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# Using The Ultrasonic Waves For Studying Thermal And Elastic Properties as a Function of Porosity in Sintered ceramic

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## Abstract

Ultrasonic pulse echo measurements on porous alumina as ceramic material with porosities ranging from (20-40)% showed effect of volume fraction of porosity on both thermal and elastic properties. A quadratic relationships, by using a least squares method, is deduced for the dependence of the shear velocity, longitudinal velocity, shear modulus, Young's modulus, bulk modulus, Poisson's ratio, Debye temperature, specific heat, and thermal conductivity on the total porosity. By these relationships, the thermal and elastic properties results of pore-free alumina were calculated. The elastic properties results of ceramic decrease substantially with increasing porosity, except Poisson's ratio which is the constant. Debye temperature and thermal conductivity decrease too substantially with increase of porosity, while specific heat increases.

## Introduction

Ultrasonic pulse echo method as nondestructive testing has become an important analytical in the world of materials. It is a way to evaluate materials without altering them and to characterize materials by the transmitted ultrasound waves(longitudinal and shear) through the materials as well as studying the characteristics of the transmitted waves. Wave characteristics include ultrasonic velocities and frequency dependence of ultrasonic attenuation and absorption(1) through which the elastic moduli , thermal properties , flaws nature and size, and other information can be determined. Most measurements cited in the literature were made by using destructive mechanical methods that require complicated shapes to measure the elastic moduli(2). In this work, the velocities of longitudinal (compressional) and shear (transverse) sound waves in sintered alumina disks were measured by using the ultrasonic pulse echo technique. This technique enables one to measure accurately and conveniently shear ultrasonic velocities. They allow calculation of the elastic moduli and thermal properties, provided that the density is known. In addition, the problem of porosity is of special importance in ceramics due to its effect on both mechanical and electrical properties(3). In the present case of porous alumina, the sound wave velocities are related to the apparent elastic moduli, $E(P),G(P),B(P),\sigma(P)$, and the thermal properties, $\theta(P),C_V(P),k(P)$, where P is the volume fraction of porosity. The apparent elastic moduli and the thermal parameters differ from the true moduli and loss than it due to the presence of the porosity. As the sintering process proceeds, the pore volume fraction decreases and the apparent elastic moduli and thermal parameters approach those of the fully dense (pore free) material. Concurrently, the apparent density increases as a result of the reduction in porosity. Thus ultrasonic velocity measurement provides information about the formation and growth of interpartical necks and pore/particle morphology which is not available from most of the common measurements(4). Alumina was chosen because there is a substantial technological interest in this material and there also is considerable data available in the literature with which these results could be compared.

## Experimental

Samples were prepared , by solid state reaction , from alumina ($\alpha$-$Al_2O_3$) powder (99.8% from Ferak) and addition of (PVA)binder which was driven off at 500°C. Disks of 2.5cm diameter and 3cm thickness were pressed at 1 ton $cm^{-2}$ by using hydraulic press. All green samples were dried at 105°C for 8h prior to sintering. The disks were sintered in air at (1200-1500)°C for different time intervals to get porosities of (20-40)%. The sintered disks were then surface ground by using emery papers followed by polishing. To measure total porosity,we must determine the geometrical (bulk) density of the sample which involves the geometrical volume (equal to the volume of the material plus closed plus open pores). The total porosity (total pore volume fraction) was then calculated as(5)

$$P=1-\frac{\rho}{\rho_o} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots[1]$$

Where $\rho$ is the bulk density calculated experimentally from the dimensional measurement and weighting or by using Archimedes method. $\rho_o$ represents the theoretical density (determined by X-ray diffraction )of the completely dense alumina, 3.98 g/cm$^3$. Ultrasonic pulse echo measurements were performed at room temperature by using (K.K.B,USK75) with longitudinal and shear wave transducers at frequency 4MHz. All measurements were performed along the axis of the disks. Sound wave velocity was determined by measuring the sample thickness with a micrometer, and dividing it by time of flight. We have obtained (7 measurements) of both longitudinal($v_L$) and shear($v_S$) velocities as a function of the total porosity for sintered alumina. Since they are directly related to the density, elastic moduli, and thermal properties, we can study their behaviour as a function of total porosity. The shear modulus (G) and Young's modulus(E) are calculated with the following formulae(6):

$$G=\rho v_s^2 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots[2]$$

$$E=G[3-(\frac{v_s}{v_L})^2]\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots[3]$$

Poisson's ratio ($\sigma$) is usually defined as(6)

$$\sigma=\frac{E}{2G}-1 \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots[4]$$

which in terms of velocities from Eqs.(2) and (3) becomes

$$\sigma=\frac{1}{2}[1-(\frac{v_s}{v_L})^2]\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots[5]$$

The bulk modulus (B) is calculated by (7):

$$B=\rho(v_L^2-\frac{4}{3}v_s^2)\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots[6]$$

Determination of Debye temperature ($\theta_D$) requires knowledge of the mean sound velocity and is given by the formula(8):

$$\theta_D = \frac{\hbar}{\kappa}(6\pi^2 \frac{\rho}{M})^{1/3}\, \bar{\nu} \text{ ......................[7]}$$

where $M$ is the molecular mass =102 a.m.u. for alumina and $\bar{\nu}$ is the mean sound velocity $\bar{\nu} = \frac{\nu_s + \nu_L}{2}$. It is interesting to compute the Debye temperature which appears in the specific heat measurement defined as(9)

$$C_v = 1944(\frac{T}{\theta_D})^3 \text{ ..........................[8]}$$

Where T is room temperature=295K.The thermal conductivity of insulator ceramic solid can be calculated by (9)

$$k = \frac{1}{3\upsilon} C_v \lambda \bar{\nu} \text{ ..............................[9]}$$

Where $\lambda$ is the transport mean free path (the distance the waves must propagate through thickness (x)of sample until their direction is randomized).It is obtained from the ultrasonic exponential attenuation $\exp(-x/\lambda)$ (10).

$\upsilon$ is the molar volume $= \frac{M}{\rho}$ ,so Eq.[9] becomes

$$k = \frac{1}{3} C_v \lambda \bar{\nu} \frac{\rho}{M} \text{ ..............................[10]}$$

## Results and Discussion

Figures(1-9) represent the approximate experimental measurements (7values) as determined by (UPE) method in addition to the values extrapolated to zero porosity presented with solid lines which were fitted to the experimental data by means of a least squares analysis. The effect of volume fraction of porosity on the elastic moduli, and thermal properties were investigated by measuring the shear velocity and the longitudinal velocity of alumina sintered at different temperatures and time intervals and applying Eqs.(1-10). However, we cannot measure these properties at each porosity experimentally ,specially at zero porosity. So we can measure them at various porosity theoretically by least squares. Figs.(1)and(2) represent the measured shear and longitudinal velocities as a function of the total porosity of alumina. Each velocity value represents an average of three measurements. Theoretical solid lines were fitted to the experimental data by means of a least squares according to:

$v_s$(km/s)= 6.417- 4.752 P.........................[11]
$v_L$(km/s)= 10.084- 6.989 P .....................[12]

Both the shear and longitudinal velocities decrease with increase porosity which was expected because the attenuation and dispersion of the echoes increase with the increase of porosity. The porosity dependence of shear, Young's, bulk moduli and Poisson's ratio can be expressed by means of a least squares as

G(GPa)=165.05-407.63 P+294.05 $P^2$ .............[13]
E(GPa)= 417.19-974.55 P+623.61 $P^2$ ...............[14]
B(GPa)= 185.14-405.7 P+243.9 $P^2$ ..................[15]
$\sigma$ = 0.299+0.014 P-0.017 $P^2$ .............................[16]

Figs.(3-5) show these relationships where the optimum values of the elastic properties of alumina at zero porosity are G=165.05 GPa, E= 417.19 GPa, B= 185.14 GPa, $\sigma$ = 0.299 . In which the shear, Young's, bulk moduli decrease with increase of porosity because voids tend to add inhomogeneous and so decrease the elastic moduli. Fig.(6) shows that Poisson's ratio remains essentially constant and its value agrees good with that for ceramic, in general $\sigma$ =0.3. Debye temperature, specific heat, and thermal conductivity are also affected with porosity. By using least squares method ,quadratic functions of the total porosity were fitted to the experimental results through the following equations

$\theta_D$ (K)= 699.94-722.35 P+70.907 $P^2$ .......................[17]
$C_v$ (J/mole.K)= 144.93+18.202 P+3145.6 $P^2$ ...........[18]
k (W/m.K)= 8.471-1.566 P+1.094 $P^2$ ..................[19]

where the values of the thermal results of alumina at zero porosity are $\theta_D$= 699.94 K, $C_v$= 144.93 J/mole.K, k= 8.471 W/m.K. We know that Debye temperature ($hf/k_B$) is a measure of the maximum phonon frequency (f) which itself is a function of the phonon velocity depending on the porosity as showed in this paper. So one can also regard Debye temperature and frequency as measures of the porosity and then the stiffness of the ceramic material. As a result we observed through Fig.(7) the decrease of the Debye temperature with increasing porosity. High void fraction systems clearly have a better insulation performance than that of low void fraction, as what would be expected since the thermal conductivity of interstitial medium (porosity) is less than that of the ceramic. At low void fraction, where ceramic particles occupies a greater portion of the system, the thermal conductivity becomes high ,i.e.; less insulation(11). Consequently we observe through Fig.(8) a decrease of the thermal conductivity slightly with increase of the porosity. The specific heat is the amount of heat which needs to be added to produce a given temperature of a system. It is a characteristic of the material of which the system is composed. Physically and actually specific heat proportion inversely with the thermal conductivity through the experimental results are cited in the literature. So the material of a better thermal insulation (low thermal conductivity), requires the greater amount of heat to raise the temperature of it. Therefore the specific heat of alumina increase with the increase of the porosity as in Fig(9).

## Conclusion

As was expected the problem of porosity is of a special importance in ceramic due to its effect on both elastic and thermal properties which are showed in this paper. This effect of porosity will also be studied ,in future work, on electrical properties. The aim of using least squares method is the capability of determining the optimum values of elastic and the thermal properties for ceramic which decrease due to the existence of porosity. Due to the inverse relations between velocity and density, the increase in the ultrasonic velocity during sintering must be related to the increasing apparent elastic moduli rather than increasing density. Thus, the increasing density associated with sintering leads(in the absence of the increasing elastic moduli) to a reduction in the ultrasonic velocity. Thus the increased ultrasonic velocity must be associated with the increasing apparent elastic moduli in ceramic. Lastly from comparing the results of the present work with the reported values of k=8-9 W/m.K, E=360-410 GPa, G=160-170GPa, $\sigma$ =0.25-0.30, B=180-190GPa(12,13), one can regard the ultrasonic pulse echo method as a successful nondestructive test to calculate the thermal and the elastic moduli.

## References

1-Tobias ,M.,(2005), J.Acoust.Soc.Am., 117(5): 2732-2741.

2-Muller, T. and Gurevich, B., (2004), Geophysics. 69:1166-1172.

3-Muller, T.and Gurevich, B.,(2005),J.Acoust.Soc.Am.117(2):1796-1802.

4-Martin, L. and Rosen, M.,(1997),J.Am.Ceram.Soc., 80:839-846.

5-Cevik, U.,(2005),supercond.Sci.Technol., 18:101-106.

6-Martin ,L. and Rosen ,M. (1998),Mat.Sci.Engi,A246: 151-160.

7-Ledbetter ,H. (1989),Physica.C, 450:162-164.

8-Golovashkin, A. (1989),Phys.Rev., 34(3): 243-247.

9-Sears, S,(1975), "Statistical Thermodynamics", 3rd,p.292,Addison-Wesley,U.S.A.

10-Schriemer ,H. (1997),Phys.Rev., 79(17) :3166-3169.

11- Heather, L. and Diller, R. (2003) J.Biomech.Enginee. 125(29): 639-647.

12-Nagarajan, A. (1971),J.Appl.Phys., 42(10):3693-3696.

13-Edwards, G. (1989),J.Ceram.Trans., 88:117-123.

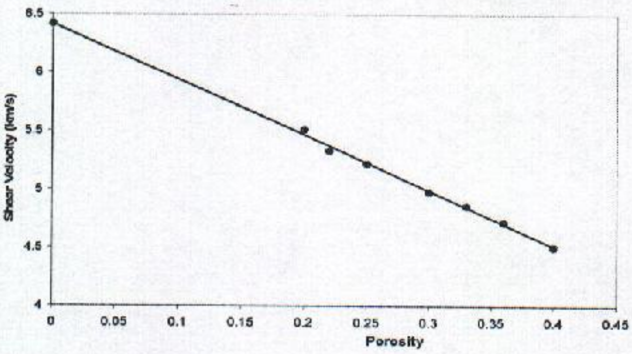

**Fig.(1): Shear velocity of alumina as a function of porosity.**

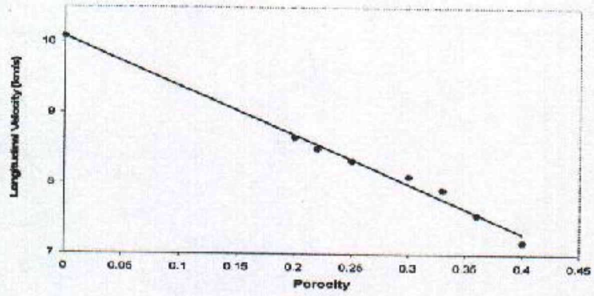

Fig.(2): Longitudinal velocity of alumina as a function of porosity.

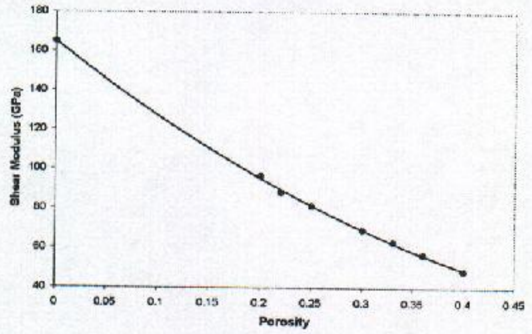

Fig.(3):Shear modulus of alumina as a function of porosity.

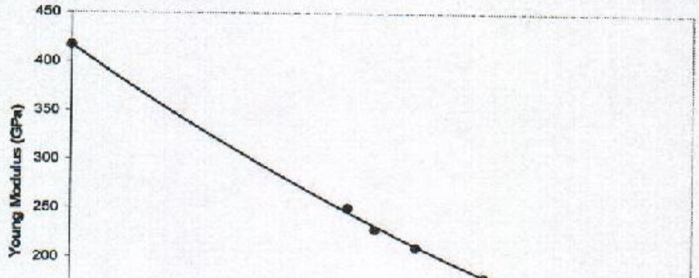

Fig.(4):Young's modulus of alumina as a function of porosity.

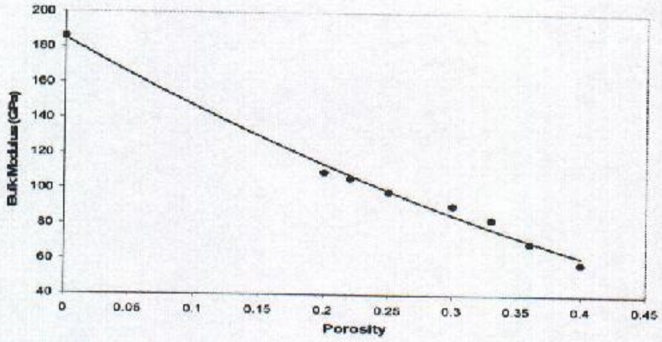

Fig.(5):Bulk modulus of alumina as a function of porosity.

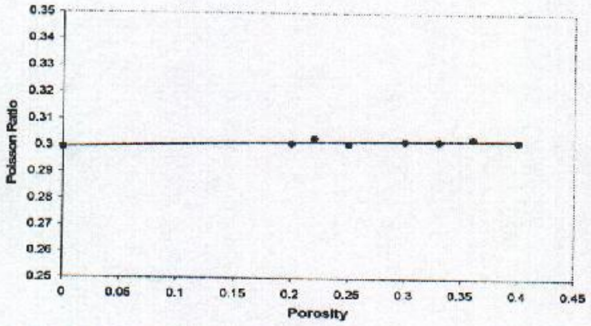

Fig.(6):Poisson ratio of alumina as a function of porosity.

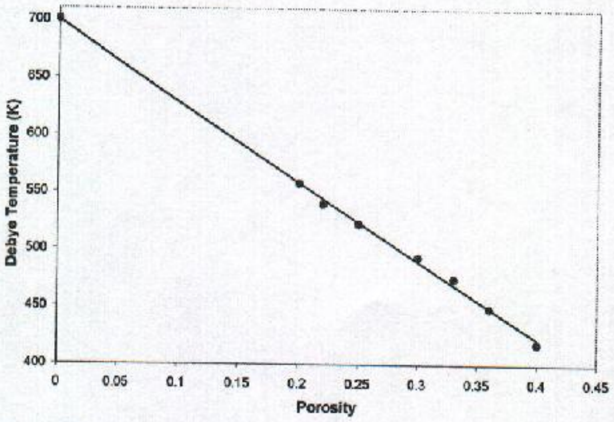

Fig.(7):Debye temperature of alumina as a function of porosity.

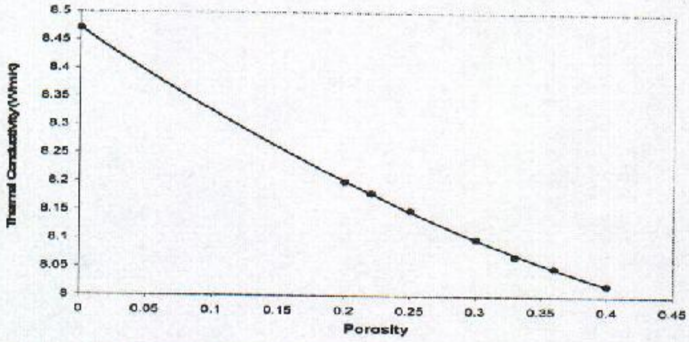

**Fig.(8):Thermal conductivity of alumina as a function of porosity.**

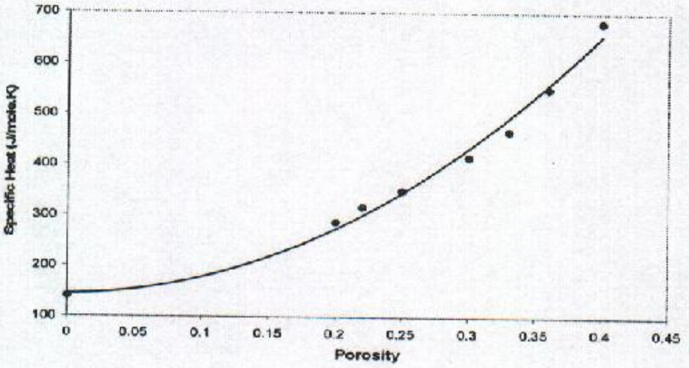

**Fig.(9):Specific heat of alumina as a function of porosity.**

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# استخدام الموجات فوق الصوتية لدراسة الخواص الحرارية و المرونة دالة للمسامية في السيراميك الملبد

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## الخلاصة

أظهرت قياسات صدى النبضة فوق الصوتية المستخدمة على الالومنا السيراميكية المسامي بمسامات ( 20- 40%)تأثير النسبة الحجمية للمسامات في الخواص الحرارية و المرونة. استنتجنا علاقات تربيعي لاعتماد معامل يونك، و معامل القص، و معامل الصلابة، و نسبة بواسن، و درجة حرارة ديباي، و الحرارة النوعية، و التوصيل الحراري على المسامات الكلية. وبهذه الدوال حسبت الخواص الحرارية و المرونة للالومنا الخالية من المسامات.خواص المرونة تقل بشكل ملحوظ مع زيادة المسامات ما عدا نسبة بواسن تبقى ثابتة. درجة حرارة ديباي و التوصيل الحراري تقل أيضا، بينما الحرارة النوعية تزداد بشكل ملحوظ مع زيادة المسامات