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EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the ^{166,167,168,170}Er(n,γ) cross-section at EAR1

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Abstract:

Research activities on Burnable Absorbers (BA) disclosed that erbium could be considered a valid substitute for gadolinium which is the most extensively used BA in nuclear power plants. Erbium capture cross sections presents several nuclear safety related improvements with respect to Gadolinium such as a minor downgrade of the power distribution and a more negative feedback coefficient. Furthermore, the Erbium capture cross sections play a role in the study of the s-process nucleosynthesis. Despite their importance, the evaluated uncertainty data (ENDF/B-VIII.0) of ^{166,168,170}Er ($>$ 1 eV) are higher than 7%, while the evaluated uncertainty (ENDF/B-VIII.0) of ^{167}Er ($>$ 1 eV) is below 3%, although this value has been proved to be questionable. A Sensitivity and Uncertainty (S&U) analysis

on an erbia doped Fuel Assembly (FA) showed the importance to perform accurate new capture cross sections measurements in order to reduce the erbium isotopes uncertainty contribute on the criticality of this type of FA. The need of an accurate revaluation of ^{167}Er capture cross section has also been confirmed by Nuclear Energy Agency (NEA) which included ^{167}Er measurement in the High Priority Request List (HPRL) database. In order to fulfil the ^{167}Er HPRL requirement, a capture cross section measurement with an erbium natural sample would be performed with liquid scintillator (C_6D_6) detector and Total Absorption Calorimeter (TAC) detectors with the aim to obtain a measurement in the thermal-epithermal region (up to 50 eV) with an uncertainty close to 2%. The capture cross section of erbium isotopes (^{166,167,168,170}Er) in the energy range that present an overlapping of the resonance structures (beyond 50 eV) would be measured by means of a experimental campaign with enriched samples using C_6D_6 detectors.

Requested protons: $9.0{\cdot}10^{18}$ protons on target
Experimental Area: EAR1

1 Introduction and motivation

In the last twenty years, the research activities in the field of burnable absorbers (i.e., material enclosed in a nuclear core with large neutron-absorption cross sections) showed that erbia (Er_2O_3) can be considered as an excellent alternative to gadolinia (Gd_2O_3), currently widely employed in nuclear commercial reactors. In fact, erbium presents lower thermal absorption cross section (minor downgrade of the power distribution), more negative temperature feedback coefficient (enhancement of the reactor core safety), higher and more energy extensive resonance integrals (better control of start-up and accidental transient phases) and a reduction of ^{239}Pu isotope content in an End Of Fuel (EOL) core inventory (enhancement of the non-proliferation actions) [1]. Furthermore, the development of the so-called Erbia Super High Burn-Up (Er-SHB) concept demonstrated that erbium could be directly mixed in all fuel pins of a Fuel Assembly (FA) at Beginning Of Life (BOL) opening the possibility for large-scale production of a higher enriched ($> 5\%$) fuel in the existing nuclear fuel production facilities without reducing the front-end phase nuclear safety of the nuclear fuel cycle [2, 3]. Despite their importance, the natural erbium isotopes (^{166,168,170}Er) capture cross sections exhibits not-negligible (> 7 %) evaluated uncertainty data (ENDF/B-VIII.0) at energy higher than 1 eV. The evaluated uncertainty data (ENDF/B-VIII.0, > 1 eV) of ^{167}Er is set to 2.3%, but this value is questionable and probably underestimated as extensively discussed in [2, 3]. S&U analysis was carried out on some FA configurations to evaluate the impact on the nuclear system criticality of the not-negligible evaluated erbium capture cross sections uncertainties at energy higher than 10 eV. The energy of 10 eV was chosen because beyond this value there is an overlapping of the erbia isotopes resonance structure for which a high-resolution measurement as one attainable at n_TOF is required. Specifically, S&U analysis was performed for an Er-SHB FA at BOL (case 1) and EOL (case 2, 60 GWd/MTU). Figure 1 shows the sensitivity spectra of the erbium isotopes, while Table 1 reports the uncertainty contribution of each capture cross section to the system criticality for neutron energy value major than 10 eV.

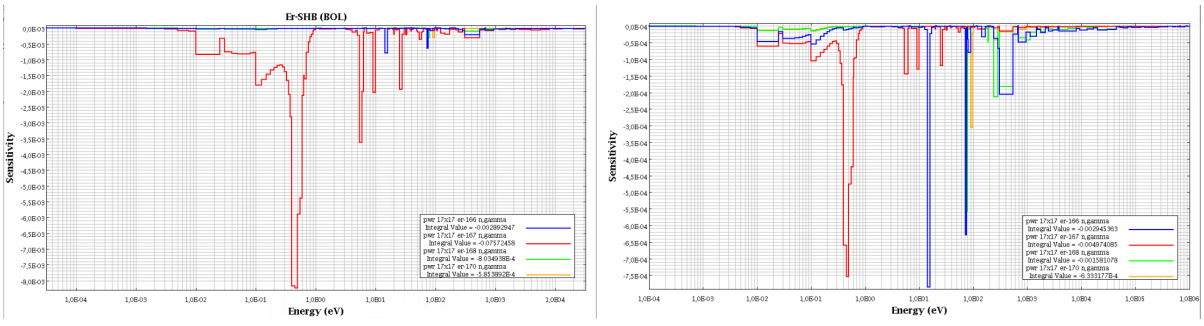

Figure 1: Sensitivity spectra of a Er-SHB FA at BOL (left) and at EOL (right).

Figure 1 shows that the relative sensitivity contributes of the ^{166,167,168}Er isotopes rise with the increase of the neutron spectrum energy (i.e., from BOL to EOL) due to the presence of resonances at higher neutron energy. Table 1 highlights that ^{166}Er and ^{168}Er are the most significant contributor to the criticality uncertainty. At the same time, the contribution of ^{167}Er cancels out at high burnup (case 2) mainly due to the transmutation of the

Isotope	Er-SHB at BOL				Er-SHB at EOL			
	Si	Si/Si,tot	ΔK/K	ΔK	Si	Si/Si,tot	ΔK/K	ΔK
	(-)	(%)	(-)	(pcm)	(-)	(%)	(-)	(pcm)
^{166}Er	-2.31e-03	24.9	1.38e-04	18	-2.28e-03	51.1	1.38e-04	15
^{167}Er	-5.79e-03	62.4	7.08e-05	9	-3.13e-04	7.0	7.08e-05	0
^{168}Er	-7.27e-04	7.8	7.67e-05	10	-1.40e-03	31.4	7.67e-05	17
^{170}Er	-4.49e-04	4. 8	5.37e-05	7	-4.68e-04	10.5	5.37e-05	6

Table 1: Erbium isotopes uncertainty contribution to criticality for a Er-SHB FA at BOL (left) and EOL (right).

^{167}Er into ^{168}Er. A further configuration (case 3) considered is the reference FA (AFA-3G) adopted in Light Water Reactors (LWRs) that use a Mixed uranium and plutonium Oxide (MOX) with an average plutonium content of 8.65% [4]. In this analysis, the average plutonium content was increased up to 12.5% in order to design an innovative system that would improve the fuel cycle efficiency (i.e., better fuel consumption) and extend the fuel cycle length (i.e., reduction of the outage phase), while preventing the void reactivity coefficient becoming positive [5]. The initial reactivity excess due to the increase in the Pu content was downgraded, doping all the fuel pins with erbia (1% at). The left panel of Figure 2 shows the sensitivity spectra. S&U outcomes revealed that the absolute integral sensitivity value of all erbium isotopes is minor that Er-SHB (BOC) system due to the competitor's presence of plutonium isotopes. The isotopes criticality uncertainty contribution was found equal to 74 pcm (^{167}Er), 20 pcm (^{166}Er), 11 pcm (^{168}Er) and 8 pcm (^{170}Er). The overall uncertainty contribution above 10 eV was equal to 12 pcm. The last case considered (case 4) is the open square FA design of the European Lead Fuel Reactor conceptual design (ELSY) with erbia (1% at) as an Integral Burnable Absorber (IBA). The right panel of Figure 2 show the sensitivity spectra. The ELSY FA S&U results showed that ^{167}Er is the isotope with the highest contribution to the criticality uncertainty. The isotope contributions were found to be equal to 46 pcm (^{167}Er), 15 pcm (^{166}Er), 8 pcm (^{168}Er) and 6 pcm (^{170}Er). The sensitivity energy range falls far beyond the 10 eV (i.e., 10^{-2} – 10^{6} keV) due to the cooling vector design choice (lead).

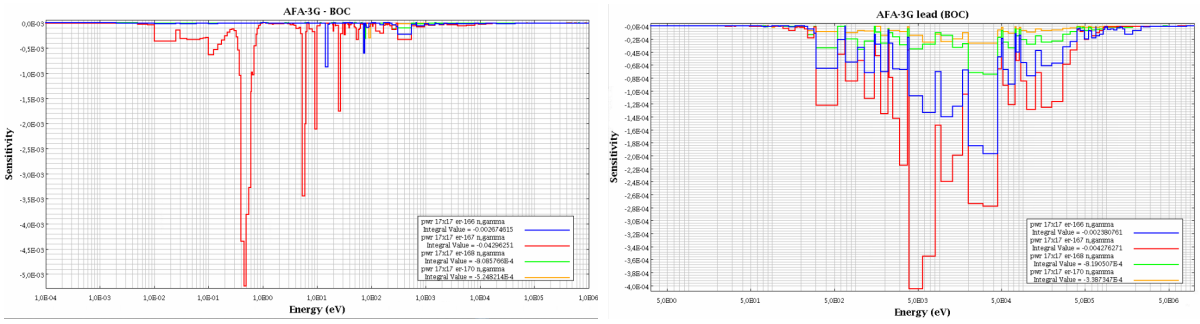

Figure 2: Sensitivity spectra of an AFA-3G (left) and ELSY (right) at BOC.

The four cases analyzed demonstrated that ^{166}Er and ^{168}Er are the most crucial contrib-

utors (case 1, 2) to the criticality uncertainty beyond 10 eV, for which a high-resolution measurement is required. ^{167}Er is the higher criticality uncertainty contributor both below (case 3) and above (case 4) the 10 eV energy threshold. Moreover, despite ^{170}Er provides a minor contribution for all cases studied, it is the isotope with the higher evaluated uncertainty (15.3%). Indeed, its revaluation is also desirable for a better definition of the nuclear safety margin and for a general reduction of the fuel weight in the design cost.
To a minor extent, accurate cross-section data on erbium isotopes could be of interest for studying the s-process nucleosynthesis around the region of rare earth elements with a mass number between 160 and 170. In fact, the abundance of ^{166,167,168,170}Er isotopes in presolar silicon carbide grains measured by Yin and collaborators [6] is in overall disagreement with the calculated abundances; this circumstance clearly calls for new (n,γ) measurements of isotopes involved in this mass region. Apart from astrophysics and nuclear reactor technology, various erbium isotopes are also used in medicine. In particular, there are some studies on the production of ^{171}Er by neutron activation of ^{170}Er [7]. Furthermore, a recent work discusses the production of ^{169}Er from ^{168}Er; this isotope is a receptor target for β-therapy applications [8].

2 Previous measurements and evaluations of Er

Table 2 presents the different experimental measurements performed up to date for obtaining the capture or transmission cross sections of ^{166,167,168,170}Er. The resonance parameters value of ^{166,168,170}Er were obtained from measurements on enriched samples only by Mughabghab [12] and Liou [13], and from measurements on natural samples by Wang [18] and Li [19]. For this reason, the evaluations of JEFF-3.3 [20], JENDL-5 [21] and ENDF-VIII [22] of these isotopes are very similar, the only considerable difference observed is the strong resonance of ^{166}Er at 171 eV included in JENDL-5 but not in the rest of the evaluations. The standard deviation in the capture cross section for ^{166}Er, ^{168}Er, ^{170}Er isotopes in the energy range between 1-3000 eV is 7-15%, while at higher energies these values increase (Figure 3). There are more measurements of ^{167}Er, but new measurements are needed to reduce the uncertainty in the capture cross section [2, 3]. Figure 4 shows that the differences in the radiative kernels ($R_k = g\Gamma_\gamma \cdot \Gamma_n/\Gamma$) for different evaluations are as high as 10% below 100 eV and higher than 20% at higher energies.

3 Er measurement at n_TOF EAR1

We propose to perform a capture campaign for the Er isotopes at EAR1 [24]. In order to fulfil the HPRL requirement of obtaining the capture cross section of ^{167}Er with an uncertainty smaller than 2% at energies below 100 eV, a precisely characterised sample is needed. A natural sample of Erbium (^{162}Er 0.1%,^{164}Er 1.6%,^{166}Er 33.5%,^{167}Er 22.9%,^{168}Er 27.0%,^{170}Er 14.9%) would be used in order to have as low as possible uncertainties in the sample characterisation. The mass of the sample would be $\sim$30 mg in order to have $3.4{\cdot}10^{-5}$ atoms/barn in the sample and a total yield below 0.2, a small yield reduces the self-shielding and multiple interaction effects. Also, in order to reduce the systematic effects two different detectors (see Figure 5) and detection techniques would be used. We

Measurement	Energy range (eV)			
	^{166}Er	^{167}Er	^{168}Er	^{170}Er
Hopkins (1958)(Cap.)[9]	Thermal	Thermal	-	-
Møller (1960)(Tra.)[10]	-	0.4-0.6	-	-
Vertebnyi (1965)(Tra.)[11]	16	Thermal-30	-	95
Mughabghab (1967)(Tra.)[12]	Thermal-600	Thermal-150	Thermal-1.5e3	95
Liou (1972)(Cap./Tra.)[13]	15-1e4	0.4-1.7e3	80-1.5e4	95-2.4e4
Kahane (1984)(Cap.)[14]	-	5-600	-	-
Knopf (1996)(Tra.)[15]	Thermal	Thermal	Thermal	Thermal
Danon (1998)(Tra.)[16]	Thermal-15	Thermal-15	-	-
Harun (2000)(Tra.)[17]	-	1e4-9e4	-	-
Wang (2010)(Tra.)[18]	15-120	0.4-120	80	95
Li (2021)(Cap.)[19]	15-100	0.4-100	80	95

Table 2: List of experimental Er stable isotopes transmission (Tra.) and capture (Cap.) measurements to obtain the neutron cross and their energy ranges.

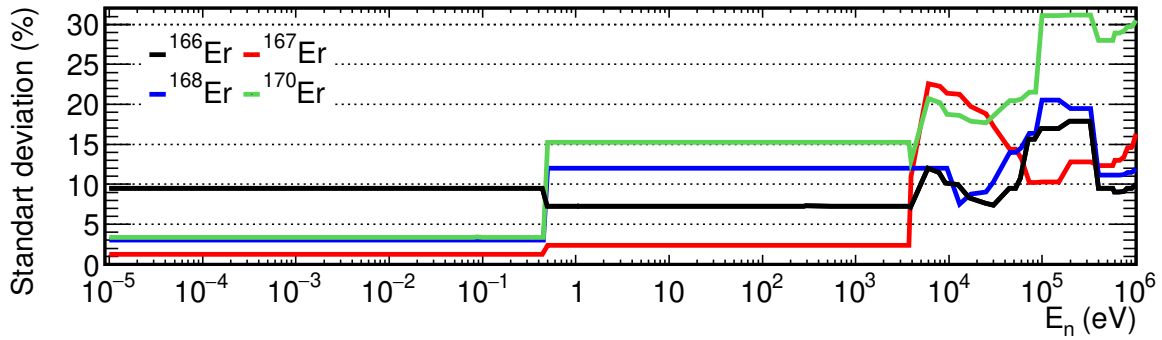

Figure 3: The standard deviation of the capture cross-section of various Er isotopes in the ENDF-VIII [22] library obtained with JANIS [23]

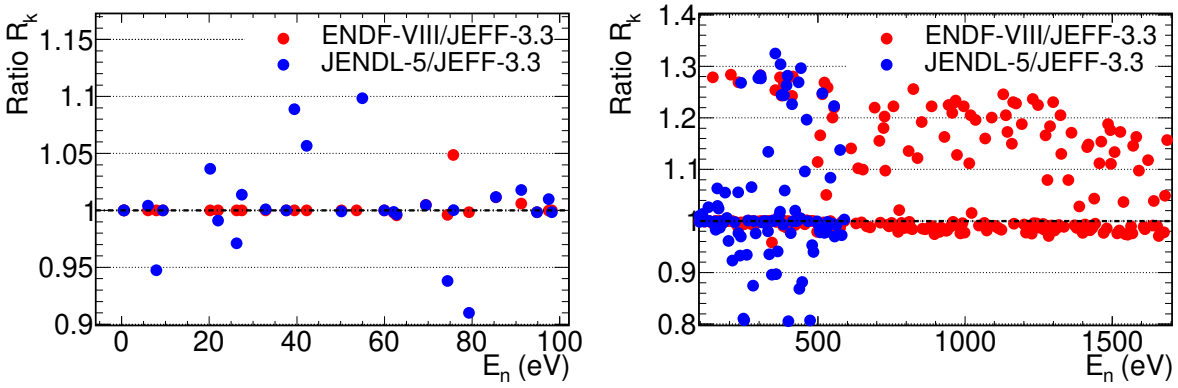

Figure 4: The ratios of the radiative kernels ($\Gamma_\gamma \cdot \Gamma_n/\Gamma$) for the resonances of ^{167}Er for various libraries JEFF-3.3 [20], JENDL-5 [21] and ENDF-VIII [22].

propose to use 3 carbon fibre C_6D_6 detectors [25] at 125º with respect to the beam and a complementary set of 5 sTED detectors [26] at various angles, see left panel of Figure 5, for the determination of the angular dependence of the γ-ray emission of the different s-waves and p-waves resonances of the Erbium isotopes. The total efficiency of the setup is ∼7%. For the analysis the Total Energy Detection (TED) and Pulse Height Weighting Technique (PHWT) [27] would be used. The other detection setup that would be used is the TAC detector [28] with an efficiency of ∼60%, see right panel of Figure 5. ^{167}Er has

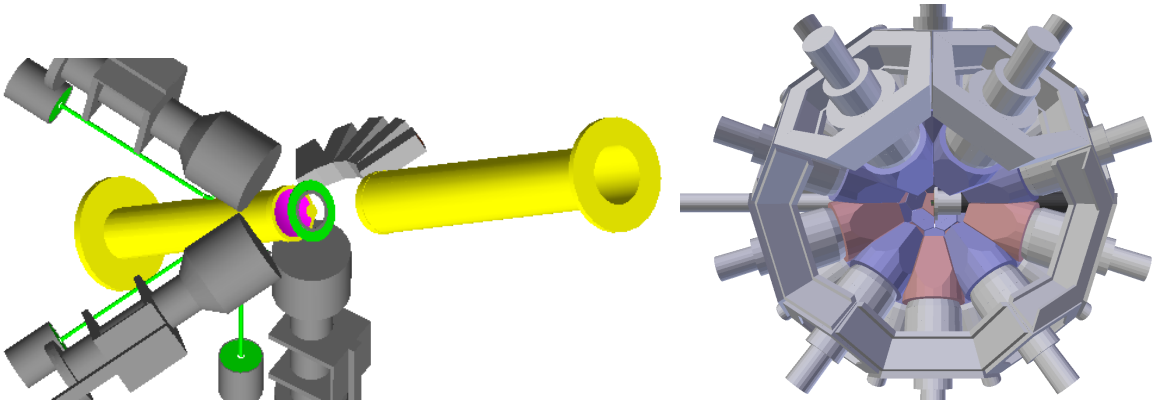

Figure 5: Schema of the setup for the measurements in EAR1 with the C_6D_6 detectors (left) and with the TAC (right).

resonances with two different spins and an isomeric state with a half-life of 109 ns. These effects would need corrections, I. Knapova et al [29, 30] have performed very interesting work on this isotope. The information on the isomeric state, the spin and the decay γ-rays cascades would be taken from this work. In Figure 6, the counting rates estimates for both detectors are presented. The estimated uncertainties due to counting statistics with these protons area lower than 2% with 1000 bins per decade for the majority of the resonances. As observed in the right panels of Figure 6 it is not possible to obtain the capture cross section of ^{167}Er with a low uncertainty using a natural sample at energies higher than 50 eV, due to the presence of other Er resonances.

In order to measure the capture cross section of ^{167}Er at energies higher than 50 eV and the cross sections of ^{166,168,170}Er samples with an isotopic enrichment of ∼97% and a mass of ∼200 mg would be used. The expected count rates presented in Figure 7 are for samples of a 97% enrichment and 1% contaminants of each other Er isotope, for example the ^{166}Er sample has 97% of ^{166}Er, 1% of ^{167}Er, 1% of ^{168}Er and 1% of ^{170}Er. Samples similar to these ones can be found commercially with an estimated price of less than 10 € per mg. The number of protons are estimated to derive capture kernels and average capture cross sections with an uncertainty on the level of a few percent in the RRR. In the Unresolved Resonance Region (URR) a 3% uncertainty due to statistics is obtained with the number of protons given for ^{166,167,168}Er in Table 3 and 100 bins per decade. The ^{170}Er cross section in the URR is lower than in the other Er isotopes and therefore more challenging to measure. The number of protons requested is $9.0{\cdot}10^{18}$ including the beam-on background measurements. Also, the normalization will be done with the saturated resonance method using the 4.9 eV ^{197}Au resonance [31].

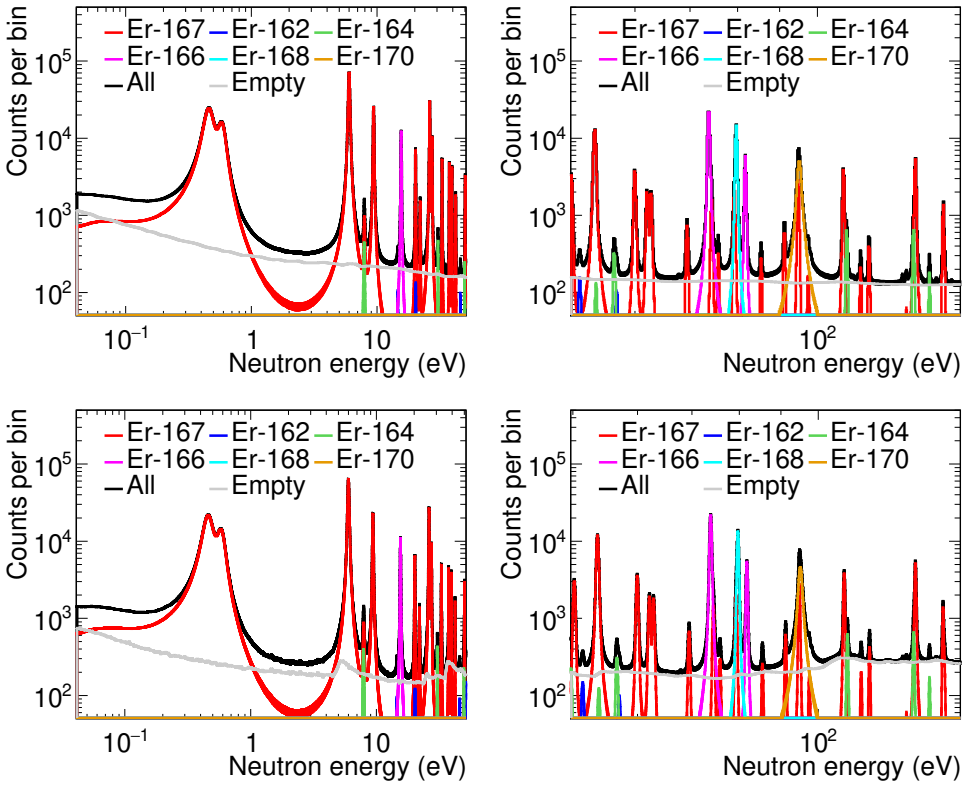

Figure 6: Counting rate estimations for a natural Er sample of 30 mg and 1 cm radius. The number of protons are $1.5{\cdot}10^{18}$ for the C_6D_6 detectors (top) and $1.0{\cdot}10^{18}$ for the TAC (bottom). In the plots the expected counts produced by each Er isotope, the empty beam-on background (Emtpy) and the sum of all the components (All) with 1000 bins per decade.

Sample	Mass	Detector	Energy range	Number protons
Natural	30mg	TAC	0.01-50 eV	$1.0{\cdot}10^{18}$
Natural	30mg	C_6D_6	0.01-50 eV	$1.5{\cdot}10^{18}$
^{166}Er	200mg	C_6D_6	10 eV-100 keV	$1.0{\cdot}10^{18}$
^{167}Er	200mg	C_6D_6	50 eV-500 keV	$1.0{\cdot}10^{18}$
^{168}Er	200mg	C_6D_6	50 eV-100 keV	$1.5{\cdot}10^{18}$
^{170}Er	200mg	C_6D_6	50 eV-50 keV	$1.5{\cdot}10^{18}$
TAC auxiliary and normalization measurements				$0.5{\cdot}10^{18}$
C_6D_6 auxiliary and normalization measurements				$1.0{\cdot}10^{18}$
Total				$9.0{\cdot}10^{18}$

Table 3: Beam time request and distribution.

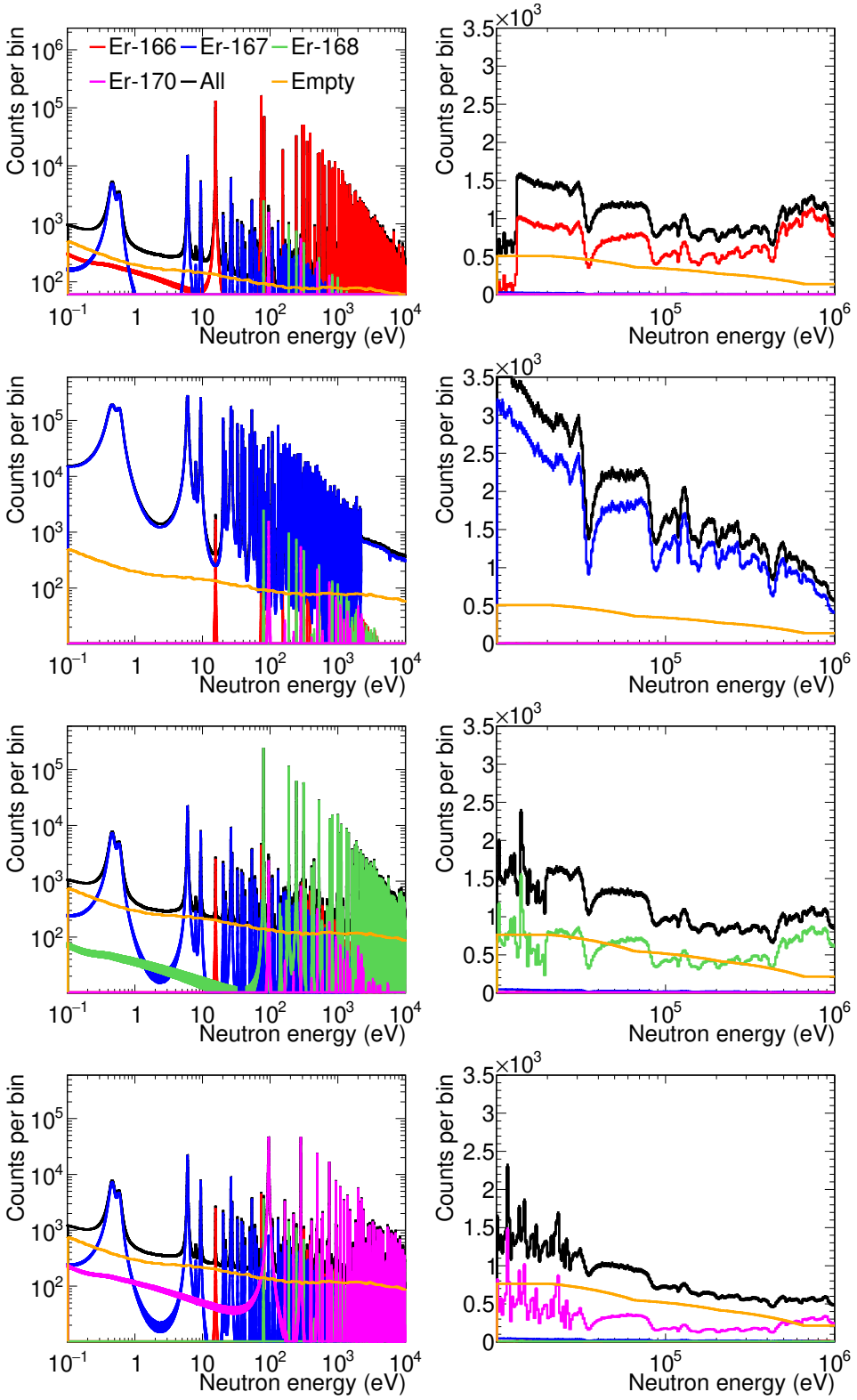

Figure 7: Counting rate estimations for different enriched Er samples of 200 mg and 1 cm radius each using JEFF-3.3. The number of protons for each sample is given in table 3. In the plots the expected counts produced by each Er isotope, the empty beam-on background (Emtpy) and the sum of all the components (All). The left plots are with 1000 bins per decade and the left ones with 100 bins per decade.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Part of the experiment	Design and manufacturing
C6D6 and TAC setup	To be used without any modification
5 stable metallic samples of Er with a total mass of ∼1 g	Standard equipment supplied by a manufacturer

HAZARDS GENERATED BY THE EXPERIMENT
Additional hazard from flexible or transported equipment to the CERN site:
Only various stable samples of Er would be transported to CERN the detectors are already at the n_TOF facility.