ORGANISATION EUROPEENNE POUR LA RECHERCHE NUCLEAIRE
EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Laboratoire Européen pour la Physique des Particules
European Laboratory for Particle Physics

Safety Commission

Technical Note
CERN-SC-2005-034-RP-TN
EDMS No. 584 649 v.2

Contamination of the Cooling Water Circuit of the n-TOF Spallation Target

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Abstract

The n-TOF facility, a spallation neutron source, uses a pure lead target for production of neutrons with a proton beam at an energy of 20 GeV. The target is immersed into cooling water. Since, 2001, a steady transfer of radioactive spallation products into the cooling water has been observed. From August 2004 on, this transfer has accelerated. The present spallation target is considered damaged. The dose rate of the ion exchanger is so high that access to the area is restricted. The cooling water has to be considered radioactive waste.
A new spallation target shall be constructed in a way that contamination of the cooling water by lead spallation products can be avoided. Some general principles, taken from a similar target at PSI, are given.

CERN, 1211 Geneva 23, Switzerland
26 April 2004

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 * 40 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/year 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.

Figure 1: Picture of the n-TOF target assembly, from [2].

From the beginning of the experimental programme, a steady transfer of radioactive spallation products into the cooling water had been observed. An ion exchanger in a bypass of the cooling water circuit concentrates the radioactive isotopes. The dose rate indicated by an ionisation chamber fixed to the ion exchanger is a measure of the total activity of the isotopes leeched from the target. Regular gamma-spectrometric analyses of cooling water samples confirmed a level of contamination, which was not negligible but relatively easy to handle. SC-RP could conclude in a report [3] that, after the end of the n-TOF programme, circulating it over fresh ion exchangers could probably clean the cooling water, which could then be released as inactive. At that time (August 2004), 30 kBq of ^{194}Hg was the most prominent contaminant in the 700 litres of cooling water. The ion exchanger resins would have to be treated and stored as long-lived radioactive waste.

2. RADIOLOGICAL ASSESSMENT

The ionisation chamber fixed to the ion exchanger is connected to the ARCON system and can be read out continuously. Figure 2 shows the development of the dose rate indicated by the chamber since May 2004. The evolution of dose rate in the months from May to July corresponds to the pattern observed in previous years. During operation, the chamber indicates a value around 5 mGy/h. Once the target is not in operation, this value decreases rapidly to a level of approximately 300 μGy/h. Long-lived isotopes, all spallation products of lead, provoke this residual dose rate.

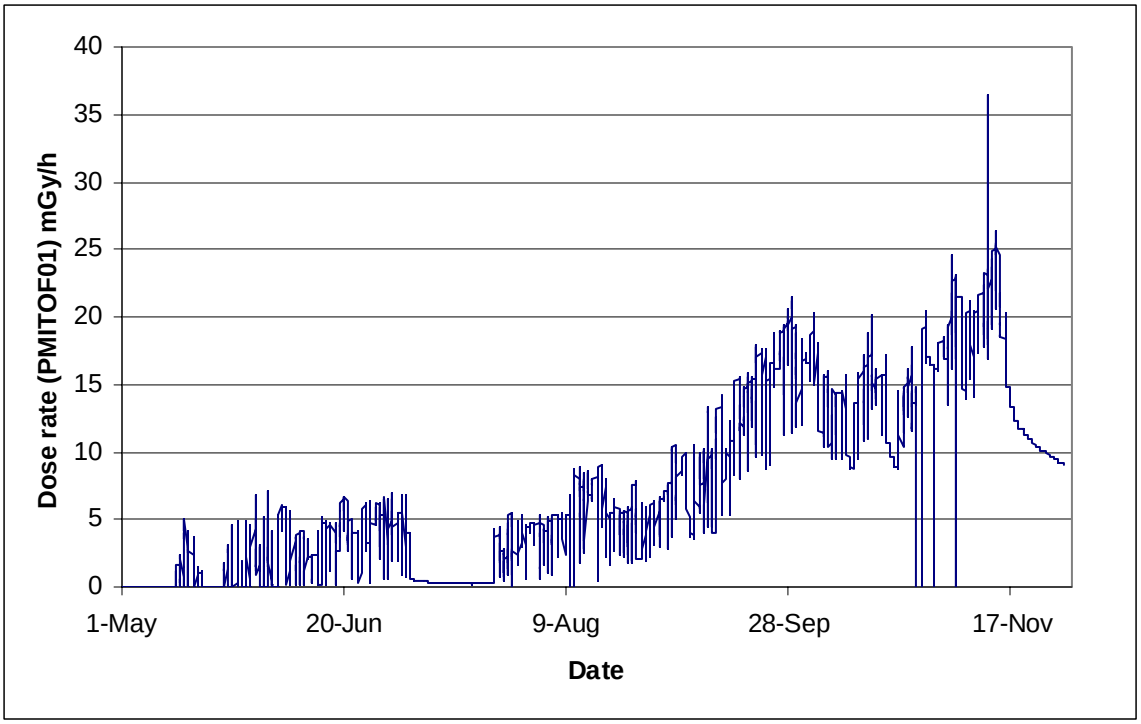

Figure 2: Evolution of the dose rate indicated by the ionisation chamber fixed close to the ion exchanger in the n-TOF cooling water circuit. From mid-August, a clear increase of the dose rate is observed.

From mid-August 2004 on, the dose rate measured on the monitor increased continuously. During the month of October, several prolonged stops of the n-TOF facility interrupted the progression, in November the increase continued.
One of the beam stops in October gave the occasion to observe the decrease of the dose rate as a function of the waiting time after the last proton beam pulse (Figure 3.) After 24 hours, the dose rate measured in 2004 was 10 times as high as at a comparable moment in 2003. This means that the isotopes retained in the ion exchanger have a longer half-live than in previous years.

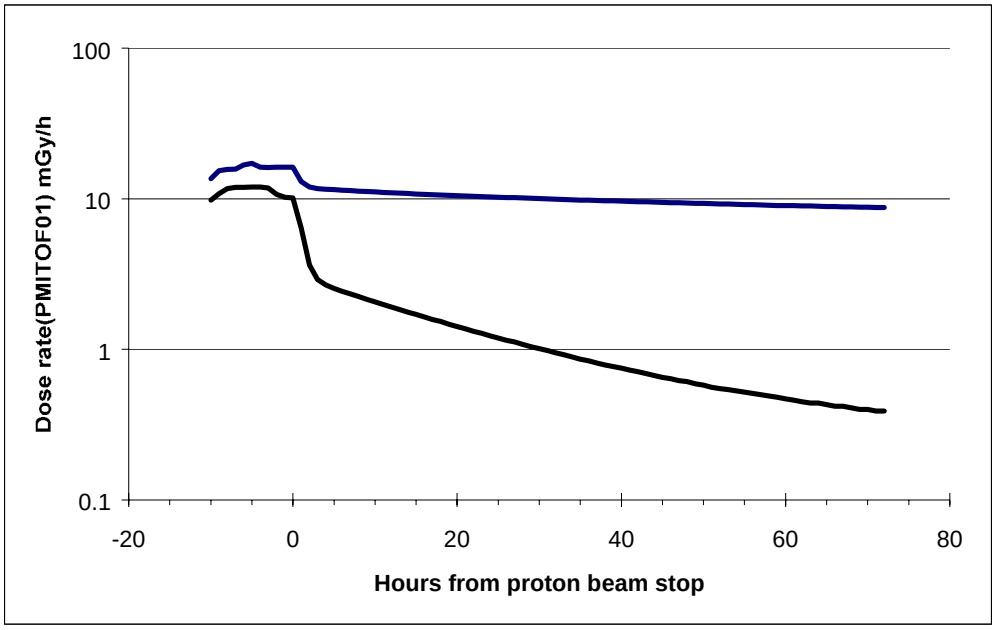

Figure 3: Dose rate measured with an ionisation chamber next to the ion exchanger in the n-TOF cooling water circuit. The proton beam is stopped at time 0. After 24 hours, the dose rate in 2004 (upper curve) is approximately 10 times higher than at the comparable time in 2003 (lower curve), while similar proton beam conditions prevailed before the end of the irradiation.

At the occasion of the beam stop in mid-October, a water sample was taken and analysed by gamma spectrometry. This was repeated on November 16, after the stop of the installation for the shutdown.
Table 1 gives the activity concentration a_i of a few selected isotopes in the water samples taken at different times.

Table 1: Some examples of gamma-emitting isotopes in the cooling water of n-TOF in 2003 and in 2004. The activity concentration of isotopes generated by spallation in lead and transferred to the cooling water has increased by two to three orders of magnitude.

Isotope	Activity concentration 11.11.2003 (Bq g^{-1})	Activity concentration 12.10.2004 (Bq g^{-1})	Activity concentration 16.11.2004 (Bq g^{-1})	Ratio Nov. 2004/ Nov. 2003
^{7}Be	99.6	84.4	74	0.74
^{65}Zn	4.49 10^{-2}	1.63	6.6	147
^{88}Y	2.88 10^{-2}	4.51	18	625
^{172}Hf/Lu	3.6 10^{-2}	6.44	23	639
^{183}Re	7.27 10^{-2}	8.83	73	1004
^{185}Os	3.46 10^{-2}	25.9	120	3468
^{195}Au	9.02 10^{-2}	59.0	360	3991

The activity concentration of ^{7}Be, produced predominantly by spallation of oxygen in water, remained more or less constant. The concentration of spallation products of lead has risen in an important fashion. One can observe that during 2004, the activity of isotopes close to the mother isotopes of lead increased most strongly. These isotopes have the highest spallation cross-sections for any lead isotope as a target nucleus. They generally have longer radiological half-lives than spallation products of water, explaining the slower decrease of the dose rate close to the ion exchanger.

At CERN, water is considered liquid waste if the activity concentration (specific activity) a exceeds the exemption limit L_E **and** the total activity A of that isotope exceeds 100 L_E. The values of L_E are defined isotope-specifically [4]. In order to consider water containing a multitude of isotopes as radioactive waste, it must fulfil the conditions (1) $\sum_i \frac{a_i}{L_{E,i}} > 1$ and (2) $\sum_i \frac{A_i}{L_{E,i}} > 100$ for a_i , A_i the specific and the total activity of isotope i and $L_{E,i}$, the respective exemption limit.
Table 2 compares the activity concentration a and the total activity A in the cooling water circuit with these limits. All isotopes in the list, with exception of ^{7}Be, are sufficiently abundant in the cooling water circuit so that the water would be regarded as radioactive waste for each of them alone.

Table 2: Activity concentration and total activity in the cooling water of n-TOF. Sample taken approximately one week after the last proton beam and then extrapolated to 24 hours after begin of the shutdown.

Isotope	Exemption Limit L_E (Bq g^{-1}) or (Bq)	Activity concentration a 16.11.2004 (Bq g^{-1})	Multiple of L_E	Total activity A in 700 l	Multiple of 100 L_E
^{7}Be	400	74	0.19	51800	1.3
^{65}Zn	3	6.6	2.2	4620	15.4
^{88}Y	8	18	2.25	12600	15.75
^{172}Hf/Lu	8	23	2.88	16100	20.13
^{183}Re	10	73	7.3	51100	51.1
^{185}Os	20	120	6	84000	42
^{195}Au	40	360	9	252000	63

3. MECHANISM OF WATER CONTAMINATION

The radioactive isotope contamination of water corresponds to the amount of spallation products in 1 g of lead per litre of cooling water over the lifetime of the facility [4]. Three hypotheses are brought forward [5]:

1. Lead has been continuously dissolved from the target surface by corrosion. It finally reached a concentration of 1 g /l. It has been activated in solution by the secondary particles emerging from the target.
2. The activated lead has been converted into lead oxide during the attempt to eliminate all water from the cooling system in the early summer 2004. The lead oxide, or at least the spallation products contained therein, has then gone into solution when the cooling water was restored for the run in summer 2004.
3. The target has been damaged by the beam impact and a macroscopic amount of activated lead has been dissolved into the water.

The first and second hypotheses assume an important transfer of lead into water by corrosion or by dissolution of lead oxide. At PSI, where different lead targets have been built or are projected, a corrosion rate of 5 10^{-3} cm/year has been determined for the Pb-Water system [6]. This means that corrosion will progress by 50 μm per year on a flat interface between lead and water. The n-TOF target has a surface of more than 32 000 cm^2, not counting the beam catcher and water penetrating between the blocks of which the target is composed. The volume of lead corroded every year is therefore 160 cm^3 with a mass of 1.8 kg. Not all of the corrosion products will be dissolved, but a concentration of 1 g/l of lead or equivalent spallation products appears in the range of the possible. The activity concentration in the cooling water could therefore be explained by hypothesis 1. However, it is not clear why the activity concentration rose steeply since summer 2004. Hypothesis 2 delivers a possible answer by locating the contamination rise after the drying and reimmersion of the target.

The validity of hypothesis 1 can be tested by a measurement of the Pb concentration in the cooling water. If it is significantly smaller than 1 g/l, then hypothesis 2 or 3 can be distinguished by a visual inspection of the target.

4. RECOMMENDATIONS FOR RADIOLOGICAL PROTECTION

The observations and measurements made in the last chapter can be summarised as follows:

1. The n-TOF target has been damaged by the proton beam interaction, probably in August 2004, leading to an important increase of the transfer of radioactive isotopes to the cooling water.
2. The cooling water and the ion exchanger in the n-TOF cooling water circuit are strongly contaminated with spallation products of lead. They present a radiological hazard in the form of external irradiation and of contamination.

The following short-term steps for radiation protection shall be taken:

1. Measurement of the lead concentration in the cooling water

2. Access to the cooling water circuit, in particular to the ion exchanger, must remain strictly limited due to the high dose rates.
3. Until further notice, no work is authorised on parts of the cooling water circuit to avoid any danger of contamination of personnel or of the area. Water released into tunnel TT2a would reach the public water system (Nant d'Avril) nearly directly.

In the next weeks, the water will continue to circulate in the cooling water system, with an interruption during the Christmas holidays. SC-RP will regularly analyse samples by gamma-spectrometry. The further procedure will depend on the results of these samplings, which may indicate a decrease of the activity concentration in water as determined by the radioactive half-life or faster, depending on the effectiveness of the ion exchanger.
During the year 2005, when the dose rate of the ion exchanger is low enough to permit an intervention, the cooling water and the ion exchanger shall be removed and stored or treated as radioactive waste. A first estimate based on the measured isotope concentrations foresees that the dose rate in 10 cm distance of the ion exchanger will amount to 760 μGy per hour in November 2005. Once the contaminated water is removed, the water tanks and pipes will present an increased risk of personnel contamination by resuspension of deposited activity. The lead target shall then be removed and visually inspected in order to identify the reasons for the target failure during 2004.

The present spallation target, in which lead is in direct contact with cooling water, represents an important liability for the conduct of the experiment and for CERN. Even if the mechanism of water contamination was understood and the cooling system decontaminated, the continued use of this target will expose CERN and the collaboration to the permanent risk of a new contamination. This event will result in high costs in personnel and material (decontamination, disposal of contaminated waste) and bears an important safety risk (external exposure, contamination hazard).
It is therefore required that before the continuation of the n-TOF research programme, a new lead target is constructed. This target shall be fully clad in stainless steel or zirkalloy. At the SINQ neutron spallation source at the Paul-Scherrer-Institute (Villigen, CH), such a target is used with a continuous proton beam power of 700 kW. No radioactive contamination other than from the spallation of water has ever been identified there in the coolant. A pulsed proton beam with a peak power of 1.2 MW will irradiate another spallation target in preparation for the UCN experiment at PSI. Also there, zirkalloy claddings are foreseen.
The cooling water circuit of the new target shall contain an ion exchanger in order to retain the spallation products of water and as a backup in case of an accidental rupture of the target cladding. Since the ion exchanger appears to create problems in conjunction with advanced coolants/moderators such as D_2O or Boric Acid, it is proposed to separate the functions of cooling and moderation: The target can be cooled with a circuit filled with light water, purified by an ion exchanger. A liquid (D_2O) or solid moderator

(borated polyethylene) can be arranged in front of the target, in direction of the extracted neutrons.

These measures will greatly improve the maintainability of the n-TOF target area. The radiological risk from the cooling circuit will be reduced to that of spallation products of water, which are generally short-lived or have, in the case of ^{3}H, a low radiological impact.

5. REFERENCES

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