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QUANTIFICATION OF EFFLUENTS IN THE PRODUCTION OF NUCLEAR FUEL

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ABSTRACT

At the Instituto de Pesquisa Energéticas e Nucleares (IPEN), the Centro de Combustível Nuclear (CCN) is responsible for manufacturing fuels for the IEA-R1 reactor and, possibly, the multipurpose reactor fuels. In order to meet the demand for both reactors, the CCN developed a new plant. The production process of the fuel generates several types of effluents - containing uranium or not - being solid, liquid and gaseous with varied physical and chemical characteristics. The objective of this work is to follow the nuclear fuel production process and to identify, quantify and characterize the effluents, especially the liquid ones, to later elaborate a plan of management of these and eventually dispose in a responsible way in the environment.

1. INTRODUCTION

For many years, Instituto de Pesquisa Energéticas e Nucleares (IPEN-CNEN/SP) worked on the development of the manufacture of fuels used internationally in reactors, aiming at the nationalization of their production for use in its IEA-R1 reactor. Since its inauguration and during almost three decades, the IEA-R1 reactor totally depended on import of the fuel elements necessary from General American ATOMICS (93% in ^{235}U) and NUKEM (20% in ^{235}U).[1]

Considering the social and strategic importance of the production of radiopharmaceuticals in Brazil, and the continued growth of expenditures on imports of the necessary radioisotopes to attend this production, the Ministry of Science, Technology and Innovation established as a goal in its Plano de Ação de 2007 [7] the construction of a multipurpose reactor, to become self-sufficient in the production of radiopharmaceuticals.

Currently, there are over 1.5 million procedures involving radiopharmaceuticals in Brazil. Radiopharmaceuticals are substances that emit radiation and are mainly used in medicine for treatment and diagnosis. Mostly radioisotopes, which are the active elements of radiopharmaceuticals, are imported generating an expense of at least 30 million reais per year.

The new reactor, called Reator Multipropósito Brasileiro (RMB), will be large and will have 30 megawatts of power, and it will have three main functions: provision of radioisotopes for application in health, industry, environment and agriculture; creation of national capacity to test and qualify nuclear materials and fuels; facilitate the installation of a national research laboratory with neutron beams.

The Centro de Combustível Nuclear (CCN), located at the IPEN, will be responsible for the manufacture of nuclear fuel for this new reactor. The current capacity of the CCN is about ten fuels per year and, to meet higher demand, the plant used will be expanded and improved and will allow the maximum production of 60 fuels per year. The new plant will operate in an integrated manner, in which all manufacturing activities will be carried out under a more coherent plan, beginning in the conversion of UF6 to the final assembly of the fuel element.

The increase in fuel production will consequently result in an increase in the amount of effluents and pollutants generated. The production process of the fuel generates several types of effluents that may or may not contain uranium in the three physical states (solid, liquid and gaseous) and with varied physical and chemical characteristics.

Due to the concern for the environment, standards have been created with the objective of controlling companies and reducing the impact on the environment. However, over the years, the business concern with fines and assessments referring to the laws not followed came to stay in the background and has been replaced by greater care with the company's image.

It was in this context that environmental management was developed, a set of actions involving public policies, the productive sector and the society in order to encourage the rational and sustainable use of natural resources. A process that ties conservation and development issues together at all levels.

Considering the historical evolution of concern for the environment until the development of environmental management, the CCN of IPEN seeks to be able to manage and control their effluents. Therefore, the objective of the present work is to construct an effluent management plan so that everyone has access and knowledge of the actions will be possible to do in relation to each effluent.

2. MATERIALS AND METHOD

In order to reach the objectives proposed in this work, the following studies and methods were carried out:

a) Survey and analysis of the environmental legislation in force in the country; [2-4]

b) Process flow analysis; [5]

c) Collection of samples of the effluents generated;

d) Characterization of all radioactive and conventional effluents (liquid and solids) produced during the manufacturing process of fuel elements, taking into account the physical parameters and specific contaminants;

e) Qualitative and quantitative survey of the effluents generated;

f) Survey the impact of these residues and effluents in the environment; [5-6]

g) Evaluation and definition of the effluent segregation in order to group those that require the same treatment, separating them from those that require specific treatments.

3. RESULTS AND DISCUSSION

The production process of element was monitored at the CCN. The conversion processes of UF_6 currently adopted for the production of fuel elements consist of obtaining U_3Si_2 through a route of preparation of intermediate compounds, among them UO_2F_2, UF_4, and U^o. The result of this step is shown in Figure 1, indicating all the liquid effluent generation points during the production process.

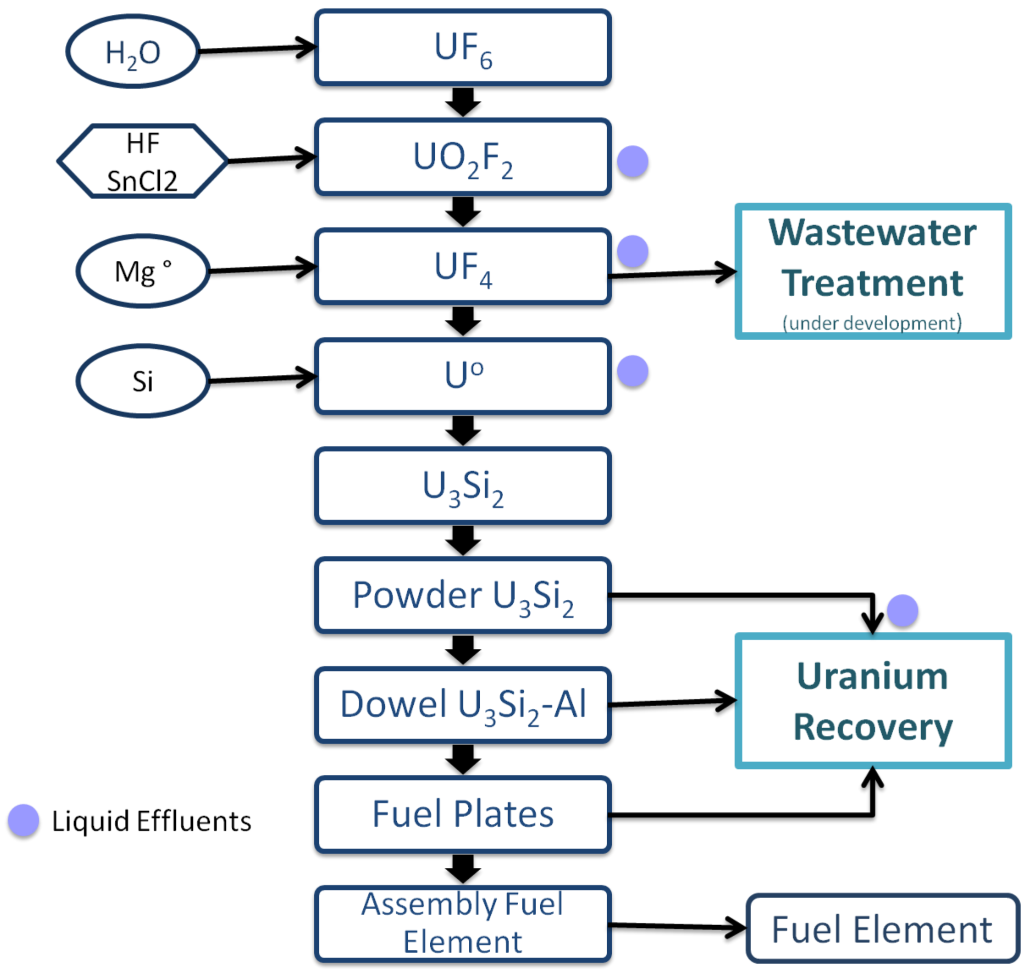

Figure 1: Stages that generate liquid effluents.

Table 1 shows the results of the liquid effluents generated in the nuclear fuel production process. In this table are provided their quantity, the process stage in which they were produced, their chemical characterization and the applied treatment.

Effluents and wastes from the productive areas of the CCN are collected at the place of origin already segregated, according to their physical, chemical and radiological composition.

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Table 1: Liquid effluents generated in the production of the fuel element.

Item	Effluent	Quantity	Step that is produced	Characterizartion	Treatment
A	Effluent UF_4	40L per boatload of Fuel Element	Production of UF_4	$[U^{+6}] = 120\ \mu g.mL^{-1}$ $[Sn^{+2}] = 20\ g.\ L^{-1}$ $[F^{-}] = 47\mu\ g.mL^{-1}$ $[Cl^{-}] = 12\ \mu\ g.mL^{-1}$ pH 2.43	In development.
B	Acid Solution (Etching U°)	300 ml for each etching. HNO_3 pure or 50%	U° production	$[U^{+6}] < 2.3\ \mu\ g.mL^{-1}$	Precipitate as ADU. Calcined to UO_3. Returns the production line as UF_4.
C	Aqueous Solution (Etching U°)	300 ml for each etching	U° production	$[U^{+6}] < 0.5\ \mu\ g.mL^{-1}$	Disposal with authorization of radiological protection after analysis of the uranium content and in accordance with the CONAMA N°20.
D	Alcohol from the superficial treatment of U_3Si_2	20 l/fuel Element	Production of plates U_3Si_2	-	Reused as technical alcohol in the production facilities of the CCN.
E	Aqueous solution from washing of granulometric sieves and equipment used in the production process	0.5 L every Fuel Element produced	Briquette production	pH 6.7	Decantation of the uranium powder. Separation of the supernatant. Control of the levels of uranium. Discard after radiological environmental protection authorized, in accordance with the CONAMA N° 20.
F	Nitric acid solution from the pickling process of fuel	HNO_3 30%	Production of fuel plate	pH 3	Solution is sent to the CCR to reuse in cleaning and decontamination of reactors, filters, dissolution of uranium (uranium recovery).
G	Sodium hydroxide solution from the pickling process of fuel plates	30 l of 2, 5 m NaOH/6 c	Production of fuel plate	pH 6	Solution is sent to the CCR to reuse in the process of dissolution of fuel cards (uranium recovery).
H	Effluent ADU	200L/10 plates	Uranium recovery	$[U^{+6}] = 8.5 \pm 1.5\ \mu g.mL^{-1}$ $[NH_4^{+}] = 8.8 \pm 0,\ 8\ g.\ L^{-1}$ $[F^{-}] = 69 \pm 1.8\ \mu g.mL^{-1}$ pH 9.2	Ion exchange and the uranium recovered with uranyl carbonate.
I	Aqueous solution from uranium recovery	15 L per batch/10plates	Uranium recovery	$[U^{+6}] = 1.96 \pm 0.12\ \mu g.mL^{-1}$	Analysis and disposal in accordance with the CONAMA N° 20.
J	Organic solution from uranium recovery. (isoparaffin triphosphate)	15 L per batch/10plates	Uranium recovery	$[U^{+6}] = 97.7 \pm 0.5\ \mu\ g.\ mL^{-1}$ $[U^{+6}] = 10.2 \pm 0.3\ \mu\ g.\ mL^{-1}$	Wash with sodium carbonate. Wash with HNO_3. Wash with water until pH7. Wash with HNO_3 3M. ~ 100 L: STOCKED.
K	Acid solution from chemical analysis performed on the fuel control CQMA and returned to CCN	Every 3 months of production produces 5 L of purified uranium solution	Fuel Element Process	$[U^{+6}] = 15\ \mu\ g.mL^{-1}$	Precipitate as ADU. Calcined to UO_3. Returns the production line as UF4.
L	Aqueous solution from the system of emergency showers and eye washes	-	Fuel Element Process	Does not apply, because so far there was no need to use the emergency system	Disposed according to the authorization of radiological protection after uranium content analysis as the CONAMA 20.
M	Aqueous solution generated in the metallographic analyses	Solution stored in a box of 2000 L	Qualification of the Fuel plate	$[NO_3^{-2}] = 0.24\ \mu g.mL^{-1}$ $[F^{-}] = 0.38\ \mu\ g.mL^{-1}$	Disposed of according to authorization of radiological protection after analysis of uranium content, activity and CONAMA 20.
N	Sodium aluminate from the dissolution of fuel cards	60-80 L batch	Uranium recovery	$[U^{+6}] = 18\ \mu g.mL^{-1}$ $[Na^{+}] = 24\ \mu g.mL^{-1}$ $[Al^{-}] = 38\ g.\ L^{-1}$	In development.

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Currently in the CCN the class of liquid effluents is considered a subdivision in radioactive and conventional effluents. Depending on the classification adopted, radioactive effluents are collected and sent for treatment and recovery of uranium and liquid effluents are provided for temporary storage in tanks prior to the disposal to the local storage area or treatment. Liquid effluent storage tanks are contained in polyethylene containment tanks.

Effluents called acid solution (B) and acid solution (K) can be segregated for treatment and recovery of uranium and fluoride by having similar chemical characteristics. We emphasize that the uranium mass enriched at 20% by weight of ^{235}U should be evaluated to avoid criticality problems.

The effluents generated in the pickling of the fuel plates such as nitric acid solution (F) and sodium hydroxide solution (G) are reused in the productive process, therefore, it is used in the dissolution of uranium slags and powders by only adjusting the Concentration of nitric acid, or simply making use of cleaning and decontamination of chemical reactors.

For the hydroxide solution it follows the same philosophy. It is used in the processing and recovery of fuel plates in the aluminum dissolution stage. Before its use, the analysis is performed to verify the NaOH concentration, and then the correction is made to the appropriate value in the process.

The alcohol (D) used in the drying process of the plates in the pickling process is also reused in the production unit as "technical alcohol". Other technologies may be applied but there are not yet under development in the CCN.

In addition, some liquid effluents in the CCN are discharged into the sewage network: aqueous solution (C), sieve-washing solution (E), shower water and eye wash (L), aqueous solution of metallographic analyzes (M).

The only organic effluent generated in the production process comes from the purification step of n-butyl uranium / isoparaffin triphosphate (J). This effluent is treated, and then reused in the purification process for another five batch of 10 fuel plates. Subsequently, it becomes a radioactive waste with uranium concentration of 10 $\mu g\ U.mL^{-1}$.

4. CONCLUSIONS

In the process of identification and evaluation of the operational processes of the CCN it was evidenced that the process of control of liquid effluents is quite significant. The current way to manage liquid effluent, while not presenting a history of contamination to the environment, needs to be improved.

Currently, part of the generated liquid effluents is released to the public sewage network after neutralization and analyzes to prove that it does not contain significant amounts of uranium compounds. Other part, it is not possible to release and is stored awaiting for final disposal, requiring adequate treatment for its final destination.

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