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RADIOPROTECTION MANAGEMENT OF DISASSEMBLY AND DECONTAMINATION OF SURFACES OF A URANIUM AND THORIUM NUCLEAR PURIFICATION FACILITY AT IPEN/CNEN-SP

Almeida, C.C., Garcia, R.H.L., Cambises, P.B.S., Da Silva, T.M., Paiva, J.E., Rodrigues, D. L.

Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
calmeida@ipen.br

ABSTRACT

This work presents the adopted steps of the planned procedure of disassembly and decontamination of the systems of a natural uranium purification facility at IPEN. The unity was designed for chemical processing of natural uranium, aiming the obtention of nuclear-grade uranyl nitrate. Afterwards, the installation operated in treatment and washing of thorium sulfate and thorium oxycarbonate dissolution, thorium nitrate obtention, and commercial fabrication of loxon mantles. The decontamination procedure applied in this unit comprehended cleaning of equipment surfaces as chemical reactors, vacuum filters, extraction, washing and reversion columns, metering pumps, venting filters, pipes, connections and valves. Physical, chemical and ultrasonic methods were used simultaneously. For facility disassembly, the local radiological protection team planned the monitoring of workers, establishing procedures for individual internal and external monitoring. Concerning the internal operation procedures of IPEN, radioactive waste control, transport of radioactive materials and authorization of use of decontaminated equipment were also subject of study.

1. INTRODUCTION

This work presents the adopted steps of the planned procedure of disassembly and decontamination of the systems that constituted the natural uranium purification facility. This unit was designed for chemical processing of natural uranium, for obtaining nuclearly pure uranyl nitrate (Nuclear Fuel Cycle – Conversion Project, 80's decade). Afterwards, this facility was used for treatment and washing of thorium sulfate, and dissolution of thorium oxycarbonate, for obtaining thorium nitrate technically pure, used for commercial purposes on fabrication of lantern mantles. The decontamination procedures of surfaces included equipment as chemical reactors, vacuum filters, extraction columns, washing and reversion, metering pumps, ventilation pumps, tubing, valves and connections. The applied methods comprised chemical, physical and ultrasonic procedures, simultaneously. For the disassembly os this nuclear facility the local radiological protection team planned and established procedures for the internal and external individual monitoring of workers. Also, control and transportation of radioactive rejects and effluents, inside IPEN. Moreover, approval and clearance of decontaminated equipment and materials.

2. KNOWLEDGE OF EXISTING MATERIALS INSIDE FACILITY

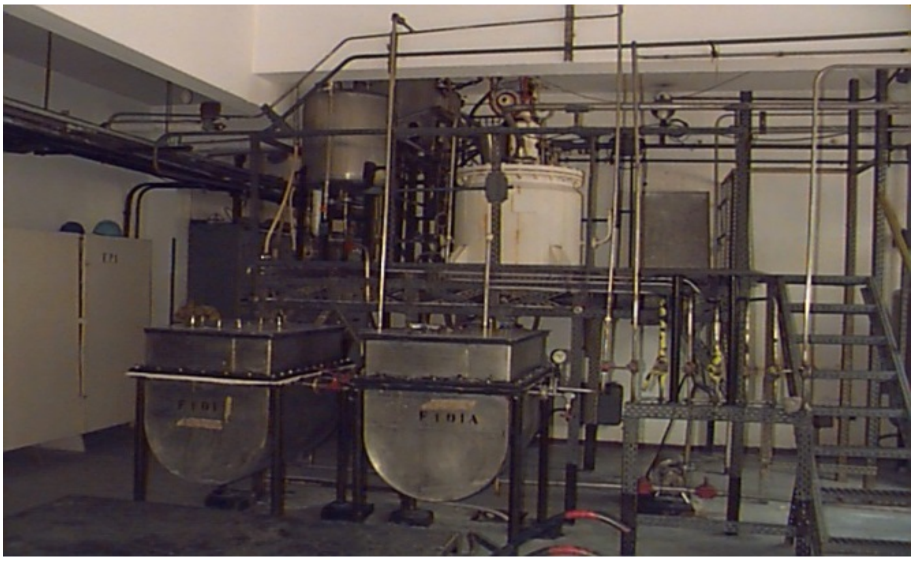

Figure 1: Materials inside facility

From june to august' 02, the radioprotection team inspected the area of pilot unity of uranium purification, and analyzed in detail the radiometric and operational records. The aim was to identify the structural needs, available resources, preliminary schedule and existing materials in the area. From this analysis, it was possible to identify radioactive sources of uranium and thorium and establish the following radioprotection rules for its handling:

- The equipment and parts considered as disposable were monitored, classified and segregated as: regular waste and radioactive waste and effluents, to receive posterior radioprotection treatment afterwards.
- The equipment and parts considered reusable in future radioactive processes were monitored, classified, segregated, packaged, labeled, transported and stored at Bloco 31. Depending on the needs of its future utilization, it could be submitted to surface decontamination processes. Its approval for uses in other restricted areas will be subjected to monitoring that consider the limits established at CNEN.NN-3.01 standard.
- The equipment and parts considered as reusable in future non-radioactive processes were submitted to the same procedures, however, the clearance to a non-restricted area will be subjected to the limits established at CNEN.NN-3.01 standard.

3. PREPARATION OF WORKPLACES

For the implementation of this planned operation, we aimed to follow the requirements of physical and radiological security, analyzing the existing structural conditions and availability of electrical power, water supply, drums and tanks for effluent storage, adequate venting systems, individual security equipment and security, changing rooms, definition of access control point, adequate place for temporary storage of contaminated material, waste and effluents. Finally, spots for the execution of procedures of surface decontamination. This initial analysis revealed the existence of shortcomings that had to be considered. For example, the lack of: operating venting systems, industrial security items and materials and equipment for decontamination processes. These items should be provided by the facility, according to orientation of the radioprotection team. To empower the course of work, workers from the operation team from CQMA and the radioprotection team of GPR gathered to enhance solutions to minimize the problems found. The main solutions found were:

- Reuse of drums and tanks that appear to be in good condition, in immersion methods of decontamination, into controlled chemical solution, aiming the removal of the untied contamination.
- Instalation of a mobile ultrasound equipment, offered by the decontamination laboratory. This equipment consist in two tanks of 100 liters, temperature control system, gases cooling, effluent gather, and mechanical stirring pump. It can be used for simultaneous processes of surface decontamination. The use of this equipment was suggested by the local radioprotection team. This suggestion was based in a study of combination and application of methodologies of decontamination in different kinds of contaminated surfaces, comprising various radionuclides used in IPEN for years. During the disassembly of the facility, processes of decontamination of parts and small and medium sized equipment were performed, and the results of the analysis of the behavior of the materials are published in a complimentary document called “Estudo e Controle de Descontaminação Radioativa em Materiais”.
- Rigid control of occupational radiation protection, considering the execution of routine tasks of monitoring, signaling, area isolation, instruction and correct use of personal protective equipment (PPE), control and elimination of radioactive reject and effluent.

4. OPERATION PLANNING

In july 2002, it was elaborated a document containing the occupational radiation protection entitled “Operação de Desmontagem, Monitoração e Triagem das Estruturas e Equipamentos da Planta Piloto de Produção de Nitrato de Tório – Procedimentos de Radioproteção, bem como seus riscos associados e procedimentos gerais de trabalho, visando principalmente a prevenção de acidentes, segurança física e radiológica dos trabalhadores e do meio ambiente”.

Figure 2: Signaling and isolation of areas

Figure 3: Signaling and isolation of areas

5. OPERATORS TRAINING

In september 2002, before the beginning of the work, a training was performed, including the operators form CQMA and the radioprotection team from GPR. The application of basic concepts of radioprotection were discussed. This training also worked as experience exchange between teams. Correct use of personal protective equipment were also discussed, and the elected PPE were:

- Apron; cotton overalls; plastic helmet; plastic cover (pants, apron and hood);
- Work boots; plastic shoe cover;
- Rubber gloves; leather gloves; surgical gloves; hoods;
- TLD dosimeters;
- Facial and semi-facil masks;
- Filters for masks (for acids and particulate).

6. RADIOLOGICAL SUVRVEY FOR THE CONTROL OF OCCUPATIONAL RADIATION DOSES

From September to December 2002, constant radiological survey was performed, before, during and after the disassembly operations. The principal objective was to establish a control of occupational radiation doses, as a function of the ambient, area classification, surface contamination, choice of spots of radiological control, definition of places for execution of segregation procedures, decontamination and temporary storage of equipment, radioactive effluents and waste and clearance of places. These procedures included:

- Direct monitoring of surface contamination, using portable monitors, aiming to identify the radiation and quantify the residual fixed contamination;
- Indirect monitoring of surface contamination, collecting smears for its evaluation in a proper count detector, aiming identify and quantify the untied and transferable particulate contamination;

- Monitoring of external radiation, using portable monitors, to identify kinds of radiation, for the control of occupational radiation doses;
- Air contamination monitoring. Specific filters were used and sent to the ambiental radioprotection sector, for analysis of radioactive particulate;
- From liquid samples collected, the radioactive effluent program monitoring were also performed by the ambiental radioprotection sector (LRA/ ipen), aiming the evaluation of concentration of radioactive particles in all the generated effluent.
- Individual monitoring of external body contamination of workers, using portable monitors, aiming to identify any deviation from normal conditions of work, to enable an immediate radioprotection action in face of an incident of superficial radioactive contamination.
- Program of internal individual monitoring of workers via “in-vitro” method. The samples were sent to the radiotoxicology lab.
- Formulation of control files and sketches of facility, for the register of monitoring data and identification of radiometric measurements spots.

Figure 4: Dose monitoring of contamination levels

7. DISASSEMBLY, MONITORING AND SEGREGATION OF EQUIPMENT AND MATERIALS

The disassembly operation, monitoring and segregation of equipment and materials located at *bloco 30* were performed from September to November 2002. Planned requirements of occupational radiation protection were followed. The works were performed daily from 8h30 to 11h30 and from 13h00 to 16h00 and the operators’ team was divided in two groups that worked in a rotating system. The adopted proceedings of radioprotection during this period were:

1) Signaling and delimitation of operational areas, to avoid access of unauthorized people;

Figure 5, 6 and 7: Disassembly, monitoring and segregation of equipment and materials

2) Isolating of floor at predetermined places, for the segregation and decontamination of equipment and parts from *bloco 30*, using impermeable plastic tarp over the floor, in order to avoid possible contamination of adjacent areas of isolated areas;
3) Monitoring, segregation and moving of equipment, parts, drums and metallic boxes containing radioactive rejects, following the daily operational plan of disassembly of Uranium and Thorium purification plant (*Bloco 30*);
4) The monitored materials considered non-contaminated were separated and stored for posterior reutilization or disposal;
5) The nuclear materials were appropriately monitored, packed, classified, signaled and transported to the safeguard sector;
6) The contaminated materials that would be submitted to surface decontamination process were classified and separated according to its size, needs, surface characterization, contamination levels and kind of impregnated contamination;
7) The radioactive rejects were properly stored in drums, containers and bags. Afterwards, monitored, classified, signaled, moved and temporarily stored at the former warehouse of *PROCON*.

8. DECONTAMINATION OF SURFACES

8.1 Parts and equipment surfaces

Decontamination methods used were specific for each kind of material. Generally, physical, chemical and combined methods were applied, starting with mild and following to harsh methods. As an example, it can be cited the use of ultrasound equipment, that speeded up the decontamination processes, as this methods evolves simultaneously the immersion of materials in chemical solutions, heating, agitation, cooling and condensing of generated gases, easing the removal of fixed contaminated particles.

Figure 8: Surface decontamination of surfaces of parts via ultrasound

Figure 9: Surface decontamination of surfaces of parts via immersing

8.1 People's skin surface

People decontamination is a subject that evolves medical and radiological specialties. The Occupational Radioprotection Service (*GPR*) owns a Laboratory of Radioactive Decontamination (*LDR*) that is adequate for the execution of these procedures. Besides that, the medical staff of *IPEN* was also supervised. As the work was operational and not an emergency, the radioprotection procedures were strictly followed, and by the end of procedures of disassembly of this nuclear facility, human contamination could not be detected.

9. RADIOPROTECTION AVALIATION

For the solution of problems and execution of operational work with proper radiological and physical security, the professional experience of *CQMA* and *GPR* teams was essential. The experience was acquired during years of operational handling of radioactive materials, as well in operational attendance of activity and radiological emergencies at places with structural deficiency. Next, we present some final comments:

1) From the reports of radiometric data produced from September to November'2002, the dose measurements, that were performed close to the parts, equipment and other specific spots of facility, varied from 1.2μSv/h (background value) to 80.0μSv/h. Considering that the operators worked 4 hours per day in a rotating system, the dose at these places would be around 6.66mSv/year. This value stands below of 3/10 of the annual limit for a worker considering the national standards.

2) The evaluation of methods employed for surface decontamination of parts and small and medium sized equipment was specific for the kind of material. At the end of the operations, it was verified that the use of conventional processes of decontamination

as aspiration, friction, abrasion, mechanical removal with humid fabric, washing of parts with running water or immersing parts into defined chemical solutions obtained results of 30 to 70% of efficiency of removal of the initial contamination, due to the structural difficulties presented at the area. Methods that combined humid, mechanic and ultrasound processes allowed efficiencies from 40 to 95% of removal of the initial contamination. From the data of *CQMA* presented about the radioactive waste generated, it was verified that, from 6 containers (total volume=9.6m^3) of solid metallic waste generated at the disassembly process of the facility, 1.5m^3 (15% of total volume) were decontaminated. From 43 drums (total volume=8.6m^3) of metallic solid waste generated, 0.86m^3 (10% of total volume) were decontaminated. Figures 10 and 11 show the results of decontamination of a lot of tubes of stainless steel that were submitted to these methods of decontamination. However, due to limiting size of the ultrasound equipment, it was not possible to apply decontamination methods in big-sized parts and equipment. These were temporarily stored at *Bloco 31*, under the standards stated previously at heading 1 and 6.

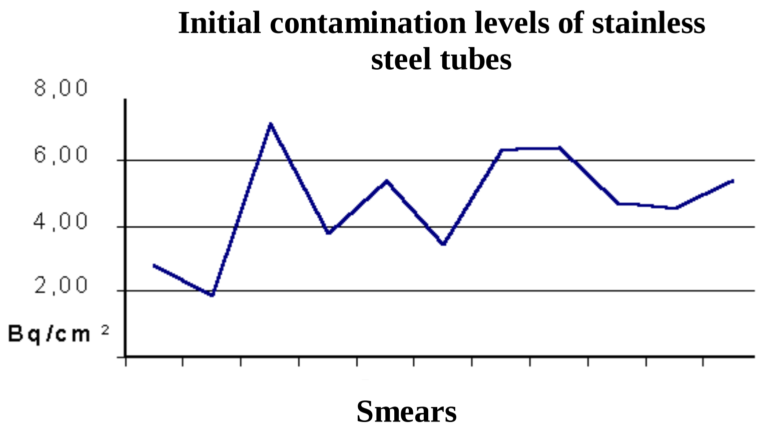

Figure 10: Initial contamination results of stainless steel tubes

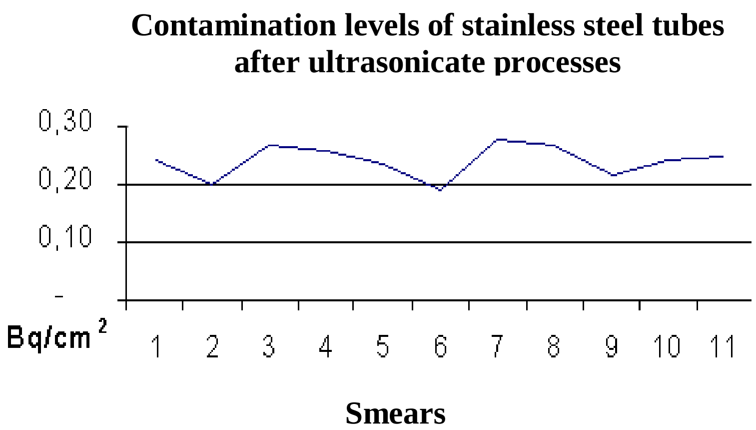

Figure 11: Contamination results of stainless steel tubes after simultaneous humid and ultrasound decontamination procedure

3) For the evaluation of surface contamination, the analysis of monitoring reports written from September to November '2002 was considered. It was found that the radioactive contamination levels of surfaces of parts, floor, equipment and various materials varied from 1.0Bq.cm^{-2} (background value) to 72.0Bq·cm^{-2}. These measurements were performed using direct monitoring methods, using a detector probe at the radiation camp, and indirect methods, using 5cm diameter smear discs. These paper discs were smeared at surfaces up to 100cm^2, and taken to measure. The count was done in one minute in scintillators. At the end of the operation of disassembly of the facility, even though various methods of decontamination were performed, spots of fixed contamination were found on surfaces of floor, wall and manholes, identified as ^{232}Th and natural U. The contamination values of alpha emitters detected were above the national standards (*CNEN-NN 3.01,Tabelas:V;VI*). Therefore, the internal areas of the facility (*Unidade Piloto de Purificação - Bloco 30 do Centro de Química e Meio Ambiente – CQMA*) are still considered, from a radioprotection view, as a **contaminated** and **restricted area**. Consequently, these areas must be controlled by radioprotection, till operational actions are taken to reduce the levels of contamination to environmental values. At table 1 and 2 the detected values of floor and walls are shown, before and after the operation.

4) For the radio toxicology analysis of the workers, the Radioprotection Supervision of the department of metallurgy of IPEN requested to the department of Radiation Metrology, *NM*, analysis of urine samples of evolved workers of the disassembly process. At December 24th of 2002, *CQMA* issued a report "*Relatório de Monitoração Interna*", #LRT – UU15/2002, that contained the results of the special monitoring. The quantified radionuclide was the natural uranium ($_{nat}$U). The results showed that the concentration was less than the method detection threshold, or 1.0 µg·L^{-1}.

5) For the analysis of present radioactive contaminants present in liquid effluents, the Radioprotection Supervision of the department of metallurgy of IPEN requested to the department of Ambiental Radiometry, *NA*, radiometric analysis, via gamma spectrometry, in liquid effluent samples. These were generated at the decontamination processes of surfaces, and the probable radionuclides were ^{232}Th and $_{Nat}$U. At Octorber 31st of 2002 the *NA* reported the following results:
   a. *Sample 1, from the decontamination tank used with immersed parts in nitric acid, that contained 2200 liters: $_{nat}$U: 51.3±3.1mg*.L^{-1}; ^{228}Ra:11.7±0.9Bq·L^{-1}. These levels satisfy the national standard (*Norma Experimental CNEN – NE – 6.05*) and could be discarded in regular wastepipe.
   b. *Sample 2, from the decontamination tank used with immersed parts in citric acid solution and ultrasound, that contained 110 liters:* no gamma radionucles emitters above the threshold, therefore, adequate to discard in regular wastepipe (*Norma Experimental CNEN – NE – 6.05*).
   c. *Sample 3, from the decontamination tank used with immersed parts in sodium carbonate solution and ultrasound, that contained 110 liters:*^{228}Ra: 41.9±2.4Bq·L^{-1}. To be discarded in a regular wastepipe, should be divided in two and discarded in different days (*Norma Experimental CNEN – NE – 6.05*).

6) To evaluate and control the concentration of radionuclides in air, the radiological protection service collected air samples during the steps of disassembly of the facility and wrote a report named "*Relatório de Análise de Urânio em Filtro de Monitoração de Ar*", emmite by the Ambiental Radioprotection Deparment – *NA*. The spot with higher radionuclide concentration denotes the air monitoring performed during the disassembly of the fume hood used during weighting processes. The value

corresponds to a mass of 25±2µg of uranium 238 for each filter. The air concentration derived from this data, for natural uranium, in the most restrictive form, is $7 \cdot 10^{-1}$Bq·m^{-3} for 2000h of work. The evaluation based on Bq·m^{-3} was calculated considering the worked hours into a dispersion of radioactive dust, and corresponds 20h. Therefore, the derived concentration of air is multiplied by a 100 factor. Then, the derived concentration for the result of this monitoring if 5.08Bq·m^{-3}, detected at the air filter by neutron analysis method, using a derived concentration of air of 70.0 Bq·m^{-3}, at an investigation level of 21.0Bq·m^{-3} and a register level of 7.0Bq·m^{-3}. The result of the performed monitoring is below the register level. Consequently, the result is satisfactory, emphasizing that the worker have worn personal protection equipment as dust mask during the operation process.

Table 1: External radiation values of *Bloco 31* before the surface decontamination processes

Spots	Description	Dose rate (µSv / h)
A	Entrance access point	1,2
B	Facility center	2,4
C	Facility center	2,8
D	Sink	1,6
E	Manhole 1	12,0
F	Corridor	2,2
G	Corridor	2,0
H	Facility center	3,6
I	Manhole 2	10,0
J	Fume hood 1	11,0
K	Fume hood 2	8,0
l	Exit access point	0,8

Table 2: Surface contamination radiation values of *Bloco 31* before the decontamination processes

Spot	Description	Activity (Bq / cm^2)
1	Entrance access point	2,65
2	Facility center	15,63
3	Facility center	18,48
4	Sink	5,45
5	Manhole 1	16,25
6	Corridor	6,08
7	Corridor	4,85
8	Facility center	15,28
9	Manhole 2	12,55
10	Fume hood 1	5,90
11	Fume hood 2	23,73
12	Exit access point	1,65

Table 3: External radiation values of *Bloco 31* after the surface decontamination processes

Spots	Description	Dose rate (μSv / h)
A	Entrance access point	0,7
B	Facility center	0,5
C	Facility center	0,6
D	Sink	0,25
E	Manhole 1	0,8
F	Corridor	0,5
G	Corridor	0,4
H	Facility center	0,5
I	Manhole 2	0,6
J	Fume hood 1	0,3
K	Fume hood 2	0,4
l	Exit access point	0,4

Table 4: Surface contamination radiation values of *Bloco 31* after the decontamination processes

Spot	Description	Activity (Bq / cm^2)
1	Entrance access point	0,30
2	Facility center	0,42
3	Facility center	0,30
4	Sink	0.23
5	Manhole 1	0.32
6	Corridor	0,24
7	Corridor	0,22
8	Facility center	0,35
9	Manhole 2	0,44
10	Fume hood 1	0,36
11	Fume hood 2	0,42
12	Exit access point	0,20

10. RESULTS EVALUATION

1) For the evaluation of the equivalent dose rate, Geiger-Muller detectors, model IPEN PI-760 were used. The background radiation was removed and the arithmetic mean method was applied. From the evaluation of the obtained results and interpretation of the standards, it was concluded that the doses, at the measured places would be (extrapolating to a year), below 3/10 of the annual limit of a worker, or 15mSv/year.
2) The surface contamination evaluation was done by indirect analysis using smear tests. 5 cm diameter paper discs were smeared on surfaces of up to $100cm^2$ and taken to

count for one minute at Eberline SAC-4 and BC-4 scintillators counters. Small contamination from ^{234Th}, ^{235}U and ^{238}U was detected [1];

3) The evaluation of air contamination was performed using an indirect method, using specific filters coupled at a Eberline RAS-1 vacuum pump. The air flow rate was $12.5 L \cdot min^{-1}$, and minimal acquisition time of 3 hours. These samples were analysed in a Camberra-type counter, model GX-20202, and activities around $8 \cdot 10^{-4} Bq \cdot m^{-3}$ of ^{238}U were detected. As the limit of annual incorporation for the most restrictive forms of ^{238}U is $7 \cdot 10^{-1} Bq \cdot m^{-3}$, the observed results showed approximately a thousand times smaller than the most restrictive limits. Therefore, it was not necessary the use of special systems for breathing , although special care for prevention of free contaminated particles spread was taken during the decontamination of surfaces [1].

11. ACQUIRED EXPERIENCE

From the acquired experience it was possible to adopt the surface decontamination operation for the decontamination that is being carried on at the M-8 building, namely, *Usina de Urânio Metálico* of the metallurgic division of IPEN. The following steps are the disassembly of the operational system and adoption of new research lines considering nuclear materials. From published works [1], the initial adopted plan made possible the elaboration of a basic procedure of decontamination, applicable at facilities were ionizing sources of radiation are handled, following the national standards and international recommendations [2,3,4]. From the authors' point of view, this experience emphasized the concept that new philosophies of radioprotection enhances of quality of life of people that work with nuclear subjects.

12. CONCLUSION

Considering the cited results, the area was considered banned, considering the measured levels of surface contamination, above the national standard CNEN-NN 3.01. Therefore, it was necessary to adopt operational procedures of decontamination, bearing in mind radioprotection criteria.

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