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ASSESSMENT OF POTENTIAL RISK AND RADIOLOGICAL IMPACT OF ACCIDENTAL RELEASE FROM THE ARGONAUT REACTOR

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ABSTRACT

In the early days of nuclear energy in Brazil, a reactor designed at the Argonne National Laboratory, originating the name ARGONAUT from the combination of the name of the Laboratory with the initials of Nuclear Assembly for University Training, reached criticality at the Institute of Nuclear Engineering. The Argonaut is a water moderated research reactor, which uses uranium enriched to 20% (235U) with prismatic graphite reflectors, designed to provide a thermal neutron flux up to 1010 $n.cm^{-2}.s^{-1}$ at an operating power of 5 kW. The presence of a nuclear research facility at the campus of Federal University of Rio de Janeiro (UFRJ) still cause concerns about radiological safety of the community around, even though this facility has been securely operating for more than fifty years. Besides, there were questioning about the potential risk of this facility to the IEN´s workforce by the Central of Harmonization Unit of Brazil (CGU). Thus, the present work aims to assess the potential risk of radiological accidents. Previously, the potential accidents evolving Argonaut reactor were considered to be the insertion of excess reactivity, catastrophic rearrangement of the core, graphite fire and fuel-handling accident. However, a recent accident scenario reassessment concluded that a severe physical damage of the core after reactor shutdown should be the emergency situation with the greater potential risk among the feasible postulated accidents.
According with the shutdown procedure, the water, used as moderator and coolant, drains out of the core and the concrete covers (each weighing 2.5 tons) are routinely removed from the top of reactor using a crane. The damage caused by the failure of the crane dropping the covers on the core would lead to breaking of the aluminum coating and the nuclear fuel plates with their release to the reactor room. This study assesses the radiological impact to workers and members of the public caused by partial inventory release to the atmosphere. Generic gaussian model was used to estimate the relative concentrations of air at ground level through the calculation of dispersion factors derived from wind data. For the dose calculation, the conversion coefficients by inhalation and plume immersion established by the ICRP were used. The results show that potential risk is above 1/10 of the limit of annual dose for workers, while they stay below the limit for members of the public, within a radius greater than 1 km.

1. INTRODUCTION

The Argonaut Reactor was designed at ARGONNE NATIONAL LABORATORY and began its first criticality in 1965, since then, it has been operating for training and research in Reactor Physics and Applied Nuclear Physics, aiming at specializing personnel in nuclear science and technology with an average of 115 operations per year.

The postulated accident occurs during the movement of the upper shields, when its fall would lead to the rupture of the coating of the combustible element that serves as a protection barrier against the gaseous / volatile and particulate fission products.

To estimate the source of this accident, the radioactive inventory in the core was calculated after 50 years of operation. The fuel element of the reactor consists of U3O8 cermet plates with uranium enriched to 19.99% of 235U.

The results of the calculation of the radioactive inventory indicated the presence of 21 actinides and 100 fission products and the amount of radioactive material that could be released in a potential accident scenario was determined [1]. These fission products are in gaseous or volatile form (halogens and noble gases) and in particulate form (transition metals), while the actinides are all in the solid form.

The objective of this report was to assess the radiological impact related to the potential risk of IEN workers in relation to the accidental release of ionizing radiation from the volatile fission products present in the eight fuel elements of the Argonaut reactor. These materials would be released into the reactor room and thence out of contention through the exhaust system.

2. METHODOLOGY

After release into the atmosphere, the radionuclides are carried in the wind (advection) and by mixing processes (turbulent diffusion). The radioactive material can be removed from the atmosphere by wet or dry deposition, as shown in figure 1. Models that take into account these processes are necessary to evaluate the concentrations of radionuclides in places in favor of the wind.

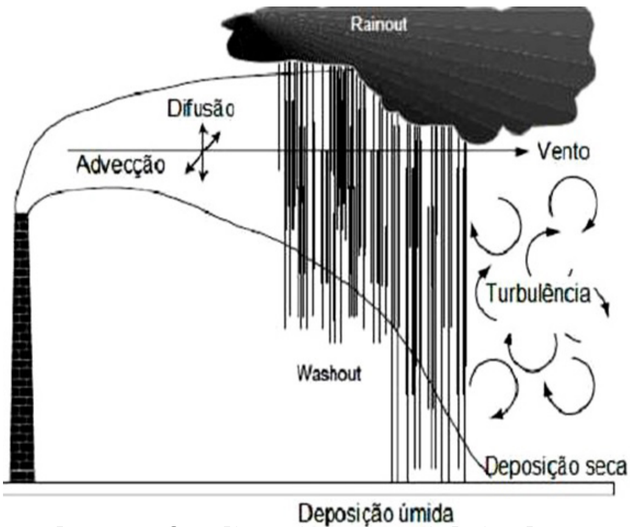

Figure 1 – Transport pathways of radioactive materials in the atmosphere. Modified from [11]

The Gaussian-based model was applied in this work to evaluate atmospheric dispersion, which is widely accepted for use in radiological assessment activities as advocated by the International Atomic Energy Agency [3].

The model is considered appropriate because it represents the dispersion of intermittent or continuous releases at a distance of up to a few kilometers from the emission source. Simplifications of the Gaussian formulation in terms of generic models were proposed to evaluate the impacts of releases of radioactive substances into the environment. These simplifications may be more or less conservative according to the scenario considered.
For a continuous release of a high point source under wind speed and constant atmospheric conditions, the Gaussian plume model can be represented by the following equation (Eq.1):

At where: $$C_A(x,y,z) = \frac{Q_i}{2\pi\sigma_y\sigma_z u}\exp\left(-\frac{y^2}{2\sigma_y^2}\right)\left\{\exp\left[-\frac{(z-H)^2}{2\sigma_z^2}\right]+\exp\left[-\frac{(z+H)^2}{2\sigma_z^2}\right]\right\}$$

CA = concentration in the air ($Bq.m^{-3}$) at the point (x, y, z) to the leeward of the source

x, y = longitudinal and transverse distance to the wind (m), respectively

z = height above ground (m)

Qi = release rate for radionuclide i (Bq.s-1)

σx, σy = diffusion coefficients (m)

μ = average wind speed (m.s-1)

H = clearance height (m)

The postulated accident considered the release of 100% of the gases / volatiles, while the emission of particulates was not considered in the impact assessment due to the high uncertainty in the calculation of the release of this source term. However, the gaseous form was released according to the 3000 m^3 h-1 flow rate of the exhaust system, which corresponds to the renewal of 1.5 times the volume of air from the Reactor Hall in one hour. This means that the entire atmosphere of the Reactor Hall is released in 40 minutes (2400 s), time taken into account in calculating the release rate.

The average speed of the wind was 2 m / s and the height of the release of 15 meters. Diffusion coefficients [4] were used for a distance of up to 150 meters from the source. In this report of accidental release, only radioactive iodine was considered in the calculation of the dose. The reasons that led to this choice are based on several factors: i) Calculation of the source term with high uncertainty for the particulates, since it involves the residence time in the atmosphere of the reactor hall and its deposition factor varies in orders of magnitude according to the meteorological parameters; (ii) The concentrations of the noble gases could not be calculated with the Gaussian model, since it is limited to materials that settle on surfaces; iii) Among the halogens, the only present in the inventory that are relevant to the individual dose, with a half-life ranging from about one hour to a few days, were the isotopes of iodine (130 to 135).

Once the source term was defined from the exhaust system information from the Reactor Hall, the exposure scenario was established for the calculation of the dose rate. Only the inhalation dose of iodine was considered, since it emits beta radiation, which is not relevant with respect to the plume immersion dose and soil deposition, thus these routes of external exposure were disregarded. Determination of the inhalation dose rate was performed according to the following equation (Eq. 2):

$$E_{\text{inh}} = C_A R_{\text{inh}} DF_{\text{inh}}$$

Einh = Inhalation Dose Rate in $mSv.h^{-1}$
CA = Concentration of radionuclide in air in $Bq.m^{-3}$
Rinh = Worker's respiration rate in $m^3.h^{-1}$
DFinh = Inhalation dose coefficient (Sv/Bq)

The concentrations in the air, for up to 150 m distance from the source and 1 m of soil height, were calculated by the Gaussian model [5], the respirator rate was established by [6] and has a value of 1.2 $m^3.h$ -1 and the inhalation dose coefficients by [7, 8].

This methodological approach aimed to establish a scenario that even conservative, adopts the precautionary principle in the evaluation of potential risk. For this reason, air concentrations and inhalation dose rates were additionally calculated considering the screening approach which is a simpler and pessimistic model by assuming that the concentration of radionuclides at the point of interest is equal concentration of atmospheric radionuclides at the point of release. This occurs in calm situations, where the winds are very weak and the plume travels by diffusion towards the receivers for a fraction of the time. Thus, we have the following equation (Eq.3):

$$C_A = \frac{P_p Q_i}{V}$$

At where:

CA = concentration in the air ($Bq.m^{-3}$) in the leeward distance of the source

Pp = fraction of the time that the wind blows towards the receiver (dimensionless)

Qi = release rate for radionuclide i ($Bq.s^{-1}$)

V = Air discharge at the release point ($m^3.s^{-1}$)

The value of 0.25 for Pp was adopted as suggested for screening models [9] NCRP 1996. The discharge was 0.83 $m^3.s^{-1}$ (3000 $m^3.h^{-1}$).

The exhaust system has a set of pre-filters and mechanical filters for particulates (3) and a gas activated carbon filter. An evaluation of the efficiency of the retention of these filters for the radioiodine is carried out, the release of which is studied in this report, emphasizing previously that in the case of noble gases these filters are not efficient.

In Thermal Reactors there are two most abundant chemical forms of Iodine: I2 (elemental iodine) and CH3I (methyl iodide). Activated carbon filters preceded by mechanical filters such as those in the Argonaut exhaust system have low yields for CH3I with moisture greater than 30%, which occurs virtually for 100% of the time. In addition, the efficiency of I2 removal by carbon filters in low concentration environments decreases sharply. The tests performed with carbon filters for Iodine also indicated that under conditions of high gas flow the retention of these radionuclides is compromised [10]. That is, the high flow in relation to the depth of the bed, greatly decreases the dwell time and, therefore, the efficiency of the filter element. Both conditions are in the case on the screen, since low iodine concentrations are released in a short time and with a high flow rate. In addition, the phenomenon of desorption and release of iodine forms in organic molecules produced in the presence of water, carbon, iodine and gamma radiation may further limit the ability of retention of activated carbon filters. Finally, we know that the Hall of the Reactor is not a watertight structure and that the clearance by cracks would also occur for other (external) areas of the IEN and at a lower height than that calculated with the Gaussian plume model.

Thus, it can be concluded that, in the present case, there is no way to estimate the efficiency of the activated carbon filter in case of an accidental release. It is known that it will be low, but it will depend on so many parameters (atmospheric, time of use, air flow, dwell time in the bed, concentration of Iodine, temperature, humidity, etc.) that does not allow us to even formulate a hypothesis for estimation of an approximate value.

Filters of activated carbon plus silver zeolites are the most efficient set for the adsorption of these forms. However, in order to ensure its efficiency, it is imperative to maintain a strict control of the relative humidity of the air, under negative pressure, with a dedicated exhaust system for accidental release. This is because the moisture present in the air would react strongly with silver thus decreasing its efficiency in the capture of iodine. Another reason is that the passage of other gases, particulates or chemical elements into continued and common use of the filters, would also deteriorate them.

The obtained results for the concentration of radioiodine in the air with the Gaussian plume model, as well as those obtained from the screening technique are presented in Table 1. Table 2 presents the iodine inhalation dose rate results based on in the results of table 1. It should be noted that the result is based on the dose value, where the dose is the product of the dose rate (mSv) for the exposure time (h), as equation below (Eq.4):

$$D = T_D * T_{Ex}$$

At where:

D = dose in mSv

TD = dose rate in mSv / h

TEx = worker exposure time in hours (h)

However, considering the principle of optimization of radioprotection with improvements in the exhaust system that could eventually be adopted to contain Iodine and assuming that it is possible to achieve efficiencies between 20 and 50% in Iodine retention, we present in Figure 2 the doses recalculated for distances of up to 150 meters between the source and the receiver.

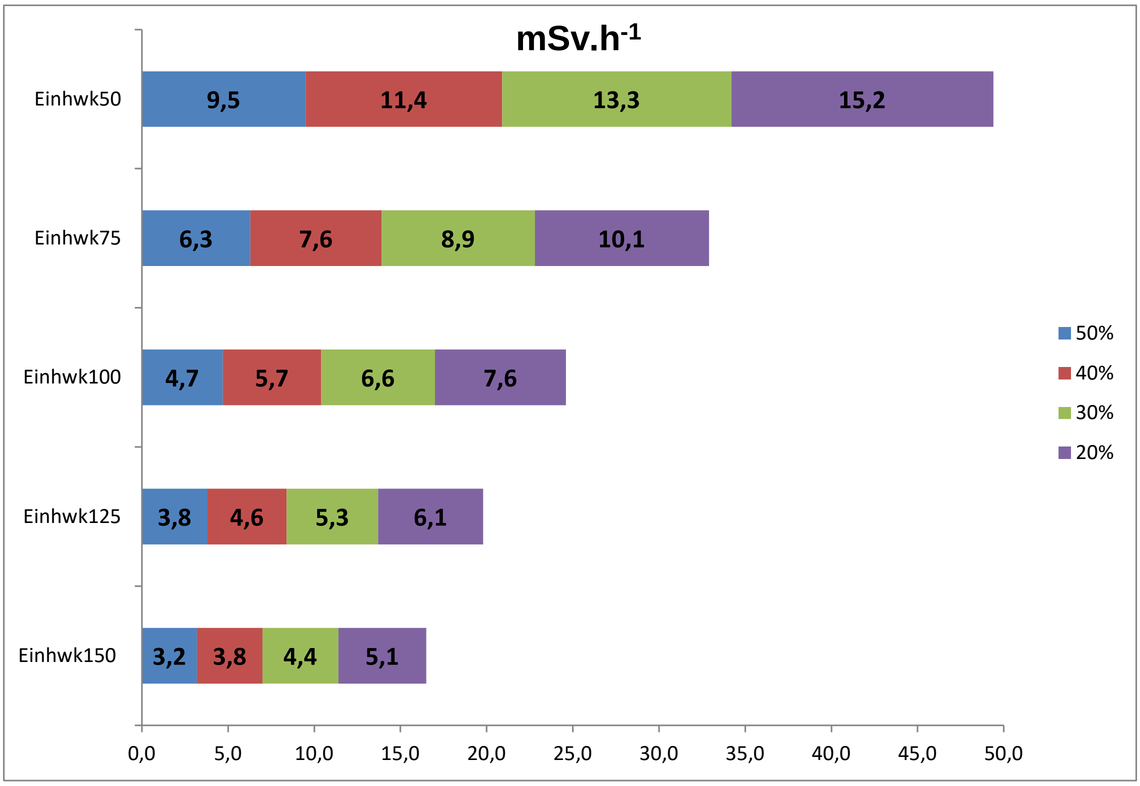

Figure 2 - Effective dose ($mSv.h^{-1}$) considering the scenarios with and without retention (20 to 50%) of Iodine in activated carbon filters for receptors located at, respectively, 150, 125, 100, 75 and 50 meters of the source.

For this purpose, only the Gaussian plume scenario was used for calculating the concentrations as previously described. On the other hand, it should be emphasized that the doses obtained in this way are based on a scenario that does not take into account the global radiological impact of the postulated accident. This is because, besides disregarding the other sources, it adopts much more modest iodine release rates, which could not be guaranteed by the current reactor control system in case of accidental release. In this way, all results should be analyzed with due caution.

Isotope	Release rate (Bq.s^{-1})	RR Bq.m^{-3}	50m Bq.m^{-3}	75m Bq.m^{-3}	100m Bq.m^{-3}	125m Bq.m^{-3}	150m Bq.m^{-3}
I-130	2,34E+07	7,05E+06	2,90E+05	1,93E+05	1,45E+05	1,16E+05	9,65E+04
I-131	2,33E+07	7,01E+06	2,88E+05	1,92E+05	1,44E+05	1,15E+05	9,60E+04
I-132	2,31E+07	6,96E+06	2,86E+05	1,91E+05	1,43E+05	1,14E+05	9,54E+04
I-133	2,32E+07	6,98E+06	2,87E+05	1,91E+05	1,43E+05	1,15E+05	9,56E+04
I-134	2,36E+07	7,12E+06	2,93E+05	1,95E+05	1,46E+05	1,17E+05	9,75E+04
I-135	2,78E+07	8,37E+06	3,44E+05	2,29E+05	1,72E+05	1,38E+05	1,15E+05

Table 1: Release rates (Bq.s^{-1}) and air concentrations (Bq.m^{-3}) of iodine isotopes in the Reactor Room (RR) and at 50, 75, 100, 125 and 150 meters from source, calculated by Gaussian (Eq1) and screening (Eq3) models.

Isótopo	TD RR mSv.h^{-1}	TD (50m) mSv.h^{-1}	TD (75m) mSv.h^{-1}	TD (100m) mSv.h^{-1}	TD (125m) mSv.h^{-1}	TD (150m) mSv.h^{-1}
I-130	5,67E+00	6,60E-01	4,40E-01	3,30E-01	2,64E-01	2,20E-01
I-131	6,23E+01	6,92E+00	4,61E+00	3,46E+00	2,77E+00	2,31E+00
I-132	9,19E-01	1,06E-01	7,10E-02	5,32E-02	4,26E-02	3,55E-02
I-133	1,26E+01	1,38E+00	9,18E-01	6,88E-01	5,51E-01	4,59E-01
I-134	3,85E-01	5,27E-02	3,51E-02	2,63E-02	2,11E-02	1,76E-02
I-135	3,21E+00	3,80E-01	2,53E-01	1,90E-01	1,52E-01	1,27E-01
Total	**8,50E+01**	**9,5**	**6,3**	**4,7**	**3,8**	**3,2**

Table 2: Total dose rates (Eq2) of the iodine isotopes, as a function of the distance from the source, in mSv.h^{-1}, according to the respective transport model.

3. CONCLUSIONS

This study evaluated the potential risk associated with the radiological impact for IEN workers, under the condition of an accident postulated in the Argonaut reactor, with release of the radioactive plume. Considering only the isotopes of iodine and the route of inhalation exposure, for receptors located up to 150 meters from the source for the period of 1 hour after the accident, doses between 5 and 250 mSv were obtained, considering the gaussian and screening models, respectively. When restricting the release in a scenario with partial retention (between 20 and 50%) of the Iodine by activated carbon filters, considering only the "Gaussian" (lower) concentrations, we obtained doses between 2.6 and 4.1 mSv.h^{-1}.

Thus, all the radiological impact estimates are above 1/10 of the annual dose limit for workers, provided for in CNEN Standard NN-3.01 [12] and Decree 877 of 20/07/1993 [2].

The approaches used produced a dose rate range corresponding to a first approximation of the problem within which the potential dose risk for the IEN workers is located. Thus, the lower doses obtained when considering the retention of iodine in the exhaust system correspond to the more optimistic scenario while those that use the value of the release point are the most pessimistic. In order to obtain more realistic dose rates, this report should also be complemented by modeling other radionuclides not currently included, especially noble gases, as well as simulations of different wind fields and atmospheric stability classes that are more representative of weather conditions.

REFERENCES

[1] Heimlich A., Lapa C.M.F., Guimarães A.C.F. & Moreira M.L. 2018. Cálculo do Inventário dos Produtos de Fissão no Reator Argonauta. Nota técnica. Rio de Janeiro. https://doi.org/10.13140/RG.2.2.10014.23366

[2] Brasil. Decreto No 877 de 20 de Julho de 1993. Regulamentação da concessão do adicional de irradiação ionizante. Brasília: Casa Civil. 1993.

[3] International Atomic Energy Agency. IAEA. 2001. Generic models for use in assessing the impact of discharges of radioactive substances to the environment. Vienna. Safety Report Series, SRS No. 19.

[4] Hosker, R.P. 1984. Flow and diffusion near obstacles, Atmospheric Sciences and Power Production (RANDERSON, D., Ed.), Rep. DOE/TIC-27601, Department of Energy, Washington, DC

.

[5] National Council on Radiation Protection and Measurements. NCRP. 1984. Radiological Assessment: Predicting the Transport, Bioaccumulation, and Uptake by Man of Radionuclides Released to the Environment, Rep. No. 76, NCRP, Bethesda, MD.

[6] Eckerman, K., Harrison, J., Menzel, H. G., & Clement, C. H. 2013. ICRP publication 119: compendium of dose coefficients based on ICRP publication 60. Annals of the ICRP, 42(4), e1-e130.

[7] CNEN. 2011. Posição Regulatória 3.01/003. Coeficientes de Dose para Indivíduos Ocupacionalmente Expostos. ResoluçãoN° 102, de 22.12.2010, publicada no D.O.U. em 10.05.2011.

[8]Food and Agriculture Organization of The United Nations, International Atomic Energy Agency, International Labour Organisation, OECD Nuclear Energy Agency, Pan American Health Organization, World Health Organization. 1996. International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, Safety Series No. 115, IAEA, Vienna.

[9] National Council on Radiation Protection and Measurements. NCRP. 1996. Screening Models for Releases of Radionuclides to Atmosphere, Surface Water, and Ground, Rep. 123 I, NCRP, Bethesda, MD.

[10] Reis, E. 1980. Testes de Filtros de Iodo. Dissertação de Mestrado. Universidade Federal de Minas Gerais. 239p.

[11] Torben Mikkelsen. Modelling of Pollutant Transport in the Atmosphere. Atmospheric Physics Division. Roskilde, Denmark, July 2003.

[12] CNEN. 2011. Norma NN-3.01. Diretrizes Básicas de Radioproteção. Resolução n. 27, publicada no D.O. de 06/01/2005.
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