UTILISATION OF THE STRAY NEUTRON FIELD PRODUCED BY A COMPACT MEDICAL CYCLOTRON

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During the routine isotope production runs at the CYCLONE 30 Medical Cyclotron operated by the Australian Nuclear Science and Technology Organisation (ANSTO) a thick copper substrate electroplated with specific target material (enriched isotope) is bombarded with a 30 MeV proton beam up to 350 μA and thereby resulting in the production of intense prompt evaporation neutrons in the vicinity of the target irradiation station. A novel irradiation rig has been developed to harness these stray neutrons and used in interesting scientific applications such as, thermal neutron activation analysis of archaeological artefacts including metallic and ceramic specimens, investigation of the neutron induced radiation damage in solid state electronic devices and testing of the integrity and remaining life of superconducting materials exposed to intense neutron environment.

1. Introduction

Since July 1991 the Radiopharmaceutical Division of the Australian Nuclear Science and Technology Organisation (ANSTO) has operated a 30 MeV H^- ion Medical Cyclotron (Model: CYCLONE 30, Manufacturer: Ion Beam Applications, Louvain La Neuve, Belgium) for the production of various important PET (Positron Emission Tomography) and SPECT (Single Photon Emission Computerised Tomography) radioisotopes for the Australian Nuclear Medicine community[1].

During the isotope production run at a cyclotron a copious number of fast neutrons are produced near the target. A method of neutron activation analysis using the un-moderated neutron field produced near the internal target of a positive ion cyclotron (Model CS 30, Manufacturer: The Cyclotron Corporation, USA) has been reported elsewhere[2]. Due to its high beam extraction efficiency (~100%) the negative ion cyclotron like the CYCLONE 30 is capable of producing a high proton beam current (~350 μA) on targets located in separate target vaults[3]. The bombardment of target plates (usually copper) with high intensity proton beam produces a high flux of fast evaporation neutrons. At our facility a neutron fluence of ~5.8 × 10^7 [$n.cm^{-2}.s^{-1}\mu A^{-1}$] with a median energy of ~2.5 MeV at 10 cm from the target was estimated with cobalt activation foils[4]. A multipurpose neutron irradiation device utilising these stray neutrons has been developed[4,5]. This paper highlights the interesting applications of the neutron irradiation device.

2. Material and Methods

2.1 Fast Neutron Irradiation Device

A fast neutron irradiation device developed at our laboratory was used for the irradiation of solid state electronic devices and superconducting materials. A frame (50cm × 10cm × 10cm) made of radiation resistant PVC (Polyvinyl chloride) sheet with height adjustable racks was secured on the target irradiation station located in the target cave adjacent to the cyclotron vault[3]. Five cobalt (^{59}Co) activation pellets (5mm diameter, 2mm thick) M1, M2, M3, M4 and M5 were attached to the centre of the racks placed at 10cm, 20cm, 30cm, 40cm and 50cm from the reference point T (proton beam impingement spot on the target) respectively (Figure 1). After the routine isotope production run (a collimated 30 MeV proton beam hitting the copper target) the Co-pellets were retrieved and gamma spectra were analysed using a High Purity Germanium detector interfaced to 8000 Channel Analyser. The area under the 811 keV photo peak of ^{58}Co produced in the pellets via the fast neutron induced $^{59}Co(n,2n)^{58}Co$ reaction with the neutron threshold energy[6] of 11 MeV was taken into account. The fast neutron fluence per unit beam current [$n.cm^{-2}s^{-1}\mu A^{-1}$] was calculated[5] and shown as function of distance from the copper target (Figure 2). At 10 cm from the target (pellet M1 in Figure 1) the fluence was evaluated to be 5.8×10^7 [$n.cm^{-2}s^{-1}\mu A^{-1}$]. The total fast neutron fluence [$n.cm^{-2}$] could be calculated by integrating the proton beam current recorded by the Health Physics Watchdog[1] over the entire target bombardment period (Figure 3).

2.2 Thermal Neutron Irradiation Device

A thermal neutron irradiation device was used for activation analysis of small metallic objects and ceramic specimens. A high density (ρ = 0.98 $g.cm^{-3}$) polyethylene moderating cylinder (30cm diameter, 35 cm high) was attached to a 2cm thick iron base plate and placed on the target irradiation station (Figure 1). The iron plate was deployed in order to slow down the primary neutrons (E_n ~2.5 MeV) by inelastic scattering. A polyethylene plug (5cm diameter, 20cm long) with a nylon vial attached to its tip was used as sample holder. A bare cobalt pellet and a pellet enclosed in a small cadmium box of 2mm wall thickness were placed into the vial and irradiated with neutrons during isotope production run. The activity of ^{60}Co produced in the pellets via the thermal neutron capture reaction $^{59}Co(n,\gamma)^{60}Co$ was assayed by gamma spectroscopy as described in earlier section. The thermal neutron fluence and cadmium ratio at the centre of the moderator were assayed to be 3.0×10^6 [$n.cm^{-2}s^{-1}\mu A^{-1}$] and 25 respectively[4]. The total thermal neutron fluence [$n.cm^{-2}$] could be calculated by integrating the proton beam

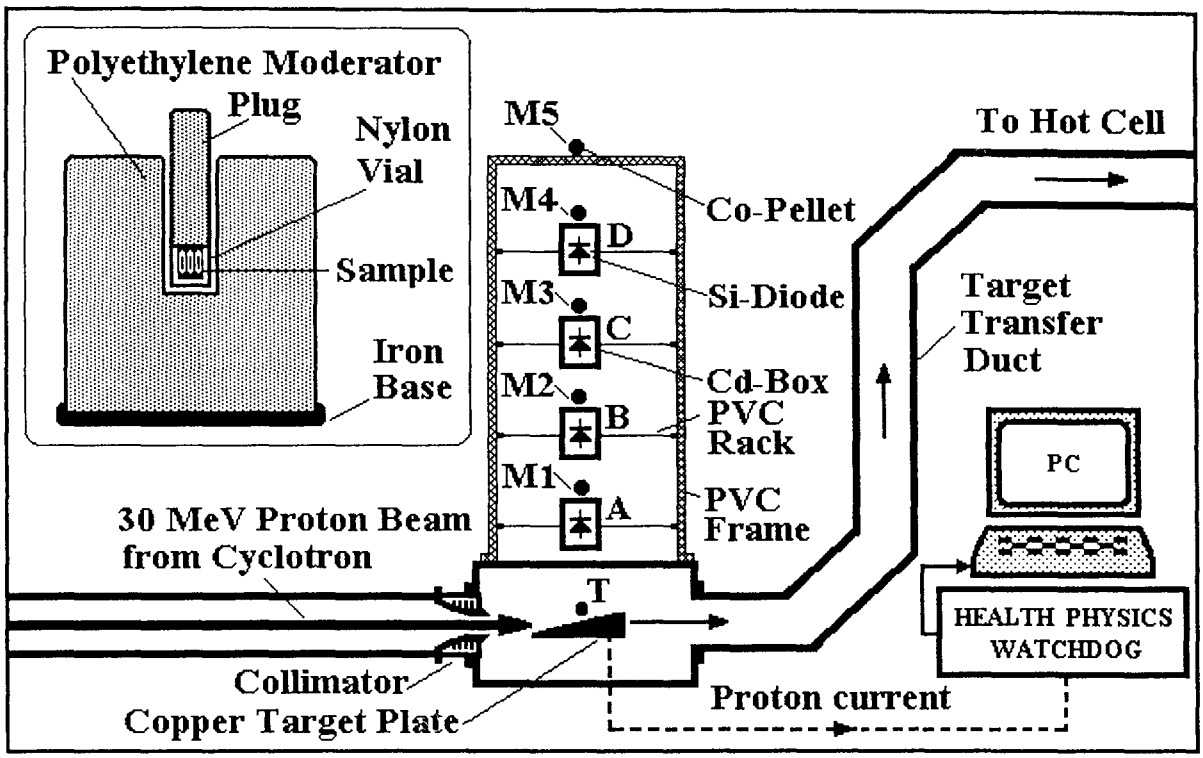

Figure 1: Schematic diagram of the neutron irradiation facility utilising the stray neutron field produced during routine operation of the CYCLONE 30 Medical Cyclotron. The figure shows the fast neutron irradiation rig placed on the target station The thermal neutron irradiation device is shown inset. The operation principle of the system is explained in the text.

current recorded by the Health Physics Watchdog[1] over the entire target bombardment period (Figure 3).

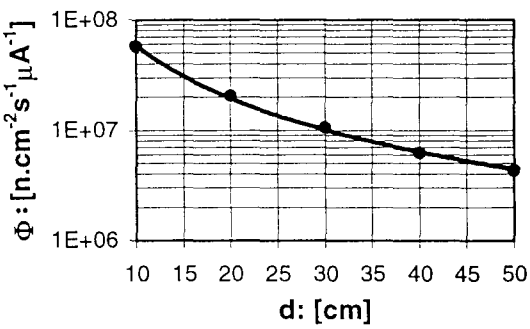

Figure 2: Showing the fluence of fast neutrons Φ evaluated with cobalt activation pellets as a function of distance from the copper target plate (Figure 1).

3. Applications

3.1 Radiation Damage Investigation of Silicon Diodes

Performance of solid state electronic devices deteriorate while operating in the radiation environment. In particular, the radiation detectors exposed to fast neutrons during their working life suffer a gradual loss of detection efficiency caused by the displacement damage of the bulk silicon material. Hence, the increasing leakage current of a reverse biased silicon diode (irradiated) is characterised as an indicator of the radiation damage[7]. The common silicon (pn) junction diodes constitute the basic building block of semiconductor radiation detectors, discrete transistors and integrated circuits (IC). In order to predict the radiation hardness of larger and complex semiconductor devices for neutrons the leakage currents of silicon diodes were measured after being irradiated with well known levels of neutron fluence in controlled conditions at the above facility.

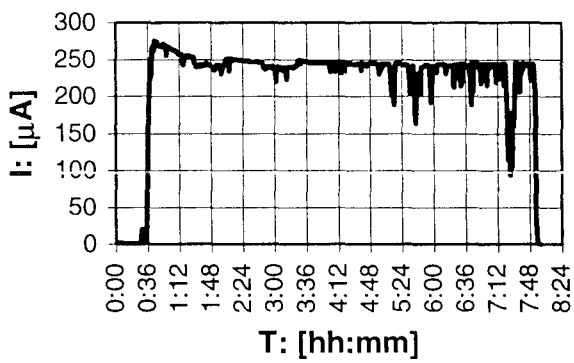

Figure 3: Showing the proton beam current I at the target as a function of target bombardment time T during a ca. 8 hour isotope production run. The data was sampled by the Health Physics Surveillance System (Watchdog) every minute.

Four batches of small signal silicon diodes (IN 4007) with 5 diodes per batch were packed in small cadmium boxes (2mm wall thickness) and placed at positions 10cm (batch A), 20cm (batch B), 30cm (batch C), and 40cm (batch D), form the target (T) in the neutron irradiation rig (Figure 1). The fifth batch was kept as control. Batch A, B, C and D were exposed to the integrated neutron fluence levels of 2.89×10^{15} [n.cm^{-2}], 5.50×10^{14} [n.cm^{-2}],

3.18×10^{14} [n.cm^{-2}] and 2.03×10^{14} [n.cm^{-2}] respectively (Figure 2).

The leakage current (nA) of the diodes were measured at room temperature (22 °C) at the reverse bias of 5V, 10V, 20V, 50V and 100V using a Pico Ammeter (Model: 617, Manufacturer: Keithley Instruments, USA) and a Digital Voltmeter (Model: 34401A, Manufacturer: Hewlett Packard, USA). The leakage currents of neutron irradiated diodes and control are shown as functions of reverse voltage in Figure 4.

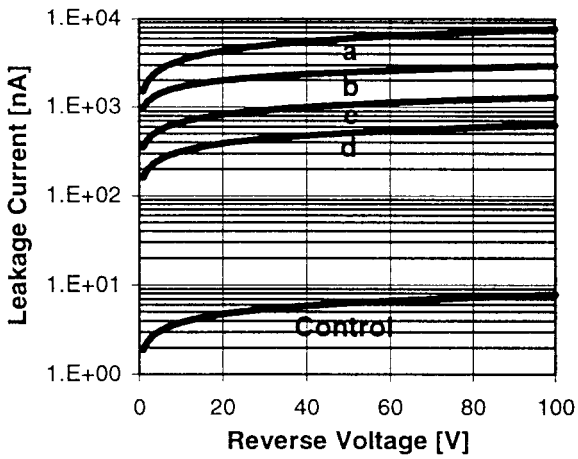

Figure 4: Showing the average reverse (leakage) current of the control (un irradiated) batch and four batches of silicon diodes irradiated with stray fast neutrons produced in the target irradiation station of the CYCLONE 30 Medical Cyclotron with the integrated fluence of (a) 2.89×10^{15} [n.cm^{-2}], (b) 5.50×10^{14} [n.cm^{-2}], (c) 3.18×10^{14} [n.cm^{-2}] and (d) 2.03×10^{14} [n.cm^{-2}] as functions of reverse bias voltage.

3.2 Radiation Damage Investigation of Superconducting Material

In a superconducting cyclotron the superconducting magnet coils carry currents up to 1000 amperes (DC) to produce a field strength of ca. 5 Tesla[8]. The superconductors are usually made of a bundle of thin filaments (ca. 50 μm diameter) of Niobium-Titanium alloy firmly enclosed in a high purity copper stabilisation matrix and refrigerated at the liquid helium temperature (~ 4.2 °K). When energised a very high electromagnetic force (Lorentz force) is generated in the coil and consequently the filaments tend to move apart. The heat generated due to friction caused by the minute movement of the filaments could raise the temperature of the superconducting wire sufficient enough to cause the loss of superconductivity (quenching)[9]. Therefore, the superconducting coils are embedded in a special epoxy resin (trade name: STYCAST) to prevent such movement. In many situations the superconducting coils are exposed to fast neutron fields[8]. Hence, it becomes imperative to estimate the effects of neutron exposure on the consistency and mechanical rigidity of the epoxy material. Three batches of sample each consisting of two sections (20 mm × 20 mm × 5mm) of the epoxy resin[8] prepared at the NSCL were enclosed in small cadmium boxes (2 mm wall thickness) and installed in the neutron irradiation facility. Samples 1 and 2 were placed at 10 cm (A) and sample 3 at 20 cm (B) from the neutron producing target (Figure 1) and irradiated. The irradiation results are summarised in Table 1. The "Year Equivalents" of the neutron exposure, expressed as the ratio of the neutron fluence in the sample and the predicted[10] integrated neutron fluence in a year (4.14×10^{14} n.cm^{-2}/yr) are also presented.

Table 1: Showing the position of the three coil samples, distances from the target (d), average proton current (I), irradiation time (T), integrated neutron fluence (Φ) and the corresponding Year Equivalent (YE) of neutron exposure. The un irradiated sample (Nr. 4) was kept as control.

Nr.	d [cm]	I [μA]	T [hr]	Φ [n.cm^{-2}]	YE [yr]
1	10	263	141	7.7×10^{15}	19
2	10	250	85	4.4×10^{15}	11
3	20	250	85	1.6×10^{15}	4
4	n.a.	n.a	n.a.	0	0

The mechanical integrity of the coil sections will be tested at the Magnet Laboratory of the NSCL shortly. The gamma spectrum of the epoxy samples recorded using a High Purity Germanium detector interfaced to a 8000 channel analyser after a cool down period of 21 days is shown in Figure 5.

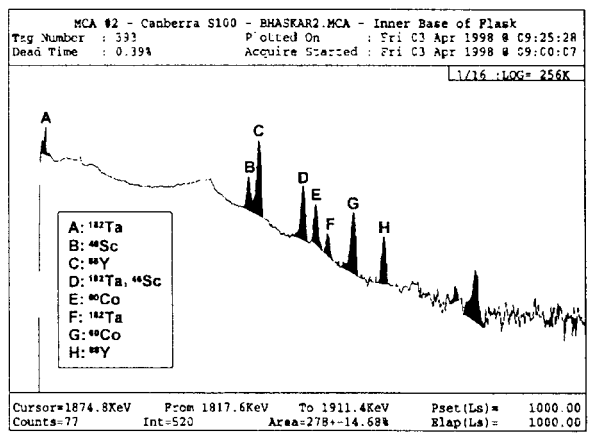

Figure 5: The gamma spectrum of a section of a superconducting-magnet coil embedded in epoxy resin irradiated with stray fast neutrons produced during routine radioisotope production process at CYCLONE 30 Medical Cyclotron. The spectrum was assayed after a three weeks cool down period. The energies of the photo-peaks A, B, C, D, E, F, G and H were evaluated to be 67 keV, 889 keV, 935 keV, 1121 keV, 1173 keV, 1222 keV, 1333 keV and 1848 keV respectively. The trace elements identified and the corresponding photo-peak energy are shown inset.

3.3 Activation Analysis of Archaeological Artefacts

Neutron Activation Analysis (NAA) using reactor neutrons (thermal) is a well established technique for the non-destructive analysis of trace elements in various materials. Activation analysis using stray neutrons produced in the internal targets of a positive ion cyclotron is described elsewhere[2]. The thermal neutron irradiation facility developed at the CYCLONE 30 negative ion cyclotron of ANSTO was used for trace element analysis of archaeological artefacts such as ancient Roman bronze coins[11] and Greek pottery[12].

Table 2: Showing the important nuclear data and the activity concentration of trace elements in the archaeological ceramic samples S1, S2 and S3 and Ohio Red Clay (ORC) Standard identified by thermal Neutron Activation Analysis (NAA) at the CYCLONE 30 Medical Cyclotron Facility during routine isotope production operations. The samples were irradiated with thermalised stray neutrons produced during routine isotope production operation at the Medical Cyclotron. The irradiated masses of the Standard (Ohio Red Clay) and the ceramic samples S1, S2 and S3 were 160 mg, 600 mg, 640 mg and 62 mg respectively.

					Activity Concentration [counts $s^{-1}mg^{-1}$]			
Trace Element	Nuclide Identified	$T_{1/2}$ [hours]	Eγ [keV]	Intensity [%]	ORC	S1	S2	S3
Na	^{24}Na	15.2	1369.6	99.9	6.92E-2	2.99E-1	3.89E-1	7.36E-1
K	^{42}K	12.4	1525.8	17.9	1.74E-2	6.03E-3	9.51E-3	2.05E-2
Sc	^{46}Sc	2016	889.7	99.9	8.69E-3	6.58E-3	8.89E-3	1.06E-2
Cr	^{51}Cr	665	319.6	9.8	1.04E-3	4.09E-3	6.03E-3	1.57E-3
Fe	^{59}Fe	1071	1292.1	43.1	2.13E-3	1.58E-3	2.07E-3	2.01E-3
As	^{76}As	26.3	559.0	44.7	4.69E-3	1.07E-3	3.09E-3	4.42E-3
La	^{140}La	40.2	1597.4	95.5	1.68E-2	6.90E-3	1.09E-3	1.91E-2
Ce	^{141}Ce	780	144.8	48.5	4.63E-3	1.21E-3	1.92E-3	5.11E-3
Sm	^{153}Sm	46.7	103.3	28.3	8.64E-2	3.72E-2	5.76E-2	1.09E-1
Yb	^{175}Yb	101	396.3	6.5	1.56E-3	4.47E-4	1.00E-3	--

Three archaeological ceramic samples (fragments) collected from ancient Greek vases (S1, S2 and S3) and the Ohio Red Clay (ORC) standard[13] (in powder from) received from the School of Archaeology at the University of Sydney were encapsulated in small nylon sample holders. The outer surfaces of the sample holders were cleaned with acetone, placed in the nylon vial and inserted in the thermal neutron irradiation rig (Figure 1 inset). The samples were irradiated during a routine 20 hour isotope production run. The target current was recorded every minute by the Health Physics Surveillance System[4]. The integrated thermal neutron fluence for the average proton beam currant of 250 μA was calculated to be 5.4×10^{13} n.cm^{-2}.

After the cyclotron shut down the sample holders were retrieved from the PVC moderator and each holder was sealed in clean plastic bag. The gamma spectra of the samples were evaluated with a 30 cc High Purity Germanium (HPGe) detector interfaced to a 8000 Channel Analyser. All samples were counted for 2000 seconds and the results are summarised in Table 2.

4. Summary and Discussion

In this paper the operation principle and application examples of a multipurpose neutron irradiation facility developed at the CYCLONE 30 Medical Cyclotron facility operated by ANSTO is presented. The intense parasitic neutron field produced by the cyclotron target when bombarded with the 30 MeV/~250 μA proton beam during the routine isotope production operation has been harnessed. The spectral hardness and intensity distribution of the neutron field were estimated with cobalt activation pellets. The primary proton beam current on the target was monitored in real-time with the Health Physics Surveillance System and used to evaluate the integrated neutron fluence accurately. The fast neutron field with a median energy of ~ 2 MeV was used to investigate the radiation hardness properties of semiconductors and remaining life analysis of the superconducting materials operating in intense neutron environments. The thermal neutron irradiation device was used for activation analysis of archaeological samples including metallic and ceramic artefacts.

Acknowledgements

The author wishes to thank Dr. P. Cross, Radiation Oncology Department, St. Vincent's Hospital Sydney, Australia and Dr. R. Ronningen and Dr. A. Zeller, National Superconducting Cyclotron Laboratory, Michigan State University, East Lansing, USA for their comments.

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