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IDENTIFICATION AND QUANTITATIVE ANALYSIS OF RADIOLOGICAL AND CHEMICAL RISKS IN A URANIUM ISOTOPIC ENRICHMENT PLANT

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Abstract: The risk associated with operating a uranium isotopic enrichment plant (UEP) is essentially related to uranium hexafluoride (UF_6), which is a toxic, radioactive, and highly reactive compound with various substances. The UF_6 accidental releases in UEP can occur from different ways, such as piping rupture, valve failure during transfer of UF_6 or from cylinders, cylinder rupture during heating or accidental fire. Thus, confinement of UF_6 is a very important safety requirement in a UEP. This article presents the results of a radiological and chemical quantitative risk assessment of a set of accidental scenarios of an isotopic recomposition system, which is part of the UEP. To achieve the goal, the first step was the identification of the most relevant events related to accident hazard scenarios in a UF_6 subsystem, then a probit function was applied to estimate the chemical risk and the death percentage caused by hydrofluoric acid, and finally a risk matrix was developed. The results of this study showed that the methodology proposed presents a promising way to solve this kind of problem and can also be used to help in emergency situations.

Keywords: UF_6, nuclear accident, isotopic enrichment, probit function.

INTRODUCTION

There is a clear need for new generating capacity around the world, both to replace old fossil fuel units, and to meet increased demand for electricity in many countries. World's electricity today is around 10% generated by about 440 nuclear power reactors. Besides, about 55 more reactors are under construction in 15 countries, equivalent to approximately 15% of the existing capacity [1].

There are two nuclear power plants in operation in Brazil, which are located at the Central Nuclear Almirante Álvaro Alberto on the Itaorna Beach in Angra dos Reis, Rio de Janeiro. They consist of two pressurized water reactors, Angra 1, with a net output of 609 MWe, and Angra 2, with a net output of 1.275 MWe. There is, moreover, in this site one nuclear power plant under construction, Angra 3, with a projected output of 1.245 MWe.

In this context, we should also point out: i) the Brazilian Nuclear Propulsion Submarine project, which started in 2012, which provides for the construction of a nuclear-powered submarine by the year 2029 [2]; ii) that Brazil has the world's sixth largest uranium reserves, and one of the few capable of mastering such advanced technology in the production of nuclear fuel; iii) that Brazil performs five out of the seven processes for the autonomy of nuclear energy production, that is, uranium mining and processing, uranium enrichment, uranium reconversion, uranium pellet and fuel assembly production and electricity generation by nuclear power plants [3].

Uranium Hexafluoride (UF_6) is generally used in many other processes, which comprise the front end of the nuclear fuel cycle (conversion, enrichment and fuel fabrication). The risk associated with the operation of a Uranium Isotopic Enrichment Plant (UEP) is directly related to the releases of UF_6, which is a toxic, radioactive and highly reactive compound with various substances. Breach of containment or equipment containing UF_6, either through equipment failure or operator error, can result in accidental releases of UF_6 [4].

The UF_6 can exist in a liquid, solid or gaseous state, depending on the temperature and pressure at which it is found. In the case of release of UF_6 in gas form during UPE operation, the reaction with water vapor present in the air (usually within one or two minutes), releases heat due to the exothermic reaction and forms small particles of uranyl fluoride (UO_2F_2) (diameter 1 to 10 μm) [5] and hydrogen fluoride gas (HF), according to equation 1. For each kg of UF_6 released there are 0.227 kg of HF and 0.875 kg of UO_2F_2 produced.

$$UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF + 218{,}55\ kJ/mol \qquad (1)$$

These chemicals are dangerous and when inhaled by humans may cause damage to lungs and kidneys. So, they may cause radiotoxicity and chemical toxicity on these organs and if a high amount is inhaled it might be lethal [6].

This article presents the results of a radiological and chemical quantitative risk assessment of accident hazard scenarios of release of UF_6 into a control room of an isotopic recomposition system of UF_6. The tool used in the identification of hazard scenarios in a UF_6 subsystem was based on Preliminary Hazard Analysis (PHA) technique [7]. Actual operational data of a typical UEP was used. After identifying the hazard scenarios, the risk was quantified based on the assessment of frequency and severity. On the other hand, to quantify the number of deaths of workers due to a given exposure to hydrofluoric acid (HF) a methodology based on probit function was used [8].

The remainder of the paper is organized as follows: Section 2 describe a brief summary of an identification of hazardous scenarios, radiological and toxicological risk calculation; Sections 3 presents the results and discussion. Finally, Section 4 presents the conclusions.

MATERIALS AND METHODS

IDENTIFICATION OF HAZARDOUS SCENARIOS

In order to develop the methodologies presented in this article, twelve (12) accident hazard scenarios of UF_6 release into a room of an isotopic recompositing system were considered, such as failures in pressure; temperature and mass measuring instruments; cylinder valve failures and pipe leaks. To identify the hazardous scenarios the PHA was used for hazardous assessment information such as: sketches of flowcharts; process data; piping instrumentation and control drawings; data sheets and procedures of typical UEP were used. Table 1 presents the hazardous scenarios identified where its causes and consequences were highlighted.

RADIOLOGICAL RISK CALCULATION

Failure rates were brought from [9] and were used to calculate failure probabilities. The estimated operating time of the equipment is considered in the concept of risk by calculating the probability of a failure event, according to Equation 2 [6].

$$P_f = 1 - e^{-\lambda t} \qquad (2)$$

A typical equipment failure rate was used for each scenario, as well as an annual estimate of the equipment operating time. The operating frequency is given by Equation. 3:

$$F_{op} = \frac{number\ of\ times\ the\ operations\ performed}{year} * \frac{duration\ of\ an\ operation}{operation} \qquad (3)$$

The frequency of radiological risk is given by Equation. 4:

$$F = F_{op} P_f \qquad (4)$$

The severity referring to the radiological risk is defined as the effective radiation dose, that is, the radiological impact is expressed in terms of radiation dose [10]. The effective dose was calculated by Equation 5 adapted from [11]:

$$D_{ef} = \frac{QAfRT}{V} \qquad (5)$$

Where:

Q - released radionuclide mass [kg];

A - radionuclide specific activity [Bq/kg];

f - compromised effective equivalent

Scenario	Accident scenario
1 – Valve failure of a 30 B cylinder (donor cylinder).	It is the most vulnerable point for UF6 release in this equipment. Structural damage to the valve can occur and there is the possibility that UF6 will solidify preventing full sealing.
2 – Heating failure of the pipe section responsible for the transfer of UF6 between cylinders.	In this part of the pipe a circuit of electrical tracers is placed whose objective is to heat the pipe, avoiding desublimation and consequently the pressurization in the line.
3 – Failure in one of the valves located in the hotbox.	Electrical heating system Failure. It is responsible for keeping a hotbox warm, so releases due to UF6 overpressure can occur.
4 – Valve failure of a 30 B cylinder (receiver cylinder).	UF6 release due to failure of the receiving cylinder valve connected to flexible tubing.
5 – Failure in the inner cylinder weighing system into the autoclave	Load cell instrument failure. Possibility of cylinder overfilling.
6 – Valve failure of a 30 B cylinder (receiver cylinder).	It is the most vulnerable point for UF6 release in this equipment. Structural damage to the valve can occur and there is the possibility that UF6 will solidify preventing full sealing.
7 - Failure in the instrument responsible for sealing the autoclave door.	Possibility of passing UF6 out of the autoclave. If there is, loss of control of the mass passage to the receiving cylinder with pressure increase. Possibility of cylinder rupture, valves rupture or piping rupture with UF6 release.
8 e 9 - Autoclave temperature controllers failure.	Pressure increases inside cylinder 30B. Possibility of cylinder rupture, valves rupture or piping rupture with UF6 release.
10 - Breakage in the liquid sampling device.	Wrong moves or bad connections. possibility of releasing UF6 into the autoclave.
11 e 12 - Pressure instruments failure	Pressure increases inside cylinder 30B. Possibility of cylinder rupture, valves rupture or piping rupture with UF6 release.

Table 1: Hazardous scenarios

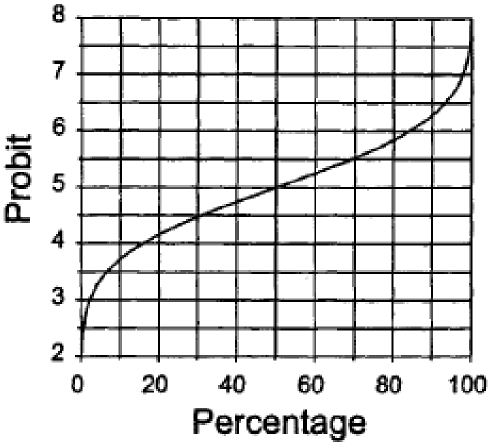

Figure 1: *Probit function*

Source: [14]

dose conversion factor due to radionuclide inhalation [Sv/Bq], [9];

R - respiration rate, [$m^{3/}s$];

T - operator contact time with the plume inside the room [s];

V - room volume [m^3].

The assessment of internal exposure in workers is based on prospective exposure parameters such as duration of exposure, concentration of radionuclides in the breathing zone, and type of radioactive aerosol materials. The specific activity A was calculated by Equation 6, as defined in [12].

$$A = 0.4 + 0.38E + 0.0034E^2 \tag{6}$$

Where:

A - specific activity [μCi/g]

E - percentage of uranium enriched in U^{235}

Equation 7 defines the radiological risk as defined [10].

$$R = C_R(FD_{ef}) \tag{7}$$

where,

C_R - radiological risk coefficient [Sv^{-1}], [13].

TOXICOLOGICAL RISK CALCULATION

In this study the probit [8] function was used in order to relate the dose of HF to its response for UEP workers. The probit function consists of representing a curve by an analytical equation. So, its application consists of calculating the value of the ordinate and estimating the percentage of fatalities, as shown in Figure 1. In the case of a single exposure the probit function is particularly suitable providing a straight-line equivalent to the dose-response curve.

The value of variable Y (ordinate) was calculated by equation 8. Table 2 presents the constants applied to the probit function for the HF.

$$Y = a + b\ln(C^n t_c) \tag{8}$$

where:

- Y – is the probit value.
- a,b e n – constants that depend on the analyzed toxic substance
- C – is the concentration, ppm/m^3
- t_c – is the exposure time, min.

Equation 9 [10] was used to determine the mean and steady-state concentration (ppm) of HF in a UPE room, given a source term Q_m and a ventilation rate (exhaustion) in the environment Q_v. The following assumptions were considered in equation 9 [8]:

- The concentration found is an average in the indoor environment of the installation. Localized conditions can result in significantly higher concentrations. Workers near an open container may be exposed to higher concentrations.
- A steady state condition is assumed.
- The non-ideal mixing factor ranges from 0.1 to 0.5 for most practical situations. For perfect mixing k = 1.

$$C_{ppm} = \frac{Q_m R_g T}{k Q_v P M} 10^6 \tag{9}$$

with:

- Q_m – material mass rate (kg/s.) resulting from material leakage
- R_g - universal gas constant.
- T – temperature (K).
- k – mixing factor for non-ideal gases
- Q_v – ventilation rate (m^3/s) in the room.
- P – internal pressure (Pa).
- M - molecular mass of the substance (kg/kmol).

RESULTS AND DISCUSSION

RADIOLOGICAL RISK RESULTS

The frequency assessment of each hazardous accidental scenario was performed by using Equation 4. Next each frequency was classified according to Table 3.

On the other hand, Equation 5 was used to assess the frequency of each hazardous accidental scenarios. Next each frequency was classified according to Table 4 [15].

Equation 7 was used to calculate the radiological risk. Then, the radiological risk was classified as low, moderate, or high. Following this, the radiological risk matrix was generated using the hazardous accidental scenarios, as shown in Figure 2. Each of the scenarios is numbered from 1 to 12 following the table 1 sequence order.

In figure 2 it is possible to observe that hazardous accidental scenarios 5, 6, 7, 8, 9, 11 and 12 were classified as moderate risk and the others were classified as low risk. In addition, in figure 3 it is possible to observe that the hazardous accidental scenarios 8, 9, 11 and 12 are responsible for approximately 98% of the total radiological risk.

TOXICOLOGICAL RISK RESULTS

As a basis for toxicological risk assessment after an accidental release of UF_6 only scenarios classified as moderate risk in figure 2 were used. Equation 9 was used to determine the mean steady-state concentration (ppm) of HF in a room where an isotopic recomposition system is located. In addition the probit function was applied to find the percentage of death. Table 5 presents the toxicological risk results.

Table 5 shows that scenarios 10, 11 and 12 have the highest HF concentration (664 ppm), consequently a higher dose (19927) and a higher death percentage (98%). Then came the scenario 8 with 627 ppm, dose rate of 18799 and 95% of deaths, scenarios 6 and 7 with 501 ppm, dose rate of 15033 and 78% of death. It is also worth mentioning that scenario 9 is the one with the lowest concentration of HF (327 ppm) and consequently the one that least contributes to death (25%).

CONCLUSION

This article presents a methodology for frequency quantification, risk and severity in case of accidental release of UF_6 in a UEP. A structured risk assessment focused on the plant's reality is very important for the plant and for the licensing bodies. The results show that the methodology is simple and easy to implement in a nuclear industrial facility.

With the radiological risk matrix generated in this study, it was possible to observe that the greatest risks are in the moderate category and that scenarios 8, 9, 11 and 12 are responsible for approximately 98% of the total radiological risk. Besides that, in relation to toxicological risk found in this study the scenarios 10, 11 and 12 have the highest HF concentration and consequently are responsible for 98% of deaths.

It can be concluded from this results that greater attention to risk management should be directed to these scenarios and the methodology proposed presents a promising way to solve this kind of problem.

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Constants	**a**	**b**	**c**
Values	-26,4	3,35	1

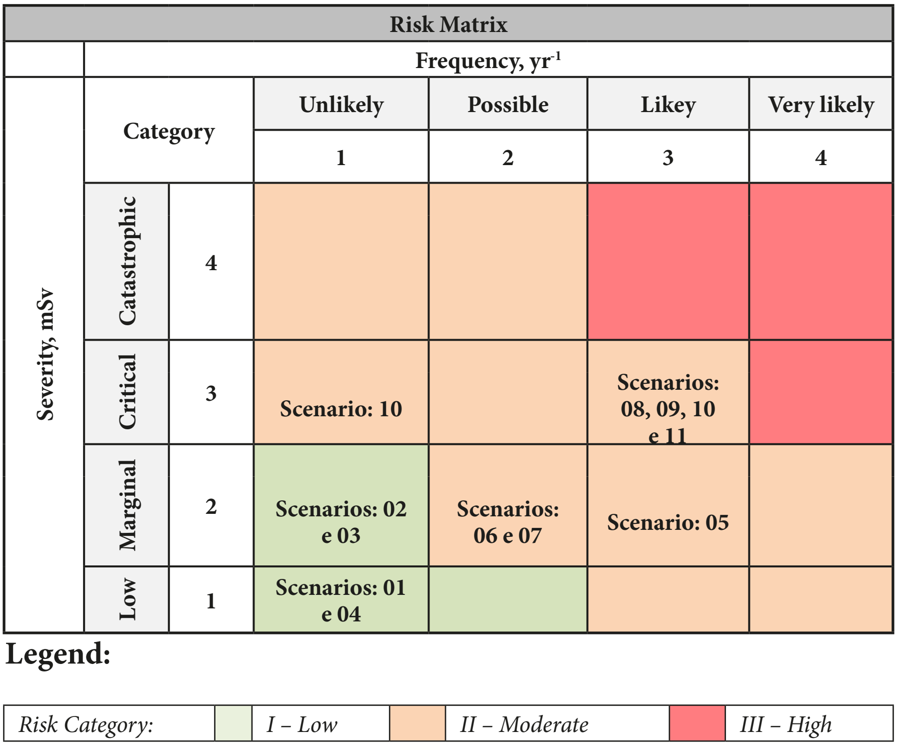
Table 2: Constants applied to the probit function [14].

Label	**Description**	**Frequency, yr^{-1}**
A – Unlikely	Chances of occurrence during plant useful life	$F < 10^{-5}$
B - Possible	Unlikely during plant useful life	$10^{-4} < F < 10^{-5}$
C - Likely	Expected to occur at most once during plant useful life	$10^{-2} < F < 10^{-4}$
D – Very Likely	Expected to occur at least once during plant useful life	$F > 10^{-2}$

Table 3: Radiological Risk frequency classification

Label	**Description**	**Frequency, yr^{-1}**
I	Catastrophic	Irreparable damage to equipment, property and/or the environment, leading to unit and/or system disorderly shutdown (slow or impossible repair); causes death or serious injury to several people (employees and/or member of the public).
II	Critical	Severe damage to equipment, property and/or the environment, leading to unit and/or system ordered shutdown; lesions of moderate severity in employees and/or members of the public (remote probability of death of employees and/or third parties); requires immediate corrective action to prevent becoming an accident.
III	Marginal	Light damage to equipment, property and/or environment (damages are controllable and/or with low repair costs); minor lesions to workers, outsourced personnel or people outside unit
IV	Low or Insignificant	No damage or minor damage to equipment, property and/or the environment; injuries and/or deaths of employees, third party and/or members of the public do not occur; cases of first aid or minor medical treatment

Table 4: Severity accidental scenarios classification



Risk Matrix						
	Frequency, yr^{-1}					
Severity, mSv	Category		Unlikely	Possible	Likey	Very likely
			1	2	3	4
	Catastrophic	4	🟧	🟧	🟥	🟥
	Critical	3	🟧 Scenario: 10	🟧	🟧 Scenarios: 08, 09, 10 e 11	🟥
	Marginal	2	🟩 Scenarios: 02 e 03	🟧 Scenarios: 06 e 07	🟧 Scenario: 05	🟧
	Low	1	🟩 Scenarios: 01 e 04	🟩	🟧	🟧

Legend:

Risk Category:	🟩	*I – Low*	🟧	*II – Moderate*	🟥	*III – High*

Figure 2: *Radiological Risk matrix*

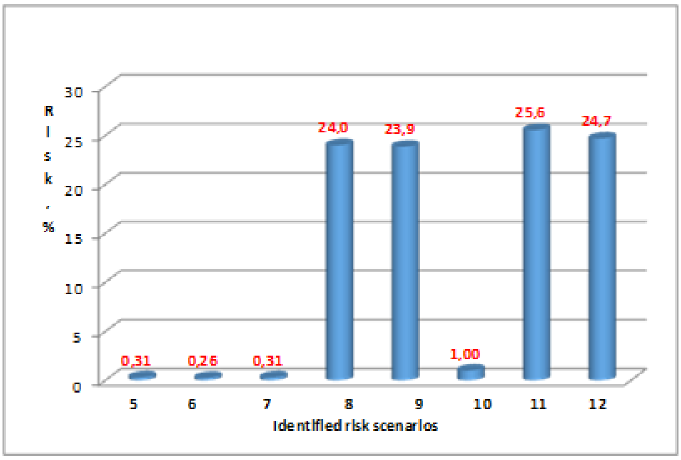


Figure 3: *Scenario contribution to individual risk*

Scenarios	Concentration	Y	Dose, C^nt	% death
5	392	5,00	11775	48
6	501	5,82	15033	78
7	501	5,82	15033	78
8	627	6,57	18799	95
9	327	4,39	9806	25
10	664	6,76	19927	98
11	664	6,76	19927	98
12	664	6,76	19927	98

Table 5: Toxicological Risk results

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