〈Technical Note〉

ANALYSIS OF ADHESIVE TAPE ACTIVATION DURING REACTOR FLUX MEASUREMENTS

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Several adhesive tapes have been studied in terms of their suitability for securing gold wires into positions for neutron flux measurements in the reactor core and irradiation facilities surrounding the core of the Open Pool Australian Light water (OPAL) reactor.

Gamma ray spectrometry has been performed on each irradiated tape in order to identify and quantify activated components. Numerous metallic impurities have been identified in all tapes. Calculations relating to both the effective neutron shielding properties of the tapes and the error in measurement of the ^{198}Au activity caused by superfluous activity due to residual tape have been made.

The most important identified effects were the prolonged cooling times required before safe enough levels of radioactivity to allow handling were reached, and extra activity caused by residual tape when measured with an ionisation chamber. Knowledge of the most suitable tape can allow a minimal contribution due to these effects, and the use of gamma spectrometry in preference to ionisation chamber measurements of the flux wires is shown to make all systematic errors due to the tape completely negligible.

KEYWORDS : Neutron Flux, Reactor Commissioning, Hot Commissioning, ^{198}Au, Activation Measurements, Adhesive Tape

1. INTRODUCTION

The OPAL Reactor is Australia's replacement research reactor and will provide services such as medical isotope production, neutron transmutation doped Si irradiation, as well as serving as a neutron source for experiments involving neutron diffraction. An accurate determination of the neutron flux not only in the core but also in these service facilities is therefore quite important. The measurement of thermal neutron flux using the reaction ^{197}Au(n,γ)^{198}Au is a standard experiment that has been thoroughly investigated and widely implemented [1]. This reaction is convenient because of the well known reaction cross section and g-factor close to 1, the relatively long half life of gold, and the lack of complications caused by the fact that gold is naturally mono-isotopic [2].

A common technique used in the mounting of samples to be irradiated in a neutron field is to use adhesive tape. These tapes are used to ensure the Au flux monitor probes stay in a fixed position for the duration of the irradiation. They may also allow small samples to be attached to pre-existing rigs or structures with minimal protrusion and difficulty. This work intends to quantify for the first time any errors caused by the neutron activation of adhesive tape. An informed choice of tape can allow a minimal error contribution to systematic error, and shorter cooling times to facilitate measurement of the flux wires.

2. METHOD

Several candidate tapes were studied, namely; *Ajec Autoadhesivos* brown packing tape, *Norton Abrasives* brown packing tape, a clear plastic tape of unknown origin, *Scotch* medium-high adhesion masking tape, a black electrical tape of unknown origin, and white polyethylene coated cloth duct tape manufactured by *Scapa*. These tapes will herein be referred to as brown packing 1, brown packing 2, clear plastic, masking, black electrical and white duct respectively. The tapes were adhered to cylindrical Silicon ingots with a diameter of 6 inches that were subsequently placed into the Si doping facility of the reactor.

Each irradiation was of 2 hours and 1 minute duration at 20 MW reactor power. The neutron flux was monitored

using approximately 100 gold wires diluted with aluminium that were attached to the same Si target. Approximately 10 of these gold wires were covered with cadmium to determine the ratio of thermal to epithermal neutrons. The final flux calculations contained a correction to account for the additional neutron flux received whilst lowering and raising the target rig.

The irradiation facility ensured an even radial distribution of the neutron flux over the surface of the flux wire rig by continuous rotation of the ingot. Several irradiations were performed with the different tapes, and the irradiation time was adjusted to account for the additional flux received when lowering the ingot into the reactor pool. Upon irradiation, the gamma ray spectrum of the tape was measured using an *ORTEC* GMX series high purity germanium (HP Ge) coaxial gamma ray detector in a lead shielded environment to suppress background activity. This system used an *ORTEC* resistive feedback pre-amplifier and conventional analogue electronic modules from *Canberra* (2026 amplifier and Multiport II MCA) without pile up rejection.

Gamma spectra were acquired initially for 1 hour from each tape to provide useful half-life information. Typical acquisition times for samples were in the order of 12 hours thereafter. A Gaussian fitting procedure was used for the analysis of the spectral peak areas.

3. RESULTS

A typical gamma spectrum for one of the activated tapes is displayed in fig. 1. As is immediately obvious, quite a few peaks are present in the spectrum corresponding to several activated components. The spectral peaks were analysed and identified with the aid of activity data taken from the online Lawrence Berkeley National Laboratory database [3].

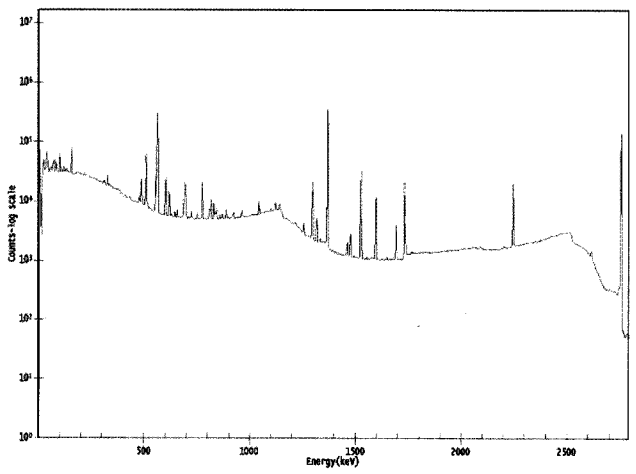

Fig. 1. Gamma Ray Spectrum for the Activated White Duct Tape Sample

Using this data, a total of 24 radioisotopes were positively identified in the gamma spectra of the 6 different types of tape. Using the spectra and the known efficiency data (measured in point source geometry) for the gamma spectrometer, approximate activities of the various components of the tape were calculated for the activity present at the end of irradiation using equation (1), which is corrected for decay during measurement time [4]:

$$A_0 = \frac{C\lambda \exp(\lambda t_{decay})}{P_\gamma \varepsilon_\gamma (1 - \exp(-\lambda t_{count}))} \quad (1)$$

where C is the number of counts in the peak of the nuclide with decay constant λ, and with relative probability of emission P_γ. The efficiency for the HP Ge detector to used detect the gamma line of interest is designated ε_γ, t_{decay} is the time between the end of irradiation and beginning of measurement, and t_{count} is the real time taken to perform the spectrum measurement. The population of radionuclides required to give such an activity at the end of the irradiation was then calculated using the familiar expression:

$$N_0 = \lambda^{-1} A_0 \quad (2)$$

Table 1 outlines the most important radionuclides in terms of activity and associated population for each adhesive tape studied. To facilitate comparison between the tapes, allowance was made for the different masses and fluxes received by the tapes by dividing the decay corrected activity and the population of nuclides at the end of the irradiation by the flux received and the mass of each tape. Obviously each type of tape has a different mass per unit surface area, thus each layer of tape used to secure the flux monitor wires may present a variable number of nuclei to the neutron flux field depending on that planar density. It may then seem instructive to multiply the results in Table 1 by the planar density of each tape to make a more dependable figure of merit for each tape. This was not done, however, as it was realised the tapes with the larger planar densities (such as the electrical tape and white duct tape) required less layers to secure the flux monitor wires than did the tapes with the smaller planar densities.

All uncertainties presented were calculated according to the procedures set forth in the International Standards Organisation Guide to the Expression of Uncertainty in Measurement [5]. Uncertainties in the results displayed in table 1 are quite low, typically less than 5% except for the nuclides which have low concentration in the tape and/or low induced activity. An exception to this is the masking tape, where there is quite a large uncertainty

Table 1. Summary of the Most Important Nuclides Present in Each Adhesive Tape as Well as Their Activity and Associated Concentration

Tape	Nuclide	Specific Activity at end of irradiation per unit flux per unit mass (Bq.cm^2.s/mg)	% of total	Concentration of radionuclides at end of irradiation per unit flux (1/mgcm^{2}s)	% of total
Brown Packing 1	**All**	**$(1.01 \pm 0.02)\times10^{-8}$**	**100**	**$(8.3 \pm 0.2)\times10^{-4}$**	**100**
	^{24}Na	$(1.01 \pm 0.02)\times10^{-8}$	99.911	$(7.8 \pm 0.2)\times10^{-4}$	94.20
	^{59}Fe	$(4.4 \pm 0.6)\times10^{-12}$	0.043	$(2.42 \pm 0.03)\times10^{-5}$	2.91
	^{51}Cr	$(1.69 \pm 0.09)\times10^{-12}$	0.017	$(5.8 \pm 0.3)\times10^{-6}$	0.70
	^{60}Co	$(5.6 \pm 0.9)\times10^{-14}$	0.001	$(1.3 \pm 0.2)\times10^{-5}$	1.60
Brown Packing 2	**All**	**$(5.09 \pm 0.07)\times10^{-10}$**	**100**	**$(4.69 \pm 0.09)\times10^{-5}$**	**100**
	^{24}Na	$(5.01 \pm 0.07)\times10^{-10}$	98.489	$(3.89 \pm 0.06)\times10^{-5}$	83.07
	^{122}Sb	$(5.0 \pm 0.2)\times10^{-12}$	0.978	$(1.69 \pm 0.07)\times10^{-6}$	3.60
	^{69m}Zn	$(8.0 \pm 0.1)\times10^{-13}$	0.157	$(6 \pm 2)\times10^{-8}$	0.12
	^{59}Fe	$(4.3 \pm 0.4)\times10^{-13}$	0.085	$(2.4 \pm 0.3)\times10^{-6}$	5.10
White Duct	**All**	**$(1.37 \pm 0.02)\times10^{-10}$**	**100**	**$(1.37 \pm 0.03)\times10^{-5}$**	**100**
	^{24}Na	$(6.0 \pm 0.1)\times10^{-11}$	43.661	$(4.66 \pm 0.09)\times10^{-6}$	33.87
	^{42}K	$(5.94 \pm 0.09)\times10^{-11}$	43.290	$(3.81 \pm 0.09)\times10^{-6}$	27.75
	^{152m}Eu	$(1.01 \pm 0.01)\times10^{-11}$	7.350	$(4.88 \pm 0.07)\times10^{-7}$	3.55
	^{122}Sb	$(4.3 \pm 0.2)\times10^{-12}$	3.167	$(1.47 \pm 0.07)\times10^{-6}$	10.74
	^{124}Sb	$(1.03 \pm 0.08)\times10^{-13}$	0.075	$(7.7 \pm 0.7)\times10^{-7}$	5.62
Electrical	**All**	**$(3.02 \pm 0.04)\times10^{-10}$**	**100**	**$(2.39 \pm 0.04)\times10^{-5}$**	**100**
	^{24}Na	$(2.57 \pm 0.04)\times10^{-10}$	84.987	$(1.99 \pm 0.03)\times10^{-5}$	83.46
	^{42}K	$(4.06 \pm 0.05)\times10^{-11}$	13.457	$(2.61 \pm 0.06)\times10^{-6}$	10.92
	^{115}Cd	$(4.12 \pm 0.09)\times10^{-12}$	1.365	$(1.14 \pm 0.03)\times10^{-6}$	4.79
	^{82}Br	$(2.25 \pm 0.08)\times10^{-13}$	0.075	$(4.1 \pm 0.2)\times10^{-8}$	0.17
Clear Plastic	**All**	**$(4.56 \pm 0.06)\times10^{-10}$**	**100**	**$(3.74 \pm 0.06)\times10^{-5}$**	**100**
	^{24}Na	$(4.55 \pm 0.06)\times10^{-10}$	99.667	$(3.53 \pm 0.06)\times10^{-5}$	94.48
	^{82}Br	$(9.2 \pm 0.2)\times10^{-13}$	0.202	$(1.68 \pm 0.05)\times10^{-7}$	0.45
	^{51}Cr	$(5.4 \pm 0.6)\times10^{-13}$	0.119	$(1.9 \pm 0.2)\times10^{-6}$	5.03
Masking	**All**	**$(2.29 \pm 0.03)\times10^{-10}$**	**100**	**$(2.2 \pm 0.3)\times10^{-4}$**	**100**
	^{24}Na	$(1.24 \pm 0.02)\times10^{-10}$	53.891	$(9.6 \pm 0.2)\times10^{-6}$	4.40
	^{69m}Zn	$(8.7 \pm 0.1)\times10^{-11}$	38.012	$(6 \pm 2)\times10^{-6}$	2.86
	^{42}K	$(7.9 \pm 0.1)\times10^{-12}$	3.466	$(5.1 \pm 0.1)\times10^{-7}$	0.23
	^{65}Zn	$(6.6 \pm 0.9)\times10^{-12}$	2.869	$(2.0 \pm 0.3)\times10^{-4}$	91.84
	^{82}Br	$(2.71 \pm 0.07)\times10^{-12}$	1.183	$(4.9 \pm 0.1)\times10^{-7}$	0.23

(15%) associated with the concentration per unit flux of ^{65}Zn, which constitutes 91.84% of the total tape composition. This is due to the fact that ^{65}Zn has a small thermal neutron capture cross section and relatively long half life, so that the activity induced by capture of neutrons in this nuclide is quite low. Thus, the large uncertainty in the concentration per unit flux can be traced to low count rate statistics. The systematic uncertainty of the differing geometry of

Table 2. Decay Data for ^{24}Na and ^{42}K [3]

Nuclide	Beta endpoint energy (keV)	Beta emission probability	Half life (Seconds)
^{24}Na	280.58	0.053% ± 0.004%	(53852.4 ± 4.3)
	1392.91	99.944% ± 0.004%	
^{42}K	1688.1	0.34% ± 0.03%	(44496 ± 108)
	2000.67	17.64% ± 0.09%	
	3525.4	81.90% ± 0.09%	

Table 3. Effective Half Life for Each Adhesive Tape

Tape	τ_{eff} (seconds)
Brown Packing 1	$(5.4 \pm 0.2)\times10^4$
Brown Packing 2	$(5.4 \pm 0.5)\times10^4$
White Duct	$(4.9 \pm 0.4)\times10^4$
Electrical	$(5.3 \pm 0.1)\times10^4$
Clear Plastic	$(5.4 \pm 0.2)\times10^4$
Masking	$(5.4 \pm 0.5)\times10^4$

the tape to the efficiency calculation has been neglected since the considerable amount of calculations [6] required to account for this effect was beyond the scope of this work.

4. ANALYSIS OF SYSTEMATIC ERRORS DUE TO TAPE ACTIVATION

The two quantities listed in Table 1 may be used to analyse the effects that influence the results obtained. The number of nuclides produced per unit flux may be used to evaluate a correction to the neutron flux due to the neutrons absorbed by the tape. This correction turns out to be very small. The largest effect is in brown packing 1, which had at the end of its irradiation a population of $(9.24 \pm 0.04)\times10^{11}$ radionuclides. This corresponds to, over the course of the 2 hour and 1 minute irradiation, a neutron loss of $(1.3 \pm 0.1)\times10^7 cm^{-2}s^{-1}$ over the surface of the 10.08 cm^2 sample. Therefore, in a neutron field of $3\times10^{12} cm^{-2}s^{-1}$ (which is typical of the irradiation facilities surrounding the core) only 0.00043% of the neutrons are lost in the tape. Note that this analysis assumes that only one neutron per radionuclide is required to produce the observed population of radionuclides. This is an underestimation, as it discounts radionuclides that were created and subsequently decayed before the end of the irradiation and the stable nuclides produced by neutron capture reactions. Nonetheless, it is certainly quite reasonable to treat the effect of absorption of neutrons within the tape as negligible.

The results for the specific activity in Table 1 are important for considerations relating to the post-irradiation cooling time for radiological protection. It has been observed that the tape was the most active component of the irradiated flux wire rig upon removal of the setup from the cooling pool for processing. The largest component to the dose was due to beta decay. ^{24}Na and ^{42}K – the most significant nuclides for activity in most samples – both emit high energy beta particles with a high probability. Thus the tape was the limiting factor on the length of the cooling time, which was typically in the order of 48-72 hours to give dose levels in the order of 1mSv/hr at zero distance from the tape. The length of cooling time was determined by the reactor radiation protection staff.

To properly account for the elemental composition of the tape and its effect on activity and cooling time, the effective half-life needs to be considered. Table 3 reports the 'effective half-life' of each tape based on the populations of the various radionuclides present which is given by the expression:

$$\frac{1}{\tau_{eff}} = \sum_i \frac{x_i}{\tau_i} \tag{3}$$

where x_i is the activity fraction of the ith radionuclide present in the sample, and τ_i is the half-life of the ith radionuclide.

The brown packing 1, brown packing 2, and clear plastic tapes all had similar effective half lives due to their similar composition. Their effective half-lives were all comparable to the ^{24}Na half life of 5.4×10^4 seconds which is the dominant radionuclide in each of these samples. The electrical and white duct tapes had slightly shorter effective half-lives than ^{24}Na due to the relatively larger activity fraction of ^{42}K present. Although ^{65}Zn has a high nuclide concentration of ^{65}Zn (half-life; 2.1×10^7 seconds), its effective half-life is comparable to ^{24}Na due to the high activity concentration of this radionuclide.

During the cooling time, the flux wires undergo over

one half-life of decay. Ideally, the ^{198}Au wires would constitute the largest component of activity in the irradiated setup to allow counting to be carried out as soon as possible within dose constraints. This is especially important for flux measurements involving a large number of gold samples to prevent the last wires to be measured decaying beyond the levels of activity measurable to the detector.

Any residual tape that is left on the wires may contribute to the measurement of the activity of the wires. The amount of residual tape present was measured for 9 wires that were covered by the brown packing 1 sample by first measuring the mass of the irradiated wires, cleaning them in an ultrasonic bath of xylene, washing with methanol followed by deionised water, drying, then weighing them again. The average mass difference due to the residual tape was 0.0934 mg. The standard deviation of the mass difference was 0.0632 mg. The greatest mass difference measured was (0.1826 ± 0.0001) mg. In order to provide an upper limit of the systematic error caused in the measurement by the residual tape, this largest of the mass differences will be used as the mass of the residual tape, and the highest measured flux of the reactor of $(2.30 \pm 0.01)\times10^{13}$ n/cm^{2}s will also be used.

There are two common ways of measuring the activity of the flux monitor wires – by gamma spectrometry or ion chamber measurements [2, 7]. Gamma spectrometry is the least affected by the residual tape, as it only measures the activity of the 411.8 keV ^{198}Au photopeak and the Compton distribution due to the many other peaks of higher energy than the ^{198}Au photo-peak. The Compton contribution to this method is unimportant, as it tends to only raise the measured background level, which is discarded in the peak analysis procedure. The relative contribution to the activity measurement of the ^{198}Au impurities in the tapes are summarised in Table 3. At worst, over the course of one hour, and assuming 100 percent efficiency, the contribution would lead to one additional count - clearly the effect is very small.

An alternate method for measuring the activation of the flux wires is by using an ionisation chamber. In this case, any activity that arises from the residual tape has the potential to contribute to the ionisation current. Table 4 compares the activities that would result from the tapes under 'worst case' conditions, i.e., after 48 hours of

Table 4. Analysis of ^{198}Au Content of Adhesive Tapes Studied. The 'Resultant Number of Decays' Value Expresses the Number of 411.8 keV Photopeak Decays that Would be Expected to Occur Over the Course of Measurement Assuming the Worst Conditions that Could be Reasonably Expected – i.e. the Largest Measured Residual Mass (0.1826 mg) and Maximum Neutron Flux (2.30×10^{13} n/cm^{2}s); All for Bare Wire Samples

Tape	^{198}Au activity at end of irradiation per unit flux per unit mass (Bq.cm^2.s /mg)	Resultant number of decays for 'worst case'
Brown Packing 1	$(7.0 \pm 0.9)\times10^{-17}$	1.0 ± 0.1
Brown Packing 2	$(5 \pm 2)\times10^{-18}$	0.07 ± 0.04
White Duct	$(2.7 \pm 0.6)\times10^{-18}$	0.040 ± 0.009
Electrical	$(2.8 \pm 0.2)\times10^{-17}$	0.42 ± 0.03
Clear Plastic	$(2.8 \pm 0.5)\times10^{-17}$	0.41 ± 0.08
Masking	$(3.3 \pm 0.5)\times10^{-17}$	0.49 ± 0.08

Table 5. Expected 'Total' Activities for the Different Tapes Based on the Effective Half Lives Given in Table 2 and Assuming the Worst Conditions that Could be Reasonably Expected – i.e. the Largest Measured Residual Mass (0.1826 mg) and Maximum Neutron Flux (2.30×10^{13} n/cm^{2}s); All for Bare Wire Samples

Tape	Total activity at end of irradiation per unit mass per unit flux (Bqcm^{2}s/mg)	Activity after 48 hours for 'worst case' (Bq)
Brown Packing 1	$(1.01 \pm 0.02)\times10^{-8}$	$(4.5 \pm 0.2)\times10^{3}$
Brown Packing 2	$(5.09 \pm 0.07)\times10^{-10}$	$(2.3 \pm 0.2)\times10^{2}$
White Duct	$(1.37 \pm 0.02)\times10^{-10}$	$(5.0 \pm 0.1)\times10^{1}$
Electrical	$(3.02 \pm 0.04)\times10^{-10}$	$(1.30 \pm 0.03)\times10^{2}$
Clear Plastic	$(4.56 \pm 0.06)\times10^{-10}$	$(2.05 \pm 0.07)\times10^{2}$
Masking	$(2.29 \pm 0.03)\times10^{-10}$	$(1.0 \pm 0.2)\times10^{2}$

cooling time between the end of irradiation and the beginning of measurement. The half lives for the tapes were taken as the 'effective' half lives given in Table 2.

Typical activities of the flux monitor wires were in the order of 10^4-10^5 Bq after 48 hours of cooling time. However, some wires were covered with cadmium during the irradiation in order to determine the thermal to epithermal neutron flux ratio, and these wires had a much lower activity - in the order of 10^2-10^4 Bq - after cooling. Thus for the case of ionisation chamber measurements, the tape may contribute a significant systematic error, at worst in the order of 10% - though more likely in the order of 1% for the typical case - for bare wires, depending on the tape used, the cooling time, the irradiated neutron flux, and the mass of the tape that is left on the wire. This is a crude estimate as it neglects the energy dependence of the response of the ionisation chamber. Cadmium covered wires, however, would obviously suffer from larger systematic error.

To avoid such an unwanted error, the most effective solution would be to clean the wires individually (to allow identification of the wires to be maintained) using an ultrasonic bath or some other non-abrasive method in order to avoid losing the mass of the wires themselves. This method is potentially quite time-consuming and for large batches may prove intractable. An alternate method of securing the flux monitor wires in place may be called for in such a case.

5. CONCLUSION

Neutron activation analysis has been performed on 6 different adhesive tapes used to protect and secure flux monitor wires into various positions within the OPAL reactor core. The most important radionuclides produced in the tapes were ^{24}Na, ^{42}K and ^{69m}Zn. The shielding of the flux monitor wires due to the tapes was shown to be negligible.

Systematic errors in flux measurement due to the activation of the residual tape on the wires have been found to be negligible when an efficiency calibrated gamma spectrometer is used for measuring the activity of the gold wires. However, when an ionisation chamber was used for activity measurement, superfluous activity is expected.

Although the use of an efficiency-calibrated gamma spectrometer may provide a successful solution, this is not always practical or possible, so an appropriate choice of tape may minimise these systematic errors. The white duct seems to be the obvious choice; it has the shortest effective half life and gives the least activity per unit mass at the end of irradiation for a given flux, allowing handling of the samples to begin shortly after the end of irradiation. Also, the lower activity per unit mass would also contribute less to the measured ionisation current, and allow more time for processes such as cleaning the wires if called for.

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