

## Fabrication of $U_3Si_2$ Powder for Fuels used in IEA-R1 Nuclear Research Reactor

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**Abstract.** Powdered uranium silicide ( $U_3Si_2$ ) 20%  $U^{235}$  enriched is an intermetallic compound used as nuclear fuel material dispersed in aluminum to be the meat of fuel elements.  $U_3Si_2$  powder is the state-of-the-art as nuclear fuel material mostly used in modern research reactors. Its recent established fabrication in IPEN replaced the previous ceramic powder  $U_3O_8$  used in the fuel of IEA-R1 (IPEN/CNEN, São Paulo, Brazil). The  $U_3Si_2$  is a compound with 92.3%wtU and 7.7%wtSi. Its production is made by induction furnace melting using metallic uranium, produced by magnesiothermic reaction, and pure silicon. The induction furnace melts under argon controlled environment using zirconia crucible. Homogenization of liquid bath at 1800°C is a compromise between crucible resistance and homogenized melting, avoiding hazardous happenings. IPEN produced its first lot of enriched  $U_3Si_2$  in September 2004, with a continuous fabrication ever since. This research work represents the ability of having fully Brazilian supply of this strategic and high cost nuclear material. The fuel quality meets the world quality standards required by International Atomic Energy Agency (IAEA) and RERTR standards. Brazilian production of  $U_3Si_2$  powder not only closed the fuel cycle, from uranium mineral to fuel element, but also allowed higher productivity of nuclear medicine radioisotopes by IEA-R1.

### 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. The production of radiopharmaceutical products made by IPEN represents nearly 98% of the total Brazilian demand. In 2006, IPEN attended more than 3 million patients. One of the most important project aims at production of Mo-99 to provide Tc-99 generators. To face this scenario, IPEN has increased continuously its production of radiopharmaceutical medicines, so IPEN upgraded the IEA-R1 research reactor power from 2 to 5 MW and its operational time from 64 to 120 hours by week. In 1997 IPEN started research activities for increasing the uranium loading inside the fuel (meat). The Brazilian silicide technology with  $U_3Si_2$  meat loadings was carried out through powder metallurgy. Earlier, until 2002, the enriched  $U_3Si_2$  powder was imported, then IPEN started to develop the conversion technology to get de  $U_3Si_2$  powder using national enriched  $UF_6$  which is produced by CTMSP (Marine Technological Center in São Paulo). Up to now, 14 silicide fuel elements, having meat density of 3.0gU/cm<sup>3</sup>, have been irradiated at the IEA-R1m research reactor. The irradiation was closely followed by visual inspections and sipping tests and after 40% burn up (average), no problems related to fuel performance arose.

IPEN is now able to execute all the other fabrication steps. Recently, in June 2007, the first completely Brazilian fuel element was put in operation in the IEA-R1 reactor core. This work describes the journey of IPEN heading for learning the silicide technology.

## Historical Background

Since 1960 decade, IPEN made efforts to develop the fabrication technology of dispersion fuel element, aiming at improving the technology for manufacturing more advanced fuels. For military and political reasons, IPEN was not able to acquire fuel elements from the international market, for many decades, to supply the IEA-R1 research reactor. The technology previously developed in the 60's was updated and from 1985 onwards, IPEN worked in assembling a small fuel fabrication facility at a laboratory level, with capacity to produce 6 fuel elements per year. This was enough to supply the IEA-R1 reactor operating at 2 MW and 40 hours by week. In 1988, IPEN provided the first fuel element fabricated in Brazil to the IEA-R1 reactor. The fissile material used was the  $U_3O_8$  powder, available in Brazil, which allowed to produce 26 fuel element with 20% enriched  $U_3O_8$  powder. Once finished this stock of uranium oxide and accounting the increasing of radioisotope demand and its consequent upgraded of IEA-R1 reactor power, the fabrication of Brazilian own fuel elements became vital. So, in 1997, to avoid rapid consumption of  $U_3O_8$ -based elements, a new higher uranium density fuel should be produced. The reason to have a more compacted core was to get better irradiation flux and also to reject less spent fuel elements in order to rationalize their storage inside the reactor fuel pool. The solution for research reactors is based on the  $U_3Si_2$ -Al dispersion with uranium loading around  $3.0 \text{ gU/cm}^3$ . In 1998, the fuel plate fabrication technology of this silicide fuel was implanted, but the  $U_3Si_2$  powder was imported from France. Between 1999 and 2000, 16 silicide fuel elements were manufactured. From 1998, the efforts to develop the  $U_3Si_2$  powder began, aiming at full nationalization of the whole production process.

## Silicide Fuel Development

The IEA-R1 Reactor of IPEN/CNEN-SP is a pool type reactor in operation since 1957. The reactor uses MTR type dispersion fuel element in a 5 X 5 core arrangement. The Nuclear Fuel Center of IPEN provides with the necessary nuclear fuel for the continuous operation of the reactor. The production line of  $U_3Si_2$  based fuel plate, as broadly indicated in figure 1, follows this sequence:

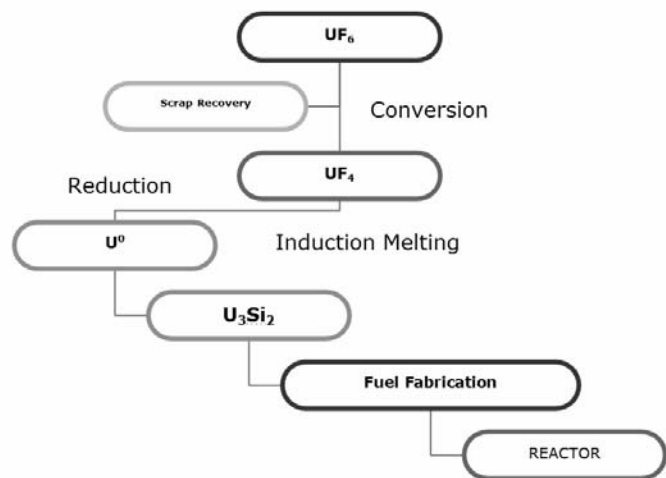


Figure 1 – Sequence of production for  $U_3Si_2$  fuel element

### *UF<sub>6</sub> conversion towards UF<sub>4</sub> [1,2]*

Essentially, the process to get UF<sub>4</sub> from UF<sub>6</sub> uses the reduction of U<sup>+6</sup> present in aqueous solution to the U<sup>+4</sup> state and its precipitation as UF<sub>4</sub> by means of the HF solution. The preparation of UF<sub>4</sub> using chemical reduction has been carried out starting from UO<sub>2</sub>F<sub>2</sub> solution resulting from the UF<sub>6</sub> hydrolysis. The solution is heated under continuous stirring to reach a temperature set, and the reducing agent is then added. Next, the hydrofluoric acid HF, as a precipitating agent solution, is slowly added. Tests have been tested using different reducing agents, such: SnCl<sub>2</sub>, CuCl, FeCl<sub>2</sub>, and Na<sub>2</sub>S<sub>2</sub>O<sub>4</sub>. The reducing agent SnCl<sub>2</sub> was the one that showed the best results and achieved an UF<sub>4</sub> precipitation in the range of 98%. However, during the UF<sub>4</sub> preparation, an amount of water is absorbed by the UF<sub>4</sub> crystal. This water could interfere in the other process steps, and to avoid problems, it must be removed.

The elimination of crystallization water from UF<sub>4</sub> is made at temperatures near 400 °C, under a constant flow of argon over the powder surface, which avoid the UF<sub>4</sub> oxidation to UO<sub>2</sub>F<sub>2</sub> or UO<sub>2</sub> and also drags out the released water vapor. The material as powder is placed in a small monel boat recipient and introduced inside of a heating chamber. After argon purging for 1 hour, the system is then heated to 400 °C under argon flow. The UF<sub>4</sub> powder remains at this temperature for 1 hour, which is a sufficient time to complete the water elimination. The subsequent cooling is carried out keeping the argon flow till the temperature reaches 100 °C. Details of the UF<sub>4</sub> precipitation procedures were presented in a previous work [6]. Figure 2 is a typical representation of the UF<sub>4</sub> morphology.

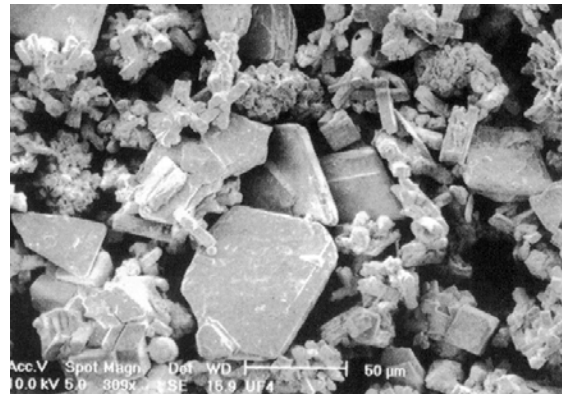


Figure 2 – UF<sub>4</sub> crystal (SEM Micrography)

### *UF<sub>4</sub> Reduction to Metallic Uranium [2,3]*

The intermetallic U<sub>3</sub>Si<sub>2</sub> 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 metallic uranium (1000 g) 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 reached amounts of 100 kg. The downscaling was difficult, since not only reduced scale for the crucible was conceived, but the reduction amount to be used was difficult to get higher 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 know the exact moment of the reaction.

We got it by use of accelerometer to get the sound waves perturbation. Optimization of this method was conquered only after almost 25 experiments with natural uranium simulations. Then we started producing 20% enriched uranium production.

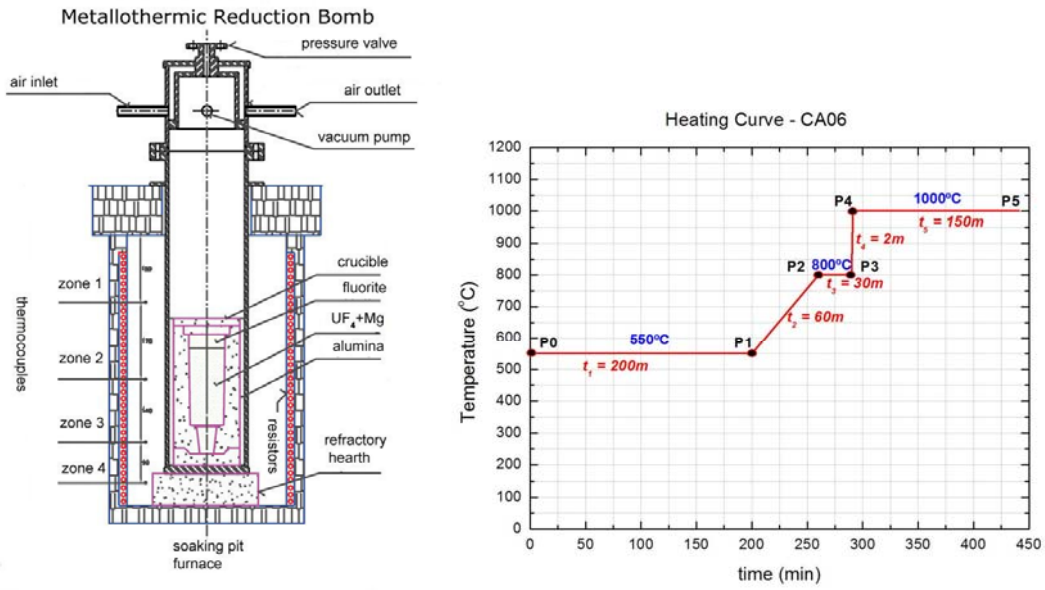


Figure 3 – Magnesiothermic reactor to reduce  $UF_4$  to metallic uranium and its thermal cycle.

The yield of this system is around 82-85% of metallic uranium. The thermal cycle used nowadays is indicated in the diagram in figure 4. The enriched material was already been produced with good operation and quality results. Figure 4 shows the enriched ingots produced by our IPEN methodology.

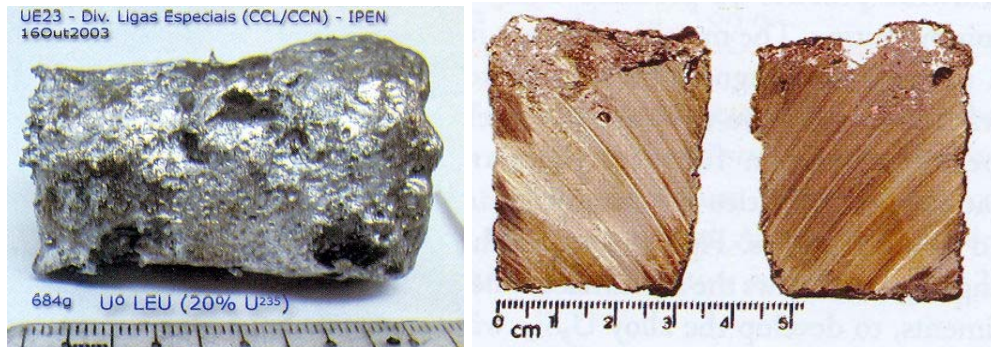


Figure 4 – IPEN enriched metallic uranium pieces.



### *U<sub>3</sub>Si<sub>2</sub> Fusion and Powder Fabrication [4]*

In 2004, after several experimental trials, IPEN arrived to the production route to fabricate the necessary alloy for nuclear fuel. From the produced uranium ingot, the uranium and hyperstoichiometric silicon addition to be remolten is placed inside an induction furnace. This furnace is then submitted to  $10^{-3}$ Torr vacuum and flushed with argon-atmosphere. Then the melting is carried out. The zirconia crucible was specially conceived so the temperature can reach more than  $1750^{\circ}\text{C}$ , at which this intermetallic could be produced. No other crucibles, than the zirconia one could cope with the aggressive environment created by uranium attack on linings. The load arrangement inside the crucible was planned to help the sequence of melting in the several stages that molten alloy passes before reaching the final intermetallic composition, as shows figure 5. More than 20 trials were carried out using natural uranium before the first LEU U<sub>3</sub>Si<sub>2</sub> were made successfully. In general, the quality of this intermetallic has fully accomplished to the needs for a nuclear material. The X-ray diffractogram (figure 7) attests the phases presents in the produced powder of this alloy.

### *Plate Fabrication*

Figure 8 show in general terms the plate fabrication and the fuel assembling and the fuel element as loaded in the reactor. The U<sub>3</sub>Si<sub>2</sub> cermet in the sandwich of the aluminum frames, is a mixture of U<sub>3</sub>Si<sub>2</sub> powder and Al powder. IPEN started manufacturing its own fuel element by using U<sub>3</sub>O<sub>8</sub>-Al dispersion fuel plates with  $1.9\text{ gU}/\text{cm}^3$ . This uranium loading represents a U<sub>3</sub>O<sub>8</sub> volume fraction of 27 %. If this same volume fraction is used by direct substitution of the U<sub>3</sub>O<sub>8</sub> by U<sub>3</sub>Si<sub>2</sub>, this results in increasing the uranium meat loading to  $3.0\text{ gU}/\text{cm}^3$ . Experiments to get higher loadings up to the technological dispersion limit of 45% in volume would reach  $4.8\text{ gU}/\text{cm}^3$ . Experiments to get this level of meat density was successfully carried out producing miniplate fabrication, but, as IPEN has no ability to fully test this miniplates after irradiation, the level of U<sub>3</sub>Si<sub>2</sub> was kept in the technological level of  $3.0\text{ gU}/\text{cm}^3$ .

Up to now, 14 silicide fuel elements, having meat density of  $3.0\text{ gU}/\text{cm}^3$ , have been irradiated at the IEA-R1m research reactor. The irradiation was closely followed by visual inspections and sipping tests and after 40% burn up (average), no problems related to fuel performance arose.

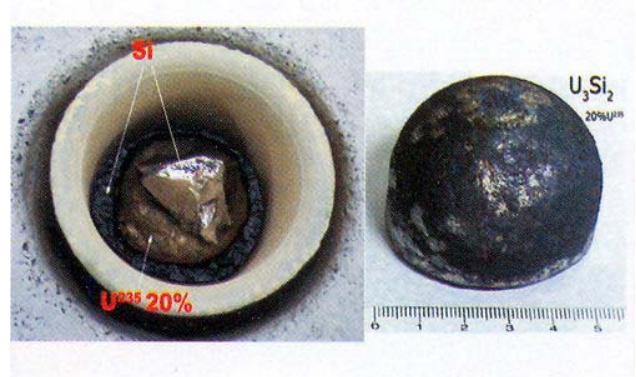


Figure 6 – Assembling of U<sub>3</sub>Si<sub>2</sub> crucible load

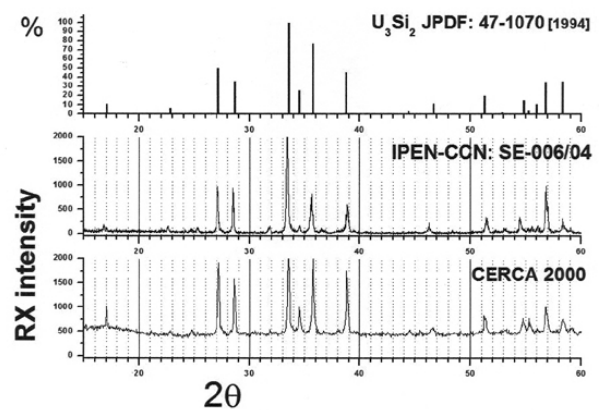


Figure 7 – X-ray diffractogram of U<sub>3</sub>Si<sub>2</sub> produced in Ipen compared with the pattern and commercial powder.

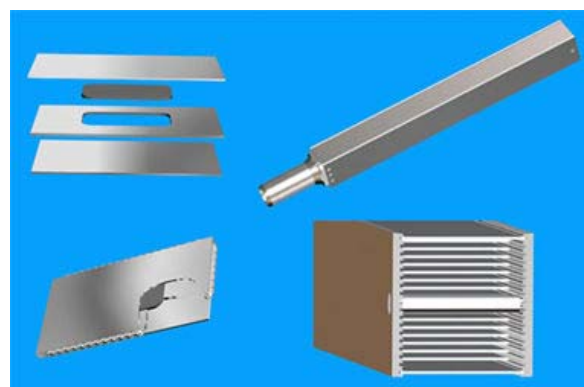


Figure 8 – Fuel Element Fabricated at IPEN.

## Conclusion

The project to achieve the technology to produce  $U_3Si_2$  as basic nuclear fuel for IEA-R1 was successfully concluded. IPEN nowadays can fabricate its own fuel elements on the basis of a wholly established sequence of production for this silicide intermetallic. Since the chemical reduction of  $UF_6$  to  $UF_4$ , followed by  $UF_4$  reduction to metallic uranium, using magnesiothermic route with good results, which is not often used abroad was also a considerable step in this technological achievement, since the amount to be produced is very small to have good metallic yields. The  $U_3Si_2$  production, by means of induction furnace, is also quite unusual among other producers of silicide around the world. Powdered product which is produced, then blended with Al as a matrix material, then compacted and frame-assembled has an optimized route. All these skills were also a successful adaptation of a previous oxide nuclear fuel material, which was used, but with its own peculiarities that were well solved by intense experimentation.

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