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# Study of $(n,\alpha)$ reactions of interest for nuclear reactors: the case of $^{19}\text{F}(n,\alpha)^{16}\text{N}$ with SCALP detector

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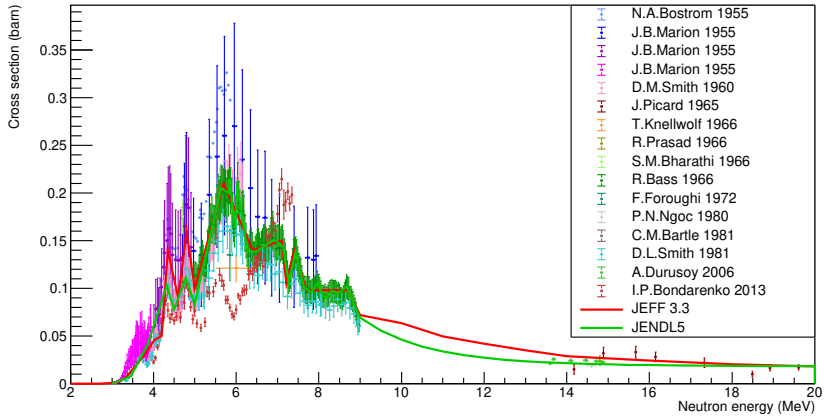
**Abstract.** The  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  cross section is of great interest for the development of the next generation IV reactors that could potentially use molten fluoride salts. Significant differences (up to a factor of 3) have been observed for this nucleus regarding the  $(n,\alpha)$  channel. In view of improving our knowledge on this  $(n,\alpha)$  reactions, the GrACE group (Groupe Aval du Cycle Electronucléaire) of the LPC Caen has developed a new detector named SCALP (Scintillating ionization Chamber for ALPha particle detection in neutron induced reactions). This paper deals with the first experiment carried out with this brand new detector at the new NFS facility (GANIL, Caen, France). After discussing the needs for new measurements of the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction, the operating procedure of the SCALP detector will be presented, as well as the experiments that have been conducted using it. Furthermore, insights into the data acquired during our experiment, as well as the ongoing data processing and associated multi-channel analysis, will be provided.

## 1 Motivations and scientific context

The Molten Salt Reactor (MSR) is one of the innovative reactor concepts retained by the international Generation IV forms. In that kind of reactors  $^{19}\text{F}$  can be found in abundance in the form of fluorine-lithium-berilium (FLiBe) for cooling and as a solvent for fissile fuel, or as a base for uranium or thorium salts. Estimated uncertainty on the total neutron interaction cross-section of  $^{19}\text{F}$  brings up to 213 pcm uncertainty on the reactor  $k_{eff}$  value[1]. The uncertainty on the cross-section of the reaction  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  alone is responsible for approximately 40 pcm to 130 pcm of uncertainty on  $k_{eff}$  depending on the type of MSR considered.[1]

Figure 1 shows the cross-section of  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction. Experimental data available in the EXFOR database are shown as dots and evaluations are represented as solid lines. One can see that there are large discrepancies up to a factor of 3 in some energy ranges. A new study is therefore recommended to improve our knowledge on the cross section of the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction.

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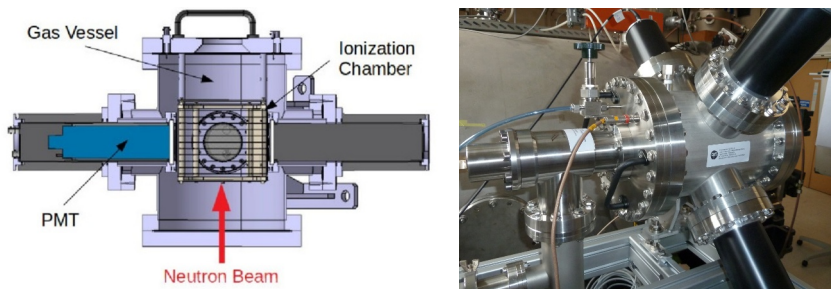


**Figure 1.** Cross section of the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction. Experimental data available in the EXFOR[2] database are shown as dots and some evaluations are represented as solid lines.

## 2 Experimental Setup

For such measurements, the GrACE group at LPC Caen has developed a new detector called SCALP[3]. SCALP is an ionization chamber filled with a scintillation gas, which allows for secondary particles detection with a very good time resolution, making the device particularly suited to neutron time-of-flight based measurements.

### 2.1 The SCALP detector

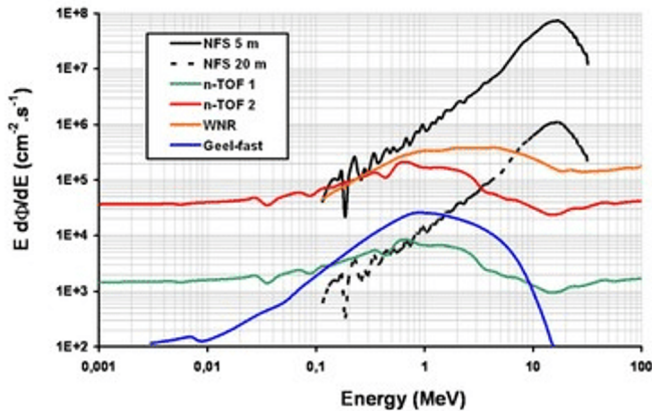


**Figure 2.** SCALP detector schematic [3] (left) and a photo of the SCALP detector (right). It includes a gas vessel containing the ionization and scintillation gas. It also includes 4 photomultipliers for measuring the time-of-flight of incident neutrons. The ionization chamber measures the energy deposited by the charged reaction products.

The SCALP detector is a closed pressure vessel. The Fig 2 shows that four photomultipliers (PMTs) are positioned around the chamber to detect photons coming from scintillation due to the interaction of charged particles produced by neutron-induced reactions with the  $\text{CF}_4$  gas. This gas was chosen for its scintillation properties to

optimize measurement resolution in addition to containing the fluorine target. The time resolution of the PMTs was measured to be 250 ps ( $1\sigma$ ) using an alpha source [3], which allows one to reconstruct the neutron time-of-flight with a very good resolution. On the other hand, the ionization chamber enclosed in the vessel was able to measure the energy deposited by charged particles with a resolution of 170 keV ( $1\sigma$ ).

## 2.2 Neutron production at NFS - SPIRAL2



**Figure 3.** Comparison of beam intensity in function of energy for different facilities[4]. The different lines correspond to other continuous neutron spectra available at other facilities, and one can see that NFS has a well-suited energy spectrum between 0.1 and 40 MeV for the measurements wanted to be performed with SCALP.

SPIRAL2 is a superconducting linear accelerator from Ganil (LINAG) in Caen, France. It accelerates a beam of light ions (p, d,  $^4\text{He}$ ) using accelerator cavities, with a beam intensity of up to 5 mA for deuterons at the frequency of 88 MHz.

NFS is an experimental area where neutrons can be produced from a primary deuteron beam and a Beryllium target. These neutrons then pass through the 30m time-of-flight (TOF) area, where various types of detector can be set up. The neutron beam supplied is a continuous energy spectrum ranging from 0.1 to 40 MeV peaked at 14 MeV, with a higher intensity compared with other facilities (Fig 3)[4].

The NFS line is well suited for the measurements planned with SCALP. A new measurement of the cross-section of the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction between the threshold energy and 20 MeV was performed with SCALP at NFS. The average deuteron intensity was set to 7.5  $\mu\text{A}$  to limit pile-up events in SCALP. The beam frequency of 730 kHz avoid the overlap of two consecutive bursts. The flight distance (the distance between the beryllium converter and the SCALP setup) was set at 28.431m to reach a high precision on the time-of-flight. For each pulse, a signal named High Frequency (HF) is generated by the machine and sent to our acquisition system. The time stamp  $t_{HF}$  corresponding to this signal correspond to the neutron departure for calculating the time-of-flight. The two neutron monitors on the NFS facility are a micromegas and a liquid scintillator. However only the micromegas was operating for this first experiment[4].

### 3 Data Analysis

The first step in the analysis was to reconstruct the neutron time-of-flight from the time stamp  $t_{PMi}$  given by each photomultiplier ( $i$ ) and the time stamp  $t_{HF}$  given by the HF during neutron production by the facility. To determine the time-of-flight using the 4 PMTs, we calculated the light-weighted time-of-flight detected by the four photomultipliers. The time-of-flight is calculated as the difference between the weighted photomultipliers time  $t_{PM}$  and the HF time.

$$tof = t_{PM} - t_{HF} \quad (1)$$

The time-of-flight distribution measured with SCALP shows that the gamma flash is very narrow. The time resolution of the gamma flash, which corresponds to the time-of-flight resolution measured by SCALP was found to be 1.3 ns ( $1\sigma$ ).

The kinetic energy of the incident neutrons detected by SCALP is calculated using the time-of-flight measured by the photomultipliers and the flight distance  $d = 28.413\text{m}$ . The spectrum ranges from 1 to 40 MeV, with a peak around 15 MeV. The neutron energy resolution directly calculated from time-of-flight and flight distance resolution is 0.4% at 10 MeV and remains below one percent between 0 and 40 MeV.

#### 3.1 Reaction identification

The reconstruction of the incident neutron kinetic energy  $T_n$  associated with the measurement of the energy deposited in the ionization chamber  $E_{dep}$  allows one to construct an identification matrix for 2-body reactions in the chamber. Indeed, in that case, the difference between  $E_{dep}$  and  $T_n$  is simply equal to the Q-value of the reaction:

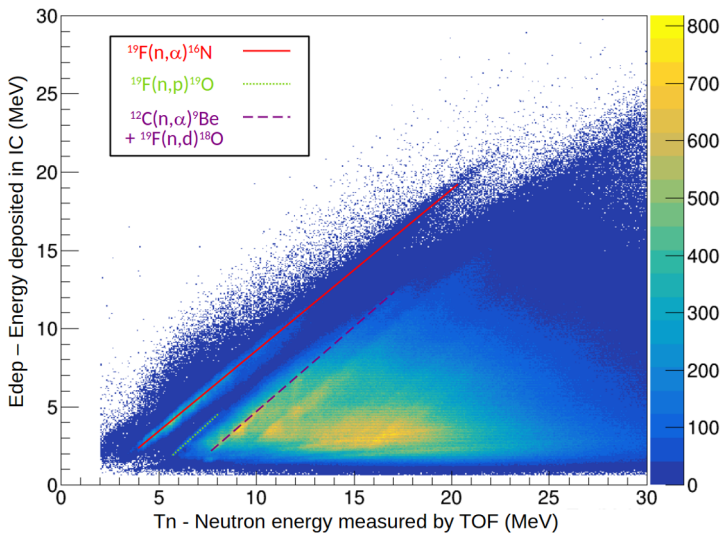
$$E_{dep} = T_n + Q \quad (2)$$

Thus events belonging to a given reaction are expected to follow a straight line in the according to the  $(T_n, E_{dep})$  matrix and each of these two-body reactions can be identified by the corresponding Q-value.

Reactions	Q (MeV)	$T_n$ threshold (MeV)
$^{19}\text{F}(n,\alpha)^{16}\text{N}$	-1.52	1.61
$^{19}\text{F}(n,p)^{19}\text{O}$	-4.04	4.25
$^{19}\text{F}(n,d)^{18}\text{O}$	-5.76	6.08
$^{12}\text{C}(n,\alpha)^9\text{Be}$	-5.70	6.18
$^{19}\text{F}(n,t)^{17}\text{O}$	-7.56	7.96

**Table 1.** Q-value for different reactions with  $^{19}\text{F}$  and  $^{12}\text{C}$ .

The  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction owns the highest Q-value, and is well separated from the other reactions allowing the identification of the events associated to this reaction without ambiguities. The same applies to the reaction  $^{19}\text{F}(n,p)^{19}\text{O}$ . On the opposite, the  $^{12}\text{C}(n,\alpha)^9\text{Be}$  and  $^{19}\text{F}(n,d)^{18}\text{O}$  reactions carry Q values of -5.70 MeV and -5.76 MeV respectively (Table 1), too close regarding the resolution of the SCALP ionization chamber, making it impossible to dissociate these two reactions. The ionization chamber is calibrated using the line of the  $^{12}\text{C}(n,\alpha)^9\text{Be}$  reaction products.



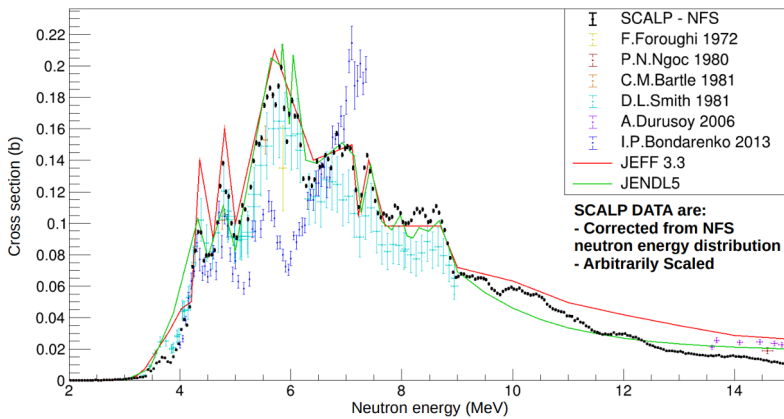
**Figure 4.** The above identification matrix corresponds to the deposited energy  $E_{dep}$  in the ionization chamber as a function of the neutron kinetic energy  $T_n$  with the number of events shown in color in z-axis. Straight lines are observed, each of which corresponds to a reaction and can be identified by the corresponding Q-value.

When plotting the deposited energy as function of the incident neutron energy (Fig 4), straight lines corresponding to two-body reactions identified by their Q values, can be observed. However, it carries a lot of noise corresponding to three-body reactions, elastic and inelastic collisions. The  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  and  $^{19}\text{F}(n,p)^{19}\text{O}$  reactions are identified and well separated from other reactions and parasitic reactions, enabling to measure their cross sections with the SCALP detector.

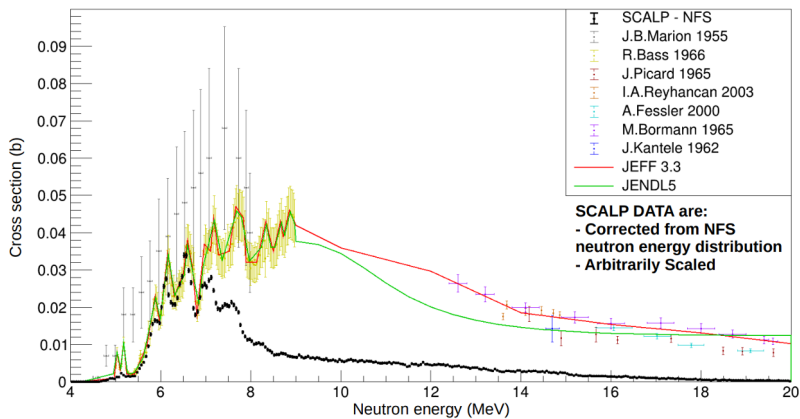
### 3.2 Preliminary comparison with some previous works

The preliminary results of the SCALP experiment for the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction are shown in the Fig 5, given the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  events in the identification matrix. The number of counts per energy bin was corrected using the energy distribution of incident neutrons, and then arbitrarily normalized with a constant factor over the whole energy range to roughly match to the magnitude of previous measurements. Error bars include only statistical uncertainties. In any case, one can see that some of the structures appearing in our data correspond to some previously observed. At around 8 MeV, SCALP measurements show structures validated by the latest evaluations (JENDL-5) that were not observed in earlier evaluations (JEFF-3.3, ENDF/B-VIII.0). Between 9 MeV and 13.5 MeV, no experimental data have been reported at EXFOR so far, so SCALP is likely to be the first cross section data set for the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction in this energy range.

As previously seen, given that the  $^{19}\text{F}(n,p)^{19}\text{O}$  reaction is identified and isolated, this reaction could be tentatively measured as a by-product of the experiment. By plotting the number of hits as a function of neutron energy we obtained the black data points of Fig 6 (arbitrarily scaled to roughly match the magnitude of previous



**Figure 5.** For the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  reaction: cross-section (in barn) as a function of neutron energy (MeV). SCALP data (black dots, with only statistical uncertainties considered) are corrected for the NFS neutron energy distribution and arbitrarily scaled. Some experimental data (colored dots) and evaluated data (lines) are shown.



**Figure 6.** Same as Figure 5 for  $^{19}\text{F}(n,p)^{19}\text{O}$  reaction.

works in the energy range). Below 7 MeV, the resonances observed by SCALP are at the same energies with those reported in the literature. However, above 7 MeV, there is a deficit in the collection of events corresponding to this reaction because the detector was designed to study  $(n,\alpha)$  reactions. In the case of  $(n,p)$  reactions, for the highest energies, protons can escape from the ionization chamber leading to an underestimation of the deposited energy and the number of events produced by  $(n,p)$  reactions.

## 4 Conclusions and future works

Some  $(n,\alpha)$  reaction cross sections of interest for nuclear reactors exhibit large uncertainties. In this context, a new measurement of the cross section of the reaction  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  was recommended. To respond to this request, LPC Caen has designed a new detector to measure  $(n,\alpha)$  cross sections with a continuous neutron energy beam using the well known time-of-flight technique. The very first experiment with this detector was carried out at SPIRAL2 - NFS (Caen, France).

The analysis of this new measurement is still ongoing. However preliminary results have shown that the  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  of interest is well separated from the other reactions. As added benefit, it is possible to measure the cross section of the  $^{19}\text{F}(n,p)^{19}\text{O}$  reaction as a by-product. To date, for the reaction  $^{19}\text{F}(n,\alpha)^{16}\text{N}$ , one sees that some of the structures observed in the SCALP data are also reported in previous works, in addition new structures are observed and some structures are in agreement with the most recent evaluations.

Soon, a full simulation of the NFS experiment will be carried out to take into account the detector response as complement to the sensitivity analysis requested for the inclusion of the systematic uncertainties, not estimated in the preliminary results. An absolute normalization of the data taken at NFS will then be carried out using the neutron flux monitors of the experiment.

Another experiment was carried out for  $^{19}\text{F}(n,\alpha)^{16}\text{N}$  cross section measurements at the nELBE (HZDR, Dresden, Germany), in which some of the parameters differ from those of NFS. So an analysis similar to the current one should be carried out from the data taken at nELBE. SCALP can also be filled with a mixture of gases containing  $\text{CF}_4$  for scintillation, plus another gas containing another target, provided that the Q values are such that the reactions for which the cross-sections are to be measured are clearly identified. In addition to the  $\text{CF}_4$  gas configuration, another measurement was performed at NFS and nELBE in a 97%  $\text{CF}_4$  and 3%  $\text{CO}_2$  configuration to measure the cross section of the  $^{16}\text{O}(n,\alpha)^{13}\text{C}$  reaction, which is of great interest to the nuclear industry. Work on the analysis of these measurements is ongoing.

## Acknowledgement

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