

# DEVELOPMENT OF LEU SILICIDE FUEL ELEMENT FOR THE IPEN RESEARCH REACTOR

M. DURAZZO, E.F URANO DE CARVALHO, A.M. SALIBA-SILVA, J.A.B. SOUZA  
*Centro do Combustível Nuclear, Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN/SP  
Av. Prof. Lineu Prestes 2242, 05508-000 São Paulo – Brazil*

H.G. RIELLA

*Departamento de Engenharia Química, Universidade Federal de Santa Catarina  
P.O.Box 476, 88040-900 Florianópolis – Brazil*

## ABSTRACT

IPEN has been increasing continuously its production of radiopharmaceutical medicines to come along with the expanding demand imposed by the Brazilian welfare. To reach its objectives, IPEN has been working in upgrading the IEA-R1 research reactor to increase its power from 2 to 5 MW and its operational time from 64 to 120 hours by week. Since 1988 IPEN has been manufacturing its own fuel element. A program for autonomous serial fuel assembly production started in the 80's, motivated by the political constraint for buying these fuels abroad. The fuel was initially based on  $U_3O_8$ -Al dispersion fuel plates with  $2.3 \text{ gU/cm}^3$ . Based on previous experience with  $U_3O_8$  fuel, a new higher uranium density fuel was developed to attend a more intense reactivity needs for continuous operation and to have a more compact core for better irradiation flux. This paper describes the developing path of silicide fuel at IPEN.

## 1. Introduction

The use of radioisotopes in medicine is certainly one of the most important social uses of nuclear energy and IPEN/CNEN-SP has a special place on the history of nuclear medicine in Brazil. Due to the federal monopoly, only the Institutes that belong to CNEN (Comissão Nacional de Energia Nuclear) can produce radioisotopes and radiopharmaceuticals for use in nuclear medicine. The production of IPEN represents nearly 98% of the total produced.

There has been a significant increase in the demand of radioisotopes over the years. Between 2002 and 2004 the increase in the demand was about 30%. Distributed to Brazilian hospitals and clinics, radiopharmaceutical products were used, in 2006, to attend more than 3 million patients, an increase of about 10% relative to the year before. To face this scenario, IPEN has been increasing continuously its production of radiopharmaceutical medicines to come along with the expanding demand imposed by the Brazilian welfare. One of the most important project aims at own production of Mo-99 in order to provide cheaper Tc-99 generators than the ones produced from imported raw materials. So, this medicine will be accessible to a bigger amount of patients. To reach the objective of this project, IPEN for years has been working on upgrading the IEA-R1 research reactor to increase its power (from 2 to 5 MW) and its operational time (from 64 to 120 hours by week). The reactor control board and ventilation system were upgraded and additional safety items were incorporated. Besides, CNEN is planning to build a new facility in order to expand and spread the use of radiopharmaceutical medicines all over the country.

Once the reactor power was planned to be raised from 2 to 5 MW, in 1997 IPEN started research activities aiming at the elevation of the uranium loading inside the fuel, in order to allow the reactor power increase. This was accomplished with the development of the silicide technology, which has been done under support of the IAEA. The Technical Cooperation Project BRA/4/047 "Fuel Improvement for the IPEN Research Reactor" was the objective to develop high-density fuel to improve the efficiency and applicability of the IPEN reactor. Until this time, fourteen silicide fuel elements ( $3.0 \text{ gU/cm}^3$ ) had been irradiated at the IEA-R1 research reactor. The irradiation was closely followed by visual inspections and sipping tests. After 40% burn up (average), no problems regarding to fuel performance was recorded. The

enriched  $U_3Si_2$  powder was imported from the international market until 2002, when IPEN started the development of the conversion technology to get de  $U_3Si_2$  powder using national enriched  $UF_6$  produced by CTMSP (Marine Technological Center in São Paulo). Nowadays, IPEN is able to fabricate the enriched  $U_3Si_2$  powder, allowing the nationalization of all the fabrication cycle of dispersion fuel for research reactors. After mining and enrichment steps, IPEN is able to execute all the other fabrication steps. Recently, on 2007 June 26, the first fuel element fabricated with national materials and technology was put in operation at IEA-R1 reactor core. This work describes the journey of IPEN heading on to acquire uranium silicide technology.

## 2. Historical Background

The beginning of the development of the fuel element fabrication technology in IPEN is very old. The work started in 1960 aiming at fabrication of the fuel for the ARGONAUTA research reactor. Between 1964 and 1965 the fuel elements were manufactured with 20% enriched  $U_3O_8$  powder provided by IAEA in the program Atoms for Peace. In spite of the low technological demand of the ARGONAUTA fuel (very low power), a seed was planted and would come to germinate 20 years later, in the decade of 80, and to bloom definitively in the decade of 90, when IPEN dominated the fabrication technology and began the production of the fuel for the IEA-R1 research reactor. The relative high power (2 MW) demanded a significant technological progress in the fabrication techniques.

Starting from 1980, IPEN intensified their efforts to develop the fabrication technology of dispersion fuel element, aiming at improving the technology for manufacturing fuels more advanced, substantially superior to the old ARGONAUTA fuel type. In that time, IPEN could not acquire fuel elements from the international market in order to supply the IEA-R1 research reactor. The growing difficulty to get fuel elements in the international market acted as an initial impelling force for IPEN to deflagrate their program for fuel element fabrication. The technology previously developed in the 60's was updated starting from 1985, with base in the recent technological advances in the area. Between 1985 and 1988, IPEN worked in assembling a small fuel fabrication facility as a laboratory level, with capacity to produce 6 fuel elements by year. This was enough to supply the IEA-R1 reactor operating at 2 MW and 40 hours a week.

On 1988 August 31, as part of the commemorations of their 32<sup>o</sup> anniversary, IPEN provided the IEA-R1 reactor with the first fuel element fabricated in Brazil, only fifteen days before the exhaustion of the reactor fuel. The fissile material used was the same  $U_3O_8$  powder previously used for the production of the ARGONAUTA fuel. There was a reserve of about 30kg of this material. Starting from 1988, after the production of the first fuel element, IPEN began a continuous production of fuel element, which continues until nowadays.

After the production of 26 fuel element, the enriched  $U_3O_8$  powder finished in 1996. So, in 1994 IPEN started developing the processes for  $UF_6$  conversion to  $U_3O_8$  and for recovering the uranium scraps generated in fuel plate fabrication. In 1996, IPEN did the conversion of about 20kg of imported enriched  $UF_6$ . IPEN was then prepared for the routine production of fuel elements starting from  $UF_6$  as raw material. In 1997, IPEN raised the fuel production capacity from 6 annual fuel elements to 10, which was the maximum considering the infrastructure available.

As mentioned before, in order to increase the radioisotope production of IPEN, the IEA-R1 reactor power capacity was increased from 2 MW to 5 MW. In 1997, the development of a new higher uranium density fuel started, in order to attend the reactivity needs for continuous operation, to have a compact core for better irradiation flux and also to have a low number of irradiated fuel elements to be stored at the spent fuel pool. The new fuel was based on the  $U_3Si_2$ -Al dispersion with uranium loading of  $3.0 \text{ gU/cm}^3$ . In 1998, the fuel plate fabrication technology of the new silicide fuel was implanted. At that time,  $U_3Si_2$  powder was imported from France. Between 1999 and 2000 sixteen silicide fuel elements were manufactured. Starting from 1998, the efforts to develop the  $U_3Si_2$  powder production technology began, aiming at the nationalization of all the production process, starting from enriched  $UF_6$  leading to the conversion to  $UF_4$ ; its reduction to metallic uranium;  $U_3Si_2$  powder fabrication and,

finally, arriving at the fuel plate fabrication and fuel element assembly. At this time, IPEN participated in an international cooperation through IAEA under the Technical Cooperation Project TC BRA/4/047.

In 1999 IPEN got the technology for  $UF_4$  production using  $SnCl_2$  as reducing agent. In the area of metallic uranium, IPEN was a valuable previous experience in producing 150kg natural uranium ingots in 90's. Based on that experience, IPEN initiates efforts aiming at scaling down the size of the metallic uranium pieces, trying to produce pieces of about 3kg using 20% enriched material as raw material for the  $U_3Si_2$  production. In 2002, the process for producing metallic uranium was dominated, which allowed the development of the  $U_3Si_2$  intermetallic. In 2004, IPEN obtained the first lot of natural  $U_3Si_2$  powder, manufactured with national technology, dominating then the "uranium silicide cycle". In 2006, IPEN consolidated the fabrication technology of the silicide fuel by manufacturing the first fuel element with fully national technology. This fuel element was put in the IEA-R1 reactor core last year, on June 26. Now, efforts have been made to produce and qualify high loaded fuel elements with  $4.8 \text{ gU/cm}^3$ .

Due to the emergent increase in radiopharmaceuticals demand and the consequent increase in the IEA-R1 power, the reactor needed an increase of fuel, from 6 ( $U_3O_8$ -Al) to 18 elements ( $U_3Si_2$ -Al) a year. In addition, a new reactor for radioisotope production was cogitated to be constructed in the Northeast region of Brazil. This decision would be very significant in near future, since IEA-R1 reactor is quite aged (50 years) and this reactor is practically the only one producer of radioisotope in the country. The new reactor (probably 20 MW) would consume about 30 annual  $U_3Si_2$ -Al fuel elements. Therefore, a demand for about 50 annual fuel elements seems to be quite realistic.

Based on this forecast demand for fuel elements and the increasing need for fuel element production to supply IEA-R1 reactor in the near future, IPEN began a project in 2001 in order to adapt the production facilities seeking on improving the producing capacity. This project is nowadays under way aiming at replacing the current facilities by a fully new one, but with a researching laboratorial style. The new facility is planned to have nominal capacity for producing 30 fuel elements in yearly basis. That will attend wholly the fuel element demand in a short period. The producing capacity of the new facility should reach 80 annual fuel elements, which would supply also the new research reactor planned to be constructed. The conclusion of this project is foreseen for 2008-2009.

Looking at the future, in 2001, IPEN started working with the U-Mo fuel in contribution to RERTR program for developing high uranium loaded fuels. This fuel could eventually substitute with advantages the  $U_3Si_2$  fuel. Nowadays this work is on course [1] and was also supported by the IAEA TC BRA/4/053 "Development of Alternative High-Density Fuel Based on Uranium-Molybdenum Alloys".

The production increased of fuel element will generate larger amount of waste (liquid, solids and gaseous) and pollutant, which has to be handled adequately. The higher fabrication scale of the new installation arise technical difficulties related to upgrading uranium recovery procedures and effluent treatment and waste disposal. As the facilities for fuel plates production are adequate to stand up the new production level, the main modifications and adjustments must be performed in the chemical processes for uranium recovery from scrap and for treatment on liquid waste and gaseous or aerosol-type pollutants, generated in fabrication plant. To face this new challenge, IPEN rely on the international skill in these subjects with support of the IAEA TC BRA/3/012 "Nuclear Fuel for Research Reactors: Improving Fabrication and Performance Evaluation in Brazil".

### **3. Silicide Fuel Development**

The IEA-R1 Reactor of IPEN/CNEN-SP is a pool type reactor operating since 1957. This reactor uses MTR type dispersion fuel element in a 5 X 5 core arrangement. The Nuclear Fuel Center of IPEN is responsible for the production of the necessary nuclear fuel to keep

the continuous operation of the reactor. Development of new fuel technologies is also a permanent concern. The Nuclear Fuel Center had produced 77 fuel elements until now, including 14 control fuel elements.

The fuel meat is fabricated according to conventional powder metallurgy techniques. The fuel element contains 18 fuel plates, each one 1.52 mm thick. Cladding and frame plates are made with the ASTM 6061 aluminum alloy. The fuel assembly is performed by a well-known picture-frame technique. The fuel element results from mechanical assembling of 18 fuel plates and other structural components.

As, in Brazil, we have not available hot cell laboratories to test irradiated fuels and as the irradiation tests abroad would be very expensive, IPEN decided to risk the testing and evaluating in pile for its own fuel, checking the performance under reactor operation. This was possible since IPEN fuel specification is conservative for dispersion fuel and the power of reactor is low. A program for fuel qualification was started with irradiation of some miniplates at the border of the reactor core just to identify any abnormal event. In July 1985 a partial fuel element with only two fuel plates (the external plates) and 16 aluminum plates was placed in the core to start fuel qualification. After this, other partial fuel element with 10 fuel plates and 8 aluminum plates was also placed in the core (November 1985). These two fuel element were identified as the precursor fuels. A periodic monitoring and evaluation was done upon them. After good results with these precursor fuels, it was decided to start loading standard fuel elements in the reactor core (August 1988), with  $1.9 \text{ gU/cm}^3 \text{ U}_3\text{O}_8\text{-Al}$ . The adopted criteria was that each IPEN fuel element had to start irradiation at peripheral positions of the core, with lower power densities, up to 4 % burnup (almost one year of irradiation) and then it could go to higher power density positions in the core. It was decided that the precursor fuels had to stay in the core up to the same time that a complete fuel element would stay. The last precursor fuel assembly was taken out of the reactor core without any operational problems. Previous paper presented details about the fuel qualification program [2]. To qualify the  $\text{U}_3\text{Si}_2\text{-Al}$  fuel the same strategy was adopted. In this case, the volume fraction of the fissile material in the dispersion was kept the same, around 27 %, resulting in a uranium loading of  $3.0 \text{ gU/cm}^3$ .

The program for silicide fuel development at IPEN had great impulse after the approval of the IAEA TC BRA/4/047 "Fuel Improvement for the IPEN Research Reactor" in 1999. The primary purpose of this program was to develop the whole fabrication process of  $\text{U}_3\text{Si}_2\text{-Al}$  dispersion fuel plate (including  $4.8 \text{ gU/cm}^3$ ), its irradiation test at the IEA-R1 reactor and post-irradiation analysis. This project gave the necessary background to IPEN to produce and qualify its own  $\text{U}_3\text{Si}_2$  powder and silicide based dispersion fuel plates for the IEA-R1 fuel element fabrication. The project steps to achieve the objectives included the following steps:

- a) to develop the process for producing  $\text{UF}_4$  starting from  $\text{UF}_6$ ;
- b) to develop the process for producing metallic uranium starting from  $\text{UF}_4$ ;
- c) to develop the process for producing  $\text{U}_3\text{Si}_2$  powder;
- d) to produce miniplates with 20% enrichment for irradiation tests;
- e) to irradiate miniplates at the IEA-R1 reactor and to perform non-destructive analysis on the irradiated fuel miniplates inside the spent fuel pool.

### 3.1 $\text{UF}_6$ conversion towards $\text{UF}_4$

Essentially, the process to get  $\text{UF}_4$  from  $\text{UF}_6$  uses the reduction of hexavalent uranium present in aqueous solution to the tetravalent state and its precipitation as  $\text{UF}_4$  by means of the HF solution. The preparation of  $\text{UF}_4$  using chemical reduction has been carried out starting from  $\text{UO}_2\text{F}_2$  solution resulting from  $\text{UF}_6$  hydrolysis. The solution is heated under continuous stirring to reach a temperature set point and then the reducing agent is added. Next, the hydrofluoric acid HF precipitating agent solution is slowly added. Tests have been carried out using different reducing agents, such:  $\text{SnCl}_2$ ,  $\text{CuCl}$ ,  $\text{FeCl}_2$ , and  $\text{Na}_2\text{S}_2\text{O}_4$ . The reducing agent  $\text{SnCl}_2$  was the one that showed the best results and achieved a level of  $\text{UF}_4$  precipitation in the range of 98%. However, during the  $\text{UF}_4$  preparation an amount of water is absorbed by the  $\text{UF}_4$  crystal. This water could interfere in the other process steps, and to

avoid problems it must be removed.

The elimination of crystallization water from the  $UF_4$  is carried out in temperatures near  $400^\circ C$ , under a constant flow of argon over the surface of the powder, which avoid the  $UF_4$  oxidation to  $UO_2F_2$  or  $UO_2$  and drag the water vapor released. The material, in a powder form, is placed in a small Monel boat and introduced inside the heating chamber. After purging with argon for 1 hour the system is heated to  $400^\circ C$  under flow of gas. The  $UF_4$  powder remains in this temperature for 1 hour, which is a sufficient time to complete the water elimination. The subsequent cooling is carried out keeping the flow of argon till the temperature reaches  $100^\circ C$ . Details of the  $UF_4$  precipitation procedures were presented in a previous work [6].

### **3.2 $UF_4$ Reduction to Metallic Uranium**

The intermetallic  $U_3Si_2$  is produced from metallic uranium. To produce metallic uranium in the required quantity (up 80 kg/year) IPEN has developed a process to produce small quantities of uranium (1000g) through magnesiothermic routine. The 1000g ingot of metallic uranium is rather small if compared to previous practice of IPEN. In 1980's, ingots of natural uranium ranged up to 100kg. The downscaling was difficult, since not only the reduced scale for the crucible was conceived, but also lower material amount to be reduced implied in difficulties to get reasonable metallic yields. The crucible itself has been planned several times to achieve the final format using a bottom extractable system to help removing the cast ingot from the crucible. The thermal profile of the furnace has already been achieved by experimentation and calculations, but to reach optimized yields it was necessary to identify the exact moment of the reaction. It was got by means of an accelerometer in order to recognize the sound waves perturbation during the reaction moment.

Optimization of this method was met only after 25 experiments with natural uranium simulations in the last 3 years. Once settled down this technology, IPEN started to produce the 20% LEU enriched uranium. The yield of this system is around 82-85% of metallic uranium. The enriched material was already produced with good operation and quality results.

### **3.3 $U_3Si_2$ fusion and powder fabrication**

Since 2000, the Nuclear Fuel Center of IPEN has been dedicating great efforts to achieve expertise in production of intermetallic alloy  $U_3Si_2$ . After facing some difficulties, as reported previously [7], in 2004 IPEN has arrived to the full experimental route to produce, in production scale, the necessary alloy for nuclear fuel.

From the produced uranium ingot, the metal was melted inside an induction furnace with silicon addition, with an adequate vacuum instrumentation and facilities for handling and melting uranium and uranium alloys. The zirconia crucible was specially designed to reach temperatures higher than  $1750^\circ C$  and to support the aggressive environment created by uranium chemical attack. More than 20 trials were carried out, using natural uranium, before the first LEU  $U_3Si_2$  were successfully made. It was produced 3 enriched  $U_3Si_2$  melting in 2005, which consisted the first own produced load of fuel plate fabrication in IPEN. In general terms, the quality of this intermetallic has fully met the needs postulated by the requirements for a routine nuclear material. The X-ray diffractogram attested the presence of the expected phases in the produced powder of this alloy.

### **3.4 Miniplate fabrication**

As previously mentioned, IPEN started manufacturing its own fuel element by using  $U_3O_8$ -Al dispersion fuel plates with  $1.9 \text{ gU/cm}^3$ . This uranium loading represents a  $U_3O_8$  volume fraction of 27 %. If this same volume fraction is used, the direct substitution of the  $U_3O_8$  by  $U_3Si_2$  would result in an increasing of uranium loading to  $3.0 \text{ gU/cm}^3$ .

The increasing of  $U_3Si_2$  volume fraction in the dispersion has a technological limit of 45 %, in order to allow fabrication of fuel plates. This means maximum uranium loading of 4.8

gU/cm<sup>3</sup>. This is the objective to be reached in the future. The increase in the volume fraction of both U<sub>3</sub>O<sub>8</sub> and U<sub>3</sub>Si<sub>2</sub> to 45 % implies in implementing modifications in the fabrication procedures in contrast to the currently adopted ones for LEU fuel. The necessary research work to adequate the fabrication procedures have been done under the IAEA TC BRA/4/047. It was fabricated 22 miniplates with approximately 120 mm length and 42 mm width fuel meats. The compacts were 20 X 40 mm. Aluminum alloy 6061 was used for the frames and cladding plates. The hot rolling temperature was 440°C. The cladding thickness over the defect zones (dog-boning) was 0.28 mm (specification states a minimum of 0.25 mm). All the fabricated miniplates showed good metallurgical bonding. In the first fabrication tests it could be observed some oxidation of U<sub>3</sub>Si<sub>2</sub> particles located near the interface core/cladding. The volume fraction of the oxidized phase was quantified by means of image analysis and the results indicated that a volume fraction between 2 and 4 % of the U<sub>3</sub>Si<sub>2</sub> particles has been oxidized. The presence of the measured volume fraction of this oxide phase, within the fuel core, was considered to be no deleterious to the fuel performance during irradiation. However, a study of the oxidation phenomenon was carried out to eliminate or minimize the oxide formation during the fabrication. The cause was determined to be the welding procedure. A modification in the welding device was done to minimize the air entrapment in the weld assembly. This was possible with the use of a glove-box with argon atmosphere. The oxidation problem was no more observed ever since.

### **3.5 Miniplate irradiation at IEA-R1 research reactor**

The irradiation device was planned to be placed at the IEA-R1 core support plate. Its overall dimensions were similar to the fuel assembly dimensions. It was projected to contain miniplates for irradiation and also to allow their removal for post-irradiation NDT to be performed at the spent fuel pool. The project assumed the possibility to irradiate up to 10 fuel miniplates at once. It was designed an internal support for the miniplates, which could be assembled inside the irradiation box. The post irradiation examination includes miniplate thickness measurement, visual inspection and sipping. The fuel miniplate thickness measurement device will be used inside the reactor pool, in the spent fuel storage area. It will be operated from the reactor pool border and will be able to measure the fuel miniplate thickness variation along its surface. This device is based on a mechanical structure for positioning the miniplate and to perform the scanning along the miniplate surface. The thickness measurement will be performed by electronic probes (LVDT probes). The results are obtained by measurement instrumentation connected to the probes and the data are stored and processed by a laptop computer.

## **4. Conclusion**

The developments involving fuel fabrication was satisfactorily completed. The next step is to fabricate full sized U<sub>3</sub>Si<sub>2</sub>-Al fuel plates with 4.8 gU/cm<sup>3</sup>. This work was already started. The experimental activities using the reactor have to be defined in written documents. Operational procedures was already written and analyzed by the reactor operational staff. The beginning of irradiation is waiting for a final approval of the Reactor Safety Review Committee for the experiment.

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