Evaluation and Reduction of the Internal Contamination for Workers due to Tritium Targets Used in a Neutron Generator Plant

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Summary

The Frascati Neutron Generator (FNG) of the ENEA Centre of Frascati (Rome) has been working since the end of 1992. For its operation several quantities of tritium have been used, up to 1850 GBq (50 Ci) per year. Measurements of surface contamination, air contamination and tritium intake are the most important bases for the evaluation of doses for workers, due to internal contamination. The report presents a review of all the analytical methods and technical solutions devised to apply the ALARA principle in this type of practice.

The FNG plant

The Frascati Neutron Generator (FNG) /1/ consists in a direct electrostatic deuteron accelerator whose beam is focalized on titanium tritide layer of the target. The neutron yield is of about $1 \cdot 10^{11}$ neutrons per second and the neutron output, at 14 MeV, is due to the D-T fusion reaction. The deuteron accelerator attains, at its maximum performance, a current intensity of 1 mA and an energy of 300 keV. The beam target is a copper cylindrical cup, 1 mm thick, that is coated on its internal side with a titanium tritide layer. The total tritium inventory of each target is about 370 GBq.

The facility is located inside a shielded room, 11.5x12 m^2 wide, 9 m high; the target is placed just at the center of this room, about 4.5 m above the floor. The plant layout is shown in fig. 1. The shielded rooms, i.e. Generator Area and Handling Area, are served by a ventilation system, performing 1 or 3 room air changes per hours at about 79 Pa of under-pressure.

FNG has been designed in order to fulfill the request of a high-energy neutron source suitable for many applications; the main ones are:

- Benchmark experiments to test neutron transport codes and data;
- Non destructive activation analysis of materials;
- Reference neutrons source for monitor’s calibration.

From radiation protection point of view, FNG presents two kinds of problems:

a) External irradiation due both to prompt radiation and to the delayed one from activated materials;
b) Internal irradiation due to the tritium intake .

Tritium contamination is detected mainly near the target, in the vacuum system and in the cooling water, as it happens at other neutron generator facilities /2, 3/.

Tritium contamination

According with Italian radioprotection law /4/, the present limit for the derived tritium concentration (DAC) as HTO in air is $3.2 \cdot 10^5$ Bq/m^3 (average over 5 years) for radiation workers . This value agreed with the most recent ICRP recommendations /5, 6/ which suggest a figure of $3 \cdot 10^5$ Bq/m^3. To fulfill this requirement accurate evaluation has been performed during the design phase. Actually cost-benefit considerations alone don't justify the choice of the low design values that were adopted and led to a very cautious approach with the consequent adoption of some safety facilities: a very conservative shield thickness, a closed loop target cooling system, a dedicated glove box for target change and a Vacuum Exhaust Cleaning Unit (VECU)
This criterion has been adopted considering the presence of some houses just outside the border of the center and the policy of the Italian licensing agency, which requires very low practical dose constrains for individuals of the public living near accelerators or similar plants. The opportunity of limiting as far as possible the tritium contamination controls mainly outside the FNG building was considered too.

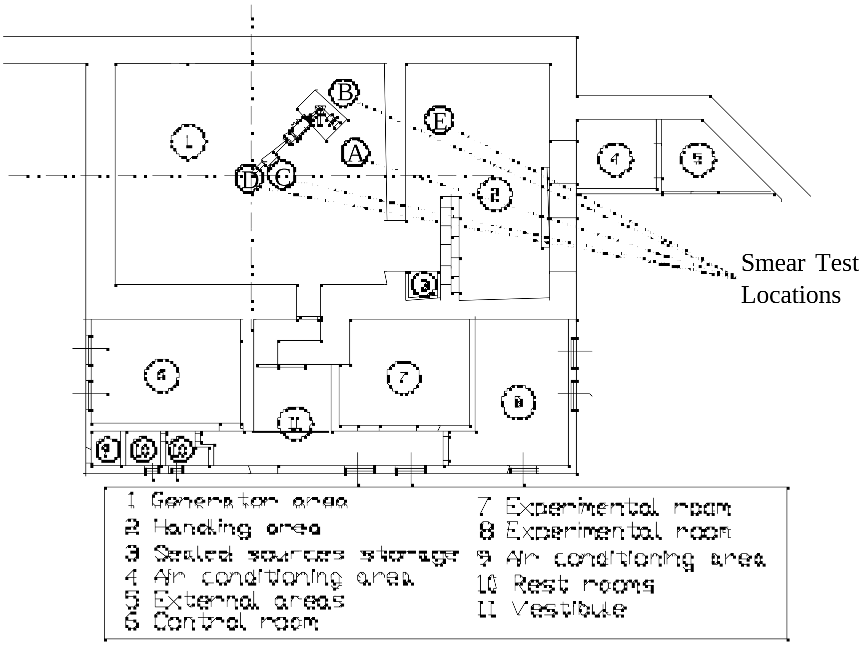

Figure 1. Plant layout

Tritiated target management

Basing on published data /7, 8/, the tritium release to the vacuum system under beam bombardment has been evaluated to be 37 GBq/h as an upper limit. A vacuum exhaust cleanup unit (VECU) has been provided to remove this tritium from the vacuum exhaust, so only 37 MBq/h are expected to be released in the main ventilation system in the more adverse conditions.

The new targets are received inside sealed boxes filled with inert atmosphere; and are stored in a glove box located in the Handling Room until their mounting on the FNG facility. The total annual release from tritium leakage in this glove box, intended also for temporary waste storage, is expected to be 6 GBq/y.

In order to reduce the possibility of surface contamination during the target change, a movable glove box suited to be fixed to the end part of the beam line is used.

Assuming a maximum of 100 target changes per year a total release of 40 GBq/y could be calculated, that will be released directly to the stack by the ventilation system /9/.

Actually during the first seven years of operation only 13 targets have been used and the expected average tritium release in the external environment is therefore about 0.8 GBq/y.

Cooling system

Tritiated target absorbs 300 W at full power beam operation, so it must be cooled to prevent a rapid loss of the tritium. The cooling is obtained by means of a turbulent water flow on the backside of the target disk. The cooling water washes off some tritium, mainly immediately after the target change. It was evaluated that each target would release about 2.2 GBq in the water, a quantity which leads to a total annual release of 220 GBq (100 targets per years operation). A closed loop cooling water system was chosen, containing 0.25 m^3 of water. The cooling water reservoir, the pump and all the valves are installed inside a ventilated box. An annual rise of tritium concentration in cooling water of about 880 GBq/m^3 was foreseen in the design phase, giving a total inventory of 2.2 TBq and a tritium concentration of 8.8 TBq/m^3 after ten years of operation, i.e. a total of 1000 targets. Actually the reduced number of targets used in the first four years of operation depicts a completely different situation and the contamination of the cooling water is considerably lower than that considered in the project.

VECU system

In the VECU, the vacuum exhaust is mixed with dry instruments air and passed through a catalytic recombiner to oxidise all the free hydrogen isotopes; a molecular sieve then adsorbs oxidised tritium.

The complete system is contained in a dedicated glove box near the generator. The VECU filter can contain a tritium quantity as high as about 18.5 TBq that corresponds to more than 500 hours of machine operation. The tritium removal efficiency is expected to exceed 99.9%.

On-line monitoring system

In order to control the tritium concentration and to verify the respect of the above design limits, a real-time monitoring network has been installed in the plant: the three components are:

A. A gamma-compensated ionisation chamber (Labserco mod. LTM 580), which is installed at the end of the VECU system;
B. A 2 litres ionisations chamber which monitor 6 sequential sampling points (Berthold mod. LB6710-2H);
C. A discriminating monitor for tritium and activated gases in atmosphere (Berthold mod. LB110).

In all the above devices the air fluxes across the measurement chamber in order to permit the detection of the low energy tritium beta decay /10, 11/.

The A monitor has two chambers, the first one is sealed and the second is opened to the fluxing air, the measurement result is the difference between the responses of the two channels.

This instrument is connected to the exhaust air from the VECU system, its response is important in order to organize the scheduled change of VECU filter. A tritium concentration higher than the usual background could also indicate that the cleaning efficiency of the VECU system is no longer acceptable and therefore some intervention is required.

The minimum detectable concentration (MDC) of this monitor is of the order of 100 kBq/m^3. This value compared with the design value of tritium concentration ranging only up to about 18 kBq/m^3 makes apparent that its main function is to early detect any unforeseen rise of tritium releases.

The B monitor, which can be connected to six different sampling points through a sequential device, is devoted to the control of the contamination of the air coming from glove boxes and from the ventilated box of the water cooling system.

The C monitor is connected to the terminal duct before the stack of the ventilation system to control the contamination of the air released to the external environment. In this monitor the air is mixed with a counting gas before it is passed through a proportional counter tube having a volume of 1.3 litres. Tritium counts are distinguished from those due to other nuclides or gamma radiation background by means of the method of pulse shape discrimination /12/.

The MDC of tritium for LB110 ranges from 5.4 kBq/m^3 in 30 s measuring time to 0.1 kBq/m^3 in 24 h measuring time. The spillover of background channel to the tritium one, which is about 7%, can significantly deteriorate this figure.

The on-line monitors so far have reported a radiation level equal to their background, some fluctuation has been clearly identified as due to other contaminant, like radon gas, coming from the soil and the building materials. This last hypothesis has been verified noting that the anomalous background was independent from the FNG operation but was strongly connected with the ventilation cycle.

The results of the on-line monitors indicate that the tritium concentration in the plant atmosphere is not higher than 5-6 kBq/m^3 that is the average sensitivity of these devices.

Two portable tritium monitors (Scintrex) are also used mainly during target changes or maintenance works which require the dismounting of some part of vacuum chamber. Their sensitivity isn't very high, 370 kBq/m^3, but they proved to be very useful in health physics assistance during such works, mainly due to their quick response and their quite small size which easily permits to sniff the tritium concentration just on the machine.

Contamination of surfaces

The surface contamination is mainly controlled with the smear-test technique /4, 9, 11/. Sometimes also the portable monitor Berthold LB1239 is used to verify the validity of the other tests.

The portable monitor has been used in the initial phase of the operation to identify some points in the generator area (see figure 1) in which proceed with periodic smear-test control. Five measurement points have been identified on the machine structures, on the floor and on the other working surfaces, these zones are shown with the capital letters from A to E in figure 1. With a monthly frequency the smear-test technique has been applied in the above areas.

The procedure is the following: standard chemical filters mainly composed by glass fibres, are wiped around each identified point inside an area of about 100 cm^2; the smears obtained in this way are then placed each inside a separated vial with a scintillating liquid solution. The vials are then measured in a scintillation counter and the appropriate conversion factor leads to the detected activity concentration, with a MDC of 0.2 Bq/cm^2 (transfer factor = 10%, measuring time = 600 s).

With the above methods a tritium contamination of the surfaces has been detected everywhere around the machine. The contamination level has been found to be very low in almost every sampling point but the machine head, where a contamination as high as 290000 Bq/cm^2 has been sometimes measured. The highest contamination in the floor of the FNG hall has been detected in the point A (see figure 1) after an assembly operation near the ion source. With the smear test technique a contamination of $1.5\ 10^4$ Bq/cm^2 was found in that circumstance. A prompt decontamination campaign was then organised using specific products and the contamination was monitored after each cleaning. The unscheduled procedure was stopped after three cleaning shifts when the higher contamination of the surface was about 62 Bq/cm^2.

In Table 1 the results relevant to some scheduled surface contamination measurements completed in the last years are reported.

Table 1. Some smear-test results related to the scheduled controls

Monthly Period	Tritium concentration on the sampling areas (Bq/cm^2)				
	A (floor)	B (floor)	C (floor)	D (target)	E (floor)
04/1995	22.6	2.7	16.1	24.6	1.9
04/1996	1	1	2	7400	1
04/1997	1	1	6.4	12000	3.2
01/1998	0.9	93	4	12000	39
02/1998	2.5	1.5	3.6	25000	71
03/1998	0.8	0.7	0.4	58	1.6
04/1998	0.4	2.1	2.7	2400	9.5
05/1998	1.3	0.6	0.3	2600	6.8
06/1998	0.3	0.1	0.2	4200	47
09/1998	1.9	5.5	0.5	15000	0.3
10/1998	1.3	0.4	15	60000	2.4
11/1998	0.8	0.5	6.2	16000	0.3
12/1998	3.6	2.1	0.1	990	1.3

The A, B, C and E areas are located on the room floor and the D area is located on the machine head, near the target holder.

The contamination detected on the machine head (D area) can be considered as a parameter to verify that the tritium diffusion and increment in the FNG hall is under control. In figure 2 a graph of the tritium concentration measured on the D area is reported showing that the contamination does not have any incremental tendency. All the higher values are promptly managed and the contamination level is reduced and maintained under control with the decontamination procedures. In fact the fluctuation of the values in the table and in the graph is mainly due to the decontamination campaigns that take place on the machine head before and after the target changing and on the floor when the surface contamination is higher than 10 Bq/cm^2.

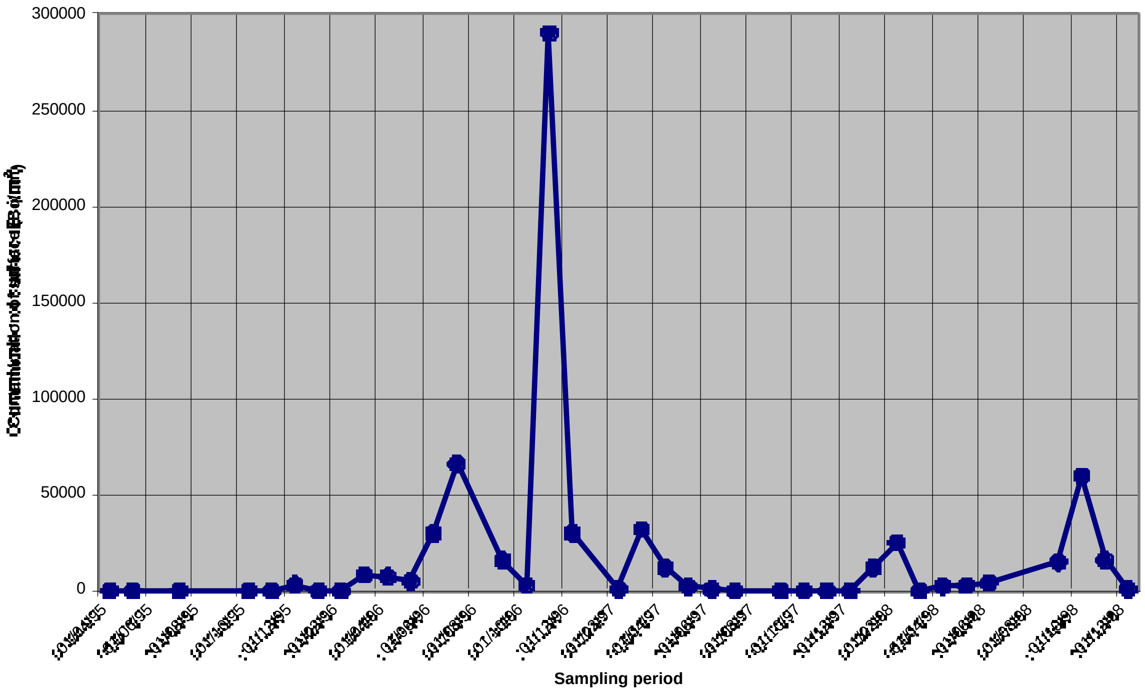

Figure 2. Contamination of the surfaces around the target

Tritium in urine assay

The internal contamination of the FNG personnel is controlled by the tritium in the urine assay method adopted in the Frascati ENEA Center. Samples of the urine of the personnel that work inside the FNG main hall are normally collected with a monthly frequency. The samples are then counted with the liquid scintillation technique. The decision limit (Lc) /13/ of the method ranges between 30 and 330 Bq/l depending on the sample condition and on the sensitivity of the counter.

The quality of the measurement procedure and of the related results is verified by participating to international intercomparisons (Procorad), generally with a yearly frequency.

The valuation of the internal dose is based on the assumption that the intake is not chronic but it could be significant only after an abnormal situation. The reference levels are then those related to the acute intakes (DARL); according to this biological model at a measured tritium concentration in urine of 4.6 kBq/l they correspond to a dose of 0.01 mSv. Owing to this parameter the derived limits for registration and investigation have been fixed at 5 kBq/l (0.011 mSv) and 50 kBq/l (0.11 mSv) respectively.

So far all the results of the measurements on the urine samples of the ENEA personnel operating at the FNG plant have been below the registration limit. The highest tritium concentration measured in this assay has been 3.6 kBq/l (0.008 mSv).

Considerations and conclusions

The problem of tritium contamination in the FNG main hall was firstly considered during the design phase and it was manifest since the beginning of the plant operation. Tritium was found on the surfaces, in the room atmosphere during the target changing and in the cooling water. Nevertheless the diffusion in the external atmosphere appear to be very low according to the measurements of the on-line monitors.

The results of the smear tests performed during the first years of machine operation have confirmed that the zones that show the highest surface tritium contamination are the machine head and the target holder. A detectable tritium concentration is found on the floor too, but only occasionally it exceed a 10 Bq/cm^2 figure. Simple decontamination procedures have been successful in reducing the removable fraction of tritium on all the surfaces. Sequential measurements have shown that the regrowth of the contamination is not important and that the surface tritium activity increases mainly after maintenance operation and target changing.

In fact the periodic procedure that has been identified as the main responsible for the tritium spread in the environment is the connection and disconnection of the movable glove box used during the target changing.

The efficiency of the radioprotection practices, as improved with the continuous implementation of the ALARA process, is confirmed by the negligible intake of tritium shown by the periodic radiobiological assay of the personnel urine.

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