EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Study of n+^{63,65}Cu reactions and their relevance for nuclear technologies and Astrophysics

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Abstract: Neutron-induced reactions in copper are relevant for emerging nuclear technologies and nuclear astrophysics. Recent sensitivity studies highlight a clear need for new cross-section data in a wide energy range, as the existing literature data is scarce and occasionally discrepant. While radiative capture is pertinent in both research fields, elastic and inelastic cross sections are particularly crucial for nuclear technology. We propose a measurement campaign of ^{63,65}Cu(n,γ) and ^{63,65}Cu(n,tot) cross sections at EAR1 using well-established C_6D_6 detectors and a fission chamber loaded with ^{235}U, respectively. Through a combined analysis of these data, the elastic cross section can be deduced. In future stages, we may examine the elastic angular distribution and explore the inelastic channel, with additional details provided in an addendum.

Requested protons: 8×10^{18} protons on target, (split into 2 runs over 2 years)
Experimental Area: EAR1

1 Introduction and scientific motivation

The interaction of neutrons with copper is relevant for both the exploration of advanced nuclear technologies and the context of stellar nucleosynthesis. Specifically, the reactions ^{63,65}Cu(n,γ) are pertinent in both cases, whereas ^{63,65}Cu(n,n) and ^{63,65}Cu(n,n') are of interest for nuclear technology. The ensuing summary provides a concise overview.
The global interest in advanced reactors, stimulated in the early 2000s by the Generation-IV International Forum, is currently steadily increasing, leveraging the aims at decarbonizing the energy systems and enhancing the sustainability of the energy sources employed for this. While all nuclear technologies contribute to decarbonization goals, fast reactors stand out for their inherent sustainability, attributed to their capacity for operating in a closed fuel cycle.
Within this framework, research reactors featuring high-energy neutron spectra are crucial for testing nuclear data and materials. The RSV–TAPIRO [1] nuclear research reactor, located at the ENEA–Casaccia Research Centre, near Rome, Italy, is a compact fast spectrum zero-power (5 kWth) reactor whose cylindrical core is made of metallic uranium-molibdenum alloy with a copper reflector. The core is surrounded by several rods, also made of copper, meant for reactor control, which is achieved by varying neutron leakage through their axial positioning inside the reflector itself. The reactor provides an almost pure fission spectrum from 1 keV to 10 MeV showing a peak at 820 keV and an average value around 1 MeV. A remarkable feature of the reactor is the extremely good characterization of these spectra in the available irradiation positions, which discloses the possibility to perform accurate integral measurements of nuclear data. Sensitivity and uncertainty studies performed with the ERANOS 2.3 code [2] (calculation tool based on deterministic approach to particle transport) revealed inadequacies in major data libraries regarding copper evaluations. These sensitivity studies facilitated the ranking of n+^{63,65}Cu reactions and the definition of the energy region of interest (refer to Table 1). Unsurprisingly, copper plays a pivotal role immediately following that of ^{235}U (not depicted in the table). Morevover, Monte Carlo simulations showed that the choice of one nuclear data library over another exerts an impact on k$_{eff}$ comparable with the effect of a control rod withdrawal. Beyond the relevance for RVS–TAPIRO [1], recent benchmark experiments on Fusion technology revealed the need for improved ^{63}Cu(n,γ) data in the resolved resonance region, particularly near thermal energy [3].

Reaction	^{63}Cu(n,γ)	^{63}Cu(n,n)	^{63}Cu(n,n')	^{65}Cu(n,γ)	^{65}Cu(n,n)	^{65}Cu(n,n')
Rank	5	1	3	6	2	4
E$_n$ (MeV)	0.01 – 0.4	0.1 – 5	1 – 8	0.01 – 0.4	0.1 – 4	1 – 6

Table 1: n+Cu nuclear reactions and related energy region of interest for RVS–TAPIRO. Their rank, i.e. decreasing contribution on k_{eff} uncertainty, is based on reactor code calculations.

For all these reasons, copper is being considered as a new entry in the NEA (Nuclear Energy Agency) "Nuclear Data High Priority Request List" [4], which lists nuclear data requests relevant for advanced nuclear systems.

From the astrophysics standpoint, the exploration of stellar nucleosynthesis has undergone substantial evolution since the seminal article by B2FH [5], as discussed in subsequent works, e.g. [6]. Notably, stable isotopes beyond iron are believed to be synthesized in Red Giant Stars (s-process) and explosive stellar scenarios (r-process). While the fraction attributable to s-process and r-process can be determined for most of stable isotopes, a few cases, including copper, remain doubtful in terms of origin.

Indeed, copper can be synthesized through various nucleosynthesis scenarios [7, 8, 9, 10, 11, 12] (not in order of respective importance): (i) the weak s-process, operating in massive stars during core-helium and carbon-shell burning stages, as well as in the explosive complete Ne-burning stage; (ii) the explosive nucleosynthesis in Type II Supernovae (SNe II); (iii) the main s-process occurring in low and intermediate mass Asymptotic Giant Branch (AGB) stars; and (iv) the explosive nucleosynthesis in long-lived Type Ia Supernovae (SNe Ia).

Advancements in our understanding of these processes rely on the availability of precise and accurate Maxwellian Average Capture cross Section (MACS) in the energy region of interest, i.e. $kT = 8 - 90$ keV. These MACS values enable stellar models to confidently estimate the contributions of weak and main s-process. Astonishingly, the MACS values of ^{63}Cu and ^{65}Cu exhibit a considerable degree of uncertainty, with deviations reaching up to a factor of 2. Therefore, KADoNiS [13] strongly recommends further investigations [13] on these isotopes.

Furthermore, considering copper as a distinctive iron-peak element, the impact of its (n,γ) cross section on the efficiency of the s–process in massive stars is significant. As illustrated in Fig. 1, the abundances within the mass range of the weak s–component synthesized in massive stars exhibit a pronounced propagation effect if only the previously evaluated cross section of ^{63}Cu and ^{65}Cu isotopes is increased or reduced by 30%. It is evident that the (n,γ) cross sections of the stable isotopes of copper play a crucial role for the quantitative description of the s–process in massive stars, i.e. for the s abundances between Fe and Y.

2 Copper nuclear data in the literature

The latest versions of major evaluated nuclear data libraries, such as ENDF/B-VIII.0, JEFF-3.3 and JENDL-5.0, show remarkable differences. For instance, variations in the reported number of resonances are observed within the energy region 50–100 keV. In addition, cross section values in the unresolved resonance region show discrepancies up to 30%.

Various types of measurements can be found in the Experimental Nuclear Reaction Data database (EXFOR) [15]: (i) time–of–flight (TOF) transmission and capture experiments, (ii) activation measurements, and (iii) elastic and inelastic experiments. Elastic cross section for $E_n < 8$ MeV is deduced from total transmission measurements, since the radiative capture channel is much smaller than the elastic channel. Relevant measurements are summarized in Table 2. It is evident that past capture experiments used γ

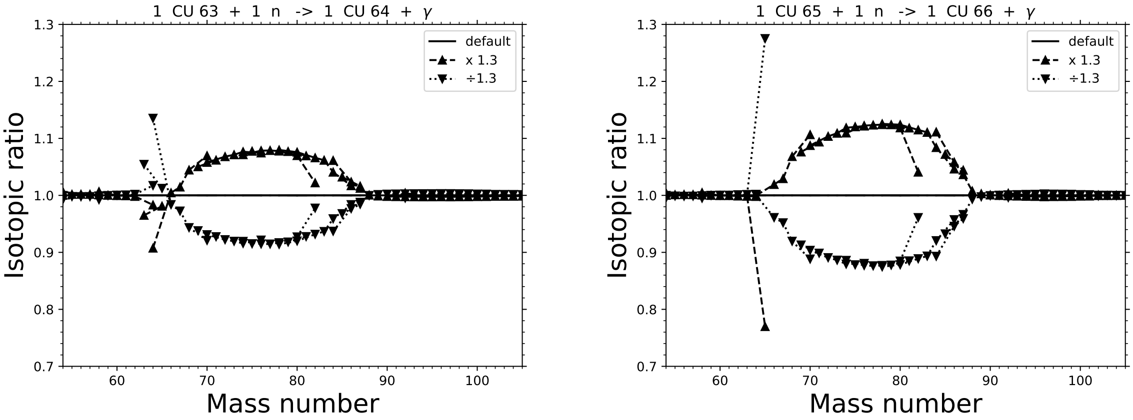

Figure 1: The effect of cross section uncertainties on the s–process efficiency in massive stars illustrated at the example of ^{63}Cu (left) and ^{65}Cu (right). Data from [14].

Ref.	type	Facility	E$_n$	remarks
[16]	TOF – capt.	GELINA	0.2–16.5 keV	plastic scintillator
[17]	TOF – capt.	ORELA	3–50 keV	C_6F_6 detector
[18]	activation	FZK	$kT = 25$ keV	MACS=58.1 mb
[19]	activation	JRC & LANSCE	$kT = 25$ keV	MACS=92 mb
[20]	activation	SARAF	$kT = 28.2$ keV	MACS=70.4 mb
[21]	TOF – tran.	ORNL	2–60 keV	energy resolution 2%
[22]	TOF – tran.	ORELA	10–150 keV	
[23]	TOF – tran.	GELINA	0.15–90 keV	
[24]	(In)Elastic	TUNL	8 –14 MeV	angular distribution

Table 2: Relevant neutron-induced cross section measurements in EXFOR. MACS values refer to the ^{63}Cu(n,γ) reaction.

detectors not particularly optimized for (n,γ) reactions, potentially introducing significant systematic errors related to neutron sensitivity. Moreover, it is worth highlighting that the MACS derived from TOF data in [17] is approximately twice as high as the value reported in [18]. In addition, the MACS in [18] does not agree with values from either [20] or [19], see Fig. 2 for a better visualization.
In summary, past measurements exhibit limitations, discrepancies, and fail to encompass the energy region of interest, especially concerning (n,n) reactions. These discrepancies highlight the need for new capture and transmission measurements across a broad energy spectrum, a requirement that can be fulfilled at n_TOF.

3 Proposed experimental setup

We propose conducting capture and transmission time-of-flight measurements on highly enriched ^{63}Cu and ^{65}Cu samples at EAR1 within the energy range pertinent to the aforementioned applications, i.e. E$_n$ < 400 keV for (n,γ) reactions and E$_n$ < 5 MeV for (n,tot).

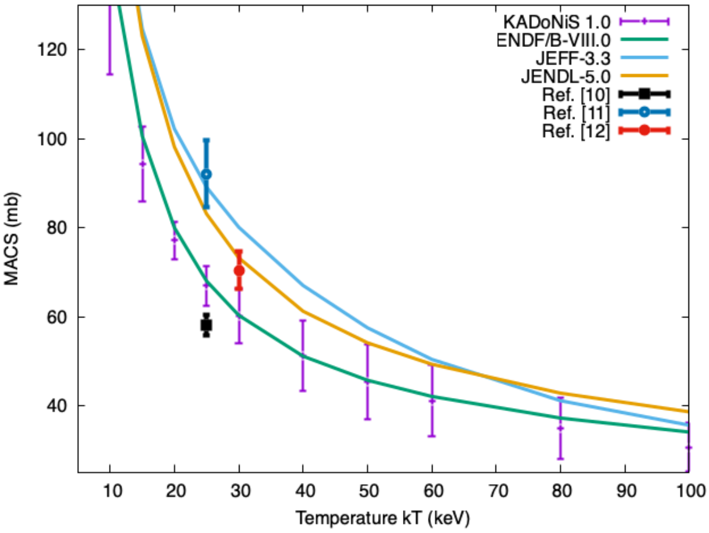

Figure 2: Calculated MACS from evaluated nuclear data files and KADoNiS 1.0 together with measured values at specific temperatures.

The combined analysis of these data sets can result in an accurate determination of the elastic cross section.

The samples will take the form of metallic discs with a 25 mm diameter, produced from two batches of material enriched in ^{63}Cu and ^{65}Cu to 99.89 wt% and 99.69%, respectively. For each isotope a sample with a total mass of 2 g will be available for capture measurements, and a total mass of 10 g for transmission measurements. The proposed partition in a thin and a thick sample is required to optimize counting rates and signal–to–bakground ratios. We have chosen EAR1 for both experiments due to its superior energy resolution and larger dynamic time–of–flight (TOF) range compared to EAR2. In fact, from experience with similar experiments at n_TOF, we expect to extract accurate resonance parameters in the epithermal region and above, aligning with the energy region crucial to the objectives outlined in this proposal.

Currently, new detection setups for studying (n,n') reactions and elastic angular distribution are under development at n_TOF. As a result, these measurements are not included in the present proposal and may become the subject of a future addendum.

3.1 Capture experiment

For the detection of the prompt γ rays resulting from radiative capture events, i.e. the electromagnetic cascade produced in the de–excitation of the compound nucleus formed in (n,γ) reactions, we will adopt the total energy detection principle in combination with the pulse height weighting technique. This approach will be implemented using an array of 4 C_6D_6 scintillators. It is noteworthy that these detectors exhibit a neutron sensitivity which is significantly lower than that of the alternative capture detector available at n_TOF, namely the 4π BaF_2 calorimenter. In addition, they are less sensitive to the γ–flash (which refers to the prompt signal caused in the detector by spallation γ–rays and relativistic particles) thereby avoiding detector blindness for a few microseconds after

γ–flash.
Finally, in addition to the enriched samples mentioned above, a metallic disc of natural copper will be utilized to verify resonance assignment to the correct Cu isotope, ensuring consistency between the data for individual isotopes and cross-section data for natural Cu. Furthermore, similar to past measurements, samples of ^{197}Au, ^{208}Pb and $^{\mathrm{nat}}$C with similar geometrical properties will be used for additional normalization and background measurements.

3.2 Transmission experiment

This proposal introduces a novel approach at n_TOF, through the combined measurement of (n,γ) and (n,tot). Transmission experiments are in principle the simplest type of neutron-induced measurements and a well-established technique [25]. The experimental determination of the transmission is based on the ratios of "Sample-in" and "Sample-out" TOF spectra, i.e. with and without the sample in the beam, respectively. A feasibility test for transmission measurements at n_TOF was proposed [26] and executed successfully, paving the way for a new frontier in neutron-induced measurements at n_TOF.
The proposed measurement setup is the same of the feasibility test [26]. It based on the measurements of the neutron flux with and without the copper sample in the beam. In the keV – MeV region, this flux measurement can be performed using an in-beam fast ionization chamber loaded with ^{235}U as converter. In fact, the ^{235}U(n,f) cross section is very high and the detection efficiency in 2-π geometry is as high as 97% for fission products. Therefore, as in the case of previous similar flux measurements at n_TOF [27], we would employ the PTB fission chamber detector loaded with 6 samples of ^{235}U for a total thickness of 1798 μg/cm^2.

4 Beam-time request

The copper isotopes are characterized by a high total cross section, dominated by the elastic channel. As a consequence, a favorable signal-to-background ratio is expected for both capture and transmission measurements.
The quantity of copper in the capture samples results from a trade-off between the need of reducing the requested beam time and the optimization of expected count rate in the keV region. The summary of the requested protons is reported in Table 3, and it is valid under the conditions that the 2 capture measurements are performed during the same measurement campaign (thus sharing background measurements) and that transmission measurements are performed during the same measurement campaign (thus sharing the same Sample-out measurement). Hereafter more details are provided.
Capture:
The beam time request of 2. $\times$ 10^{18} protons for each enriched Cu sample and 0.3 $\times$ 10^{18} for the natural Cu sample (this latter measurement can be performed either in EAR1 or EAR2) is calculated so to achieve a final accuracy better than 5% in the unresolved resonance region. A conservative value of 20% for the detection efficiency is assumed and a correction factor is applied in order to take into account the area of the samples, which

is smaller than the dimension of the neutron beam. As in previous capture measurements, the estimation of the different components of the background requires a total number of 0.6×10^{18} protons. In addition, the normalization of capture data and the validation of the measurement at high energy and the cross-check of the flux stability is achieved by a cyclic measurement of a gold sample with 500 mg. This further study requires a neutron intensity corresponding to 0.1×10^{18} protons.
Transmission:
The beam time request of 1.0×10^{18} protons for each sample is calculated so to achieve a final accuracy better than 5% in the energy region $E_n > 100$ keV with 100 bins per energy decade.

Exp.	Sample	Protons	Comments
Capture	^{63}Cu	2.0×10^{18}	
Capture	^{65}Cu	2.0×10^{18}	
Capture	^{nat}Cu	0.3×10^{18}	EAR1 or EAR2
Capture	Empty-sample	0.2×10^{18}	background study
Capture	Pb	0.2×10^{18}	background study
Capture	C	0.2×10^{18}	background study
Capture	^{197}Au	0.1×10^{18}	normalization
Transmission	^{63}Cu	1.0×10^{18}	"Sample-in"
Transmission	^{65}Cu	1.0×10^{18}	"Sample-in"
Transmission	Empty-sample	1.0×10^{18}	"Sample-out"
		8.0×10^{18}	

Table 3: Summary of requested protons.

5 Summary and outlook

We request a total of 8×10^{18} protons on target to carry out the proposed measurement campaign on the stable copper isotopes ^{63}Cu and ^{65}Cu. It consists of transmission (3×10^{18} protons) and capture (5×10^{18} protons) experiments to be performed at EAR1.
In subsequent phases, we contemplate investigating the elastic angular distribution and exploring the inelastic channel. Additional specifics on these aspects might be presented in a forthcoming addendum.
Summary of requested protons: 8×10^{18} protons on target.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Part of the experiment	Design and manufacturing
C_6D_6, SiMon	☒ To be used without any modification ☐ To be modified
PTB fission chamber	☐ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☒	pressure = 1.1 bar, volume = 2 l
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
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	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			