EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of the neutron capture and fission reactions of plutonium-241 at the n_TOF experimental area 1 (EAR1)

[26/09/2023]

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Abstract

The plutonium-241 is an important fissile nucleus of the U/Pu fuel cycle. However, its short half-life of about 14 years induces a very high radioactivity that challenges sample preparation and cross section measurements. In addition, ^{241}Pu decays to ^{241}Am which is an issue because of its large capture cross section and its α radioactivity. For these reasons only one capture measurement is available and the Nuclear Energy Agency recommends to improve the accuracy of both the capture and fission cross sections. The experimental setup for this proposal is a combination of the Total Absorption Calorimeter (TAC) for detecting gamma-rays and a Fission Chamber (FICH) for identifying fission events. A similar setup was successfully used to measure other fissile actinides at n_TOF recently. The measurement will provide neutron-induced capture cross section and capture-to-fission ratio as well as valuable information on the distribution of energies and multiplicities of the prompt gamma-rays emitted after capture and fission reactions.

Requested protons: 5.3 x 10^{18} protons on target
Experimental Area: EAR1

Motivation

Plutonium is produced in current power reactors using uranium fuel. ^{239}Pu is produced by neutron capture on ^{238}U followed by two β decays. ^{240}Pu and ^{241}Pu are produced by successive neutron captures on ^{239}Pu (Figure 1). At the end of a reactor cycle, when most of the ^{235}U has been used, the fissile isotopes of plutonium (^{239}Pu and ^{241}Pu) contribute significantly to the production of energy. In the case of an innovative reactor using plutonium fuel, the contribution of ^{241}Pu is important from the beginning of the reactor cycle.

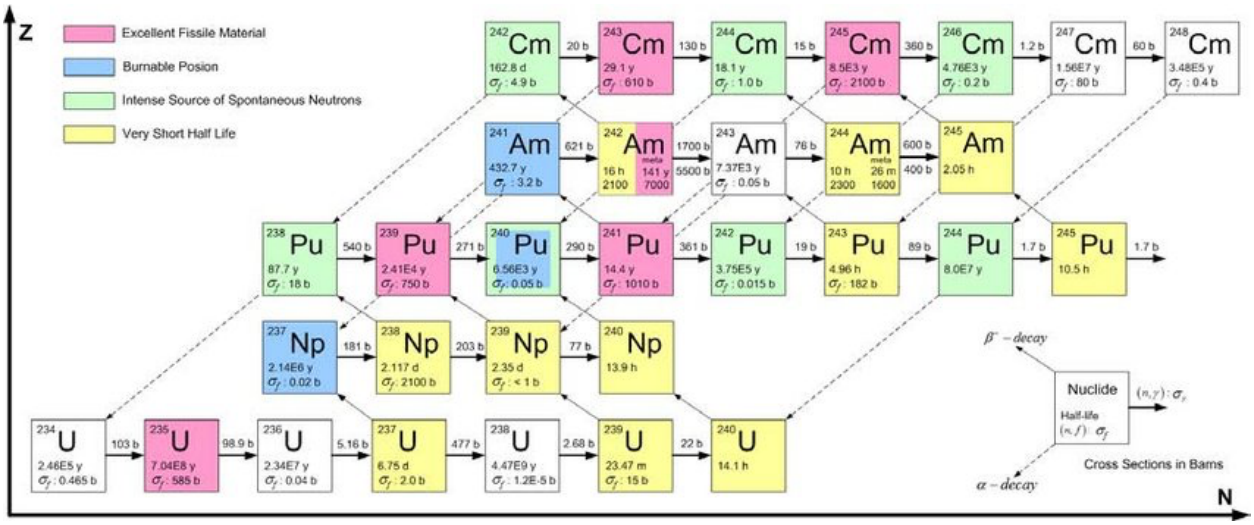

Figure 1 – Chart for the production of transuranic elements in a nuclear reactor [1]

The ^{241}Pu cross sections are not well known because of its relatively short half-life (~14 years) and the associated difficulties (availability, radioactivity) as well as the ^{241}Am build-up that challenges accurate measurements. The Nuclear Energy Agency therefore recommends to improve the accuracy of ^{241}Pu capture and fission cross sections from 0.1 eV to ~1 MeV in its High Priority Request List (HPRL) for nuclear data [2]. The requested accuracy for capture is as low as 2% - 3% at low energy for the current power reactors (PWR, EPR) and between 11% and 14% in the fast range for innovative Sodium Fast Reactors (SFR), see Table 1. The current uncertainties of the capture cross section in the evaluated libraries are about 10% up to 25 keV and increasing with energy up to 20%-30% at 500 keV depending on the library. Therefore, the NEA request implies a challenging reduction of the uncertainties.

Table 1 – Summary of ^{241}Pu(n,γ) cross section accuracies requested in the HPRL [2]

(n,γ) cross section	Thermal reactors		Fast reactors
Energy range	EPR, PWR	VHTR	SFR
0.10 – 0.54 eV	3%	2%	
183 keV – 1.35 MeV			11% - 14%

The fission cross section of ^{241}Pu is at least three times larger than the capture cross section (Figure 2). In order to perform an accurate measurement of the capture reaction it is thus necessary to detect gamma-rays from both capture and fission reactions while identifying fission events independently. In the proposed experiment, gammas from the (n,γ) and (n,f) reactions are detected by the 4π Total Absorption Calorimeter (TAC) while fission events are identified by a dedicated fission chamber containing ^{241}Pu samples placed in the centre of the TAC.

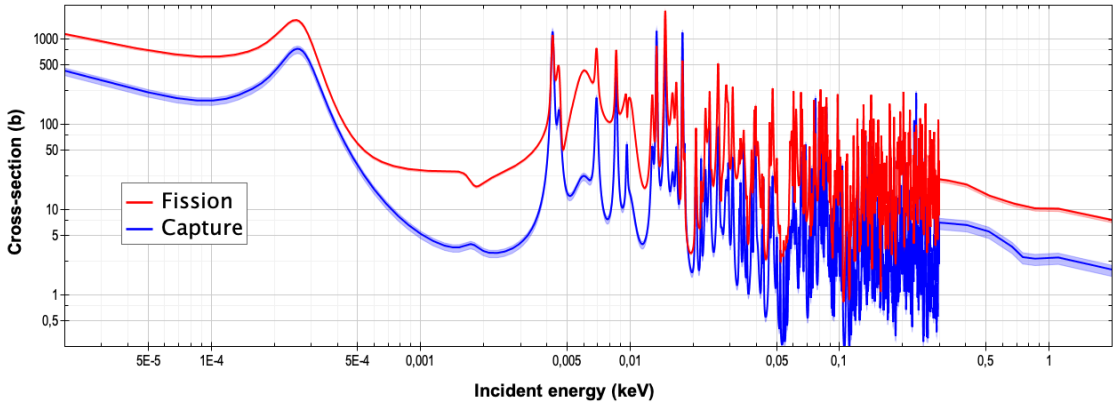

Figure 2 – Capture and fission cross sections of ^{241}Pu from the JEFF-3.3 library [3]

The proposed measurement will benefit of the excellent and unique signal-to-background ratio available at n_TOF for small radioactive samples. The aim is to improve the accuracy of the ^{241}Pu capture cross section up to a few hundreds of eV for which only one measurement is available with an accuracy estimated at ~10% in this energy region [4].

In addition, the n_TOF measurement will provide information on the gamma-ray spectra and multiplicities separately for the fission and capture reactions. Further analysis of these gamma-ray spectra will complement the study recently performed on the fertile isotopes of uranium isotopes ^{234}U, ^{236}U, and ^{238}U [5].

Complementary fission measurements at GELINA (JRC-Geel) with the same fission chamber and samples will confirm and improve the accuracy of the fission cross section up to ~1 MeV.

Experimental setup

The TAC [6] has been already used in fission tagging configurations for previous successful capture measurements of fissile actinides at n_TOF, e.g., ^{233}U [7], ^{235}U [8] and ^{239}Pu [9].

The fission chamber that will be used for tagging the fission events will be adapted from the one used in the ^{233}U experiment [10]. The detector is a fast and compact multi-plate fission chamber that may contain up to 14 actinide deposits of about 1-2 mg each. Its fast response allows to deal with high activities (> 1MBq per sample) in order to avoid signals pile-up. It is compact enough (9 cm-diameter x 12 cm) to be placed in the (scattered-)neutron absorber at the centre of the TAC (Figure 3). The new version of that fission chamber will be more compact (half as many samples) and further optimized for high count rates (faster electronics) and low background (less material and less gas in the beam).

The preparation of the ^{241}Pu samples is challenging because of the radioactivity of ^{241}Pu and ^{241}Am, and the difficulty to find enriched material with low contamination from other Pu isotopes. The samples will be prepared and characterized at JRC-Geel using legacy material enriched in the 80's. The raw material needs to be chemically processed to remove the ^{241}Am and ^{237}Np daughter nuclides (see estimations in Table 2 assuming a reduction by a factor 1000). The plutonium will be deposited as a 4 cm-diameter active spot (i.e., 12.566 cm^2) on 10 µm-thick aluminium backings using molecular plating. It is planned to make 6 to 8 samples with deposits of ~150-200 µg/cm^2 of plutonium for a total mass of about 10 mg of ^{241}Pu. Preliminary characterisation and purification tests have been performed and will continue in 2024. The actual target preparation will start early 2025 after a last Am purification. The measurements must start as soon as possible after the samples are ready in 2025 and before the ^{241}Am build-up becomes an issue, first because of the large ^{241}Am capture cross section, which is about 10 times larger than ^{241}Pu at 0.3 eV, and second because of its contribution to the α background in the fission chamber.

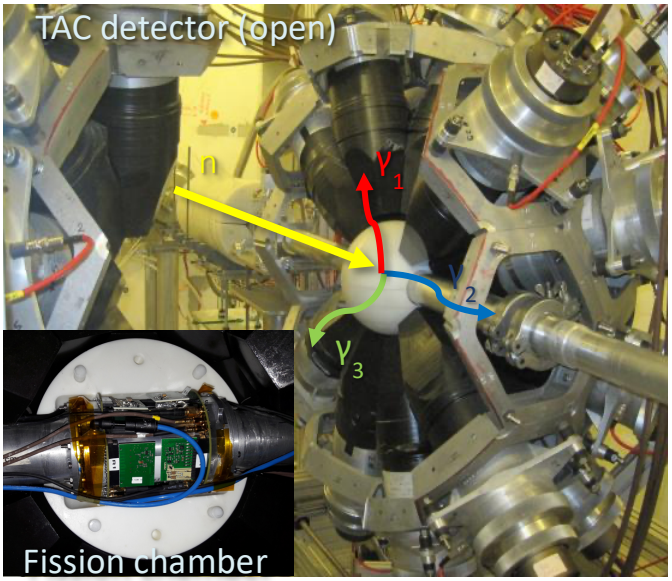

Figure 3 – Picture of the TAC (opened) with the neutron absorber (white sphere) containing the fission chamber (shown in the inset) [7]

Table 2 – Sample composition measured in 1985 and calculated in 2025 after Am and Np purification. The values calculated in 2026 illustrate the ^{241}Am build-up.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{244}Pu	^{241}Am	^{237}Np
1985	0.02%	0.87%	3.38%	**93.22%**	2.51%	0.001%	0%	0%
2025	0.07%	4.3%	16.6%	**66.3%**	12.4%	0.005%	0.4%	0.02%
2026	0.07%	4.3%	16.6%	**63.3%**	12.4%	0.005%	~3%	0.02%

The ^{241}Pu essentially decays via beta radioactivity (99.998%) with a Q-value of 20.82 keV that allows to populate the ^{241}Am ground state only (the first level is at 41.18 keV), thus without gamma emission. The alpha-background is dominated by the contributions from plutonium isotopes (mainly ^{238}Pu and ^{240}Pu) for a total activity of ~2 MBq (per mg of plutonium), and of course by ^{241}Am after a few months when the build-up exceeds 1% (Table 3). For ~10 mg of plutonium the alpha-background will be similar to the one we had in the ^{233}U measurement [10] and thus under control if the ^{241}Am build-up stays low enough. This is a strong constraint for completing the measurement as soon as possible after the last Am purification, ideally within a couple of months.

Table 3 – Alpha-background contributions from a 1 mg plutonium sample (composition calculated in 2025 four months after the last Am purification leading to the production of 1% of fresh ^{241}Am)

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{244}Pu	^{241}Am	**Total**
Atom %	0.07	4.3	16.6	65.3	12.4	0.005	~1	~100
α-activity (MBq) /mg of Pu	0.43	0.10	1.40	0.06	0.02	< 0.01	~1.3	~3.3

The samples will be mounted in the fission chamber at JRC-Geel and the complete "experiment-ready" detector shipped to CERN for data taking after a first test at JRC-Geel. The n_TOF measurement must be completed within a few months after the last Am purification in order to keep the Am-241 build-up below 2%. Figure 4 shows the capture cross sections from JEFF-3.3 [3] scaled to the sample content expected at the start of the measurement. The contributions from ^{238}Pu and ^{244}Pu are negligible. However, the contributions from the ^{241}Am build-up and from the other isotopes of plutonium (^{239,240,242}Pu) will have to be carefully accounted for to deduce the ^{241}Pu capture yield. These contributions will be subtracted either by pure (evaluation-based) simulation or by scaling the simulated yields on a few isolated resonances clearly identified. The contributions from ^{240}Pu and ^{242}Pu will be subtracted thanks to their large and isolated first resonance. The additional information from the gamma-ray energies provided by the TAC will help discriminate between the different contributions, especially between ^{241}Am (and ^{237}Np if any) on one side and ^{241}Pu (and ^{239}Pu) on the other side as their binding energies differ by ~0.8 MeV (Table 4).

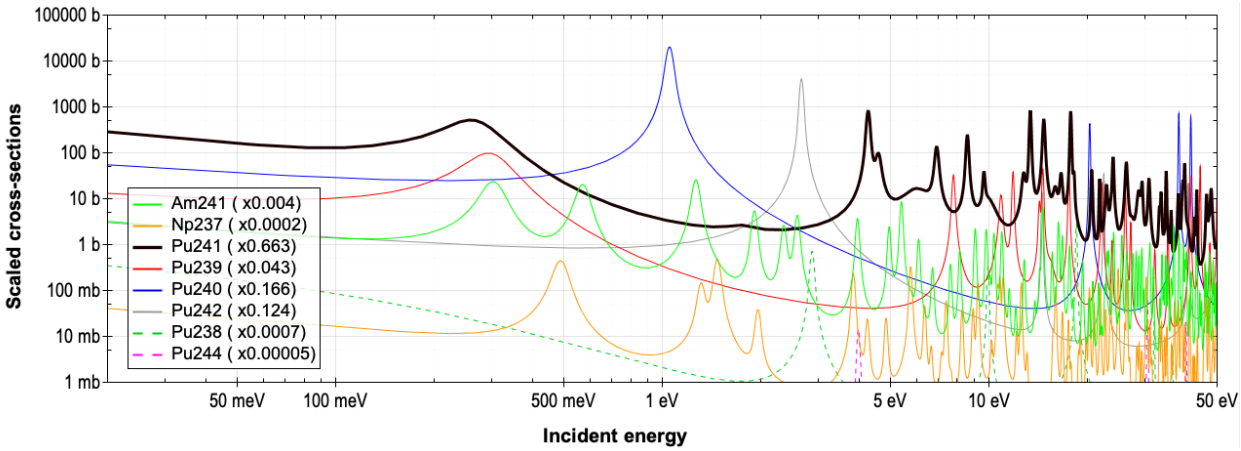

Figure 4 – Capture cross sections scaled to the sample composition calculated in 2025 after Am and Np purification

Table 4 – Binding energies of ^{241}Am, ^{237}Np and the main Pu isotopes

	^{241}Am + n	**^{237}Np + n**	**^{239}Pu + n**	**^{240}Pu + n**	**^{241}Pu + n**	**^{242}Pu + n**
S_n (MeV)	5.54	5.49	6.53	5.24	**6.31**	5.03

Beam request

The measurements should be carefully scheduled to minimize the ^{241}Am build-up in the plutonium samples. Several configurations will be used:

- ^{241}Pu fission chamber in the TAC for the main measurement
- C sample in the TAC for measuring the background generated in the TAC by neutrons scattered on the samples
- Dummy fission chamber in the TAC for measuring the background generated in the TAC by the neutrons scattered on in-beam materials (except samples)
- Au and Ir samples in the TAC for normalizing the capture yields to saturated resonances

Additional configurations will be measured during periods without beam:

- Beam-off background measurement with and without the ^{241}Pu fission chamber
- Calibration measurements with gamma-ray sources

Figure 5 shows the expected number of counts per bin and per bunch of 7 x 10^{12} protons in the TAC induced by ^{241}Pu capture (in blue), fission (in red) and elastic scattering (in green) reactions. The corresponding cross sections from the ENDF/B-VIII.0 library [11] have been used together with the following assumptions:

- The samples consist of a total of 10 mg of ^{241}Pu deposited on a 4 cm diameter spot.
- The beam interception factor is estimated to be 95% (for a beam diameter of 3.5 cm).
- The number of protons per bunch is 7 x 10^{12}.
- The fission chamber efficiency is 90%.
- TAC events with multiplicity higher than 2 are selected, which implies the following efficiencies: 60% for events from capture, 70% for events from fission, 1% for events from scattered neutrons.

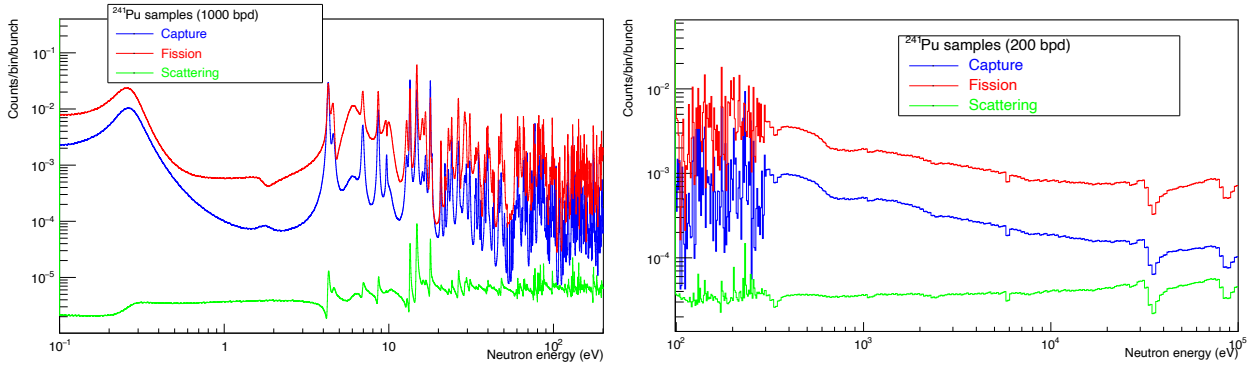

Figure 5 – Count rate estimates based on ^{241}Pu cross sections from ENDF/B-VIII.0 assuming standard bunches of 7 x 10^{12} protons. The plot on the left is a zoom on 1000 bpd results between 0.1 eV and 200 eV. The plot on the right is a zoom on 200 bpd results between 100 eV and 100 keV.

The most constraining energy region in terms of counting statistics and target accuracy is the resolved resonance range from a few eV to a few tens of eV. The expected number of ^{241}Pu capture events per bin for 4 x 10^{18} protons (5.7 x 10^5 bunches) are summarized in Table 5 from 0.1 eV up to 10 keV using 1000 bins per decade (bpd) in the resonance region up to 100 eV and 200 bpd beyond. These estimations show that 4 x 10^{18} protons for the configuration with the Pu-241 fission chamber in the TAC will provide enough counting statistics to allow reaching the target accuracies providing the systematic uncertainties are well under control for subtracting contributions from background and contaminants.

Table 5 – Estimation of the average number of ^{241}Pu capture events per bin for 4 x 10^{18} protons

Energy (eV)	Binning (bpd)	Average number of capture counts/bin	Statistical uncertainty (%)
10^{-1} – 1	1000	1709	2.4*
1 – 10	1000	486	4.5*
10 – 10^2	1000	514	4.4
10^2 – 10^3	200	514	4.4
10^3 – 10^4	200	183	7.4

* Outside the energy range (from 0.5 eV to 3 eV) where the total counting rate will be dominated by the first large resonances of ^{240}Pu and ^{242}Pu

The background generated in the TAC by neutrons scattered on the samples is at least ten times lower than the capture signal below 10 keV (Figure 5). Accordingly, a statistical uncertainty of 10% on a 100 bpd binning is enough to determine this contribution using a graphite sample. The number of protons for that is estimated to be 0.5 x 10^{18}.

The measurement of the background generated in the TAC by neutrons scattered on in-beam materials (except samples) will require another 0.5 x 10^{18} protons for the measurement using a dummy chamber (without samples).

Finally, 0.2 x 10^{18} protons are necessary for the normalization measurements using ^{nat}Ir (saturated resonances at 0.65 eV and 1.3 eV) and Au (saturated resonance at 4.9 eV) samples.

The alignment and verification of the setup collimation will require 0.1 x 10^{18} protons.

The requested protons for measuring these configurations are summarized in Table 6.

Summary

We propose to measure simultaneously the neutron capture and fission reactions of ^{241}Pu at the n_TOF Experimental area 1 (EAR1). This measurement will provide the neutron capture cross section and the capture-to-fission ratio as a function of neutron energy up to a few hundreds of eV. The goal of the measurement is to reach a total uncertainty better than 3% on the capture-to-fission ratio as a breakthrough to support future evaluation work. These new data combined to the fission cross section measurement performed at JRC-Geel will be decisive to fulfil the NEA HPRL request at low energy. In addition, the n_TOF measurement will provide information on the gamma-ray spectra and multiplicities separately for the ^{241}Pu fission and capture reactions. The measurements will be carried out with a set-up that combines the 4π BaF_2 Total Absorption Calorimeter and a dedicated fission chamber containing samples prepared and characterized at JRC-Geel. Considering the sample mass and the targeted uncertainties a total of 5.3 x 10^{18} protons is requested to perform this experiment in EAR1 in 2025.

Table 6 – Summary of requested protons for the Pu-241 capture/fission measurements at EAR1

Measurement	Sample	Number of protons
Pu-241 capture and fission	^{241}Pu fission chamber	4.0 x 10^{18}
Background from neutron scattering on the sample	C	0.5 x 10^{18}
Background induced by the fission chamber	Dummy fission chamber	0.5 x 10^{18}
Normalization	^{nat}Ir, Au	0.2 x 10^{18}
Alignment of the setup	-	0.1 x 10^{18}
Total		**5.3 x 10^{18}**

References:

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
If relevant, write here the name of the fixed installation you will be using TAC	☒ To be used without any modification ☐ To be modified
If relevant, describe here the name of the flexible/transported equipment you will bring to CERN from your Institute Fission chamber + samples	☐ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing
[Part 2 experiment/ equipment]	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[insert lines if needed]	

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m^3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]

	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			