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The LICORNE neutron source and measurements of prompt γ-rays emitted in fission

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Abstract

The emission of prompt gamma rays is one of the least measured and least well-understood parts of the fission process. Knowledge of prompt fission gamma spectra, mean energies and multiplicities are important for reactor gamma heating and hence linked to reactor safety. At the IPN Orsay we have developed a unique, directional, fast neutron source called LICORNE, intended initially to facilitate prompt fission gamma measurements. The ability of the IPN Orsay tandem accelerator to produce intense beams of ^{7}Li is exploited to produce quasi mono-energetic neutrons between 0.5 - 4 MeV using the p(^{7}Li,^{7}Be)n inverse reaction. The available fluxes of up to 7×10^7 neutrons/second/steradian are comparable to existing installations, but with two added advantages: (i) The kinematic focusing produces a natural neutron beam collimation which allows placement of gamma detectors adjacent to the irradiated sample unimpeded by source neutrons. (ii) The background of scattered neutrons in the experimental hall is drastically reduced. The dedicated neutron converter was commissioned in June 2013

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1. Introduction

The calculation of reactor core temperatures is a difficult problem because of the different nuclear reactions taking place in the core and the complex processes by which heat is generated, transported and evacuated. The majority of the reactor power originates from neutron-induced fission reactions in the fuel and is deposited very close to where the fission takes place via rapid loss of kinetic energy of the fission fragments. However, about 10% of the energy released in the core is in the form of gamma rays and can travel larger distances (many centimetres) from the initial reaction site. During reactor operation gamma heating processes dominate for all non-fissile materials in the reactor core (e.g. structural materials, core shielding, reactor instrumentation, etc.). When the reactor is shut down, gamma heating is the dominant energy deposition process for all core materials and thus the problem of gamma heating is directly related to reactor safety.

Currently, benchmark models of gamma heating in reactor cores underestimate heating effects by up to 30% when compared to experimental results (Rimpault, 2006). This is mainly due to insufficiently accurate nuclear data and possibly also deficiencies in the modelling. The gamma rays originating from neutron capture, inelastic scattering and beta decay of the fission fragments are fairly well understood. However, about 40% of this energy is prompt gamma emission (< 1 ns) and here, the available data for thermal neutron induced fission date from the early 1970's (Verbinski et al., 1973; Peelle and Maienschein, 1971).

However, these data have the potential for improvement and such measurements are at the top of the high priority nuclear data list of the NEA/OECD (NEA, 2006). Since future generation IV reactor concepts will mostly use fast neutron spectra, prompt gamma emission data in fast neutron induced fission are very important. However, currently very little data of this kind exists. For fast neutron induced fission the spectral shape, mean multiplicities and energies are expected to be different because excitation energy and angular momentum of the fissioning compound nucleus will change, along with the resulting fission yields, average neutron energies and multiplicities. There are particular technical challenges associated with prompt gamma emission measurements for fast neutron induced fission. Firstly, fission cross sections are typically three orders of magnitude lower than those for thermal neutron induced fission. To produce high fluxes the target and gamma detectors need to be very close to the neutron source. However, since conventional quasi-mono-energetic neutron sources emit neutrons isotropically gamma detectors will require heavy shielding to avoid being blinded by neutrons form the source, which is often highly impractical.

With these constraints in mind we developed the LICORNE directional neutron source (Lebois et al., 2014; Wilson et al., 2013) which overcomes the last severe constraint by using reactions which produce neutrons in inverse kinematics to produced focused beams of neutrons.

2. The LICORNE neutron source

Natural collimation of neutron beams can be achieved if the neutrons are produced using a reaction in inverse kinematics where the projectile is much heavier than the target. Neutron production via this method thus combines the best features of white neutron sources (collimated beams) and conventional quasi-mono-energetic neutron sources (high neutron fluxes at short distances).

The main advantage of inverse kinematics is the natural forward collimation of the reaction ejectiles. This opens up the possibility of placing gamma detectors very close to the source without them being irradiated with source neutrons. In addition, the lack of emission at most angles means that the source is a very low background source.

For reactions which eject neutrons this will induce large enhancements of neutron fluxes at 0 degrees in the laboratory frame. The p(^{7}Li,^{7}Be)n reaction is one of the most commonly used in direct kinematics to produce mono energetic neutrons, especially below 0.7 MeV. However, in inverse kinematics with a Li-beam a mono-energetic neutron of 1.5 MeV is produced at bombarding energies at the reaction threshold of 13.09 MeV. The results of 2-body relativistic kinematics calculations are shown on figure 1. The kinematic curves for a given bombarding energy have two distinct peaks in the laboratory frame corresponding to forward emission and backward emission of neutrons in the centre of mass frame. The relative size of the principal (high energy) peak and satellite (low energy) peak is governed by the relativistic kinematics of the focusing and the angular distribution of emission in the centre of mass frame, which changes quite dramatically as a function of ^{7}Li bombarding energy.

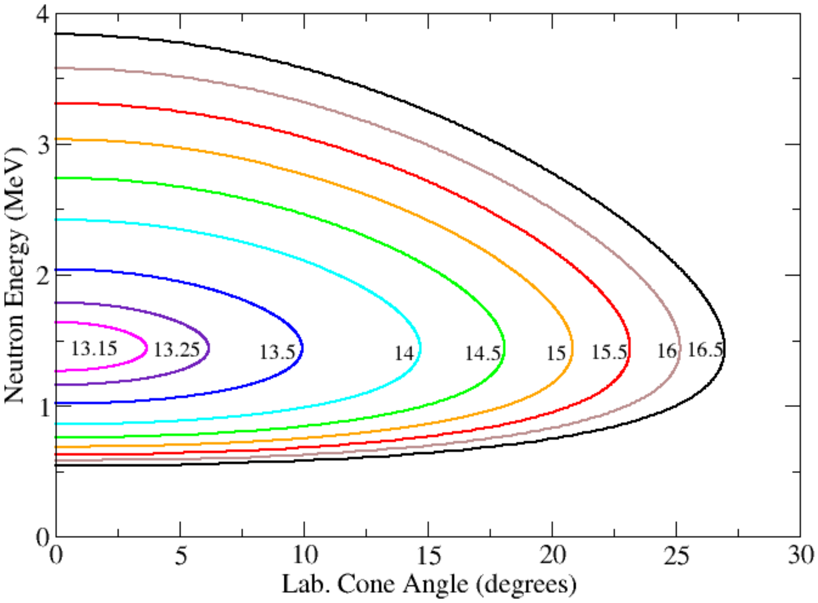

Fig. 1. The series of kinematic curves for the p(^{7}Li,^{7}Be)n inverse reaction as a function of the ^{7}Li bombarding energy. The angle of neutron emission in the laboratory frame is shown on the x-axis, while the energy is shown on the y-axis. Curves for ^{7}Li bombarding energies between 13.15 MeV and 16.5 MeV are shown in different colours.

The gain from the focusing and natural collimation can be expressed in terms of neutron flux enhancement over the non-inverse reaction. Near the threshold the enhancement factor is maximal (> 100) since the emitted neutrons move with the centre of mass of the system, which follows the ^{7}Li beam direction. As a consequence, close to the threshold energy, it is possible to produce very narrow (< 5 degrees) cones of neutrons. With increasing ^{7}Li bombarding energy, the cone broadens and the number of neutrons in a given solid angle decreases so the enhancement factor drops to around 20 at 16.5 MeV.

However, the huge gain in intensity due to the kinematic focusing is offset by corresponding losses from two other factors. Firstly, the available beam current of ^{7}Li is much lower than that available for protons in the non-inverse reaction because of the relative difficulty of extraction of ^{7}Li-ions from the ion source. Secondly, the energy loss of ^{7}Li across a given target will be higher than that for protons due to its higher atomic number.

3. LICORNE converter design

The LICORNE neutron converter sits in an aluminium chamber of diameter approximately 17 cm. It is designed with a rotating polypropylene target wheel of diameter of 8 cm. The rotation is necessary to increase the irradiated surface area by a factor of 25 with respect to a fixed target. Polypropylene is not very resistant to radiation damage, and therefore the rotating target prolongs the lifetime of the target by a similar factor.

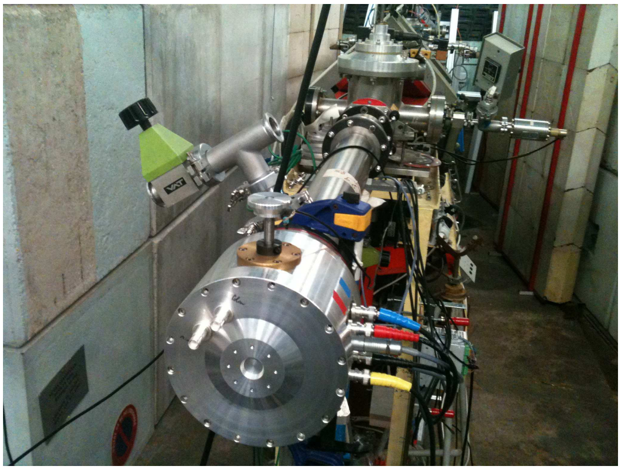

Fig. 2. The LICORNE neutron convertor installed at the Tandem accelerator of the IPN Orsay during commissioning in June 2013.

Between one and ten self-supporting polypropylene targets can be stacked on the target wheel. A version of the target wheel for more fragile targets has three support arms, which are made of tungsten to prevent nuclear reactions with ^{7}Li occurring. The beam current and time structure can be measured at the beam stop, which consists of a 50µm gold foil. This measurement coupled to neutron flux measurement in the experimental area can serve as an online monitoring of hydrogen content in the target and can indicate the appropriate time to change targets before significant quantities of hydrogen are lost.

The exit window of the LICORNE convertor front face is made of aluminium and is only 0.3 mm thick. A mini camera and an illuminating LED are included for beam tuning and inspecting the targets from inside without having to break the vacuum of 10^{-5} bar. The beam spot is tuned by placing a retractable phosphorescent quartz target in the path of the beam to ensure that the beam spot is sufficiently diffuse (typically 8mm diameter). This ensures that the power density in the polypropylene is sufficiently low to keep temperature rises to only a few degrees since the polypropylene is very sensitive to heating with a melting point of 160℃. Macroscopic structural changes almost certainly occur at lower temperatures and it is currently an open question how much the polypropylene deforms and/or becomes thinner under a combination of surface tension, heating effects and radiation damage. What is clear is that under irradiation at the highest primary ^{7}Li currents (> 100 nA) the targets liberate hydrogen from radiation damage to the polymer chains at a significant rate. To maintain the highest neutron fluxes requires that the target be replaced once every few hours. Figure 3 shows the results of a long irradiation at high average beam currents over several days. The polypropylene turns from optically transparent to an opaque brown colour, presumably due to the remaining carbon after sufficient H_2 atoms have been formed and evaporated. The irradiated portion of the surface is brittle and the initial surface tension of the disk is lost creating an overall wrinkled surface. The observed target degradation is the combination of radiation damage, heating and surface tension effects. More precise quantification of these effects needs to be carried out.

Fig. 3. Image of a stack of 8µm polypropylene targets after several days of irradiation at high beam currents.

4. Available fluxes

The fluxes available from the LICORNE source depend principally on the primary ^{7}Li beam current and the polypropylene target thickness. The current sputter source of the IPN tandem can produce primary ^{7}Li beam currents of up to 200 nA, however currents greater than 500 nA may be achievable with source improvements. For the purposes of flux estimations we have assumed 100 nA of primary current which should be easy to maintain over long periods of time. Using a thin polypropylene target, available fluxes are typically 10^7 n/s/steradian, comparable to conventional isotropic neutron sources but with the added complication of a bi-energetic neutron spectrum.

A Monte-Carlo code has been developed for the purpose of predicting neutron spectra as a function of ^{7}Li bombarding energy at any point in space. The Monte-Carlo code was validated by simulating the detected spectra in, EDEN, a liquid scintillator detector placed at different angles with respect to the beam axis. The neutron angular distribution and spectral shapes are well reproduced. This gives us confidence in the predicted spectral shapes seen by samples at short distances and covering larger solid angles.

To obtain the highest neutron fluxes while still preserving the beam directionality a thick polypropylene target can be used. This creates a white neutron source producing a neutron spectrum constrained by the kinematics of the reaction to be between 0.5 and 4 MeV and peaking at 1.5 MeV. For a 28µm thick target the energy of the ^{7}Li ions drops from 16.5 MeV to 13 MeV during their traversal. The principal advantage is that the reaction rate is increased and the total flux available is 7 times higher (approx. 7×10^7 n/s/steradian) than those obtained with the thin targets. If the ^{7}Li beam is pulsed, then time-of-flight can be used at short distances to determine the neutron energy. However, the maximum beam current available in pulsed mode drops by a factor of 4. The energy resolution of such time-of-flight measurements would be dominated by the contribution of the 1.5 ns beam pulse width, so up to 10 energy bins for a flight distance of 20 cm should be possible. The fluxes available with LICORNE are comparable with other accelerator-based neutrons sources although the neutron spectra at the highest fluxes have a broad energy distribution peaking at 1.5 MeV. However, LICORNE has the added advantages of a natural directionality and a much lower background.

5. Spectroscopy of fast neutron induced reactions

Since LICORNE produces neutron cones with opening angles of less than 25 degrees an entire new class of experiments is now possible. High-resolution gamma-ray detectors can be placed around the sample to be irradiated, but outside the neutron cone. It will thus be possible to perform high-resolution spectroscopy of neutron-induced reactions. In 2014, the first coupling of LICORNE and the ORGAM Germanium array will take place. At least two types of future experiment are possible:

(i) Spectroscopy of fission induced by fast neutrons will allow the study of exotic fission fragments from fertile isotopes such as ^{238}U and ^{232}Th. Since pulsed neutron beams are available, individual fission fragments and their partners can be cleanly identified by the time-correlations present in decay of isomeric states. A first experiment will be performed in early 2014 to study production of exotic nuclei from fission of a sample of around 100g of ^{238}U. The fission rates induced should be many tens of kHz.

(ii) Spectroscopy of fast-neutron induced capture reactions could also be performed. The possibility to detect gamma-ray cascades which sum to the nuclear binding energy plus the incident neutron energy will allow the study of nuclear gamma-ray strength functions and nuclear level densities. Gamma ray strength functions describe the way in which the nucleus absorbs and re-emits gamma rays and are known colloquially as the “colour” of the nucleus. While the gamma-ray cascade summing technique has been used successfully to study capture reactions with thermal neutrons, no experimental data are currently available for capture reactions with fast neutrons due to the difficulty of having high fluxes and neutron beam directionality – a problem that LICORNE has now solved.

6. Conclusion

LICORNE is a dedicated facility to produce intense, naturally collimated, quasi-mono-energetic neutron beams at the IPN Orsay. The kinematic focusing of the neutron allows gamma detectors to be placed near the irradiate sample and opens up a whole host of new possibilities for the study of neutron-induced reactions, in particular nuclear fission. The study of gamma-rays emitted in fission is for both fundamental and applied physics purposes. Fission gamma rays are important for the gamma-heating problem in innovative nuclear reactors. Detection of fission gamma rays with arrays of high-resolution germanium detectors will also allow the tagging and study of exotic nuclei far from. To summarize, LICORNE is operational since June 2013 and offers some interesting new experimental possibilities.

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