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Radiation Protection Issues for running the n-TOF Facility

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Abstract

The n-TOF facility, a spallation neutron source, has been commissioned at CERN in the year 2000 as an experiement. For being operated as a facility (similar to ISOLDE or AD), a consolidation programme is proposed. The spallation target area has to be equipped with a monitored ventilation system to limit and to account for releases of radioactive aerosols. The present installation and equipment of the experimental area seriously limits the type and activity of radioactive samples that can be employed therein and should be upgraded to a work sector for handling unsealed radioactivity.
This report summarises the shortcomings of the facility from the point of view of radiation safety and makes recommendations for a consolidation programme before starting a new experimental programme in the year 2006 and thereafter.

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1. INTRODUCTION

In 1998, plans were made for a spallation neutron source at the CERN Proton Synchrotron (PS) [1]. The source and a time-of-flight neutron spectrometer, named n-TOF, were built and first proton beam was taken in 2000. The main commissioning phase extended throughout 2001. A proton beam with a momentum of 20 GeV/c hits a production target made from lead (80 * 80 * 30 cm^3) and generates neutrons by the spallation reaction. The nominal proton intensity is 7 10^{12} in one pulse, and 4 pulses per supercycle of 16.8 s duration i.e. 1.6 10^{12} s^{-1} or 6 10^{15} h^{-1}. In 2004, CERN's Research Board authorised a total of 1.6 10^{19}protons for n-TOF. The spallation neutrons emerging from the lead target are moderated by cooling water in contact with the target and then enter the evacuated time-of-flight tube in an angle of 10^o to the proton beam direction.
After commissioning, the experimental programme lasted from 2001 until today. Neutron capture and fission cross sections of interest for future scenarios of nuclear energy generation or nuclear waste management were determined, in addition experiments with a focus on astrophysics or nuclear structure physics were conducted.
During the 4 years of exploitation of the facility, radiological safety issues were coming up in two areas:

1.) The target area with the spallation target is not ventilated. Due to spallation in air, radioactive isotopes with half-lives ranging from hours to months are produced. The air is released unmonitored into the environment. Conservative models, which have to be applied when no monitoring is available, predict that the releases from n-TOF exceed those of all other facilities at CERN. Furthermore, the cooling water circuit of the neutron production target is contaminated with radioactive isotopes diffusing out of the lead. The majority of the activity is concentrated in an ion exchanger.
2.) The experimental area, where targets are exposed to the neutron beam, is constructed like an experimental area in an accelerator facility, not like in a nuclear laboratory. This fact limits the type and the activity of radioactive sources to be handled therein and restricts the experimental programme to sealed radioactive targets.

The limitations of the n-TOF facility listed here can be overcome by a number of technical consolidations. The following two chapters deal in detail with the radiological safety issues in the target area and in the experimental area. Experiences from the last years are described and documented with results from measurements. The limitations of the research programme coming from these safety issues are highlighted. Then, the necessary improvements are described. Once realised, they will allow handling more important quantities of radioactive isotopes in the experimental area. A table of the limiting activities is given.

2. THE N-TOF TARGET AREA

2.1. Air Activation and gaseous Radioactive Releases

In every high-energy accelerator, activation of air by spallation is a safety issue. Beam loss in the accelerators and transfer lines, but specifically in the target areas lead to secondary particle cascades traversing air and creating activation products by spallation, and to a lesser extent, by slow neutron capture. The air from an accelerator tunnel will eventually be released into the environment, carrying with it any activation products. The release of radioisotopes is taken account of in the operating licence of a facility. There, certain annual release limits will be stipulated. It is the duty of the establishment to construct its installations in a way that the release limits are respected under normal operation conditions. Usually, air in an accelerator tunnel is exchanged by a ventilation system, the exhaust of which is forced through a particle filter into a stack. The concentration of radioisotopes in air is measured downstream of the filter with a radiation monitor. From the activity concentration and the flow rate in the ventilation stack, the total emission can be deduced. This procedure is required in CERN's Safety Code F [2] in order to assess releases. The Environmental Monitoring section of SC-IE group is operating a network of emission monitors and is able to demonstrate compliance with emission limits for all installations at CERN with exception of n-TOF.

2.2. Air activation in the n-TOF tunnel (TT2a)

During the construction of n-TOF in the former transfer tunnel TT2a it has been decided not to install a ventilation system with a filtered and monitored stack, in spite of the high beam power incident on the n-TOF target. Furthermore, the secondary particles leaving the n-TOF target are not caught in a beam dump, but they have a considerable path length in air, creating more activity in air per incident proton than in other installations.
In November 2001, the air activation in the n-TOF tunnel was measured during operation by taking aerosol samples on filters and performing a subsequent γ-spectrometry [3]. The results are given in Table 2.

Table 2. Results of an aerosol concentration measurement in the n-TOF tunnel in November 2001 at nominal beam intensity (2.8 10^{13}p per supercycle). Remark: the measurement was performed at 25% of this intensity.

	Activity concentration	
	Measurement position	
Isotope	70 m from target	140 m from target
	Bq m^{-3}	Bq m^{-3}
^{7}Be	928	620
^{24}Na	308	336
^{39}Cl	2120	480

Additional information could be gathered from the measurement: the presence of ^{41}Ar in the experimental area (200 m from target) but the absence of shorter-lived isotopes indicates a migration time of air between 3 and 8 hours, or approximately 4 air exchanges per day. With the volume of the tunnel (l=200 m, A=12 m^2), 2400 m^3, a release of 230 MBq of ^{7}Be, of 77 MBq of ^{24}Na and of 370 MBq of ^{39}Cl per month at nominal beam intensity can be estimated, more than from any other facility at CERN.

The secondary particle cascade hitting the slightly inclined tunnel wall downstream of the target generated so much background radiation in the experimental area that design levels of radiation background were exceeded there by a factor of several 100 (but remaining at levels without concern for radiation protection). This incited the n-TOF collaboration to implement additional shielding immediately downstream of the target. Monte-Carlo calculations of air activation before implementation of the shielding confirmed the aerosol measurements above within a factor of two [4]. According to the same calculations, a concrete shield of 14.4 metres thickness would reduce the production (and thus concentration) of radioactive aerosols in tunnel TT2a by a factor of 10.
The additional shielding was installed in spring of 2002. In addition, access door 203 was moved 40 m downstream, away from the target, thus separating the area in which most activity is generated in air from the remainder of the tunnel. The aerosol measurements were repeated in July 2002 [5] and the results are presented in table 3. One observes an important reduction of activity concentration at door 104 (140 m from the target). Close to door 203, however, the observed concentrations remain identical to those measured in the year 2001.

Table 3. Results of an aerosol concentration measurement in the n-TOF tunnel in July 2002 after installation of additional shielding at nominal beam intensity (2.8 10^{13}p per supercycle).

	Activity concentration	
	Measurement position	
Isotope	Door 203	Door 204
	Bq m^{-3}	Bq m^{-3}
^{7}Be	860	27
^{24}Na	290	4

The installation of the additional concrete shield and the displacement of the access door have attenuated the production and the propagation of radioactive aerosols by the amount expected from the simulations. In spite of this, there is still a significant amount of activity produced, calling for a monitored ventilation system of the target area. With exception of dedicated measurements with an aerosol sampler the emissions are uncontrolled. It is indispensable that this situation is rectified and that a filtered and monitored ventilation system is installed in the n-TOF target area.

2.3. Contamination of the cooling water

The n-TOF spallation target, a lead block is immersed into a cooling water circuit. The demineralised cooling water is in direct contact with the lead block. Mobile radioisotopes migrate into the cooling water and lead to its contamination. An ion exchanger is retaining a large number of radioisotopes in ionic form; only soluble elements and tritium remain in the circulating water. Measurements performed in the shutdown 2003/2004 revealed a tritium concentration of 2.5 MBq/l with a sampling uncertainty of approximately 20% after 3 years of operation [6]. The 700 l of cooling water contained 1.75 GBq of ^{3}H. A γ-spectrometry of the resins after the same duration of operation revealed that they contain, apart from numerous other contaminants, the isotope ^{194}Hg ($t_{1/2}$ = 444 years) in an amount of 30 kBq, exceeding the level of radioactive waste by a factor of 150. The water has to be considered radioactive waste and can only be released after thorough cleaning with ion exchangers and by giving due consideration to the tritium content. The resins have to be treated as long-lived radioactive waste and an elimination pathway for them has to be defined.

3. THE N-TOF EXPERIMENTAL AREA

3.1. Radiological issues of unsealed radioactive sources

International recommendations in radiological protection often refer to sealed radioactive sources. The concept of a sealed source is to encapsulate the radioactive material in a protective shield. The capsule protects the source from external influences and prevents dispersion of its content under normal conditions of use. An International Standard ISO 2919 [7] lists requirements for the resistance of sealed radioactive sources against the external factors temperature, pressure, mechanical impact, vibration and puncture. A resistance against each of the external influences is graduated in 5 classes. The exact classification required for a sealed source will depend on the environment in which the source is to be used and the radiotoxicity of its contents. Higher resistance classification obviously goes hand in hand with heavier encapsulation. A source is considered as a sealed source when its resistance against external influences has been tested by an approved service. This service will establish a certificate for every individual source, noting the isotope, the activity and the verified classification according to the standard. Every radioactive source, which is not accompanied by such a certificate, has to be considered as an unsealed source.

The n-TOF experimental programme includes the measurement of neutron capture and neutron-induced fission cross sections on radioactive isotopes. The isotopes under study have to be prepared as targets. Especially during measurements of fission cross sections the radioactive targets cannot be encapsulated. But also in capture cross section measurements the type and thickness of encapsulation is limited by considerations of shielding against the (n,γ)-radiation and parasitic neutron interactions. Finally, some of the targets to be used at n-TOF have been prepared in other research laboratories without applying the same procedures as used for the production of sources by the specialised industry and without access to approved testing facilities for the establishment of a

certificate according to ISO 2919. One can state that the majority of radioactive targets used at n-TOF in the years 2001-2004 had the form of unsealed radioactive sources. It has been shown during 2004, that conformity certificates for sealed radioactive sources for use at n-TOF can be obtained from accredited laboratories. These sources can be used in the experimental area in its present form as long as the criterion developed in section 3.4 is respected.
Unsealed radioactive sources do not offer protection against dispersion of the radioactive material even during normal conditions of use. The result of handling such a source may be contamination of storage rooms, laboratory rooms and finally the experimental area and the subsequent incorporation of radioactive isotopes by personnel working in these areas, leading to a personal dose from internal irradiation.
These consequences can be extremely severe for material with high radiotoxicity (e.g. actinides) and a high activity. In order to offer a protection during the handling of unsealed radioactive sources, which is commensurate with the risks, CERN's Safety Code F adopts a classification system originally developed in Switzerland [8]. For every radioactive isotope, a so-called authorisation limit for its activity (symbol L_A) is defined. The inhalation of radioactive material with an activity L_A would entail a committed dose from internal irradiation of 5 mSv. The use of unsealed sources with an activity of less than L_A is unrestricted. An activity exceeding L_A must be handled and stored in specific work sectors. For a mixture of isotopes or an assembly of different radioactive sources, one can calculate the ratio of total activity to the compound authorisation limit by

$$\frac{A_{tot}}{L_{A,comp}} = \sum \frac{A_i}{L_{A,i}},$$

where the index i runs over all isotopes contained in the mixture or assembly.

3.2. Work sectors for the safe handling and storage of unsealed sources

A work sector designates a room in which precautions have been taken to protect the personnel from incorporation and to protect the environment from dispersion of radioactive material. CERN's Safety Code F, in line with the Swiss Ordinance for Radiological Protection [8] and the Ordinance for Handling of Unsealed Radioactive Sources [9] define 3 types of work sectors, from Type C to Type A with increasing technical and organisational requirements, allowing handling higher and higher quantities of unsealed radioisotopes.
Table 4 shows the activity limits and table 5 the technical requirements for the three types of work sectors.

Table 4: Activity limitation for handling and storage in the three types of work sectors

Work Sector	**Handling**	**Storage**
Type C	$A < 100\ L_A$	$A < 10\,000\ L_A$
Type B	$A < 10\,000\ L_A$	$A < 10^6\ L_A$
Type A	A < individually authorised amount	

Table 5. : Various requirements for construction, ventilation, installation, hygiene, and working clothes in work sectors for handling unsealed radioactive sources. The table has been compiles from information in the Swiss Ordinance for radiological protection (ORap)[8] and the Ordinance for handling unsealed radioactive sources (OSrc) [9].

Requirement	**Work Sector Type C**	**Work Sector Type B**	**Work Sector Type A**	**Reference**
Construction				
Fire resistance	F30, T30	F60, T30, R30	F90, T60, R60	OSrc 5
Floor	Continuous and impermeable floor coating			
	Plinths welded to floor	Floor coating raised 10 cm to wall		
Walls	Washable paint	Continuous and impermeable coating		
Ventilation, Waste water				
Ventilation	-	Artificial ventilation, more than 5 air renewals per hour, Rooms with higher hazard potential are under pressured with respect to room with lower hazard.		OSrc 11
			Under pressure secured in case of power failure. Fire retention system obligatory	
Fume cupboards		Air flow > 0.5 m^3/s with an opening of 20 cm		
Glove boxes		Under pressure of 50 Pa with respect to environment		
Waste water	Isolated work sector: no requirements: More than one work sector on site: control may be required	Control of waste water may be required	Control of waste water obligatory	OSrc 13
	Treatment may be required			

Table 5, continued

Requirement	Work Sector Type C	Work Sector Type B	Work Sector Type A	Reference
Installation, Instrumentation				
Furniture,	To be limited to the strict minimum, easily decontaminable			OSrc 27
Signalisation	Usual signposts for controlled radiation areas plus indication of special hazards such as high dose rates			OSrc 22
Measurement apparatus	At least one suitable device for contamination measurements Dose rate meters, if necessary			OSrc 19.1
Extremity control	A hand monitor	A hand-foot monitor		OSrc 19.3
Hygiene				
Changing room	-	Changing room in front of entrance		OSrc 6
Access to work sector	-		Access only via changing room	
Showers	-		Shower and decontamination equipment	
Decontamination wash basins	In immediate proximity of work sector, needs to be operable without hands			OSrc 8
Working clothes				
Wearing	Obligatory	Specifically marked working clothes, not to be worn outside of the work sector. Specific shoes or overshoes.		OSrc 25
Closets		For working clothes inside the work sector, for private clothes outside the work sector		
Control, washing	Contamination control. If guideline value CS (ORap annex 3, column 12) is exceeded, cleaning only by an authorised establishment			

3.3. Protection of the Public and the Environment

As a general rule, establishments using radioactive sources or operating devices, which generate ionising radiation, have to take the necessary technical and organisational precautions in order to protect the environment and the public. For this purpose a critical group of the public is identified, a hypothetical group of persons for whom the individual impact of ionising radiation from the establishment is maximised by their living place and habits. By definition, the radiation dose of the critical group is a conservative estimate of dose to an arbitrary member of the public. The radiation dose to the critical group is evaluated using approved, standard models [10,11], taking into account direct radiation and radioactive releases.

Under normal operation conditions, the annual radiation dose to the critical group must not exceed the value of 300 μSv. In fact, the establishment has to demonstrate by measurements and calculations of the radiation impact that operation of the facility leads to a smaller dose than this guideline.

In case of major incidents, occurring with a probability of less than 10^{-2} per year, the dose to the public from one incident must not exceed the value of 1 mSv per year, the dose limit for members of the public. It is the duty of the establishment to demonstrate that this limit is under no circumstance exceeded. Under an incident one has to understand a situation which may develop in such a way that even radioactive sources, which are regarded as sealed for normal operational conditions are no longer retaining the activity contained in them. The 1-mSv-dose limit criterion can be converted into an absolute activity limit for all types of sources, which are not specifically protected against the type of incident under study.

Certain activities (e.g. nuclear power generation) may have very severe consequences for the public in the case of incidents occurring with a very low probability. Once it has been established that a certain incident in such a facility has an annual probability of less than 10^{-4}, the competent safety authority will decide upon the necessary safety requirements, bearing in mind the protection of the public. The decision process will involve representatives of the public as the main stakeholders. Emergency response operations are set up and regularly reviewed and trained in the surroundings of such a facility.

3.4. Activity limitations in different scenarios

With the information from the previous sections, activity limits can be calculated for every radioactive isotope that is to be handled in the form of an unsealed source in a work sector of a given type. In table 6, maximum activities and masses of radioactive isotopes are given for handling in a work sector of type B and considering the 1 mSv-criterion for protection of the public in case of an incident. In this case, the activity limit is derived from a calculation of the environmental impact during an accidental release at ground level from the CERN site [12]. The calculation assumes a worst-case scenario in which the total activity of the source is dispersed at ground level and the critical group is situated in 700 m distance in downwind direction from the release point. Persons on the

CERN site in the vicinity of the release point might receive doses in excess of 1 mSv as a consequence of such an incident.
At CERN, where no specific precautions are taken for limiting the consequences of incidents with very small probabilities, an amount of activity not exceeding the value derived from the 1-mSv criterion can be authorised for handling in either sealed form or in a work sector of type A. This criterion therefore represents the ultimate activity barrier, all other possible technical and organisational requirements for a work sector of type A taken into account.

Table 6. List of representative isotopes for study at n-TOF. The authorisation limit L_A and the derived activity limit for a work sector of type B is given in Bq and in mg (if not specified otherwise). The last two columns list the activity limit derived from the exposure limit for the population in the case of a major incident, entailing dispersion of the source content. An entry of 0.0 means that the authorised amount is less than 0.1 mg.

Isotope	Authorisation Limit L_A	Work sector type B (10 000 L_A)		Protection of public (1 mSv criterion)	
	Bq	Bq	mg	Bq	mg
Pb-210	5.00E+03	5.00E+07	0.0	2.22E+10	7.8
Bi-207	2.00E+06	2.00E+10	11.9	1.27E+11	75.6
Bi-210	8.00E+04	8.00E+08	38.0 g	2.69E+11	12 kg
Po-210	2.00E+03	2.00E+07	0.0	7.46E+09	0.0
Ra-226	2.00E+03	2.00E+07	0.5	6.99E+09	191.1
U-233	7.00E+02	7.00E+06	19.7	6.80E+09	19 g
U-234	7.00E+02	7.00E+06	29.7	6.99E+09	29 g
U-235	8.00E+02	8.00E+06	99.5 g	7.81E+09	97 kg
U-236	8.00E+02	8.00E+06	3.3 g	7.63E+09	3 kg
Np-237	3.00E+02	3.00E+06	115.0	1.06E+09	40 g
Pu-238	2.00E+02	2.00E+06	0.0	5.32E+08	0.8
Pu-239	2.00E+02	2.00E+06	0.9	4.90E+08	212.5
Pu-240	2.00E+02	2.00E+06	0.2	4.88E+08	57.8
Am-241	2.00E+02	2.00E+06	0.0	5.81E+08	4.6
Am-243	2.00E+02	2.00E+06	0.3	5.95E+08	80.7
Cm-244	3.00E+02	3.00E+06	0.0	9.01E+08	0.3
Cf-252	4.00E+02	4.00E+06	0.0	1.22E+09	0.1

4. SUMMARY AND CONCLUSIONS

The n-TOF facility in its present state does not present the technical features necessary for limiting and controlling the radiological risks associated with its operation. Two upgrades should be considered in a consolidation programme before continuing the research programme at n-TOF:

1.) The spallation target area needs to be equipped with a ventilation system. The used air has to be evacuated via particulate filters into a monitored stack. Filtering will reduce the aerosol concentration in the released air. The releases from the stack will be monitored, allowing the demonstration of compliance with release limits.

2.) The experimental area needs to be modified and equipped with the technical features of a work sector for handling unsealed radioactive sources. A type B work sector would allow handling a very limited range of actinide isotopes. The additional technical requirements for a type A work sector are minor and would open research at the installation to isotopes where the activity is only limited by the criterion of protection of the public from the consequences of a major incident.
In the experimental area in its present state, only certified sealed sources can be used up to this activity limit.

This note has been focussed on the technical aspects for radiological safety at the n-TOF facility. They will be accompanied by precise procedures to safely import, transport, store and handle sources and targets with high specific radioactivity and radiotoxicity. The success of the protection measures needs to be demonstrated by a monitoring programme. CERN's environmental monitoring programme of radiological parameters is already able to cope with the specific radioisotopes used at n-TOF. A comprehensive monitoring programme for the potentila contamination of the laboratories and the internal dose of the personnel working at n-TOF remains to be designed.

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