of 25 ^{o}C – 2000 ^{o}C and a resonator cavity resonates at five modes of frequency values, 0.615 GHz, 1.412 GHz, 2.214 GHz, 3.017, GHz and 3.820 GHz respectively.

It is computer-automated system in measuring the resonant frequencies of the cavity with and without the sample, f and f_o, respectively, the quality factors of the cavity with and without the sample, Q and Q_o, respectively. The changes in these quantities, f, f_o, Q, and Q_o, are related to the shifted parameters Δf and ΔQ to the two parts of the complex permittivity, ε' and ε'' of the sample by the simple perturbation formula derived by Nakamura & Furuichi (1960).

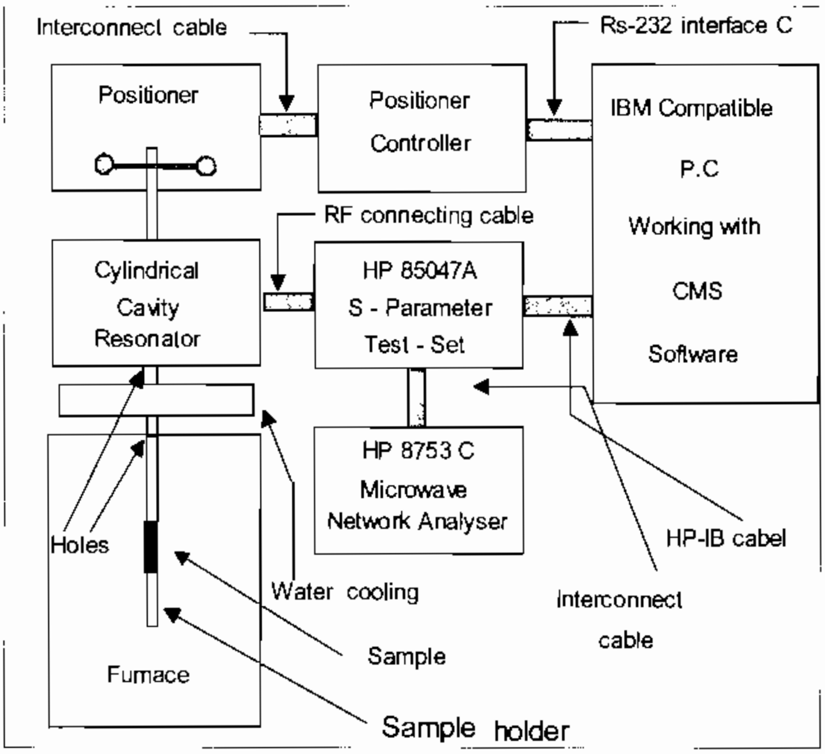

Fig. (1). The full computerized measurements system.

It is computer-automated system in measuring the resonant frequencies of the cavity with and without the sample, f and f_o, respectively, the quality factors of the cavity with and without the sample, Q and Q_o, respectively. The changes in these quantities, f, f_o, Q, and Q_o, are related to the shifted parameters Δf and ΔQ to the two parts of the complex permittivity, ε' and ε'' of the sample by the simple perturbation formula derived by Nakamura & Furuiehi (1960).

$$\varepsilon' = 2j^2(\chi_{on})\frac{a^2}{b^2}\frac{\Delta f}{f} + 1 \tag{1}$$

$$\varepsilon'' = j^2(\chi_{on})\frac{a^2}{b^2}\Delta\left(\frac{1}{Q}\right) \tag{2}$$

Where χ_{on} is the root of the zero order Bessel function of the first kind. a and b are the sample and the cavity volumes respectively. Δf and $\Delta(1/Q)$ are the change in resonant frequency and reciprocal of Q respectively upon the insertion of the sample into the cavity.

Results And Disscution

High purity (>99.0%) SiN powder was used for these measurements using the two techniques. Silica tubes of the same material and same diameter, 5mm, are filled by SiN to move the samples between the furnace and the cavity during the measurements.

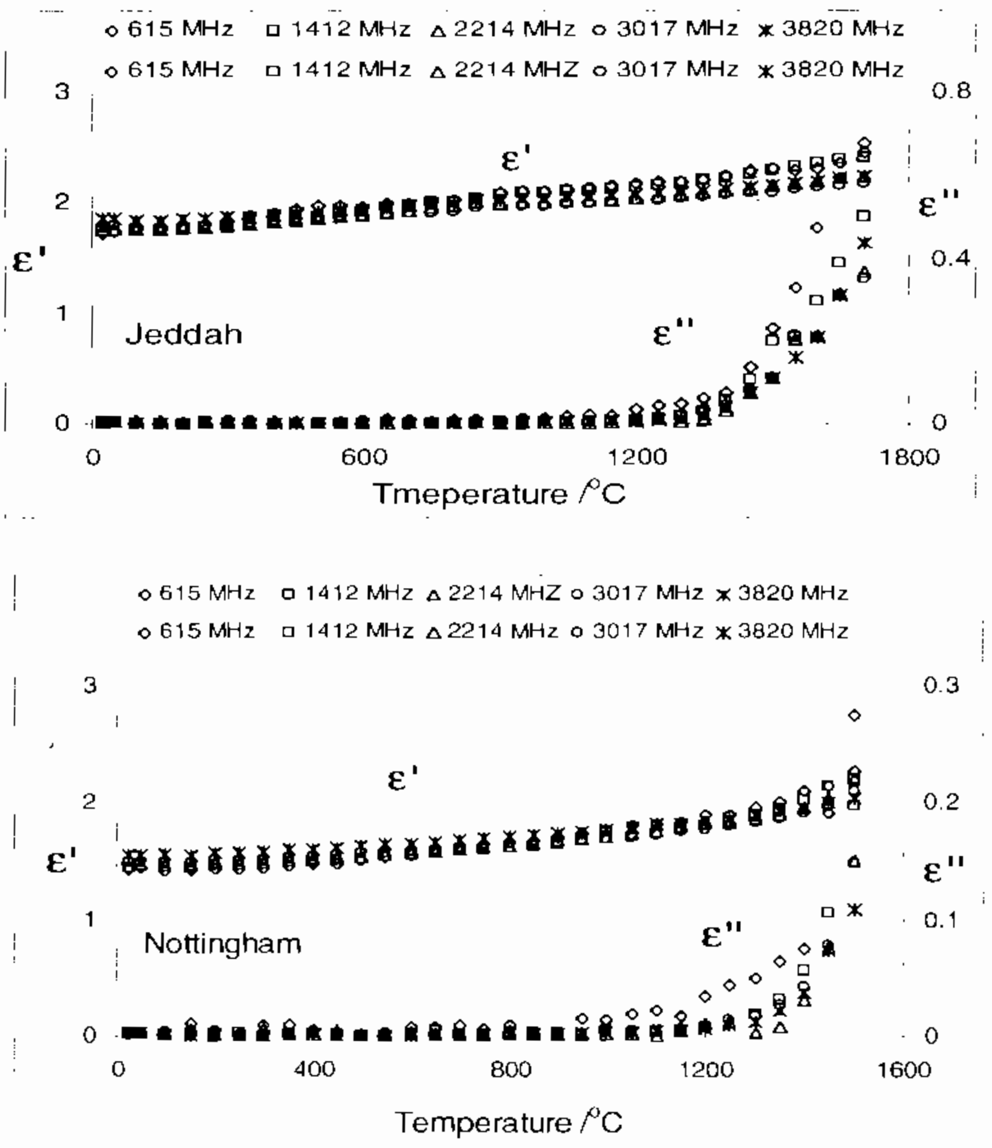

Fig. (2). The real and the imaginary parts of the complex permittivity of SiN at high temperature and different frequencies.

The two parts of the complex permittivity, ε' and ε'', of SiN shown in Fig. 2. are determined from the measured shift in frequency, Δf, and Q-factor, ΔQ after substituting them in equations (1) and (2). Fig. 2 shows the two parts of the complex permittivity of SiN where they have nearly the same behavior when changing with temperature for the two measurement systems with a very small different values of ε' and ε'' due to the difference in the quantities filling the tubes holding the powder samples. An abrupt increase of the imaginary part, ε'', shown in Fig. 2, indicted that the SiN interacts with microwaves and absorbs microwave energy when temperature reach 1200 °C.

Bruce (1991) has obtained the electrical conductivity, σ, shown in equation (3).

$$\sigma_{AC} = 2\pi f \varepsilon_o \varepsilon'' \tag{3}$$

Where ε_o is the permittivity of the free space and f is the frequency when the sample is inserted inside the cavity.

Moore (1974) suggested that the frequency dependence of the electrical conductivity of SiN investigated in terms of the frequency exponent *n* shown in equation (4).

$$\sigma(\omega) = \sigma_o + A\,\omega^n \tag{4}$$

Where σ_o is the dc conductivity and $n^1 1$ is a temperature dependent parameter . Exponent *n* was calculated by using the slope of log σ vs. log $2\pi f$.

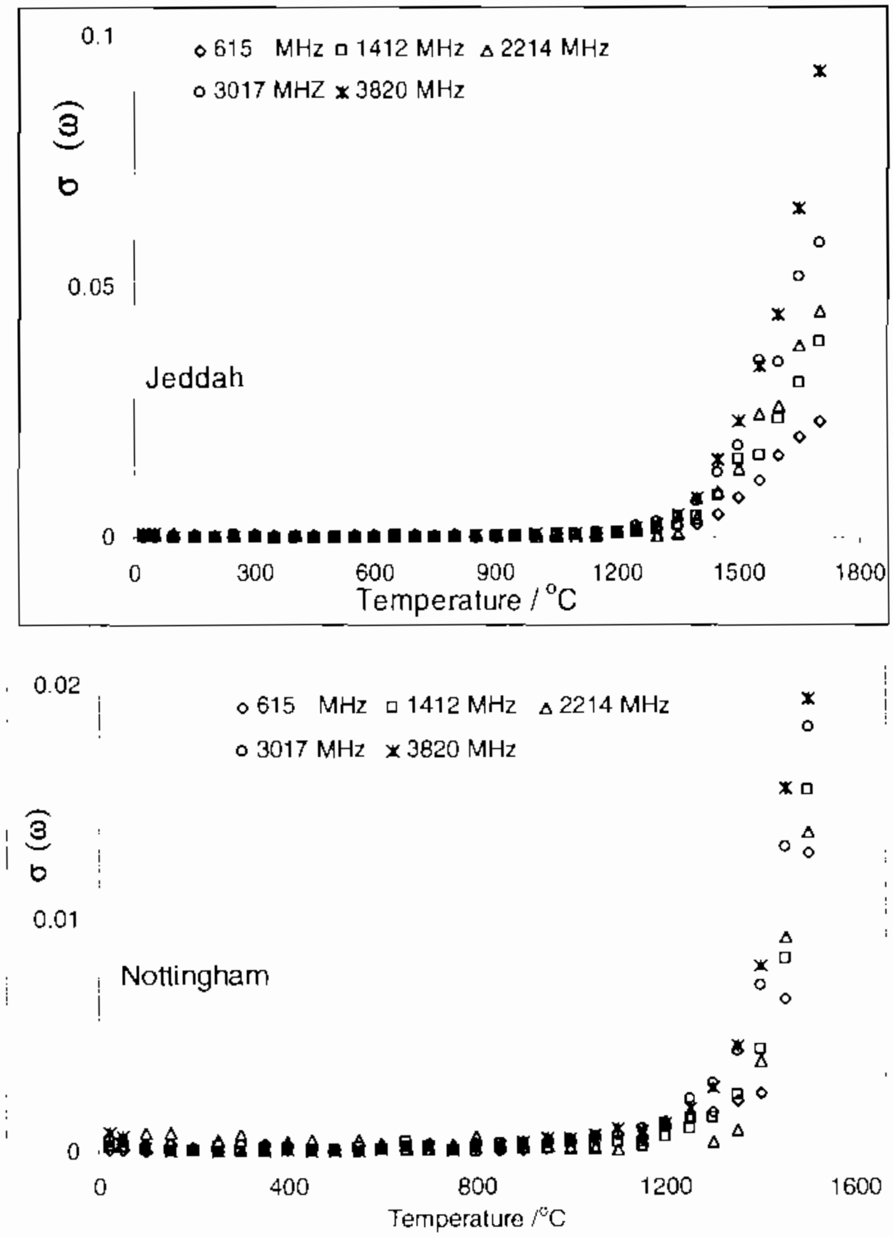

Fig. (3). The electrical conductivity of SiN at high temperature and different frequencies.

The activation energy E_A can be calculated by using the slope of equation (5) (Strickler & Carlson (1965)).

$$\sigma = A\,e^{(E_A/kT)} \tag{5}$$

Where A is a pre-exponential term, E_A is the activation energy, k is the Boltzmann constant, and T is the absolute temperature.

The electrical conductivity calculated by substituting the complex permittivity imaginary part, ε'', illustrated in Fig. 2 in equation (3). The change of electrical conductivity with temperature and different frequencies is shown in Fig. 3.

The temperature dependent parameter; n, indicated in equation (4), calculated by taking the slope of equation (3). The change of this parameter with temperature and different frequencies is illustrated in Fig. 4.

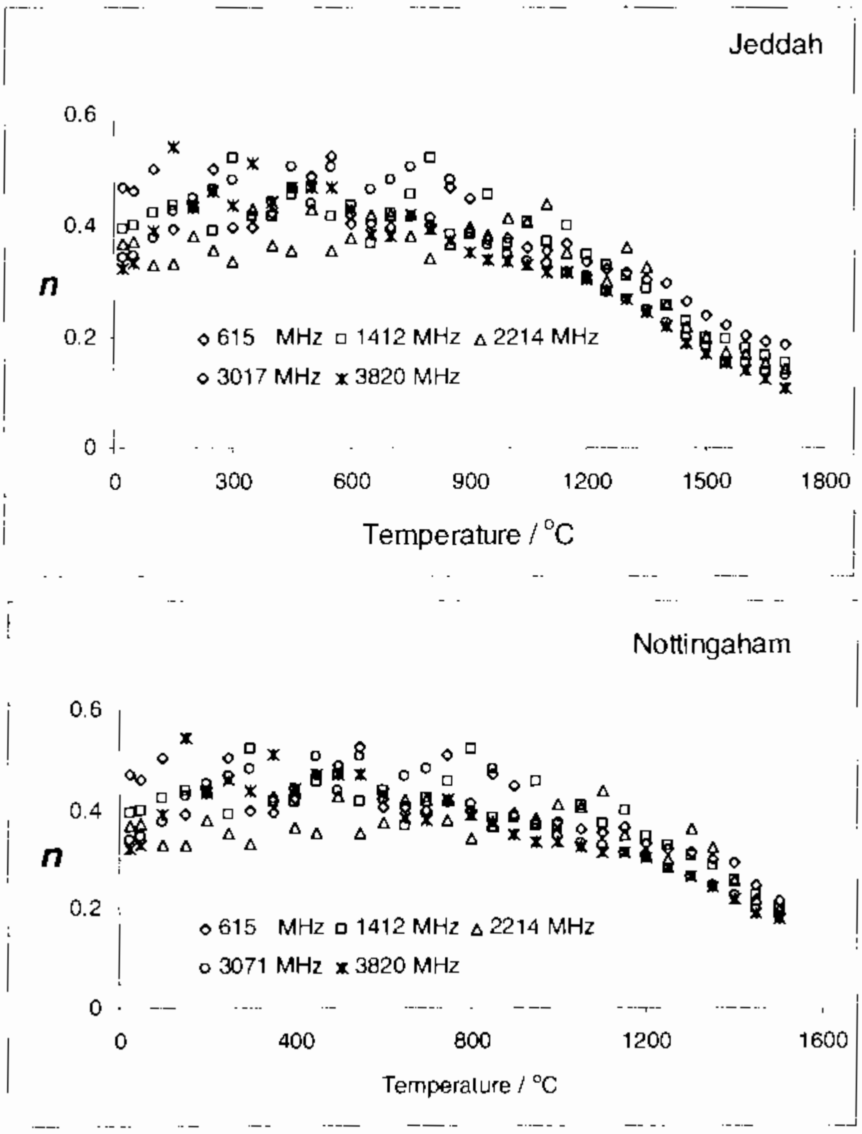

Fig. (4). The temperature dependent parameter; n, of SiN at high temperature and different frequencies.

The change of the natural logarithm of electrical conductivity of SiN versus the inverse temperature (1/T) at different frequencies is shown in Fig. 5.

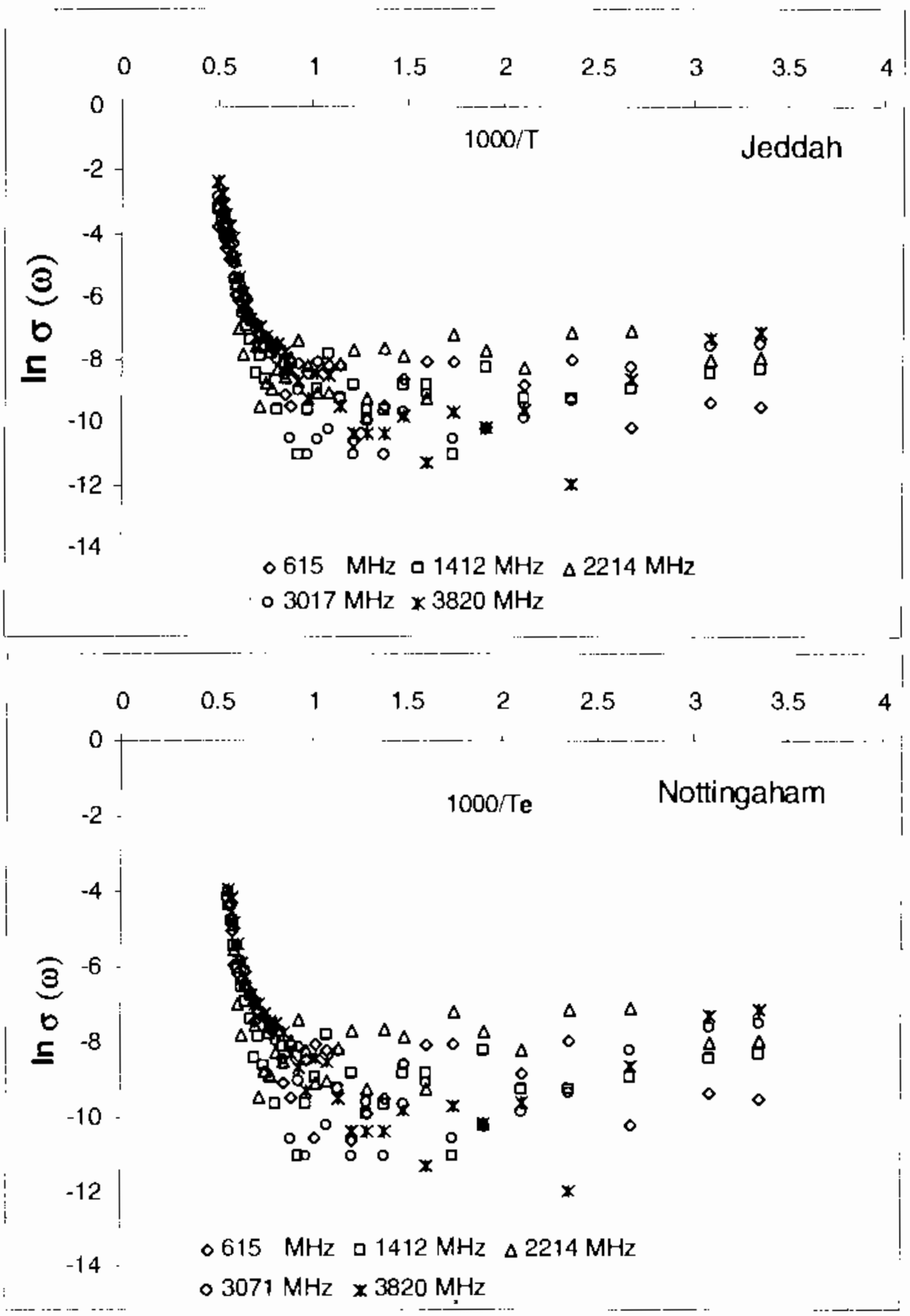

Fig. (5). The natural logarithm of the electrical conductivity of SiN versus the inverse temperature (1000/T) at different frequencies.

The activation energy E_A has been calculated by taking the natural logarithm of the two sides of equation (5) and using the slope of the natural logarithm of the electrical conductivity of SiN versus the inverse temperature (1000/T) indicated in Fig. 5. The variation of the activation energy of SiN with the variation of temperature and different frequencies is shown in Fig. 6.

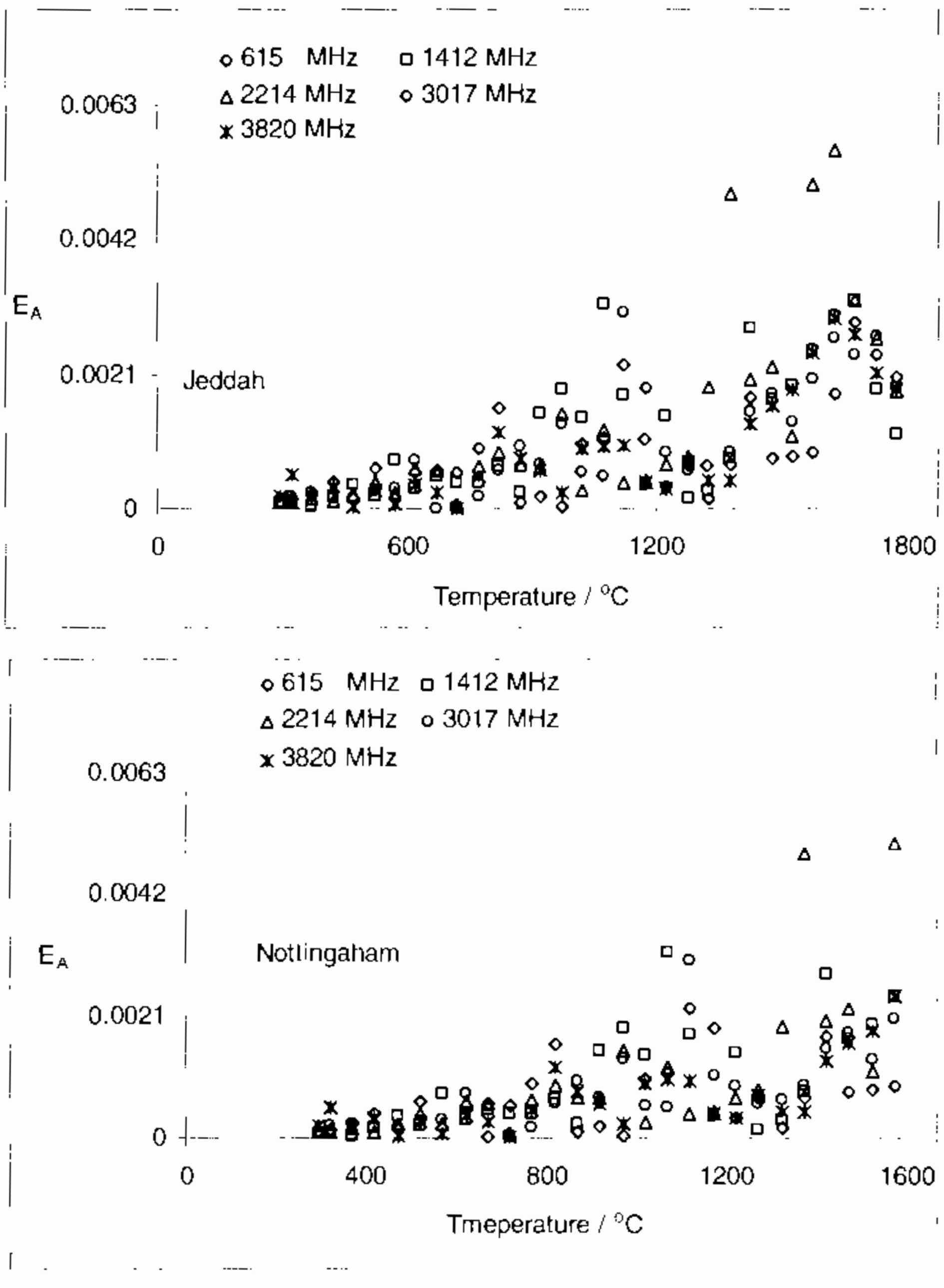

Fig. (6). The variation of the activation energy of SiN at the variation temperature and different frequencies.

The variation of the electrical conductivity with the variation of temperature shown in Fig. 3 indicated that these results are in agreement with the results of the complex permittivity two parts, ε' and ε'', shown in Fig. 2 for the two measurement techniques. The electrical conductivity changes with temperature only when the temperature > 1200 °C. It highly increases with temperature increasing after the temperature reaches 1200 °C. The change of the electrical conductivity and the change of the complex permittivity two parts, ε' and ε'', with frequency at $T > 1200$ °C is very small as shown in Fig. 2 and 3. The results of the frequency dependence of the electrical conductivity shown in terms of the frequency exponent n shown in equation (4) and the activation energy shown in Fig. 4 and 5 agree with the results shown in Fig. 2 and 3.

Conclusion

The measurements and the calculated results of the two cavity perturbation techniques have the same characters at high temperature and different frequencies. The small difference in the values of the complex permittivity two parts, ε' and ε'', is due to the difference in the quantities filling the tubes holding the powder samples. The results of dielectric properties show that SiN interacts with microwave radiation and absorb microwave energy when the temperature reaches 1200 $^{\circ}$C. This means that this result is helpful in microwave processing of silicon nitride at high temperature.

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قياسات الميكروويف للعازل الكهربي لمادة السيليكون نترايت عند درجات حرارة عالية

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المستخلص. سوف يتم قياس خواص العازل الكهربي لمادة السيراميك (SiN السيليكون نترايت) في مدى ترددات الميكروويف بواسطة جهازي الفجوة الرنينية (Cavity perturbation technique) حيث تم بناء الجهاز الأول في جامعة نوتنجهام ببريطانيا والأخر تم بناؤه في جامعة الملك عبد العزيز بالمملكة العربية السعودية ، ويعمل هذين الجهازين في مدى ترددات معينة في مجال ترددات الميكروويف (٦١٥ ميجاهيرتز و ١٤١٢ ميجاهيرتز و ٢٢١٤ ميجاهيرتز و ٣٠١٧ ميجاهيرتز و ٣٨٢٠ ميجاهيرتز) عند درجات حرارة عالية تبدأ من ٢٠ إلى ٢٠٠٠ درجة مئوية . نتائج هذه القياسات سوف تعطى فكرة واضحة عن التغيرات التي تحدث لمادة السيراميك SiN في المدى الصناعي لترددات الميكروويف و عند درجات حرارة عالية جدا وذلك نتيجة لقياس مركبات معامل السماحية (ε) لهذه المادة وهو ما يعرف بخواص العازل الكهربي للمواد (Dielectric properties) . باستخدام الجزء التخيلي من معامل السماحية المركب سوف تحسب معاملات الخواص الكهربية التي تعتمد على التغير في التردد والتغير في درجات الحرارة كالموصلية الكهربية ، σ وطاقة التنشيط ، E_a، ومعامل التردد الأسى n ، 0 ، إن دراسة التغيرات للخواص الكهربائية المقاسة والمحسوبة يساعد على فهم التفاعل الذي يحدث بين جزيئات المواد وأشعة الميكروويف مما يؤدى إلى التحكم في عمليات استخدام الميكروويف في تصنيع مواد السيراميك.