2013 International Nuclear Atlantic Conference - INAC 2013
Recife, PE, Brazil, November 24-29, 2013
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-05-2**

CHARACTERIZATION OF CARBON, SULFUR AND VOLATILE COMPOUNDS IN NUCLEAR FUEL U_3Si_2-Al

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ABSTRACT

The scope of this work is to describe the characterization of Carbon, Sulfur and Volatile Compounds in nuclear fuel U_3Si_2-Al used in a research pool type reactor with 5 KW power capacities, located in São Paulo, Brazil. This reactor produces a large range of radioisotopes for radiopharmaceutical needed in Brazil nuclear medicine. The fabrication of the fuel U_3Si_2-Al plate is the key of the whole assembly production and its quality directly affects the safety and reliability of the fuel assembly performance. For this reason, it is very necessary to analyze the Carbon, Sulfur and Volatile Compounds to avoid damage in the fuel plate. The Carbon and Sulfur are characterized by the method of radio frequency furnace gas extraction system coupled with infrared cell detector. The Volatile Compounds are characterized by the method of heat gas extraction coupled with gravimetric technique. These methods are recommended by American Society for Testing Materials ASTM for nuclear materials. The average carbon and sulfur analyzed are 30 µg/g and 3 µg/g, respectively. The average for Volatile Compounds is 40 µg/g. These results represent satisfactory performance of the fuel inside the nuclear reactor. A statistical laboratory program has been set to validate the data generated in the nuclear fuel material to specify any agreement with the recommended ASTM methods.

1. INTRODUCTION

The nuclear reactor IEA-R1m located in Instituto de Pesquisas Energéticas e Nucleares IPEN, São Paulo, Brazil, is a research pool type reactor with 5 MW power capacity, that produce a large range of radioisotopes to supply the radiopharmaceutical needed in the Brazilian nuclear medicine. The fuel elements for this reactor, has been produced by IPEN since 1984. The reactor fuel, are plates made of U_3O_8-Al dispersion with 3.2 gU cm^{-3} and U_3Si_2 – Al that can reach 4.8 gU cm^{-3}. The silicide dispersion fuel of U_3Si_2 – Al is currently recognized as the best performance fuel for research and test reactors with a typical fuel loading up to 4.8 gU cm^{-3} for dispersion fuel. The fabrication of the fuel U_3Si_2 – Al is the key of the whole assembly production and its quality directly affects the safety and reliability of the assembly performance [1,2].

The quality control of nuclear fuel is a major item to assure that all these components will work as technical specifications in a nuclear plant. The Carbon, Sulfur and Volatile Compounds are items that make part of this quality control. The Carbon and Sulfur are analyzed by the method of Radio Frequency gas extraction combustion coupled with Infra-red detector. The Volatile Compounds are analyzed by the method of heat gas extraction coupled with gravimetric measurement. These methods are recommended by American Society for Testing Materials ASTM for nuclear materials [3].

2. CARBON AND SULFUR ANALYSIS

The characterization of Carbon and Sulfur in nuclear fuel has been done using the analyzer made by LECO model CS-400. This analyzer is a microprocessor based, software driven instrument for wide-range measurement of carbon and sulfur content of metals, ores, ceramics and other inorganic materials. The CS-400 uses the HF-400 induction furnace and measures carbon and sulfur by infrared absorption. A pre-weighed sample of approximately 1 gram is combusted in a stream of purified oxygen. The carbon in the sample is oxidized primarily to carbon dioxide CO_2 with some carbon monoxide CO possibly being produced. The sulfur is oxidized to sulfur dioxide SO_2. These gases are swept, along with the oxygen, through a dust filter and drying reagent into an infrared cell IR where sulfur is detected as SO_2. The gases are then routed through a heated catalyst which converts CO to CO_2 and SO_2 to sulfur trioxide SO_3. The SO_3 is removed by a filter and CO_2 is measured in separate IR cell. Carbon measurements will be made based on the range selected. The low carbon range features a longer path-length IR cell, while the high carbon range utilizes a shorter path-length IR cell. The difference in path length assures optimum resolution for the range selected. The CO_2 and SO_2 absorb IR energy at precise wavelength within the IR spectrum. Energy at these wavelengths is absorbed as the gases pass through respective IR absorption cell. Changes in energy are then observed at the detectors. The absorption of IR energy is attributed to only CO_2 and SO_2 depending on the cell, and its concentration is then determined. Each IR cell serves as both a reference and measure chamber. This analyzer has been used for qualify all the stain steel that been used in the nuclear cycle, besides are used for many proposes in the nuclear area in IPEN. Figure 1 shows the LECO CS-400 analyzer.

Figure 1: Characterization of Carbon and Sulfur by Radio Frequency gas extraction combustion coupled with Infra-red detector method, LECO CS-400.

2.1. Volatile Compounds analysis

During the manufactured process of the nuclear fuel, permanent gases like Hydrogen, Nitrogen, Oxygen and Water are trapped in the interstitials spaces of the fuel. These trapped gases are known as Volatile Compounds residual gases. In the manufacturing process, the fuel is set up at high sintering temperatures surrounding by a reducing gas environment, gases like Argon / Hydrogen or Nitrogen / Hydrogen are common sintering gases. During this cycle, some gases get trapped and will be released during the operation conditions of the reactor. They may pressurize the clad, alter the thermal conductivity of cover gas, or react with the clad and cause damage. Hence, it is of importance to have a precise knowledge of the Volatile Compounds content.

The characterization of Volatile Compounds in nuclear fuel has been done using the analyzer made by IPEN model GB-IPEN-101. The analyzer is a home-made gas extractor system built in IPEN laboratories. The GB-IPEN-101 consist of a metal glove-box of 0.5 m^3 volume capacity within control atmosphere that avoid humidity and oxygen airborne contamination using an inner gas atmosphere at positive pressure. The glove-box has a glass visor that allows to the analyzer operate with rubber gloves inside the box without contamination. Inside the glove-box is set a precision balance made by Mettler model H20T where can be measurement variation of mass at the order of 0.00001 g. The glove-box is link to a resistive furnace outside the box through a quartz glass tube. The nuclear fuel samples to be analyzer are carrying on by a quartz glass crucible with special hooks drive by a glass pointer inside the middle of the furnace. The resistive furnace has a capacity to heat until 500 ^oC, this temperature is controlled by a computing LabJack U12 and a solid state relay with a precision of ±1 ^oC. The glove-box and the quartz tube has independently its gas supply and both does not contamination with the airborne of the lab (Figure 3). The GB-IPEN-101 has been used to measurement the stoichiometry ratio O/U in UO2 nuclear fuel as well [4]. Figure 2 shows the GB-IPEN-101 analyzer.

Figure 2: Characterization of Volatile Compounds by heat gas extraction coupled with gravimetric measurement method, GB-IPEN-101.

Figure 3 show a schematic plot of the GB-IPEN-101. The parameters used to analyses the Volatile Compounds in the nuclear fuel U_3Si_2 – Al by GB-IPEN-101 are the follow: furnace temperature, 120 ^{o}C, atmosphere in the glass quartz tube, Helium, mass sample 200 mg and time in the furnace 1 hr. The fuel sample is set inside the quartz glass crucible, and then the sample is weighted before and after this procedure inside the glove-box by the analytical balance with controlled atmosphere. The GB-IPEN-101 analyzer gives the following data m_f and m_i that are final and initial mass, respectively, the equation 1 calculate the Volatile Compounds (VC) inside the sample in micrograms per grams of fuel.

$$VC\left(\frac{\mu g}{g}\right) = \frac{m_f - m_i}{m_i} \cdot 10^6 \tag{1}$$

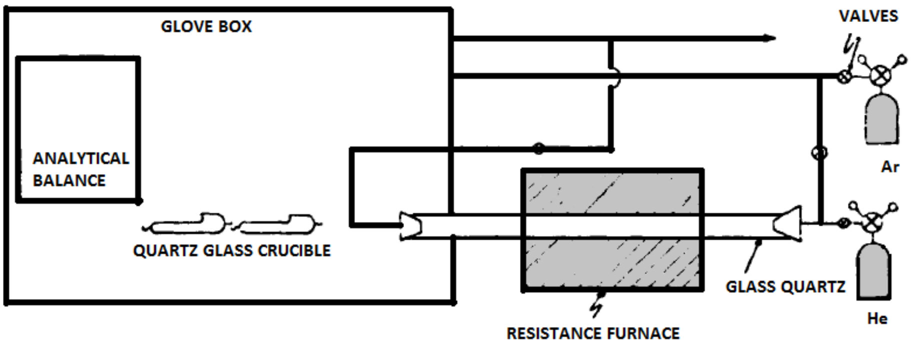

Figure 3: Schematic plot of the GB-IPEN-101

2.2. Statistical laboratory program

A statistical laboratory program has been set to validate the data generated in the nuclear fuel material to specify any agreement with the recommended ASTM methods. The validation program is applied in the IPEN laboratories for assessing how the results of a statistical analysis will generalize to a reliable data set. The validation program set many statistical parameters such as selectivity, linearity, detection limit, quantification limit, precision and accuracy, these parameters are definite by two agencies in Brazil, these are, ANVISA (Agência Nacional de Vigilancia Sanitária) and INMETRO (Instituto Nacional de Metrologia, Normalização e Qualidade Industrial) [5,6].

2.1.1. Results and discussion

Table 1 present the results of three nuclear U_3Si_2 – Al fuel elements where the Carbon, Sulfur and Volatile Compounds were analyze. Carbon and Sulfur were determined using method of Radio Frequency gas extraction combustion coupled with Infra-red detector. Volatile Compounds were determined using method of heat gas extraction coupled with gravimetric measurement. The results show that all fuel elements are below the maximum limit established by the nuclear quality control manual.

The statistical results show a precision and accuracy in the range of 10%, these results are compatibility with the ASTM mandatory regulation for the nuclear fuel in the manual of the quality assure for U_3Si_2 – Al nuclear fuel.

Table 1: Carbon, Sulfur and Volatile Compounds in U_3Si_2 – Al nuclear fuel

Fuel element	01	02	03
Carbon (µg/g)	33 ± 2	35 ± 3	26 ± 2
Sulfur (µg/g)	2.0 ± 0.1	6.0 ± 0.2	3.0 ± 0.2
Volatile compounds (µg/g)	40 ± 5	36 ± 2	42 ± 3

3. CONCLUSIONS

The average Carbon is 30 µg/g and Sulfur is 3 µg/g. The average for Volatile Compounds is 40 µg/g. These results represent a satisfactory performance of the fuel inside the nuclear reactor. The quality control of the nuclear fuel with reference to gases is completed with the analysis of Hydrogen, Nitrogen and Oxygen that has been reported elsewhere. All the nuclear fuel is mandatory to test the quality control of gases in the fuel to assure a well performance of the fuel inside the nuclear reactor IEA-R1m located in Instituto de Pesquisas Energéticas e Nucleares IPEN, São Paulo, Brazil.

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