



24th COBEM - 2017



24th ABCM International Congress of Mechanical Engineering  
December 3-8, 2017, Curitiba, PR, Brazil

COBEM-2017-1661

## PRODUCTION AND CHARACTERIZATION OF $Sr_2AlZrO_{5,5}$ CERAMICS TO ENCAPSULATE TEMPERATURE SENSORS IN THE PETROLEUM EXTRACTION INDUSTRY

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**Abstract.** In petroleum extraction wells, various sensors are used to monitor different parameters such as flow, pressure and temperature. However, the petroleum is highly corrosive, which can cause equipment failure, damaging the process and generating high maintenance costs. Advanced ceramics appear as an alternative especially to environments with high temperatures. Zirconium oxide is a widely used material because it presents superior mechanical properties when compared with other ceramic materials. This work has as an objective to produce new ceramics of complex cubic perovskite structure  $Sr_2AlZrO_{5,5}$ , for the encapsulation of temperature sensors. The constituent oxides were homogenized and calcined at  $1150^\circ\text{C}$ . The crystalline structure of the compound was determined by X-ray diffraction which showed the formation of the ordered complex cubic perovskite structure. The powder was compacted and sintered at temperatures of  $1350^\circ\text{C}$  and  $1500^\circ\text{C}$ . Mechanical properties of the sintered pellets were analyzed by Vickers microhardness. After these tests the samples are immersed in onshore and offshore crude petroleum and will again be subjected to the Vickers Microhardness test, XRD, in addition to optical microscopy and SEM in order to verify the stability of the ceramic in crude petroleum.

**Keywords:**  $Sr_2AlZrO_{5,5}$ ; encapsulation; temperature sensor; petroleum industry

### 1. INTRODUCTION

In ceramic materials, the interatomic bonds are totally or predominantly ionic but have some covalent character, which gives the ceramics a high stability. These materials also have high melting temperatures, are hard but fragile, and can be both thermal and electrical insulator. Crude petroleum is a highly corrosive product so the degradation of equipment used in the wells is a serious problem in the crude petroleum industry. Thus, an alternative to solve this problem is to encapsulate these equipments with materials inert to this corrosion. There is a group of ceramic materials that have simple perovskite structure or with little distortion of it, which are called complex perovskites. Transition metal oxides of perovskite structure are known for their variety of electronic, magnetic, optical and catalytic properties (ARAKAWA, 1993; CALLISTER, 1999; BELLEVILLE, 2010).

In this work we produced the complex perovskite ceramics  $Sr_2AlZrO_{5,5}$  and studied the behavior of sintering and its mechanical and microstructural properties. The objective of this work is to obtain a ceramic of high mechanical

strength and homogeneous microstructural characteristics, besides chemical stability in the corrosive environment of petroleum to be used as encapsulation of temperature sensors.

## 2. EXPERIMENTAL DETAILS

The ceramic powder was obtained by mixing the  $\text{SrCO}_3$ ,  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  oxides in a mortar for 30 minutes. The homogenized mixture was deposited in a cylindrical matrix and compacted uniaxially in a hydraulic press under pressure of  $4\text{ton} / \text{cm}^2$  for 5min. The pellets were calcined at a temperature of  $1150^\circ\text{C}$  for 24 hours. The calcined pellets were then milled and subjected to X-ray diffraction analysis. The ceramic powder was powdered again compacted by uniaxial pressing at a pressure of  $10\text{ton}/\text{cm}^2$  for 5minutes. After compaction, the pellets were sintered in two different thermal cycles. In the first, the samples were sintered using a heating ramp of  $10^\circ\text{C} / \text{min}$  until reaching the plateau of  $1350^\circ\text{C}$  during 24h. In the second, the samples were initially heated, at a rate of  $15^\circ\text{C} / \text{min}$ , until reaching  $700^\circ\text{C}$  and remaining for 1 hour at this level; Then the temperature was increased to  $1050^\circ\text{C}$  with a heating ramp of  $15^\circ\text{C} / \text{min}$ , with 0.5 hour remaining at this level; The furnace temperature was again increased to  $1500^\circ\text{C}$ , ramp of  $10^\circ\text{C} / \text{min}$ , remaining at this temperature for 2 hours for sintering the samples; Finally the furnace was cooled to  $800^\circ\text{C}$ , remaining 0.5 hour at this stage, and then cooled to room temperature. After sintering, the pellets were submitted to the Vickers microhardness test and immersed in onshore and offshore for subsequent analysis of optical microscopy, x-ray diffraction and Vickers microhardness in order to verify the stability of the ceramic (ARAÚJO, 2015).

The samples were cooled without forced ventilation. The sintered samples were polished with sand papers of anded with 220, 400, 600, 800, 1200 and 1500 meshes of silicon carbide until they presented the mirrored appearance and were subjected to Vickers microhardness analysis by optical microscopy. Optical microscopy was performed using an Olympus BX51M optical microscope to analyze the surface of the sintered samples. Scanning electronmicroscopy (SEM) was used to study the microstructures of sintered ceramics. For the surface observation the samples were covered with a thin layer of graphite in a carbon coating unit (Cressington, model 108carbon / A). The microhardness tests were performed on the sintered and polished samples. The microhardness was determined through the HVS-5 model microdurometer which applied a 1000 grams charge through a diamond pyramid shaped indenter for 10 seconds. Ten readings were taken and the two most discrepant were removed. The microhardness was obtained by the arithmetic mean of the remaining 8 readings. For the chemical and physical stability tests of sintered ceramics in crude petroleum, finely polished samples were immersed in onshore crude petroleum of northeast region of Brazil, provided by PETROBRAS, a premier petroleum company of Brazil. Samples were taken out from the petroleum after 30 and 60 days and subjected to the optical microscopy for the evaluation of surface degradation, if any.

## 3. RESULTS AND DISCUSSION

Figure 1 shows the x-ray diffraction of the calcined  $\text{Sr}_2\text{AlZrO}_{5,5}$  ceramics. It can be observed, according to the jcpds literature and database of materials with the complex cubic perovskite structure (LAPA, 2004; OLIVEIRA, 2013), by the relation of the peak intensities and angular positions of them, that the ceramic presents a singlephase characteristic of an ordered complex cubic perovskite structure.

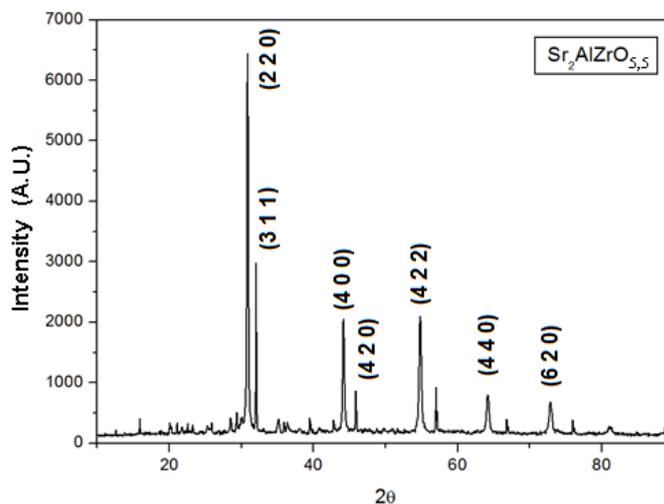


Figure 1: X-ray diffraction of calcined  $\text{Sr}_2\text{AlZrO}_{5,5}$  powder.

Table 1 shows the x-ray diffraction parameters of the  $\text{Sr}_2\text{AlZrO}_{5,5}$  system. From these data the experimental lattice parameter was determined using the highest intensity peaks. The theoretical lattice parameter found in the  $\text{Sr}_2\text{AlZrO}_{5,5}$  system was 8,5786 Å and the experimental value calculated through the diffractogram analysis was 8,4487 Å. The difference between the experimental value and the theoretical value is 0.98%. The difference between the experimental and theoretical values of the lattice parameter occurs because in this theoretical model the atoms are represented as rigid spheres, which does not correspond to reality, since they have vibrations in the atomic positions similar differences have been observed in values of theoretical and experimental network parameters of some other types of a 2BB'O6 perovskite oxides such as  $\text{YBa}_2\text{NbO}_6$ ,  $\text{ErBa}_2\text{sbo}_6$ ,  $\text{DyBa}_2\text{SbO}_6$ , etc., (yadava, 2001; lapa, 2004).

Table 1: X-ray diffraction parameters of  $\text{Sr}_2\text{AlZrO}_{5,5}$

$2\theta$	D (Å)	(h k l)	Intensity (I) (A.U.)	I/ $I_0$
30,8955	<b>2,8992</b>	<b>2 2 0</b>	<b>6447,2797</b>	<b>1,0000</b>
32,1748	<b>2,7798</b>	<b>3 1 1</b>	<b>2969,1863</b>	<b>0,4605</b>
44,2288	<b>2,0462</b>	<b>4 0 0</b>	<b>2018,7770</b>	<b>0,3131</b>
45,7640	<b>1,9810</b>	<b>4 2 0</b>	<b>852,6721</b>	<b>0,1322</b>
54,8329	<b>1,6729</b>	<b>4 2 2</b>	<b>2099,6629</b>	<b>0,3256</b>
64,2146	<b>1,4493</b>	<b>4 4 0</b>	<b>808,8589</b>	<b>0,1254</b>
72,9992	<b>1,2950</b>	<b>6 2 0</b>	<b>694,2705</b>	<b>0,1076</b>

After the XRD analysis and determination of the complex cubic perovskite structure the calcined material was ground in the agate mortar and subjected to particle size analysis. This study was carried out to analyze the grain size distribution, considering that its homogeneity is a fundamental factor for the sintering, microstructure and mechanical properties of the final product. The particle size distribution curves of the  $\text{Sr}_2\text{AlZrO}_{5,5}$  ceramics are shown in figure 2. From this curve the parameter D50 was obtained which is related to the median of the distribution corresponding to the mean particle diameter ( $D_m$ ).

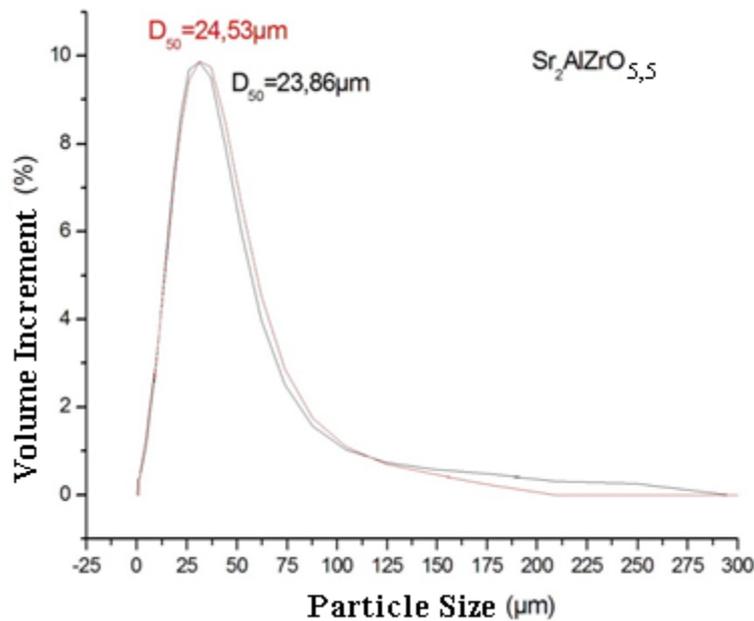


Figure 2: Granulometric distribution of the  $\text{Sr}_2\text{AlZrO}_{5,5}$  system after milling

The granulometric analysis showed homogeneity in particle size. The  $\text{Sr}_2\text{AlZrO}_{5,5}$  system had an average particle size of approximately 24µm. In order to analyze the microstructures of the sintered perovskite the scanning electron microscopy technique was used.

Figure 3 presents the typical results of sem microstructural analysis of the sintered ceramics. As seen from the figure 3, sintered  $\text{Sr}_2\text{AlZrO}_{5,5}$  presents typical microstructure of sintered  $\text{Sr}_2\text{AlZrO}_{5,5}$  ceramics bodies with uniform particle size distribution and surface morphology .

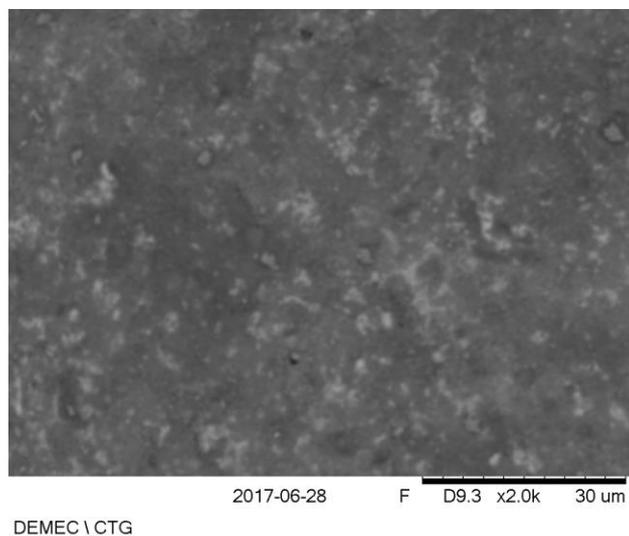


Figure 3: Results obtained from the sem of the  $\text{Sr}_2\text{AlZrO}_{5,5}$  system

Figure 4 illustrate elemental element analyzes of the  $\text{sr}_2\text{alzro}_{5,5}$  ceramic, performed by eds together with the scanning electron microscopy .

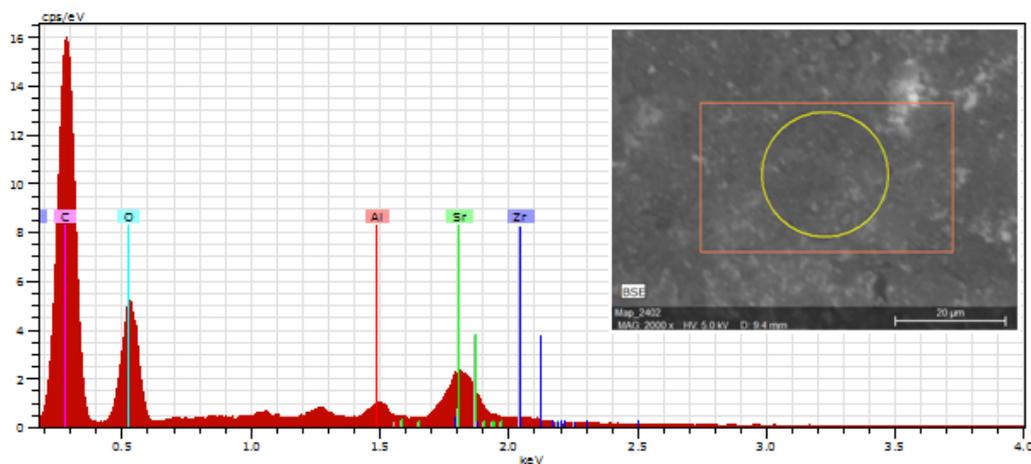


Figure 4: eds analysis of  $\text{sr}_2\text{alzro}_{5,5}$  ceramic

Through the analysis of the eds we verified that the only element elements present in the samples are constituent chemicals used for the production of this ceramic. That is, the analyzed samples were not contaminated during the production process. With the vickers microhardness test of the sintered ceramic  $\text{Sr}_2\text{AlZrO}_{5,5}$  , it was possible to determine a mean value of the ceramic hardness. Ten indentations were made and the 2 most discrepant values were discarded, obtaining an arithmetic mean with the remaining 8 values. The average hardness value determined in this test was 240.12 hv for the sintered ceramics at 1350°C and 254.38 for the ceramics sintered at 1500°C. The value of average hardness found in the system produced in this work is considered good when compared to other ceramics of similar properties already researched (YADAVA et al, 2001; LAPA, 2004; OLIVEIRA, 2013).

Optical microscopy was performed with the objective of analyzing the surface of samples and verify whether or not it has suffered chemical attack due to contact with crude petroleum. Figure 5 presents the typical optical micrographs of the  $\text{Sr}_2\text{AlZrO}_{5,5}$  ceramic before and after immersion in crude petroleum. When compared these optical micrographs after 60 days of immersion in oil, it was verified that the samples do not present visual elements that suggest alterations in their surface. These results show that the studied ceramics did not suffer chemical attack by the crude petroleum, which indicates that it presents a good chemical stability when submitted to petroleum in the analyzed period.

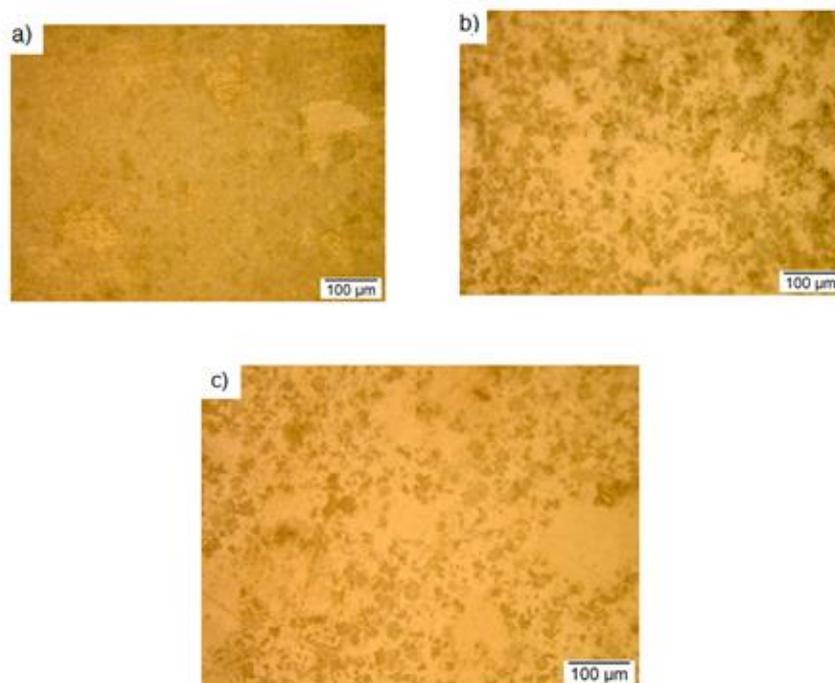


Figure 5: Optical microscopy of  $Sr_2AlZrO_{5,5}$  ceramic a) before immersion in oil; b) after 30 days immersed in oil earth c) after 60 days immersed in oil earth.

Table 2 presents the chromatography of crude petroleum, used in this work, oil extracted from the ground in the coast of the state of rio grande do norte, in the northeast of brazil.

Table 2 – Crude Petroleum Chromatography

Analyzed product	Results	Methods
<b>Water and sediments</b>	0,30	D 4007
<b>Relative density</b>	0,8852	D 5002
<b>Sufidric gas - vapor phase (ppm)</b>	1750	D 5705
<b>Water (Karl Fischer) wt %</b>	0,8392	D 4377
<b>Nacl (mg/L)</b>	7,4	D 3230
<b>API grade density</b>	26,90	D 4057

The result of the chromatography shows that the crude petroleum selected has a small content of water and sediments, in addition to a reasonable salt content (sodium chloride). The result shows that this crude petroleum is of the medium type (26,90 ° api) by the API (american petroleum institute) classification.

#### 4. CONCLUSIONS

In this work a new complex cubic perovskitas oxide  $Sr_2AlZrO_{5,5}$  was produced and characterized by XRD for phase identification. The diffractogram shows the formation of the ordered complex cubic perovskite structure with an

experimental lattice parameter of 8,4487 Å. After the XRD analysis and determination of the perovskite complex cubic structure the calcined material were ground in the agate mortar and subjected to particle size analysis. The granulometric analysis showed homogeneity in particle size and mean particle size of the  $\text{Sr}_2\text{AlZrO}_{5,5}$  system of approximately 24µm. Scanning electron micrographs showed typical microstructure of sintered  $\text{Sr}_2\text{AlZrO}_{5,5}$  ceramics bodies with uniform particle size distribution and surface morphology. Through the analysis of the EDS we verified that the analyzed samples did not suffer contamination during the processing since the only elements that are present in the sample are chemical elements used for the production of these ceramics. It was observed that the increase in temperature did not significantly increase the value of the average hardness of the ceramics. The value of average hardness found in the system produced in this work is considered good when compared to other ceramics of similar properties already researched. In chemical stability tests, it was verified that the samples do not present visual elements that suggest alterations in their surface. These results show that the studied ceramics did not suffer chemical attack by the crude petroleum, good microstructural homogeneity, and good chemical stability when submitted to crude petroleum present strong potential of  $\text{Sr}_2\text{AlZrO}_{5,5}$  ceramics for the production of inert ceramic encapsulation for temperature sensor, used in crude petroleum extraction.

## 5. ACKNOWLEDGEMENTS

Authors are highly thankful for FACEPE and CNPq for financial support.

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