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

Neutron capture on ^{50}Cr and ^{53}Cr for criticality safety

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Abstract

In nuclear technology, criticality safety benchmarks allow performing sensitivity analysis of the nuclear data used as input for nuclear reactor calculations. Based on such studies, the Nuclear Energy Agency (NEA) has recently called in its High Priority request List (HPRL) for new measurements on neutron capture of the neutron capture cross section on ^{50,53}Cr isotopes in the 1 to 100 keV energy range, as chromium is a major component of stainless steel. In response to this request, we propose to perform such measurements at the n_TOF EAR1 measuring station with a set of carbon fiber C_6D_6 detectors using high purity targets specially designed to reduce the multiple interaction effects, which are behind the disagreement of previous measurements. The n_TOF experiment will be complemented by transmission and activation campaigns at the GELINA and HISPANOS facilities, respectively.

Requested protons: 4x10^{18} protons on target
Experimental Area: EAR1

NEUTRON CAPTURE ON ^{50}CR AND ^{53}CR FOR CRITICALITY SAFETY

1. MOTIVATION

Nuclear reactors operate in such a way that the neutrons created by fission are enough to make or the leakage or absorption so that the number of neutrons produced by fission remains constant, and hence the fission chain reaction is self-sustained. In this case, the effective multiplication factor k_{eff}=(Neutrons produced in one generation)/(Neutrons produced in the previous generation) is 1 and the reactor is *critical*, with a reactivity (departure from criticality) $\rho=(k_{eff}-1)/k_{eff}=0$. A critical reactor needs to be maintained really near to critical; if subcritical ($\rho<0$), the chain reaction cannot be sustained; while, if supercritical ($\rho>1$), the reactor would soon be out of control.

In this context, criticality safety relates to accident prevention and protection from an uncontrolled nuclear fission chain reaction, or a criticality accident. By means of running, analyzing and simulating benchmarks in experimental nuclear reactors, the criticality safety analysts and evaluators are capable of evaluating the nuclear data (mainly reaction cross sections) used as input in the calculations and quantify overall uncertainties through various types of sensitivity analysis [ICSBEP].

Selected criticality benchmarks with large amounts of Cr (e.g., PU-MET-INTER-002, and HEU-COMP-INTER-005/4=KBR-15/Cr) show high sensitivity to capture in Cr, which is dominated by ^{50}Cr and ^{53}Cr [Trkov:2018, Koscheev:2017]. Chromium is indeed an abundant (11-26%) [ref] element in the stainless steel and other alloys used as structural material, mostly in the vessel. Currently the discrepancies between evaluations (e.g., BROND-3.1, ENDF/B-VII.1, ENDF/B-VIII.0 and JEFF-3.3) and cross section compilations [Mughabghab:2006, Bao:2000] are at the level of ~30%, although this is not reflected in estimated uncertainty of the evaluated files (e.g., JEFF-3.3 uncertainty is around 10% which is inconsistent with the observed spread in evaluations). Such 30% uncertainty in Cr(n,γ) has an effect as large as 1000 pcm in reactivity, and therefore an entry has been made to the Nuclear Energy Agency (NEA) High Priority Request List (HPRL) in 2018 to measure the capture cross sections of ^{50}Cr and ^{53}Cr in the region between 1 and 100 keV with an accuracy between 8% and 10%.

Reacting to the new requests of the NEA HPRL is the best contribution that the n_TOF collaboration has to offer to the field of nuclear technology, in this case to nuclear safety, one of the five topics selected within H2020 in relation to nuclear energy. Hence, with this proposal we aim at measuring ^{50}Cr and ^{53}Cr in the region between 1 and 100 keV with an accuracy between 8% and 10% at n_TOF EAR1 [Guerrero:2013] combining the C_6D_6 detection system [Plag:2003] and a set of high quality chromium samples designed to overcome the limitations and issues faced in the previous experiments performed elsewhere.

2. LIMITATIONS OF THE PREVIOUS DATA AND SOLUTION PROPOSED

2.1 Previous measurements and status of evaluations

Chromium is a major component in stainless steel and other alloys present in the nuclear reactor cores, and hence has received the attention of the nuclear data community since many years. Indeed, neutron capture measurements on ^{50}Cr and ^{53}Cr have been performed at the major neutron beam facilities worldwide: RPI in USA [Stieglitz:1971], FZK in Germany [Beer:1975], ORELA in USA [Kenny:1977, Guber:2011] and GELINA in Belgium [Brusegan:1986]. Some details on these measurements are summarized in Table 1.

Table 1. Information about the existing ^{50}Cr and ^{53}Cr neutron capture experiments by time-of-flight available in the literature. The values of c_{MS} refer to the size of the correction in the capture yield by multiple interactions (scattering) before the capture takes place (see Section 2.2).

		Beer:1975	**Stieglitz:1971**	**Brusegan:1986**	**Kenny:1977**	**Guber:2011**	**This proposal**
	Facility	FZK	RPI	GELINA	ORELA	ORELA	n_TOF
	L (m)	0.7	27	60	40	40	185
	E_n (keV)	1-300	1-200	1-200	1-200	0.01-600	1-100
	Detector	Scint. Tank	Scint. Tank	C_6D_6	C_6F_6	C_6D_6	C_6D_6
^{50}Cr	**n (10^{-3} at/barn)**	18	8	7	5,0 / 8,0	-	0,6 / 5
	1-C_{MS}(1-10 keV)	0,8	0,65	0,6	0,55	-	**0,1**
^{53}Cr	**n (10^{-3} at/barn)**	14	14	12,0 / 60,0	8,0 / 12	14	1,2 / 8
	1-C_{MS}(1-10 keV)	0,7	0,7	0,65	0,55	0,7	**0,12**

Considering that in chromium neutron scattering features a cross sections three orders of magnitude larger than capture, the following must be noted:

- Only the most recent measurements, Brusegan et al. and Guber et al., used the C_6D_6 detector type that minimizes the neutron sensitivity, which means that all previous measurements suffer certainly from the problematic neutron scattering background.
- All measurements aimed at covering a wide energy range, trying to acquire statistics in the full resonance region that extends up to several hundreds of keV. Consequently, thick targets were used to produce enough reactions at high neutron energy, where the cross section becomes smaller. This means that at low neutron energy, where the cross section is larger, there are multiple interactions within the target and hence a dominant contribution from multiple interaction capture reactions that, as discussed in the next section, significantly alter the shape of the measured yield and hence needing really large and often inaccurate corrections.

The status and comparison of the current evaluations is revised in Ref. [Trkov:2018], where it is concluded that indeed new data are needed for both isotopes, despite the recent data from ORELA at ORNL [Guber:2011] for ^{53}Cr and ^{nat}Cr.

2.2 Multiple interaction issues

If one considers that only one neutron interaction takes place inside a target, then the measured capture yield is related to the neutron cross sections as

$$Y \approx Y_0 = [1-\exp(-n\sigma_{tot})]\, \sigma_\gamma/\sigma_{tot}$$

However, if the target thickness or the scattering cross section are large enough, then the neutron capture can occur also after one or several neutron scattering interactions [Choudry:1969]. Neutrons lose their energy in each scattering event, hence enhancing the capture yield in the high energy tail of the measured resonance. This is illustrated in Figure 1 for the case of a measurement at GELINA with a 0.007 atoms/barn ^{55}Mn target (enriched to 10.5%) [Kopecky:2013], where the calculated yields correspond to calculations with the code REFIT. The problem is that accurate resonance parameters are very difficult to extracts form a yield which shape is dominated by multiple interactions captures, and that explains the sizable deviations between different measurements of ^{50}Cr and ^{53}Cr in the region of interest for criticality safety. Previous experiments aiming at measuring neutron capture up to energies of several hundreds of keV had to use thick targets to gather amount statistics at high energies, but neutron scattering in the tens of keV region is about three orders of magnitude larger than capture and hence those data suffer from very large multiple interaction corrections in the low energy region. This correction, defined as the fraction of the capture yield produced after more than one scattering [$c_{ms}=1-(Y_0+Y_1)/Y)$] is larger than 50% and up to 80% (see Table 1, and Figure 2) in all previous experiments in the keV region.

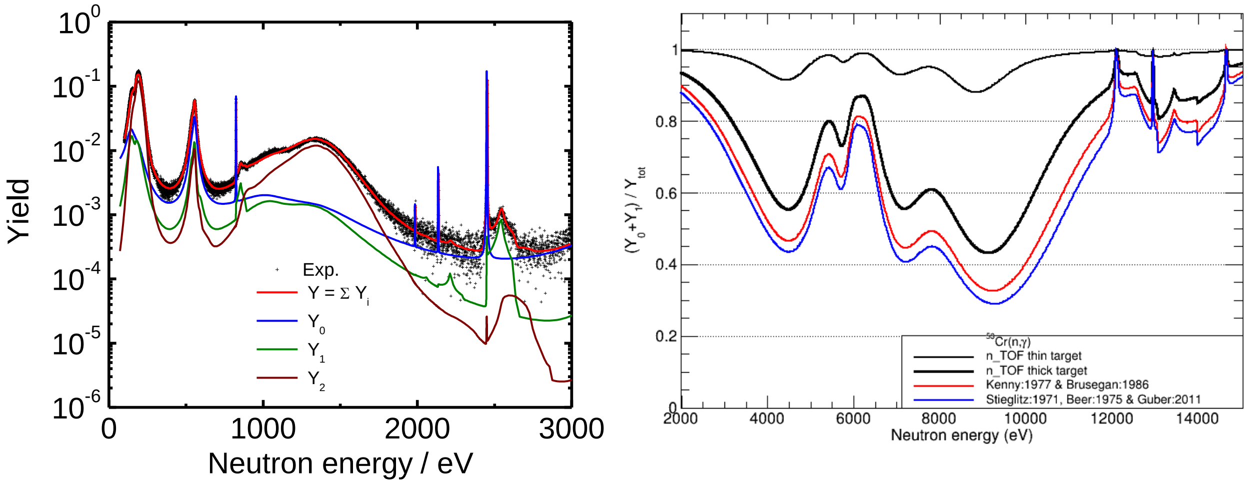

Figure 1. Effect of the multiple interactions in the target before the capture reaction takes places. Left: comparison of the measured ^{55}Mn capture yield measured and the individual contributions to the total yield calculated with REFIT. Right: calculated contribution of the Y_0 and Y_1 components to the total yield for different ^{50}Cr(n,γ) experiments.

2.3 The (very) thin target approach at n_TOF

As the region of interest in the NEA request related to criticality safety is below 100 keV, and considering that the accuracy of previous experiments is limited due to multiple interaction corrections to the capture yield, in this proposal we will implement a thin/thick target approach with the aim of keeping the correction (c_{MS}) at a maximum of ~10%. This require that two different targets are used, a thinner one at low (<15 keV) and a thicker one for high (>15-100 keV) energy. In addition, the sample material will be in metallic form instead of oxide, which was the case in all previous experiments. This allows to significantly reduce the multiple interaction in the sample, in this case by elastic scattering in oxygen.

3. ^{50,53}Cr(n,γ) MEASUREMENT AT n_TOF

3.1 Experimental set-up and samples specifications

The measurement will take place at the neutron time-of-flight facility n_TOF, at Experimental Area (EAR1) with a nominal flight path of 185 m. The large flight path is necessary to have enough resolution for the high energy (tens of keV) resonances in ^{50}Cr and ^{53}Cr.

Regarding the detection system, the array of four carbon fiber C_6D_6 detectors is envisaged, as it features the lowest neutron sensitivity (~10^{-4}) compared to the other neutron capture detection systems available at n_TOF, the TAC and the commercial C_6D_6.

The material for producing the samples will be leased from *ORNL Isotopes* which offers highly enriched ^{50}Cr (96,42(2)% enriched) and ^{53}Cr (97,2(2)% enriched) material as metal, instead of the usual oxide form. The targets' thicknesses have been defined as to keep the multiple interaction correction at maximum of ~10% in the peak of the resonances, using a different thickness to low (<15 keV) and high (15-100 keV) energy ranges. To this end, the ^{50}Cr targets will have thicknesses of 0.6×10^{-3} and 5×10^{-3} atoms/barn, with 1.2×10^{-3} and 8×10^{-3} atoms/barn for the case of ^{53}Cr. Since the thickness is limited, the statistics will be maximized by making large diameter (40 mm) targets that cover the full size of the beam (~35 mm), eliminating in this way as well the uncertainty due to the target alignment and the calculation of the beam interception factor (BIF).

3.2 Beam time estimate

The aimed accuracy is 10%, hence the contribution from statistics must be kept significantly lower than that, i.e. around 3-5%. The beam time requirements have been calculated with using the neutron flux in EAR1, the C_6D_6 system detection efficiency and the JEFF-3.3 evaluated cross sections, then convoluting the results with the EAR1 resolution function. The resulting counting rates per pulse indicate that the aimed statistical accuracy can be reached with the delivery of a total of 4x10^{18} protons, distributed according to Table 2.

Table 2. Beam time request and distribution.

Sample		Protons
^{50}Cr	**Thin**	5x10^{17}
	Thick	8x10^{17}
^{53}Cr	**Thin**	5x10^{17}
	Thick	17x10^{17}
Backg. & Norm.		5x10^{17}
Total		**4.0x10^{18}**

The expected counting distributions are displayed in Figure 2, where the expected background level is shown as well. The background will be determined from beam-off and beam-on runs with graphite and lead targets, while the normalization will be assessed via the saturated resonance method with the 4.9 eV ^{197}Au resonance.

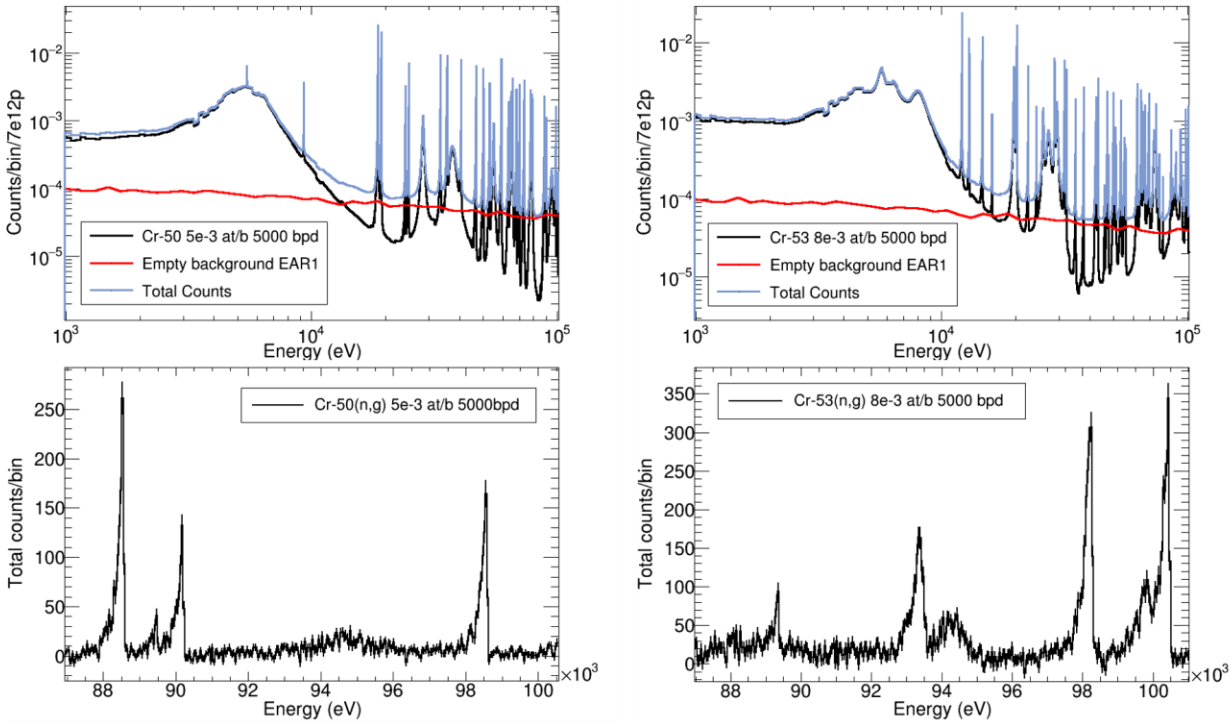

Figure 2. Counting rate estimation for the ^{50}Cr and ^{53}Cr thick targets. Top: overview. Bottom: statistics expected at the high energy range of interest, where the cross sections are lower.

3.3 Complementary experiments

The experiment proposed herein will be complemented with transmission measurement at the JRC GELINA facility in Geel, Belgium. This will allow a combined resonance shape analysis that will certainly improve the accuracy of the resonance parameters, especially because of the use of metal instead of oxide targets. The transmission measurement will also include a ^{nat}Cr targets, to help identifying the resonances from Cr isotopes other than ^{50}Cr and ^{53}Cr.

In addition, the fact that ^{51}Cr is unstable, $t_{1/2}$=27.702(4) days, allows for neutron activation experiments. The MACS (Maxwellian average Cross Section) of ^{50}Cr will be measured at the classical *k*T~25 keV using the ^{7}Li(p,n) neutron spectrum at the HISPANOS neutron source in Seville, Spain. Moreover, attempts will be made to generate at *k*T~5 keV neutron beam using the ^{18}O(p,n) reactions, as this would give information about the dominant ~7 keV ^{50}Cr resonance.

Summary of requested protons: 4.0x10^{18} protons at n_TOF-EAR1

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