EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Beta-delayed neutrons from oriented ^{137,139}I and ^{87,89}Br nuclei.

09/23/2013

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Abstract

We propose a world-first measurement of the angular distribution of beta-delayed neutrons and gamma radiation from oriented $^{137,\,139}$I and ^{87,89}Br nuclei, polarised at low temperature at the NICOLE facility. Beta-delayed neutron emission is an increasingly important decay mechanism as the drip line is approached and its detailed understanding is essential to phenomena as fundamental as the r-process and practical as the safe operation of nuclear power reactors. The experiments offer sensitive tests of theoretical input concerning the allowed and first-forbidden beta-decay strength, the spin-density of neutron emitting states and the partial wave barrier penetration as a function of nuclear deformation. In ^{137}I and ^{87}Br the decay feeds predominantly the ground state of the daughters ^{136}Xe and ^{86}Kr whereas in ^{139}I and ^{89}Br we will explore the use of n-γ coincidence to study neutron transitions to the first and second excited states in the daughters ^{138}Xe and ^{88}Kr. These measurements will use the new "Versatile Array of Neutron Detectors at Low Energy" (VANDLE) and high purity Ge detectors for neutron-gamma coincidence and monitoring of the nuclear orientation experiment. This instrument had its first use in on-line experiments using fission fragments at HRIBF and in direct reaction studies at NSCL and Notre Dame University. The experiments described in this proposal have direct relevance to a recently initiated Coordinated Research Project (CRP) at the International Atomic Energy Agency (IAEA) in Vienna on the development of a "Reference Database for Beta-delayed Neutron Emission Evaluation".

Requested shifts: 34 shifts, 17 in 2014 and 17 in 2015

1. Introduction:

Particle emission, including the β-delayed processes, is basic to physics as the drip-lines are approached. Importantly, all the r-process nuclei are delayed neutron emitters and any nuclear model used in astrophysical simulations must account correctly for beta-delayed neutron (βn)-emission.

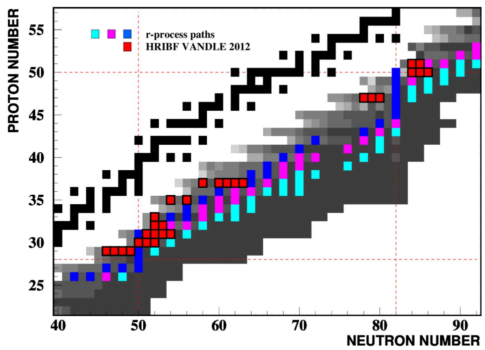

Figure 1 Summary of βn emission for nuclei Z>40 as predicted in FRDM+QRPA [1]. Various r-process paths (blue-pink squares) were measured by VANDLE at HRIBF [3].

As we move further from stability the decay window Q_β-B_n increases and βn-emission becomes a dominant decay mode, sensitive to properties of precursor, parent and daughter nuclei [1,2]. It depends on the β-decay properties of the precursor nucleus; the β-decay strength function and the relative strength of first forbidden and allowed components. It involves properties of the emitter nucleus including deformation, level spin density distribution, partial-wave barrier penetration and branching of neutron-gamma emission from the excited parent states which finally determine the strength and partial-wave make-up of the neutron emission to states in the daughter.

Conventional spectroscopy provides over-all information concerning energies, level structures and intensities, however there are severe limitations in establishing details of the processes, the contributions of different partial waves and the structure of the nuclei involved, their deformation and wave-function make-up.

Angular distribution studies offer a hitherto unexploited measurable with clear sensitivity to variables such as spin and parity of the levels concerned and the angular momenta (partial waves) involved in the emission. A continuous emission spectrum is sensitive to the density of states as a function of spin. When, as in nuclear orientation, the angular distribution is measured with controlled, large and variable degree of nuclear polarisation, these sensitivities are enhanced.

We propose a world-first measurement of angular distribution of beta-delayed neutrons from oriented $^{137,\ 139}$I and ^{87,89}Br nuclei, oriented at low temperature at the NICOLE facility. The aim of this pilot experiment is to make direct observation of the magnitude and sign of the departure from spherical symmetry of the continuous βn spectrum and establish whether the observed distribution agrees with predictions of the models used in the following particulars:

- Do the average sign and magnitude of the anisotropy at a given neutron energy agree with theory?
- Does the average anisotropy demonstrate the predicted dependence upon neutron energy?

For this purpose we will first use oriented precursors ^{137}I and ^{87}Br and study simple neutron emission from ^{137}Xe and ^{87}Kr which feeds solely the 0^+ ground state of ^{136}Xe and ^{86}Kr daughter nuclei.

The experiment will also measure the anisotropy of the competing γ transitions in the emitter nucleus, gaining valuable direct information as to their multipolarity and the neutron emitting levels.

In a second experiment we will explore the viability of including n-γ coincidences to separately study neutron transitions to the ground, first and second excited states in the daughters ^{138}Xe and ^{88}Kr of emitters ^{139}Xe and ^{89}Kr, following the β-decay of oriented ^{139}I and ^{89}Kr. Angular distribution of the neutron transitions, measured in this way, would provide an important test of the theoretical predictions of the partial wave composition of the neutron waves in dependence on their energy and the spins of the initial and final states.

The experiments described in this proposal have direct relevance to a recently initiated Coordinated Research Project (CRP) at the International Atomic Energy Agency (IAEA) in Vienna on the development of a "Reference Database for Beta-delayed Neutron Emission Evaluation". The details of the CRP can be found on IAEA's webpage: http://www-nds.iaea.org/beta-delayed-neutron/. The IAEA-CRP started formally in August 2013 for duration of four years. Primary objective of this CRP is to create a reference database for microscopic (P_n, $T_{1/2}$, neutron spectra, etc.) and macroscopic (or aggregate) quantities, the former for applications in nuclear structure physics and r-process in nuclear astrophysics, and the latter for reactor technology. In the decay of βn precursors, it is often the case that ground-state spin-parities are not firmly assigned, as applies to ^{89}Br, ^{137}I and ^{139}I in the proposed experiments. The spins and parities of the neutron decaying states (or neutron resonances) are known poorly almost in all cases. Novel angular distribution measurements described in this proposal should make it possible to acquire knowledge about the hitherto tentative or unknown quantum properties of the levels and radiations involved in βn decays.

2. Theoretical Background:

The process of βn-emission is illustrated in Figure 2 for the case of ^{137}I. In medium and heavy nuclei this process is usually described by a statistical model, outlined below.

From a precursor nuclear ground state of spin J, atomic number Z, Gamow-Teller (GT), allowed β-decay is assumed, feeding states in the neutron emitting parent having spins J_i = J, J+1, J-1. The spectrum of populated states in the neutron emitter is determined by the absolute β intensity per MeV of levels at the energy E^*. This density is calculated from the β-decay strength function [4] and the statistical rate function [4,5]. Here Q_β is the maximum energy available in the beta-decay. E_i^* is the energy of the neutron emitting state $E_i^* = B_n + E_i^f + E_n A/(A+1)$ where E_n is the kinetic energy of the neutron.

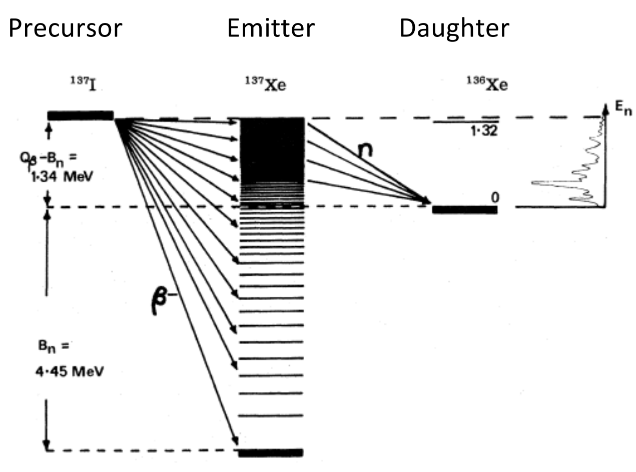

Figure 2. Schematic β-delayed neutron emission from ^{137}I. Latest values of Q_β=6.027 MeV, B_n=4.026 MeV and $Q\beta$-B_n = 2.001 MeV http://www.nndc.bnl.gov/ensdf/

B_n and A are the binding energy of the neutron in the emitter, and its mass. E_i^f is the energy of the i^{th} final state in the daughter nucleus.

The decay from each neutron emitting state is a competition between neutron and γ-ray emission. We assume E1 γ-rays and calculate the γ-decay width adopting a model by Hardy [6]. The n-γ branching ratio is calculated by estimating the partial neutron width in terms of the transmission coefficients [7], dependent on neutron partial waves L allowed by angular momentum selection rules and the density of states at the energy and spin of the neutron emitting state. The total neutron emission probability P_n and the intensity $I_n^{if}(E_n)$ of emitting neutrons from the initial state *i* to the ground and excited states *f* of the daughter nucleus can then be estimated.

For a specific case of an even-even daughter nucleus with lowest states 0+, 2+ and 4+ the expression for the energy dependence of the intensity of neutron emission reads

$$I_n^{warm}(E_n) = \sum_m \omega(J,m) I_\beta(E^*) \left[I_n^m(E_n L, 1/2) + \sum_{L,I_c} I_n^m(E_n L, I_c) \right] \quad (1)$$

in terms of partial waves and channel spin I_c, the vector sum of the particle spin and the spin of the final state. Summation is made over all allowed L and I_c combinations. m denotes the emitter spin J_i and ω is their statistical weight. The first term in the square bracket is for the 0^+ ground state and the channel spin I_c=1/2. In this case there is only one L value to consider, whilst the excited daughter states have non-zero spin allowing two values of the channel spin I_c and a range of neutron L values. The superscript "warm" indicates that no orientation of the precursor is included. We follow the usual assumption of an incoherent sum of neutron partial waves with different values of L. This is based on the presence of a large number of unresolved transitions for which phase sensitivity on average cancels.

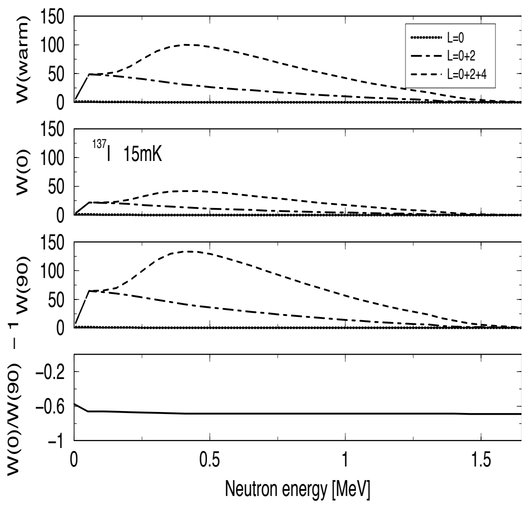

Figure 3 Prediction of angular anisotropy of neutron emission from oriented ^{137}I

The orientation of the long-lived precursor β-emitter can be described by the usual B_λ orientation parameters [9]. The change in orientation produced by the β-decay is included through the de-orientation parameters U_λ. Each neutron decay path has an angular property, which is described using the R_λ of Satchler [10], which depend upon the spins of the states involved, the partial wave(s) L and the spin of the emitted particle. Summations are made over all allowed L and I_c combinations. The angular distribution is given as

$$I_n^{cold}(E_n,T,\theta) = \sum_{\lambda=0}^{\lambda_{max}} B_\lambda R_\lambda Q_\lambda \left\{ \sum_m U_\lambda(m) \omega(J,m) I_\beta(E^*) \left[I_n^m(E_n, L, \frac{1}{2}) + \sum_{L,I_c} I_n^m(E_n, L, I_c) \right] \right\} P_\lambda(\cos\theta) \quad [2]$$

Here, λ is even, T is the absolute temperature and θ is the angle of observation. $P_\lambda(\cos\theta)$ are Legendre polynomials.

The anisotropy of the angular distribution is defined by the ratio of cold to warm intensities at a given angle and temperature. We show in Fig. 3 predicted anisotropies for βn from ^{137}I at the temperature of 15 mK. W(warm), W(0^0) and W(90^0) denote normalized neutron intensities defined above at 1K, 15mk and 0^0 and 15 mk 90^0, respectively. Similar results for the other proposed nuclei will be shown.

3. Nuclear orientation experiment

The beams of ^{137,139}I, ^{87,89}Br will be implanted into an iron foil at the cold finger of the NICOLE dilution refrigerator and cooled down to temperatures ~15 mK (cold). Spectra will be also taken at 1K (warm).

The contribution of different neutron partial waves to the total observed anisotropy is a function of the degree of orientation, i.e. of sample temperature, through the B_λ factors.

Basic properties of the proposed isotopes are summarized in Table I.

Table I. Precursors of β emitters:*

Isotope	$T_{1/2}$ (s)	T_1 (s)	I^π	P_n [%]	Isotope	$T_{1/2}$ (s)	T_1 (s)	I^π	P_n [%]
^{137}I	24.5	3	$(7/2^+)$	7.1	^{87}Br	55.6	2	$(3/2^-,5/2^-)$	2.6
^{139}I	2.28	3	$(7/2^+)$	10.0	^{89}Br	4.36	2	$(3/2^-,5/2^-)$	13.8

*T_1 is spin-lattice relaxation time at 15 mK which must be <~ $T_{1/2}$ for a successful OLNO experiment. The P_n values were taken from http://www.nndc.bnl.gov/ensdf/ .

^{137}I: This isotope has a well-estimated magnetic moment close to 3 n.m. (compare $^{129-135}$I) and a strong hyperfine interaction of 115T in iron so that degrees of nuclear polarisation in excess of 80% are accessible at NICOLE on-line temperatures. Nuclear spin lattice relaxation has been observed in other 7/2+ iodine isotopes and is much shorter than the ^{137}I half-life even at the lowest temperatures.

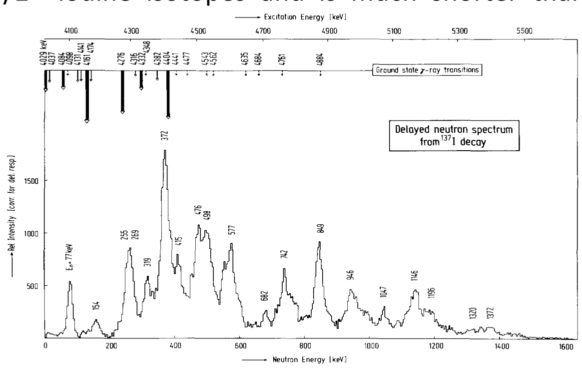

Figure 4 βn spectrum from ^{137}I [11]

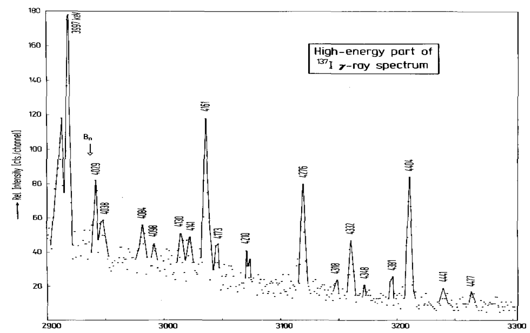

Figure 5 γ-ray spectrum from ^{137}I [11]

The βn decay scheme is shown in Figure 1 and the neutron and (partial) γ-ray spectra [11] in Figures 4,5. β-decay takes place to states in the ^{137}Xe which decay either entirely by γ emission to lower states or, above the energy required to give access to the ground state of the daughter ^{136}Xe, in competition with neutron emission which makes up 7.1% of the total decay rate. This case is simple in that very close to all neutron transitions feed the ground state of the daughter.

Allowed GT beta decay of the $(7/2^+)$ ^{137}I feeds positive parity states of spin 5/2,7/2,9/2 in the emitter ^{137}Xe which exhibit γ-decay dominated by E1 transitions to lower states (over 50 high energy E1 transitions have been identified [12]). There is the possibility that there is also considerable first forbidden β strength. The experiment will give evidence on the relative allowed/first forbidden β strength from the anisotropy, and hence multipolarity, of the observed γ-transitions. Strong forbidden β strength will also show up in the presence of unpredicted neutron partial waves. Above the threshold, neutron emission to daughter states in ^{136}Xe competes with γ decay. Assuming the density of neutron emitting spin states as a function of neutron energy, given by a specific model, direct prediction of the average neutron anisotropy at any particular neutron energy and its variation with neutron energy is available (see section 2). These can both be compared with experimentally determined neutron anisotropies.

^{87}Br: The second accessible experiment involves orientation of the precursor ^{87}Br which β-decays to the emitter ^{87}Kr, feeding the daughter ^{86}Kr by neutron emission. Neutrons form 2.6% of the total decay rate. The physics variables are the same as the first example, however the two emitters have very different nuclear deformation, 0.24 for ^{137}Xe and 0.06 for ^{87}Kr. This difference is expected to influence the competing neutron partial wave amplitudes. The spins involved for the Br decay are ^{87}Br (3/2,5/2) states in ^{87}Kr 1/2, 3/2, 5/2 whilst in the daughter the ground state has spin 0 as for ^{136}Xe. The neutron and γ-ray warm spectra were measured by Nuh et al [13] and discussed by Kratz et al [14].

^{139}I and ^{89}Br. Although they are expected to have similarly favourable hyperfine interaction strength as the lighter isotopes, polarisation of these isotopes is somewhat more difficult because, at the lowest temperatures, their half-lives are comparable to their relaxation time. Complete thermal equilibrium may not be achieved. However, the degree of orientation obtained can be directly estimated from the observed E1 transition anisotropies. Their study is important because they exhibit considerable feeding to excited states in the daughter nuclei ^{138}Xe and ^{88}Kr. An advantage is their higher neutron yield (see Table I). Neutron spectra, measured with limited energy resolution, were reported by Rudstam and Shalev [15].

4. Detectors and data acquisition system

These measurements will use the "Versatile Array of Neutron Detectors at Low Energy" (VANDLE) [16,17] and high purity Ge detectors for neutron-gamma coincidence and monitoring of the nuclear orientation experiment. This instrument was recently commissioned and used in on-line experiments using fission fragments at HRIBF and used in direct reaction experiments at NSCL and Notre Dame University. VANDLE is capable of detecting neutrons over a wide range of energies (100 keV to 6 MeV) due to the innovative use of digital electronics. Because of its modular geometry it is also easily reconfigurable and portable. In this experiment we will use forty eight 60x3x3 cm^3 plastic scintillator bars placed around the NICOLE implantation spot in cylindrical geometry at 50 cm distance. Small plastic detectors with silicon photomultipliers for light readout will be employed as time-of-flight start detectors and VANDLE bars will detect neutrons and provide "stop" signals. Two high-efficiency HPGe detectors will be used for β-n-γ coincidences. This allows measurement of the gamma transition anisotropies in the daughter nucleus as well as in the emitter. Temperatures will be measured using a standard nuclear orientation thermometer.

The intrinsic efficiency of one VANDLE module was obtained using the (d,n) calibrated neutron source at Ohio University [18]. For the typical energies in beta-delayed neutron emission, 100-2000 keV, the efficiency range is in between 5% at 100 keV to 50% at 1 MeV. The angular acceptance with 48 bars in this configuration will be ~20% of 4π, giving an array efficiency of 1-10% in the energy range of interest. The efficiency of beta detection used as beta trigger is about 5%.

The entire system is instrumented with Pixie-16 digital electronics because they offer benefits of low energy threshold, simplicity, and stability. We have demonstrated that with a 250 MHz digitization rate we can obtain the sub-nanosecond time resolution required for time-of-flight measurements [19].

The system will be calibrated using ^{8}He beam. We estimate 2 shifts will be needed for the set-up.

5. ISOLDE beams

Bromine and iodine beams virtually free of isobaric contaminants can be produced by negative surface ionization. At ISOLDE-SC beams of ^{87,89}Br and ^{137,139}I with yields of $9x10^5$ to $2.9x10^7$ ions/uC were produced from ThO_2 and UC_x targets with a LaB_6 (MK4) negative surface ion source [20,21]. Our experiment requires a minimum beam intensity of $1x10^5$ ions/s, hence it is readily feasible with established ISOLDE target and ion source technology, even if the on-line yields show to be lower than the book values.

Although we strongly prefer a negative ion source, in case this cannot be scheduled (e.g. incompatibility of negative ions with jointly scheduled experiments), alternative solutions exist. The experiment does not require ultimate beam purity since it has built-in selectivity in the fact that only beta-neutron coincidences will be exploited. Hence, isobaric contaminants that do not emit beta-delayed neutrons are only a concern if they were so intense that they saturated the beta detector thus inducing too many false start signals for the TOF system or give seriously interfering gamma spectra. We estimate that we can run with 20% purity beams. The isobars closer to stability (^{87}Kr, ^{87m}Sr, ^{89}Kr>^{89}Rb, ^{137}Xe, ^{137m}Ba and ^{139}Xe>^{139}Cs>^{139}Ba, respectively) do not emit neutrons and those further from stability (Se/As and Te/Sb, respectively) are weakly produced and/or slowly released. Hence a standard UC_x target with neutron converter (to minimize production of isobars close to stability) and "hot plasma source" (MK5/VD5) should deliver exploitable beams too. On mass 87 and 137 bromine and iodine, respectively, are expected to represent 20-50% of the β activity (scaled from relative yields measured at HRIBF and ALTO). On mass 89 and 139 this would however drop to few percent.

Additional selectivity can be gained by exploiting molecular sidebands such as ^{40}CaBr$^+$ and ^{40}CaI$^+$ [22,23]. Such beams were already observed previously at ISOLDE, namely ^{40}Ca^{90}Br$^+$ contamination on mass 130 [24] due to tiny calcium contaminations in the target material. The intensity of these sidebands could be significantly boosted by adding a ^{40}Ca mass marker to the target. The iodine isotopes would be separated at mass settings (177 and 179) with negligible background of quasi-isobars. Enriched ^{40}Ca should be used to prevent ambiguous combinations such as ^{42}Ca^{x-2}I$^+$ or ^{44}Ca^{x-4}I$^+$.

Thus, such a target and ion source combination would provide sufficiently clean beams at least for ^{87}Br, ^{137}I and ^{139}I. For ^{89}Br the obtainable beam purity (atomic ions vs. molecular ions) needs to be determined experimentally.

Summary of the shifts: 34 shifts in total

We request 30 shifts (15 shifts in 2014 and 15 shifts in 2015) with a ThO_2 (or UC_x) target with MK4 negative surface ionization source. In addition, 2 shifts of 9Li or 8He with warm fridge before the experiment are needed for calibration.

Alternatively at least the first 15 shifts could be made with a UC_x target with neutron converter, ^{40}Ca mass marker and "hot plasma" source MK5/VD5.

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DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: *(name the fixed-ISOLDE installations, as well as flexible elements of the experiment)*

Part of the Choose an item.	Availability	Design and manufacturing
NICOLE	☒ Existing	☒ To be used without any modification
VANDLE	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
Ge detector	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

Hazards named in the document relevant for the fixed NICOLE installation.

Additional hazards:

Hazards	*[Part 1 of the experiment/equipment]*	*[Part 2 of the experiment/equipment]*	*[Part 3 of the experiment/equipment]*
Thermodynamic and fluidic			
Pressure	[pressure][Bar], [volume][l]		
Vacuum			
Temperature	[K]		
Heat transfer			
Thermal properties of materials	Cryostat of Ge detectors		
Cryogenic fluid	Liquid nitrogen: 1 bar, 20 l		
Electrical and electromagnetic			
Electricity	[voltage] [V], [current][A]		
Static electricity			
Magnetic field	[magnetic field] [T]		
Batteries	☐		
Capacitors	☐		
Ionizing radiation			
Target material	[material]		
Beam particle type (e, p, ions, etc)	8He, 9Li, 87Br, 89Br, 137I, 139I		
Beam intensity	1E5-1E6 ions/s		
Beam energy	60 keV		
Cooling liquids	[liquid]		

Gases	[gas]		
Calibration sources:	☐		
• Open source	☐		
• Sealed source	☐ [ISO standard]		
• Isotope			
• Activity			
Use of activated material:			
• Description	☐		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope			
• Activity			
Non-ionizing radiation			
Laser			
UV light			
Microwaves (300MHz-30 GHz)			
Radiofrequency (1-300MHz)			
Chemical			
Toxic	[chemical agent], [quantity]		
Harmful	[chemical agent], [quantity]		
CMR (carcinogens, mutagens and substances toxic to reproduction)	[chemical agent], [quantity]		
Corrosive	[chemical agent], [quantity]		
Irritant	[chemical agent], [quantity]		
Flammable	[chemical agent], [quantity]		
Oxidizing	[chemical agent], [quantity]		
Explosiveness	[chemical agent], [quantity]		
Asphyxiant	[chemical agent], [quantity]		
Dangerous for the environment	[chemical agent], [quantity]		
Mechanical			
Physical impact or mechanical energy (moving parts)	[location]		
Mechanical properties (Sharp, rough, slippery)	[location]		
Vibration	[location]		
Vehicles and Means of Transport	[location]		
Noise			
Frequency	[frequency],[Hz]		
Intensity			
Physical			
Confined spaces	[location]		
High workplaces	[location]		
Access to high workplaces	[location]		
Obstructions in passageways	[location]		
Manual handling	[location]		
Poor ergonomics	[location]		

0.1 Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): *(make a rough estimate of the total power consumption of the additional equipment used in the experiment)*

VANDLE power supplies 0.5 kW

VANDLE data acquisition 0.5-1 kW

Data acquisition server 0.5 kW

Total is: 1.5-2 kW