ULTRASONIC VELOCITY MEASUREMENTS IN ALUMINA-ZIRCONIA CERAMIC COMPOSITE SYSTEM

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Abstract: Among novel structural materials, composites are attractive because they combine the properties of two o more individual components. As an important example, in alumina-zirconia ceramic composite system, the incorporation of α-Al_2O_3 into stabilized zirconia increases significantly the mechanical and thermal properties. In this work an alumina-zirconia ceramic composite have been prepared with different α-Al_2O_3 contents from 10 to 95 wt% in order to investigate the enhancement in mechanical properties of this composite. The different composition were monitored by precise ultrasonic velocity measurements. In order to find out the factors affecting the variation in wave velocity, the ceramic composite have been characterized by X-ray diffraction and scanning electron microscopy. It was found that the ultrasound velocity changed considerably with respect to the ceramic composite composition. Mainly, we studied the behavior of the physical material property hardness, an important parameter of the ceramic composite mechanical properties, with respect to the variation in the longitudinal and shear wave velocities. Shear wave velocities exhibited a stronger interaction with microstructural and sub-structural features as compared to that of longitudinal waves. In particular, this phenomema was observed for the highest α-Al_2O_3 content composite. Interestingly, an excellent correlation between ultrasonic velocity measurements and ceramic composites hardness was observed.

Introduction: The study of ultrasonic wave propagation in metals and composites gives information on the microstructure, mechanical and physical properties of the material. The quantitative assessment of microstructural changes and properties can be carried out through the measurement of ultrasonic parameters such as attenuation and velocity [1]. The ultrasonic wave velocity depends on the elastic constants and density of the body while ultrasonic wave attenuation depends on microstructure and crystalline defects [2]. The wave propagation in a composite is affected by different material parameters such as density, stiffness, chemical composition and microstructural features [2]. Hing et al. determined the elastic properties of ZrO_2-Al_2O_3 ceramic composite system from ultrasonic velocity measurements and found that the wave velocity increases up to a maximum for about 3 wt% of unstabilized ZrO_2 dispersed in Al_2O_3 matrix and decreases monotonically thereafter [3]. The increase in moduli, shown by an increased in the ultrasonic velocity, is attributed to phase transformation of the unstabilized ZrO_2 from tetragonal to a monoclinic phase, which leads to a toughening and strengthening effect [3] .The ZrO_2-Al_2O_3 ceramic composites display different properties depending on the raw materials, chemical composition and preparation route. The development of diverse methods for fabricating transformation-toughness ceramics such as ZrO_2-Al_2O_3, mullite- ZrO_2, Si_3N_4- ZrO_2 and others, has received significant interest recently. The ZrO_2-Al_2O_3 ceramic composite have been studied most widely among them. It has been observed that the microstructure of the matrix material and the zirconia particles dispersed in the alumina matrix are so important in order to produce optimally tough transformation-toughened composite materials that increase the mechanical and thermal properties of the composite [3-5]. In this work, we investigate the influence of α-Al_2O_3 seeding on the sinterability of ZrO_2-Al_2O_3 and monitored by precise ultrasonic velocity measurements. The ceramic composite is characterized by X-ray diffraction and scanning electron microscopy in order to find out the factors affecting the variation of the longitudinal and shear wave velocity.

Results: Homogeneous sols of pseudoboehmite and $ZrO_2(Y_2O_3)$ were prepared by a mecanochemical treatment employing HNO_3 as a peptisizing agent. Pseudoboehmite with average formula $Al_4O_3(OH)_6$ and specific surface area 227 m^2/g was obtained from a basic

aluminum sulfate derived from alunite mineral (U.G. Process) [6]. The basic salts of such process were completely hydrolyzed in an aqueous ammonia medium at 70-80°C at pH 9-10. Suspensions were prepared with pseudoboehmite sols seeded with 2.5 mass% α-Al_2O_3 of 0.20 μm (Taimicron, TM10). Tetragonal zirconia powders (TOSOH, TZ-3YS) with average particle size of 0.26 μm were added in adequate proportions for each composition. The suspensions were ultrasonically stirred and thereafter spray dried using a YAMATO Mini-spray dryer ADL31. In this way, mixtures of TZ-3YS and α-Al_2O_3 seeded pseudoboehmite with compositions of 100, 90, 70, 50, 30, 15 and 0 mass % of ZrO_2 were prepared. The mixed powder was compacted into half cylindrical shaped 40×20×10 mm samples by isostatic compression at 200 MPa. The samples were presintered at 1250°C and finally sintered at 1550 °C for 1h [7]. The samples were identified with the letters SDI followed by a number that indicates the Al_2O_3 content except the sample that contains 100 % wt of ZrO_2 as shown in Table 1. The density of the sintered samples was measured by the Archimedes method. The theoretical density was estimated with the rule of mixtures. The microstructure and phases content of the sintered samples were determinated by scanning electron microscopy (SEM, Jeol mod. 6400) and X-ray diffraction (XRD, Phillips 5000) respectively.

Sample ID	Composition wt% ZrO_2-Y_2O_3/Al_2O_3
ZTY	100/0
SDI10	90/10
SDI30	70/30
SDI50	50/50
SDI70	30/70
SDI85	15/85
SDI95	5/95
SDI100	0/100

Table 1.- Samples identification of ceramic composites.

Next, we will describe the experimental set up and procedure we used for the ultrasonic measurements in the ceramic composite samples with different %wt Al_2O_3. Figure 1 shows a schematic diagram for the experimental arrangement used to study the behaviour of the ultrasonic velocity in these samples. The generator provides adjustable excitation pulses which are received by an amplifier with a large adjustable gain [8]. In a water tank , the displacement of the focused ultrasonic transducer in the plane X-Y is undertaken by two step- by-step motor ordered via an interface by a microcomputer. The numerical scope, which permits sampling in an adjustable temporal window, is also ordered by a microcomputer via an interface. So the signals collected are sent to a microcomputer in order to process them. On the other hand the ceramic composite samples are fixed on two supports and submerged in water for its inmersion ultrasonic inspection. The computational software permit to order different focused ultrasonic transducer displacements with regard to the samples, sampling, processing, data acquisition and visualization of the obtained results.

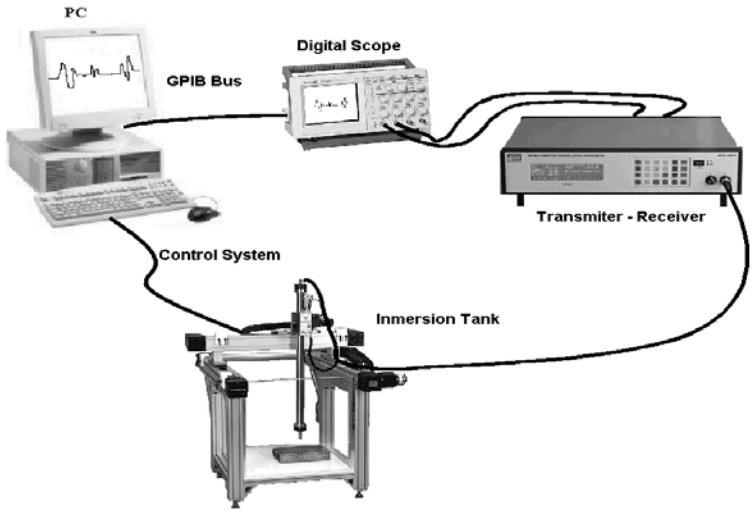

Figure 1.- Experimental set up for the ultrasonic measurements

The hardness is defined as the material resistance to a local penetration [9] . It is a complex material physical property to interpret, and one on which material mechanical and physical properties depend. Hardness was measured on the ceramic composites using a Vicker´s hardness tester. The Vickers hardness is given by, $HV = 2p \sin(\theta/2) / D^2$ where p is the applied load (Kgf), D is the impression diagonal and θ is the angle between two opposed faces (136°). Hardness measurements have been made in "line" direction (along sample length) on all the ceramic composite material samples. Figure 2 shows the measured Vicker´s hardness of the ceramic composites as a function of % wt Al_2O_3 . The obtained hardness results were different depending on the composition of the two phase material. It was observed that the hardness is higher on the Alumina- Zirconia composite ceramic as the % wt Al_2O_3 increases from ≈ 1285 Hv in the ZrO_2 phase until ≈ 1792 Hv in the Al_2O_3 phase. This can be explained by the fact that the alumina (reinforcement) dispersed within the zirconia matrix increases so the composite material became harder and stronger increasing its mechanical properties and also due to the fact that the pure Alumina ceramic is a higher material resistance to a local penetration than the pure Zirconia ceramic.

In Figure 3, XRD patterns of sintered α-Al_2O_3 seeded composites are presented. The characteristic peaks of tetragonal (t) and alfa (α) phases of Zirconia and Alumina are depicted respectively. It is observed that as the % wt Al_2O_3 increases the characteristic peaks of Zirconia tetragonal (t) phase decreases, while the α-Al_2O_3 phase increases. The t-ZrO_2 peaks increases in relation to the as-received $ZrO_2(Y_2O_3)$ powder signals, due to the increment in density obtained during the sintering and also the increment of the % wt $ZrO_2(Y_2O_3)$ in the two-phase composite material that promotes to have greater tetragonal phase stabilization. This phenomena can be seen clearly in Figure 4 in which the variation of the sintered density of the samples as a function of the % wt Al_2O_3 is presented. The pure $ZrO_2(Y_2O_3)$ attained a density of 6,0400 gr/cm^3 after sintering. Addition of alumina in to the $ZrO_2(Y_2O_3)$ promoted a decrease of the sintered density until it reaches a minimum value of 3.9010 gr/cm^3 for pure Al_2O_3 phase. This phenomena can be also explained by the fact of the differences of the internal friction between α-Al_2O_3 seeds , the pseudoboehmite and $ZrO_2(Y_2O_3)$ particles in the sample packing process during the cold isostatic pressing, where the soft agglomerates are deformed by the applied pressure.

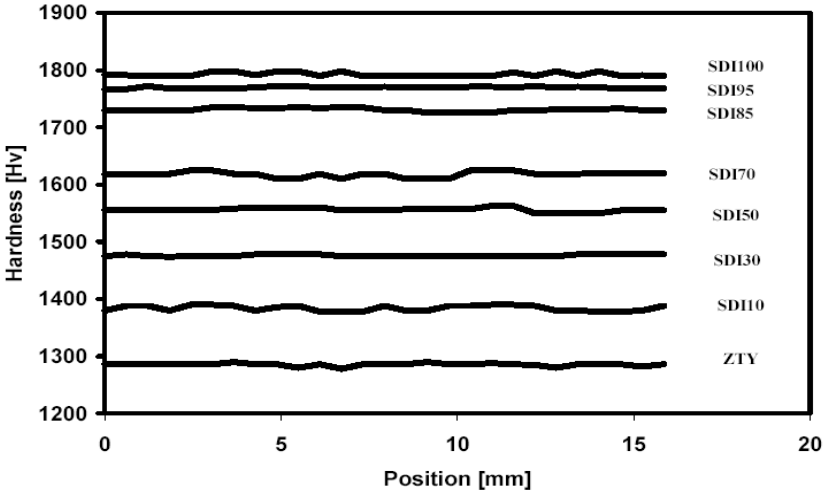

Figure 2.- Vicker´s hardness for ZrO_2-Y_2O_3/Al_2O_3 composites

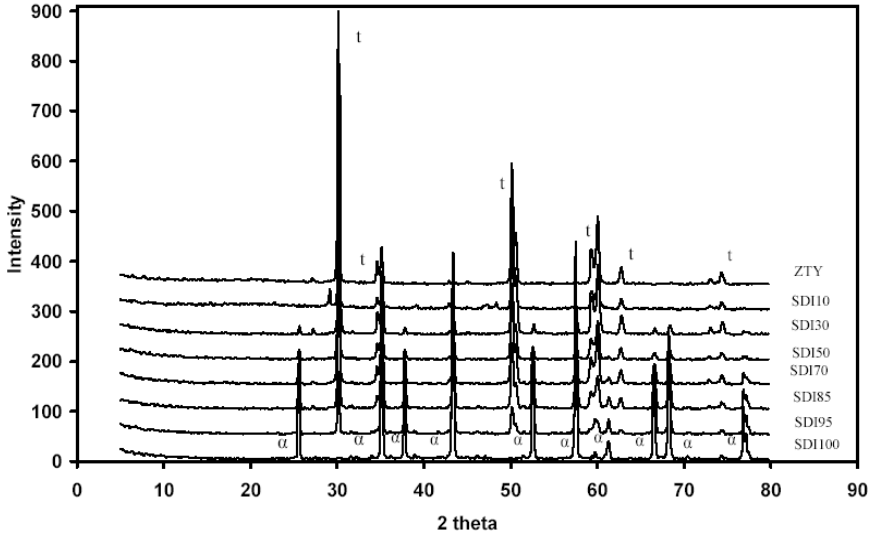

Figure 3.- XRD patterns of sintered ZrO_2-Y_2O_3/Al_2O_3 composites.

The ultrasonic velocity measurements were performed using a 5 MHz immersion focused beam transducer. The shaped samples (half cylindrical) were selected in order to obtain the longitudinal and shear velocities modes in the sample sample[11]. The ultrasonic measurements were taken in immersion at oblique and normal incidence angles using a focused transducer [12] . Ultrasonic velocity is determined by measuring the time taken for the ultrasonic waves to travel through the material thickness. In normal incidence was measured the time-of-flight of the longitudinal waves in the ceramic composite samples. In oblique incidence was measured the time-of-flight of the shear waves in the ceramic composite samples. For the longitudinal wave velocity measurements the principle consists of measuring the wave time of-flight to cover the sample thickness, which it is measured with precision. The longitudinal wave velocity measurements on the axis as a function of the %wt Al_2O_3 are given in Figure 5. Comparing to the variation of the ceramic composite samples sintered density, the longitudinal wave velocity as a function of the % wt Al_2O_3 is shown in Figure 6.For the shear wave velocity measurements the principle is the same as

the longitudinal wave velocity measurements. The shear wave velocity measurements on the axis as a function of the %wt Al_2O_3 are given in Figure 7. Comparing to the variation of the ceramic composite samples sintered density, the shear wave velocity as a function of the % wt Al_2O_3 is shown in Figure 8.

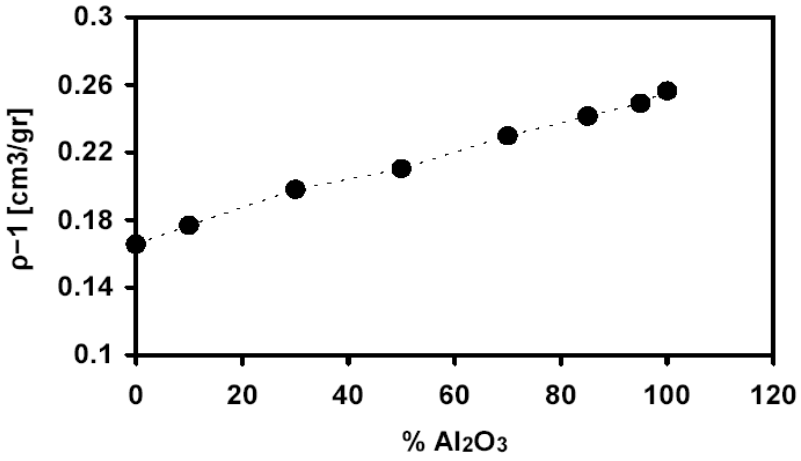

Figure 4.- Variation of the sintered density of the samples as a function of the % wt Al_2O_3.

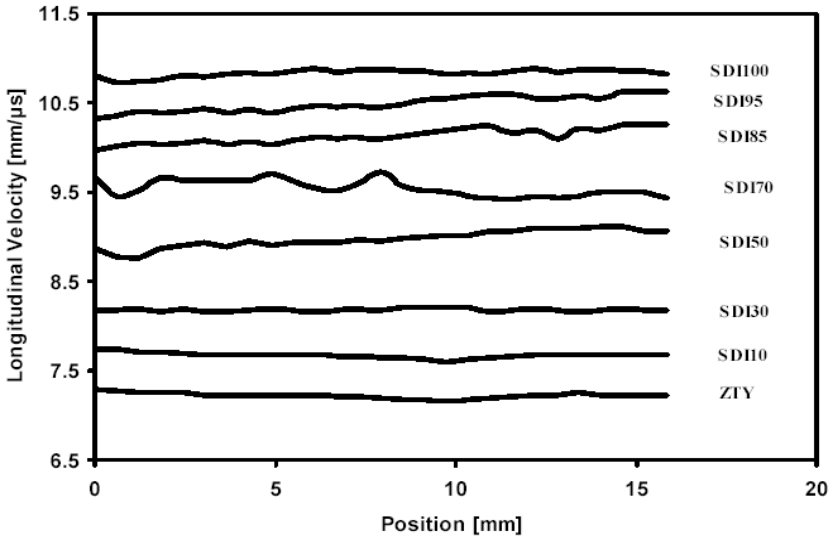

Figure 5.- Longitudinal wave velocity measurements on the axis as a function of the %wt Al_2O_3.

Discussion: By analyzing Figs. 5 and 7 representing the longitudinal and shear waves velocity variation with respect to the axis as a function of the %wt Al_2O_3, it is clearly observed the same trend as the hardness curves shown in Fig. 2. These curves begin with a maximum and decrease gradually as a function of the Alumina content decrease until it reaches the Zirconia phase (ZrO_2), exactly as for the hardness curves. Concerning shear waves, the same verification took place except for more dispersion of the data in particular in the % 50 wt Al_2O_3 and higher. This phenomena can be explained by the fact that longitudinal velocity measurements are more

accurated compared to shear velocity measurements for determining the hardness curve [13]. Furthermore, shear velocity wave is approximately 0.5 longitudinal velocity wave. Therefore, the wavelength of shear wave is less as compared to the wavelength of longitudinal wave so shear wave is expected to have higher interaction with the ceramic composite microstructure compared to that of longitudinal waves [11]. Therefore, it can be deduced from the results of the longitudinal and shear velocity curves compared to the hardness curve (Figs.2, 5 and 7) that it is possible to obtain the material hardness from its longitudinal or shear waves [14]. Similary by analyzing Figs. 4, 6 and 8 representing the longitudinal and shear waves velocity variation of the sintered density of the samples as a function of the % wt Al_2O_3, it can be observed the same trend as the sintered density curves shown in Fig. 4. These curves begin with a minimun and increase gradually with the inverse of the sintered density of the ceramic composite samples as a function of the % wt Al_2O_3 until it reaches the Zirconia phase (ZrO_2), exactly as for the sintered density curves. Again concerning the shear waves, the same verification can be made except for more dispersion of the data in particular in the % 50 wt Al_2O_3 and higher. This phenomena can be explained as we mentioned above by the fact that shear wave velocities exhibited a stronger interaction with microstructural and sub-structural features as compared to that of longitudinal waves. Therefore, it can be deduced from the results of the logitudinal and shear velocity curves compared to the the sintered density curves (Figs.4, 6 and 8) that it is possible to determine the material sintered density from its longitudinal or shear waves.

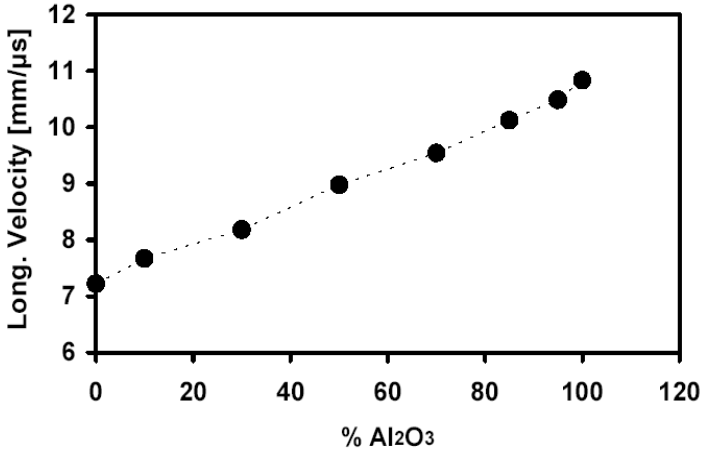

Figure 6.- Longitudinal wave velocity versus the variation of the sintered density of the samples as a function of the % wt Al_2O_3 .

Conclusions: The hardness and sintered density of ZrO_2-Al_2O_3 ceramic composite were determined from longitudinal and shear velocity measurements. The results were found to be dependent upon the content of the Al_2O_3 and ZrO_2 phases (changes in material chemical composition and density). In both cases, the longitudinal and shear velocities increase up to a maximum for about 100 %wt Al_2O_3 phase and decreases monotonically thereafter until it reaches the ZrO_2 phase. The increase in hardness as a function of the %wt Al_2O_3 phase, as shown by an increased in velocity, is attributed to variation in the chemical composition and density of the ceramic composite material under study. The possibility was investigated of an ultrasonic normal and oblique incidence immersion technique for velocity measurements of longitudinal and shear ultrasonic waves through a ceramic composite with different Al_2O_3 and ZrO_2 content (two-phase material) due to the following material properties hardness and sintered density. This experimental work demonstrates that it is possible to obtain the material qualitative hardness and

sintered density from the longitudinal or shear velocity wave measurements. However, it was observed that shear wave velocity exhibited a stronger interaction with microstructural features as compared to that of longitudinal wave velocity.

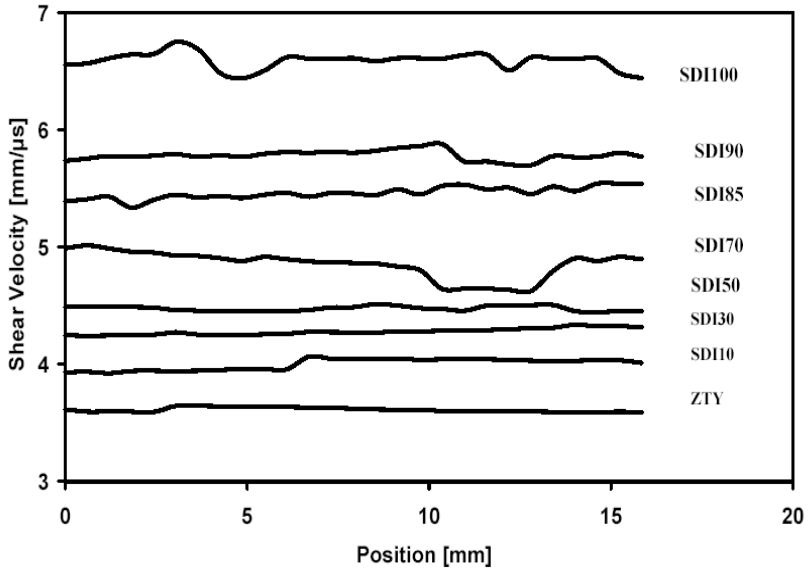

Figure 7.- Shear wave velocity measurements on the axis as a function of the %wt Al_2O_3.

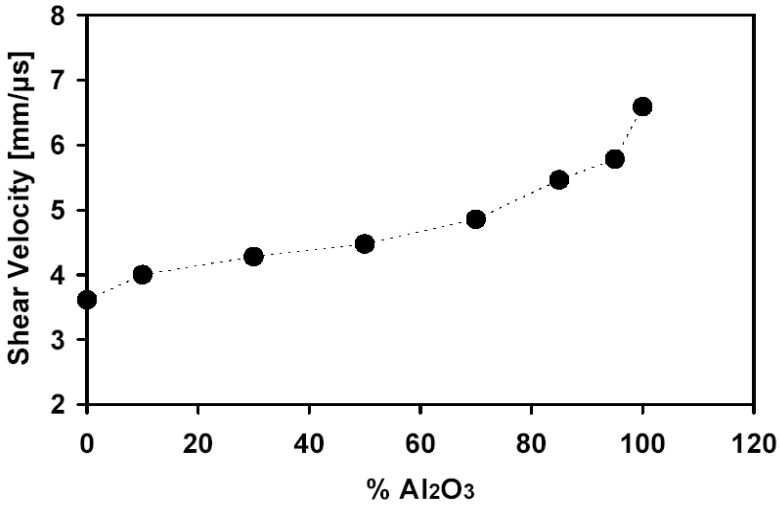

Figure 8.- Shear wave velocity versus the variation of the sintered density of the samples as a function of the % wt Al_2O_3 .

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