23rd ABCM International Congress of Mechanical Engineering
December 6-11, 2015, Rio de Janeiro, RJ, Brazil

STUDY OF THE STABILITY OF $CA_2ALZRO_{5,5}$ CERAMICS IN CRUDE PETROLEUM AND PRODUCTION OF SUBSTRATES FOR TEMPERATURE SENSORS FOR PETROLEUM EXTRACTION INDUSTRY

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Abstract. *In petroleum production, various types of sensors are used in order to monitor important parameters such as temperature, pressure and flow. The temperature sensors, Resistance Temperature Detectors, generally these devices are built with metal detector elements as resistance temperature encapsulated in inert ceramics. In this study it was produced complex cubic perovskite ceramics for encapsulation of temperature sensors thermomechanical process. The stoichiometric amounts of the constituent chemicals are homogenized, in a ball mill, compacted and calcined at 1150°C. The crystal structure of the compound was determined by X-ray diffraction. The powder was compacted and sintered at a temperature of 1300°C. The microstructure and mechanical properties of the sintered discs were studied by OM, SEM, and Vicker's microhardness test. After these tests the samples were immersed in ground offshore crude petroleum for 60 days and subjected to XRD, OM and Vickers hardness test again to verify the stability of the ceramics in crude petroleum environment. Stability Study on crude petroleum showed no change in the structure and microstructure of the ceramic after immersion in crude petroleum. Therefore, the ceramic Ca2AlZrO5,5 has the potential to be used as encapsulation of temperature sensors in petroleum wells.*

Keywords: *$Ca_2AlZrO_{5,5}$ ceramics, stability in petroleum, inert encapsulation, temperature sensors*

1. INTRODUCTION

Ceramics are multiphasic materials containing metallic and non-metallic elements that form covalent and/or ionic chemical bonds. The existence of several ceramic phases enables combinations of metal atoms, forming many structural arrangements. Thus triggering the acquisition of ceramic materials for a wide application in industry. (Callister Jr., W. D, 1999) (Chandler, M, 1967) (Richerson, D. W., 1982)

The ceramic materials have many properties, they may be good conductors of electricity, super conductors, insulators, magnetic, among others, thus providing various ends in the advanced technology due to, as well, their characteristics of being resistant to high temperatures and corrosive environments, and also for being hard, brittle and for that they have many applications in the field of materials. (Padilha, A. F , 1998) (Callister Jr., W. D, 1999) (Arakawa , T., 1993)

In the petroleum production, for example, various types of sensors are used in order to monitor some important parameters and these sensors are subjected to harsh operating conditions. Therefore they must present an inert and stable behavior in these working conditions. The temperature sensors which are most suitable for the petroleum industry are the Temperature Detectors by Resistance (TDR). Generally these sensors are constructed with metals as temperature detectors by encapsulated resistance in inert ceramics. (Lapa, C., 2004)

The ceramics of complex perovskite structure present the A2BB´O6 or A3B2B´O9 formulas, resulted from the alternation of cations B and B' in the octahedral sites of the unit cell of the simple perovskite. (CHIANG, Y at all, 1997) (Chandler, M, 1967) The main purpose of this research is to produce new ceramics of Ca2AlZrO5,5 cubic complex perovskite structure for the encapsulation of temperature sensors, presenting their characteristics and behavior.

2. METHODOLOGY

The acquisition of the ceramic powder was done by mixing the constituent oxides CaO, Al2O3 e ZrO2. Initially stoichiometric calculations were done to determine the quantity of these oxides necessary to be used for the formation of tablets. The stoichiometry of the reactions was determined from equation 1:

$$2CaO + \tfrac{1}{2} Al_2O_3 + ZrO_2 \rightarrow Ca_2AlZrO_{5,5} \qquad (1)$$

The oxides were weighed on an analytical balance and mixed in a high energy ball mill for 24 hours to homogenize. The homogenized mixture was then deposited in a cylindrical instrument of A2 AISI (HRC 58) type, abrasion resistant with 30 mm diameter. Then the powder was uniaxially compacted in a hydraulic press under a pressure of 5ton/cm² for 5 min, thereby obtaining a tablet of approximately 5 mm thick.

After the compression, the tablets were taken to the high temperature kiln to be calcined. This process was carried out at a temperature of 1150°C for 24 hours. Then the calcined tablets were again taken to the high energy ball mill in order to obtain the powder again and perform the X-ray analysis to identify the ceramic phase.

The ceramic powder was again compacted by uniaxial pressing, however now under a pressure of 10 ton/cm² during the same time of 5 minutes. Then the tablets were taken to the high temperature kiln. This procedure was done at a temperature of 1300°C for 24 hours so that the sintering could take place in solid phase.

After sintering, the tablets were sanded in order to acquire the conditions necessary to be observed in optical microscopy.

The tablets were metallized with carbon to be submitted to Scanning Electron Microscopy (SEM) for a visualization of the characteristics of the grains, as well as EDS to identify the chemical components which are present. They were also subjected to the test of Vickers microhardness.

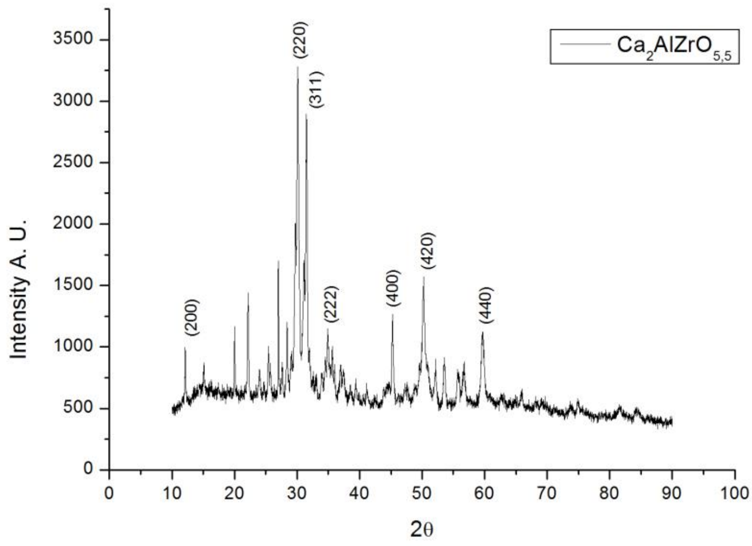
After these steps the pellets were immersed in Sergipe petroleum of sea and land for 60 days. The samples were examined by optical microscopy were also subjected to tests again diffraction of x-rays and Vickers hardness.

3. RESULTS AND DISCUSSION

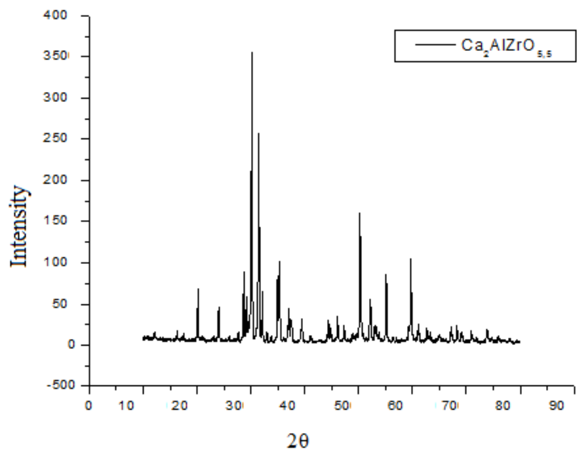
3.1 X-Ray Diffraction

The powder of the calcined sample was subjected to a X-ray diffraction analysis in Siemens D-5000 equipment with copper target (Cu radiation - Kα, with wave length of α = 1.5406 Å) with a scan of 10-90° with an exposure time of 1,0s for the determination of the crystal structure.

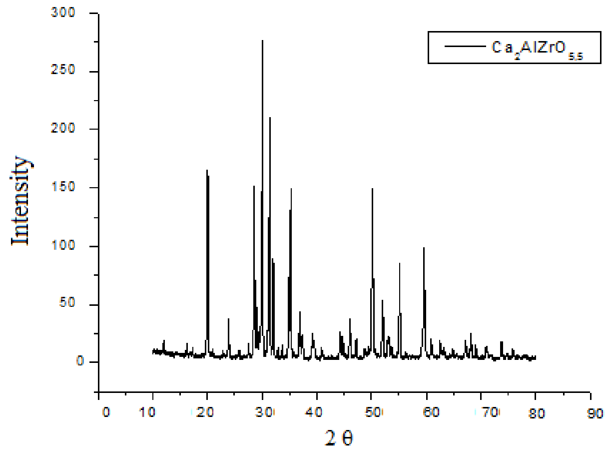
The structural characterization of the compounds after immersion in crude petroleum was analyzed by testing X-ray diffraction. The experimental lattice parameter was found to be 8.3792 Å.



(a)



(b)



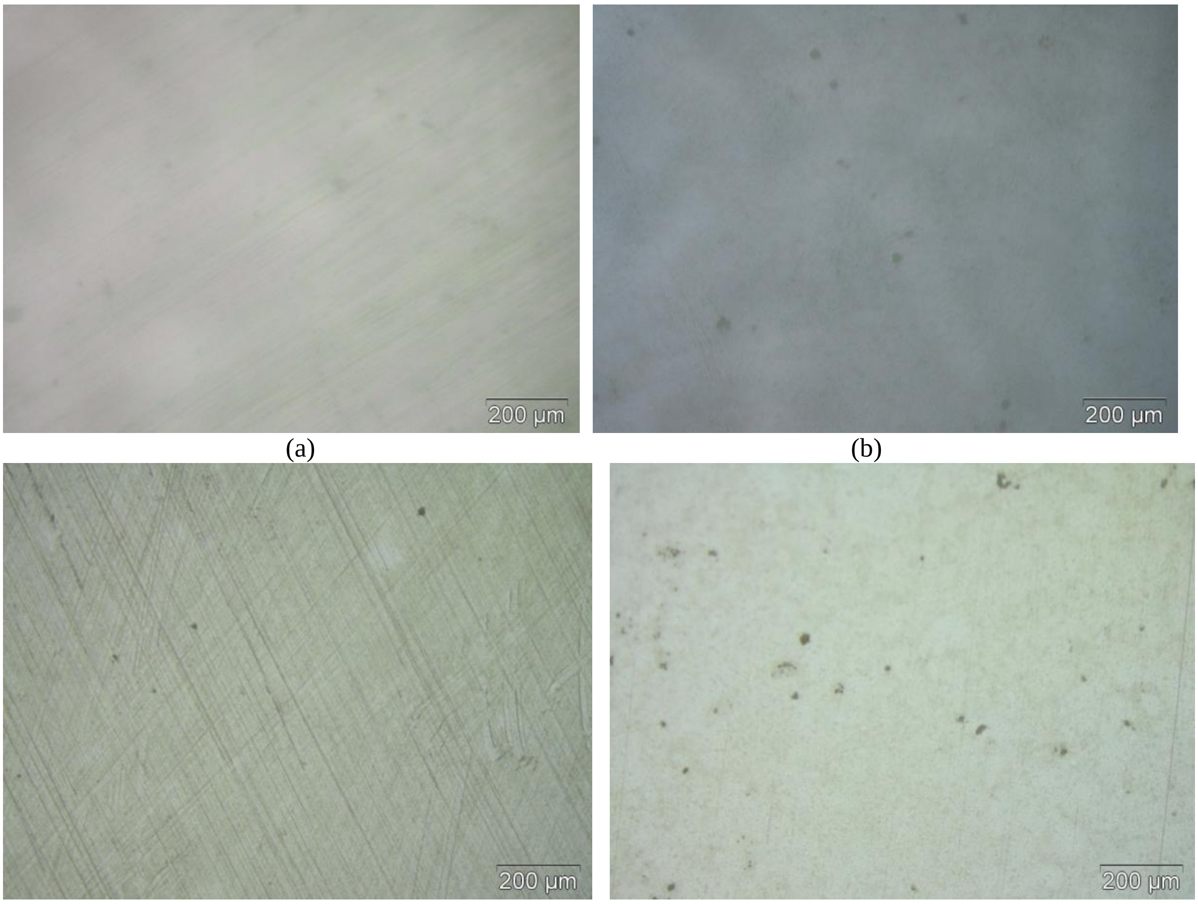
(c)

Figure 1. Sample diffraction $Ca_2AlZrO_{5,5}$ (a) calcined. (b) 60 days after immersion in petroleum sea; (c) 60 days after immersion in petroleum earth.

Through the diffractograms shown in Fig. 1 (a) (b) (c) it can be seen that even after the direct contact with the crude petroleum sea and land, the crystalline structure of the compounds did not change, showing that there was no change to structures of the compounds prior to contact with the petroleum.

3.2 Optical Microscopy

Optical microscopy was performed to analyze the surface of the samples and see if she suffered or not chemical attack due to contact with petroleum. After the sanding of the tablets it was possible to have a display of the sample surface through an optical microscope. The figures show the surface of samples before and after being immersed in petroleum.



(a) (b) (c) (d)

Figure 2. Optical microscopy of the sintered sample to the nearest 100x (a), (b) prior to immersion in petroleum (c) after 60 days immersion in sea petroleum (d) after 60 days immersion in petroleum earth.

When compared with the optical microscopy after 60 days immersion in petroleum, it was found that samples show no visual evidence to suggest changes in its surface. These results show that the compounds studied did not suffer chemical attack by the petroleum, which indicates that they have a good chemical stability when subjected to petroleum during the period.

3.3 Vickers Microhardness

To make the Vickers microhardness test the tablets were sanded so that their surface would be sufficiently reflective. The samples were subjected to the microhardness testing by a hardness tester that applied a load of 500 grams by an indenter shaped as a diamond pyramid for 10 seconds. 10 indentations were made on the sanded surface and the 2 most discrepant were discarded. Of the 8 remaining results an arithmetic average was done in order to obtain the average hardness of the tablets.

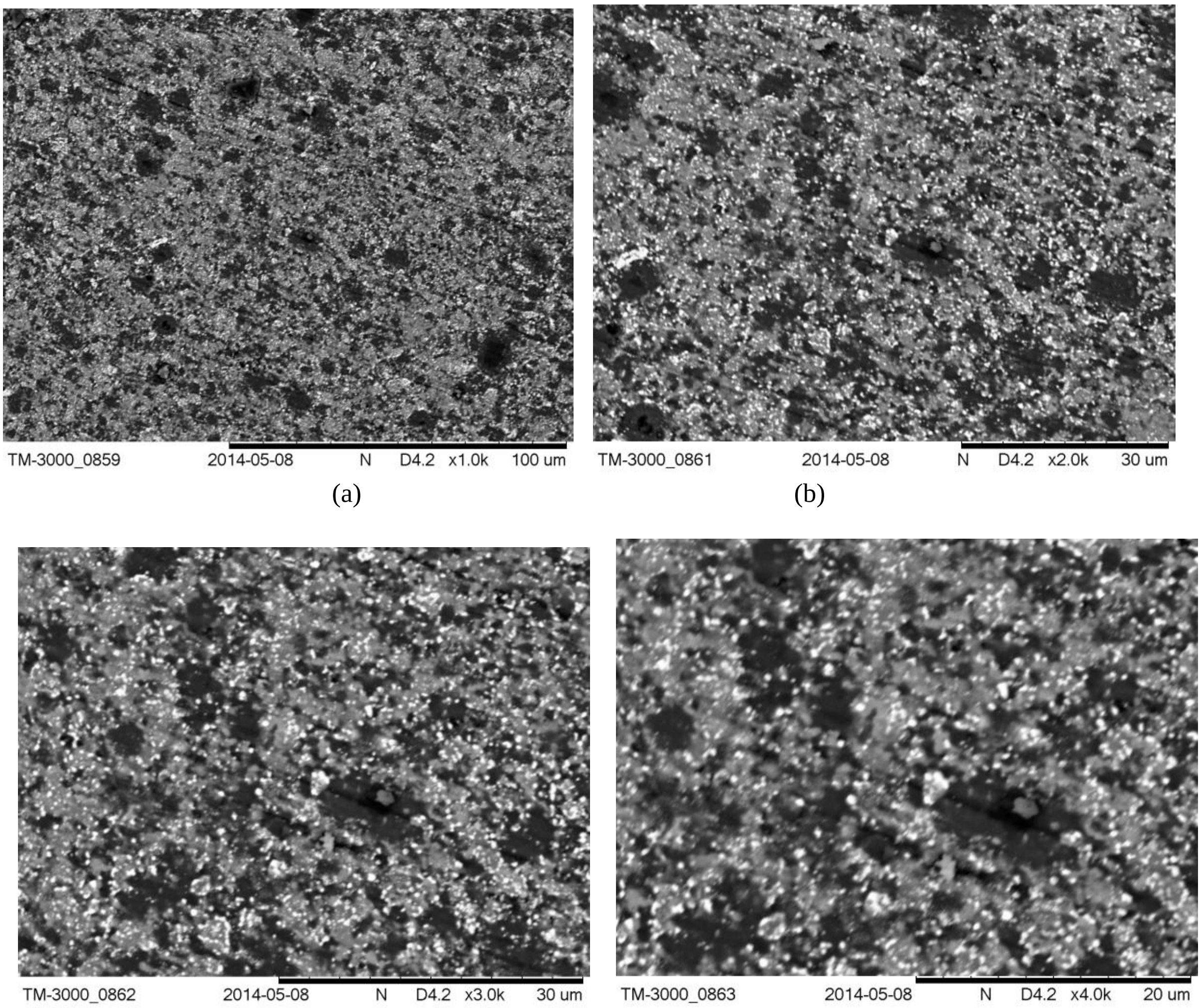
With the Vickers microhardness tests of sintered $Ca_2AlZrO_{5,5}$ it was possible to determine an average hardness number of 160 Kgf/mm². This number is considered reasonable compared to other ceramics with similar properties already researched.

After 60 days of immersion in petroleum samples were retested by the Vickers microhardness test. The tablets immersed in sea of petroleum had an average hardness of 320 kgf / mm^2. In the wafer immersed in the petroleum dirt increase was even greater, 350 kgf / mm^2. This variation may be due to petroleum impregnation. These samples soak acted positively increasing surface microhardness. This increase can only be explained in more detailed investigation using scanning electron microscopy with backscattered electrons.

3.4 Scanning Electron Micorscopy (SEM) and EDS Analisys

To analyze the microstructure of complex cubic perovskite $Ca_2AlZrO_{5,5}$ was used the technique of scanning electron microscopy using secondary electrons. As these ceramics are insulators, before the SEM they were metallized with carbon.

The figures below show typical results of microestrutrural SEM analysis of ceramics sintered at a temperature of 1300 ° C for 24 hours.



(a) (b)

(c) (d)

Figure 3. Results of SEM with magnification (a) 1000x (b) 2000x (c) 3000x (d) 4000x

The analysis of these results shows that there was a good uniformity in particle size and distribution. All ceramics sintered at this temperature showed the same surface morphology. The increase in magnification 1000-4000 times shows the gradual evolution of the microstructure.

Chemical analysis of ceramic compounds was performed by EDS with the scanning electron microscope, shown in Fig 4.

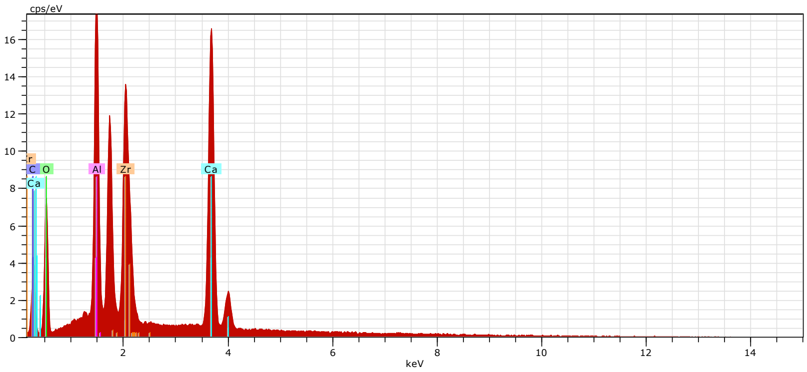


Figure. 4. $Ca_2AlZrO_{5,5}$ SDS sample sintered at 1300ºC.

Through EDS analysis we verified that the only chemical components that appear in the sample are the initially mixed to obtain the ceramic powder. Meaning the sample suffered no contamination in any of the trials.

4. CONCLUSIONS

This work was produced $Ca_2AlZrO_{5,5}$ ceramics by thermomechanical process. After calcination, the crystal structure of the compound was determined by X-ray diffraction showed that the formation of the complex cubic perovskite structure with ordered lattice parameter 8.3792 Å.

The experimental value of the lattice parameter is 11.50% greater than the theoretical value of the lattice parameter (7.515 Å). This difference occurs because in this model the atoms are represented as hard spheres, but in fact they are not, because they have vibrations in atomic positions.

The optical microscopy of the sintered $Ca_2AlZrO_{5,5}$ ceramic at 1300°C showed a good microstructural homogeneity.

The average hardness of samples increased considerably after immersion in petroleum in the analyzed period, the largest increase in the sample immersed in the land of petroleum.

The scanning electron microscopy showed a good distribution and particle size homogeneity of the sintered compound.

EDS showed the presence of only Ca, Al, Zr and O, and has no presence of impurity elements.

Stability Study on crude petroleum showed no change in the structure and microstructure (optical microscopy) of the ceramic after immersion in crude petroleum in 60 days. Therefore, the ceramic $Ca_2AlZrO_{5,5}$ has the potential to be used as encapsulation of temperature sensors in petroleum wells.

5. ACKNOWLEDGEMENTS

Author Rebeka Oliveira Domingues acknowled CAPES for the master fellowship.

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