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History of nuclear fuel cycle development, facility decommissioning and site restoration at IPEN — CNEN/SP, Brazil

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INTRODUCTION

With the decommissioning of the pilot plants of the nuclear fuel cycle at the Nuclear and Energetic Research Institute - IPEN-CNEN / SP an important period of the Institution is being concluded. From its foundation until the 1990s, the technological domain of the various stages of the fuel cycle was perhaps the most important activity of IPEN, in terms of the number of researchers and technicians involved, as well as the financial resources used.

IPEN was founded in 1956 and its growth was centered in the IEA-R1 Reactor, a pool type reactor in operation since 1957. IPEN is located at the west of Sao Paulo city, inside the Campus of the University of Sao Paulo –USP. IPEN occupies an area of nearly 500.000 m2 (20 % buildings). It is associated to the University of Sao Paulo for teaching purposes. Through a partnership with USP, IPEN conducts a post-graduation program. The IPEN research centers have been engaged in multidisciplinary areas such as nuclear radiation applications, radioisotope production, nuclear reactors, nuclear fuel cycle, radiological safety, dosimetry, laser applications, biotechnology, materials science, chemical processes and environment. An example of a large national impact IPEN activity has been the production and supply of radiopharmaceuticals. About 2 million diagnostic and therapeutic nuclear medicine procedures per year have been performed in 2004 with products supplied by IPEN.

Nowadays, the main IPEN's facilities include: the nuclear research reactor IEA-R1m that reached criticality in 1957 (built with United States support under the Atoms for Peace Program) and has been upgraded recently to operate at 5 MWth; a Zero Power Reactor IPEN/MB-01 (critical assembly); two Cyclotrons (CV-28 and Cyclone 30 MeV –for radioisotope production); two electron beam accelerators of 1.5 MeV for irradiation applications in the industry and engineering; two Cobalt-60 Irradiators (11,000 and 5,000 Ci); dispersed fuel fabrication facilities (for research reactors); laboratories for chemical and isotope characterization, micro structural and mechanical tests [1].

IPEN had an important role in the development of uranium and thorium fuel cycles in Brazil. Since its foundation, IPEN has played a decisive role in the development of the nuclear science and technology in Brazil. It was created with the main purpose of performing research and development of nuclear energy peaceful applications. The Institute recent history has shown a major participation in the technological development of all steps of the nuclear fuel cycle. Nuclear fuel cycle R&D activities in the IPEN, from uranium purification to hexafluoride conversion and fuel fabrication for research reactors, besides thorium and zirconium purification, were accomplished in pilot plant scale and most facilities were built in the 70-80 years, destined to the technological domain of the several stages of the fuel cycle. The facilities were used to promote human resources, scientific research and better understanding of fuel cycle technologies.

The pursuit of autonomous technological development was emphasized in the Institution for several decades, having experienced a considerable impact when the Brazil-Germany agreement was signed in 1975. Initially, the IPEN's researchers felt unmotivated by the option of acquiring technology. However, the agreement was beneficial in terms of training and qualification opportunities for technical teams.

IPEN had a new stimulating period when cooperation with the Brazilian Navy began in the early 1980s. Autonomous technological development had once again become important and mobilized a large contingent of people at the Institute. One example of the important engagement of IPEN in the technological development in the nuclear fuel cycle area is the isotopic enrichment of uranium by ultra centrifugation, nowadays in process of industrial implantation. This significant achievement was performed in cooperation with the Brazilian Navy. Another important achievement was the IPEN-MB 01, a research reactor built in IPEN also in cooperation with Brazilian Navy, using UO2 pellets produced at the IPEN's pilot plant.

Nevertheless, in the nineties, radical changes in the Brazilian nuclear policy determined the interruption of the fuel cycle research activities and the plant-pilot's shut-down. Unfortunately, those changes interrupted decades of autonomous research and development efforts in the area of the nuclear fuel cycle at IPEN, with significant losses for the country. Despite the existence of a nuclear industry currently consolidated in Brazil, it is necessary to distinguish clearly between autonomous development and acquisition of technology.

Since then, IPEN has faced the problem of the dismantling and/or decommissioning its Nuclear Fuel Cycle old pilot plants. Most Nuclear Fuel Cycle Facilities had the activities interrupted until 1992-1993. Those facilities already played their roles of technological development and personnel's training, with transfer of the technology for institutions entrusted of the "scale up" of the units. Most of the pilot plants interrupted the activities more than twenty five years ago, due to the lack of resources for the continuity of the research [2].

HISTORY OF URANIUM DEVELOPMENTS AT IPEN

This section summarizes the progress of research concerning the uranium fuel cycle set up at the IPEN from the raw yellow-cake to the uranium hexafluoride, besides the reconversion of the hexafluoride to ammonium diuranate (ADU) and, at a later period, to ammonium uranyl carbonate (AUC). Also it is presented the progress in the obtainment of thorium compounds from concentrates produced by the monazite processing.

About sixty years ago, IPEN began a systematic R&D program on establishing of the uranium and thorium technology as part of the Brazilian program for developing nuclear energy for peaceful uses. The main activities were focused on the recovery of uranium from ores of domestic resources, purification of uranium and thorium raw concentrates and their transformation in compounds with purity and properties suitable for further fabrication of fuel elements for research reactors. The strategy adopted to dominate the uranium cycle was, like other countries, to develop processes on a laboratory scale, bench and pilot, to finally start industrial production.

The first studies for the installation and operation of a pilot unit for uranium purification at IPEN were initiated in the Division of Radiochemistry (DRQ) of the IEA (by that time IPEN was named IEA –Atomic Energy Institute), under the guidance of Prof. Dr. Fausto W. Lima, in 1959[3,4]. After preliminary laboratory-scale studies, a pilot plant was installed and operated, which went into operation in 1960 and continued until 1963, when its first stage was closed. The starting product was sodium diuranate (SDU), and then produced by the industrial processing of the monazite in São Paulo, by a private company (Orquima). The final product should be a high purity ammonium diuranate (ADU) with characteristics appropriate for conversion to uranium oxides and these in uranium tetrafluoride. It was also envisaged in a long-term program to obtain uranium hexafluoride. This first facility for uranium purification was based on an ion-exchange process. During its operation, the pilot plant produced approximately 4 tons of ammonium diuranate (ADU). This ammonium diuranate was converted into uranium dioxide (UO2) and used in the manufacture of fuel elements by the Division of Nuclear Metallurgy (DMN) of the IEA [5], founded in1962 under leading of Prof. Dr. Tharcísio D. Souza Santos. Those fuel elements were destined to the construction of a sub-critical unit, named RESUCO, to be installed in the Nuclear Energy Institute of the Federal University of Pernambuco, in Recife.

At that time a sodium diuranate (SDU) obtained by the industrial processing of the monazite and containing, among others, sodium, thorium, rare earths (RE), iron, phosphorus and silicon as main impurities was used as uranium concentrate. It should be noted that this uranium concentrate obtained from monazite is different from most concentrates obtained from other ores. The presence of thorium, RE and phosphate demands special considerations about the decontamination that depends on the chemistry process employed [4]. The sodium diuranate was dissolved with nitric acid for the preparation of a uranyl nitrate solution. The main concern was the removal of Th and RE. The loaded strong cationic resin was washed with water and then with diluted HNO3 and eluted with ammonium sulfate. ADU was precipitated by flowing the elution into ammonium hydroxide and keeping the reaction medium at a pH not less than 6.5 to maintain the co precipitated sulfate at a minimum level (< 0,5% U3O8) [6]. Following completion of the first stage of its existence, this pilot plant was dismantled [3].

With the completion of construction of the building of the Chemical Engineering Division - DEQ, in December 1966, under guidance of Prof. Dr. Alcídio Abrão, the installation of new laboratories was initiated as well as the construction of a new purification pilot plant by solvent extraction [7, 8]. This unit was based on the conventional liquid-liquid extraction technique. The facility comprised a section for yellow cake dissolution and three pulsed columns with perforated plates for the extraction of uranyl nitrate –UN - with TBP-varsol,

scrubbing and stripping of pure U. The development of U purification by pulsed columns was carried some years before the assembling of the pilot plant [9].

Besides the development of purification processes related to monazite concentrates, some alternative techniques were evaluated for the extraction of U, Mo and V from sulfuric acid liquors since, by that time, it was being developed an acid leaching process for the uranium ore from Poços de Caldas, State of Minas Gerais [10]. The dissolution of the impure yellow cake was accomplished with 2M HNO3 at ~100℃ and the concentration was adjusted to 300 gU/L. The extraction was done in countercurrent with org/aq ratio of 2.2. The loaded organic phase (TBP-varsol) with 135 gU/L was scrubbed with HNO3. The organic phase was stripped with water, resulting a purified uranyl nitrate solution.

A pilot plant was built to perform the precipitation of DUA by bubbling anhydrous NH3(g) into uranyl nitrate with 100 gU/L, at 60℃. The unit could be operated in a batch as well as a continuous way. The process pH was an important parameter to be controlled depending on the use of the ADU. For production of UF4, it was important to keep the pH between 4.0-4.5, to avoid oxide agglomeration. However, for obtaining of UO2 with better ceramic reactivity it was important to keep the pH at 7.0-7.5 [10]. The calcination of ADU to UO3 (for UF4 conversion) was accomplished in a belt furnace, with the ADU loaded in trays of stainless steel, at 500℃.

After some preliminary studies, a pilot plant facility for UF4 production was set up. The establishment of this unit had the collaboration and technical assistance from the International Atomic Energy Agency –IAEA. The UF4 pilot plant operated using the moving bed process. The starting material was UO3, reduced to UO2 by cracked NH3, and anhydrous hydrogen fluoride for the conversion to UF4 [11-13]. A laboratory scale development work was also carried out to obtain UF4 by alternative method via uranium dioxide reaction with hydrofluoric acid (wet method). A semi-pilot unit for this technology was also installed, with the construction of a reactor in polyethylene with mechanical agitation with a capacity of 100 kg UF4/batch.

An alternative method to ADU route for UO3 obtainment was reached with the construction of a denitration pilot plant. This facility used a fluidized bed process for conversion of uranyl nitrate solution to uranium trioxide and recovery of nitric acid. The bed of UO3 was fluidized by upward flowing of air in a 3"Φ tube (reaction chamber) heated by external electric heaters. Uranyl nitrate solution is injected at the bed level by a nozzle. UO3 seeds were previously placed to form the bed and the decomposition of uranyl nitrate occurred around the seeds. The spherical form of UO3 was preserved. Obtaining of UO3 directly by denitration of uranyl nitrate avoids precipitation, filtration and drying ADU which occurred in the conventional process.

The next step was the construction of an electrolytic fluorine generator and a pilot plant for UF6 production, having the UF4 produced in the moving bed reactors as starting material. During its operational life, the unit produced tens of metric t of UF6, used in developments of ammonium uranyl carbonate –AUC precipitation process. The development of the AUC process was of fundamental importance in the production of the UO2 fuel pellets for the IPEN-MB 01 research reactor that was totally design and constructed by Brazilians in a cooperation of IPEN and Brazilian Navy.

In parallel with the uranium related developments, a program for thorium purification and production was conducted at IPEN. The alkaline process for breaking up the monazite had been practiced since 1948, in São Paulo city by the private company Orquima. The processing of the monazite sands aiming the export of rare earths and other materials gave origin to thorium concentrates suitable for purification by solvent extraction. A pilot plant was built. The production and purification of thorium compounds was carried out at IPEN for about 18 years. During this period, the main product sold was the thorium nitrate with high purity (nuclear grade), having been produced over 170 metric tons of this material in the period, obtained through solvent extraction. The raw materials used were some thorium concentrates obtained from the industrialization of monazite sands. The thorium nitrate was supplied to the domestic industry and particularly used for gas portable lamps (Welsbach mantle). The thorium compounds produced permitted to accomplish several studies with a view to conversion of nitrate nuclear-grade thorium oxide suitable for the manufacture of fuel pellets, manufacture of mixed oxide pellets (U,Th)O2, obtaining of thorium tetrafluoride and its reduction to metallic thorium and studies of some properties of the UO2-ThO2 solid solutions [14, 15].

The process developments, besides construction and operation of pilot plants, gave rises to several additional related developments such as treatment of effluents, construction of a fuel reprocessing laboratory, production of Sol-Gel microspheres, analytical procedures for quality control, as well as, the development of further metallurgic processing of uranium and thorium compounds and studies for strategic materials like Zr and RE.

DISCUSSION AND CONCLUSION

Immediately after the nuclear R&D program interruption, in the nineties, the uncertainties related to an eventual retaking of the Program created some political hesitation about the facilities dismantling decision. As the retaking of the R&D Nuclear Program had been discarded, the decommissioning seemed to be the obvious choice. The appropriate facilities maintenance had been also harmed by the lack of resources, with evident signs of deterioration in structures and equipments. The existence of these facilities also implicates in the need of constant surveillance, representing additional obligations, costs and problems.

With the decommissioning of nuclear fuel cycle facilities at IPEN [2, 16], not yet concluded, an important cycle of the institution's life is being closed. The first years of a new research center like IPEN in the sixties were full of problems and challenges. This work is a modest record and a tribute to the achievements of those pioneers who began their studies on the nuclear fuel cycle and related materials in Brazil.

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