Progress in Nuclear Science and Technology Volume 4 (2014) pp. 358-362

ARTICLE

Beam monitoring of high energy proton flux by the activation foil technique

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Activation of thin Al foils is a well-established technique for measuring the intensity of high-energy proton beams via the ${}^{27}Al(p,3pn){}^{24}Na$ reaction. The aim of this work was to investigate the various factors that influence the reliability of the results: the cross sections, the competing production of ${}^{24}Na$ by secondary neutrons generated in the foil and spallation processes that can cause loss of the produced nuclei from the foil. The technique was applied for the calibration of an ionization chamber used as beam monitor for a 280 GeV/c mixed proton/pion beam at CERN. The calibration factor obtained experimentally was compared with the theoretical value deduced by Monte Carlo simulations. An additional experiment was conducted employing the alternative reaction ${}^{nat}Cu(p,x){}^{24}Na$, whose cross section is known with low uncertainty over a wide energy range.

Keywords: activation measurements; beam monitoring; calibration; Monte Carlo; spallation process; competing reactions; cross section; high-energy accelerator

1. Introduction

The activation of Al foils via the ${}^{27}Al(p,x){}^{24}Na$ reaction is a well-known technique to measure the intensity of high-energy proton beams and obtain an absolute calibration of in-beam instrumentation [1]. However, the reliability of this reaction is influenced by various factors: the cross section, the competing production of ${}^{24}Na$ by secondary neutrons generated in the foil and spallation processes that can cause loss of the produced nuclei from the foil.

The activation foil technique was used at the H4IRRAD test area [2] at CERN to calibrate an Ionization Chamber (IC), which is in use since many years as beam monitor at the CERF (CERN-EU Reference Field) facility [3]. H4IRRAD is a mixed field irradiation test area conceived to perform electronic equipment testing for LHC and other accelerator-related applications. For this experiment the test area was operated with a 280 GeV/c hadron beam (~94% protons, 5% pions and less than 1% kaons [4]) from the super proton synchrotron (SPS).

The IC has been calibrated with two activation experiments: firstly, with the activation of Al foils via the ${}^{27}\text{Al}(p,x){}^{24}\text{Na}$ reaction mentioned above; secondly, with the activation of natural Cu foils via the alternative monitor reaction ${}^{\text{nat}}\text{Cu}(p,x){}^{24}\text{Na}$.

In the data analysis of Al activation, a particular attention was devoted to understand the influence of the different factors affecting the reliability of the technique. The calibration factor obtained experimentally was compared with the theoretical value deduced from Monte Carlo simulations.

2. The activation foil technique

2.1. Theoretical basis

The production of a radionuclide of interest at time t is expressed by the well-known formula:

$$n(t) = \frac{N\sigma\phi}{\lambda} \left(1 - e^{-\lambda \cdot t_{IRR}}\right) \cdot e^{-\lambda \cdot t_{WAIT}}$$
(1)

where n(t) is the number of atoms per unit volume of the radionuclide of interest at time t (cm⁻³), N is the number of atoms per unit volume of the target (cm⁻³), σ is the production cross section of the selected radionuclide (cm²), ϕ is the particle flux density (cm⁻² s⁻¹), λ is the decay constant (s⁻¹), t_{IRR} and t_{WAIT} are the irradiation and waiting time (i.e. the time elapsed from the end of the irradiation until the foil is counted). The specific activity (in Bq/cm³) induced at time t is given by a(t) = λ ·n(t).

If L₁, L₂ are the transverse dimensions of the target and Δx is its thickness, the total activity is then given by A(t) = a(t)·L₁·L₂· Δx . If N_x = N· Δx is the surface atomic density (cm⁻²) and $\phi' = \phi \cdot L_1 \cdot L_2$ is the number of particles

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per unit time (s⁻¹) traversing the foil, then

$$A(t) = N_x \sigma \phi' \left(1 - e^{-\lambda \cdot t_{IRR}} \right) \cdot e^{-\lambda \cdot t_{WAIT}}$$
(2)

Eq. (2) relates the activity A(t) of the produced radionuclide, to be measured by γ spectroscopy, with the number of beam particles per time unit ϕ ' which has traversed the foil.

2.2. The ${}^{27}Al(p,x){}^{24}Na$ reaction

The ²⁷Al(p,x)²⁴Na reaction is one of the most extensively used reactions for monitoring proton fluxes. ²⁴Na has a half-life of 14.957 h and decays by β^{-} emission producing two γ rays of energies of 2.754 MeV and 1.369 MeV.

However, the production of ²⁴Na from ²⁷Al is sensitive to secondary particles, because of the competing ²⁷Al(n,α)²⁴Na reaction. Data in the literature are quite contradictory about the influence of this effect. Some authors showed it has little importance. Stheney [5] measured a contribution of less than 1% per 200 mg/cm² foil thickness. Cumming *et al.* [6] proposed a value of 0.25% per 100 mg/cm². Some others estimated a bigger importance. Brandt *et al.* [7] reported that this effect has an influence of (1.1 ± 0.5) % per 100 mg/cm². Grover [8] showed a stronger dependence on foil thickness, of about 3.3% per 100 mg/cm². Therefore in this work it was decided to evaluate this contribution independently, both via Monte Carlo simulations and experimentally.

2.3. The $^{nat}Cu(p,x)^{24}Na$ reaction

The ^{nat}Cu(p,x)²⁴Na reaction was chosen as a promising reaction for beam monitoring. This reaction shows most of the desired properties proposed by Cumming [9] for a material to be an ideal candidate for applications in high-energy proton beam monitoring. First, the absolute cross section of the reaction is known at very high energy (up to 800 GeV) with low uncertainty, less than 4% [10]. Secondly, the short half-life of the reaction product results in high specific activity of the foil, as seen for the ²⁷Al(p,x)²⁴Na reaction. Lastly, being the material natural Cu, uniform foils are readily available.

3. Cross section of the activation reactions

For the sake of simplicity, the beam was considered as composed of 100% protons. This approximation is reasonable because of the small contamination of pions (5%) and kaons (less than 1%) in the beam [4].

3.1. ${}^{27}Al(p,x){}^{24}Na$

The cross section calculated by Cumming *et al.* [11] at 28 GeV is equal to (8.3 ± 0.5) mb. At higher energies, only extrapolated data are available. At 300 GeV Kaufmann *et al.* [12] determined a cross section of (8.04 ± 0.58) mb. Assuming that the values of the cross section at 280 and 300 GeV are the same, one can take a

value of (8.0 ± 0.6) mb.

3.2.
$$^{nat}Cu(p,x)^{24}Na$$

The ^{nat}Cu(p,x)²⁴Na reaction cross section was measured at energies of 30, 150, 400 and 800 GeV by Baker *et al.* [10]. An energy-independent cross section value of (3.59 ± 0.14) mb was obtained. Being the energy of the impinging protons included in the range, the same value and related uncertainty can be assumed.

4. Monte Carlo simulations

The Monte Carlo simulations were carried out with FLUKA 2011 [13-14]. The code allows switching off the transport of neutrons so that, by eliminating their contribution to the reaction of interest, the net contribution of the protons to the induced activity of 24 Na was calculated.

A first set of simulations was performed in order to evaluate the induced activity in the foils. A second set of simulations was performed with the same settings, with the exception of the transport of neutrons. This was turned off via the PART-THR card, whose momentum cut-off was set to 280 GeV/c.

The values of activity estimated in the simulations were corrected for the value of the cross section of the $^{27}Al(p,x)^{24}Na$ reaction. In fact, the cross section predicted by FLUKA is 4.5 mb. The discrepancy with respect to the experimental value above can be partially explained by the fact that the standard FLUKA version does not take into account, for residual nuclei production purposes, the quasi-elastic channel, which has some importance for the isotopes near the target. Actually, the standalone nuclear interaction generator of FLUKA includes this contribution, yielding a comprehensive cross section of about 6 mb, reasonably closer to the available measurement, considering the limited fraction of the reaction cross section associated to the channel under discussion.

It was then decided to re-normalize offline the activity due to the primary protons by applying the cross section value of 8.0 mb instead of 4.5 mb, i.e. by multiplying the FLUKA results by a factor of 1.77.

5. Calculation of the theoretical calibration factor

To estimate the theoretical calibration factor, one can calculate the charge q collected on the plates of the IC:

$$q = \rho_{AIR} \cdot \frac{p_{ATM,CERN}}{p_{ATM,0 m}} \cdot L_{IC} \cdot S \cdot \frac{e}{W_{AIR}}$$
(3)

where ρ_{AIR} is the air density (0.001127 g/cm³), $p_{ATM,CERN}$ and $p_{ATM,0 m}$ are the atmospheric pressures at CERN altitude (400 m a.s.l.) and at sea level (963 and 1013 mbar, respectively), L_{IC} is the length of the sensitive region of the IC (32 mm), e is the electron charge, W_{AIR} is the W-value for protons in air (34.2 ± 0.5 eV [15]), i.e. the average energy required to produce an ion pair. S is the energy deposited in the air volume of the IC by the particles per unit path mass length. This quantity does not include the energy transported by the delta rays that escape from the air volume of the IC, but takes into account the energy lost by the particles through nuclear reactions. It is important to underline that S differs from the stopping power.

The only data available in literature on the energy deposited by protons in air is the stopping power. Data are available for energies up to 10 GeV. For 10 GeV protons the stopping power in air is 2.05 MeV·cm²/g. Therefore a dedicated FLUKA simulation was run to assess the energy deposited by 280 GeV/c protons. The simulation was conducted simulating protons with a momentum of 280 GeV/c passing through a simplified model of the IC. The beam was considered as composed of 100% protons.

The results of the FLUKA simulations give an energy deposition value of $7.741 \cdot 10^{-3}$ MeV per proton. Using this value in Eq. (3), one obtains an expected charge deposited in the IC by a primary particle of $3.442 \cdot 10^{-17}$ C. Taking into account the electronics of the IC converting the charge in counts (whose sensitivity is equal to 1.19 cts/pC [16]), the theoretical calibration factor is 24410 ± 901 particles/ct.

This value is calculated on the hypothesis that the air is completely dry. This is a stringent simplification, because the presence of humidity in the air can lead to significant changes in pressure, deposited energy and W-value [17]. The value of the active length of the IC is also affected by some uncertainty, since it was designed 40 years ago and no technical drawings are available.

6. Experimental set-up

Hyper-pure Al and Cu foils were used (99.999% Al, 99.99% Cu), with dimensions $50 \times 50 \text{ mm}^2$. The foil thicknesses were $(2.00 \pm 0.02) \text{ mm}$ and $(0.50 \pm 0.01) \text{ mm}$ for Al, $(0.25 \pm 0.01) \text{ mm}$ for Cu. Sandwiches of three foils of the same thickness were fixed on both ends of a hollow aluminum tube two meters upstream of the IC. In order to evaluate the contribution of background radiation to the foil activation, additional foils were attached to the tube but exposed out-of-beam.

The in-beam foils were exposed in sandwiches of three to check if a cross-contamination of the foils from recoil nuclei of the reaction was significant. In fact nuclei produced in the spallation process can recoil in the forward direction and leave the foil. In this case they would not contribute to the foil activity and introduce an error in the calibration factor estimation.

7. Results

7.1. Experimental results

The gamma spectrometry measurements of the irradiated foils were performed with very low

background coaxial HPGe detectors by Canberra. Table 1 sums up the irradiation time for each foil, the integrated counts of the IC and the measured activity. The data acquisition and analysis was carried out using the Canberra® Genie-2000 and the PROcount-2000 software, which are comprehensive environments for data acquisition display and analysis. They include analysis algorithms, which provide nuclide identification, correction, weighted mean interference activity. background subtraction and efficiency correction. They also take into account geometrical effects. self-absorption and decay during the measurements.

Table 1. Irradiation configurations and induced activity for each foil. U = upstream, C = central, D = downstream, B = background.

| E 11 | Irradiation Integrated | | Activity | |
|--------------|------------------------|---------------------|-----------------|--|
| Foil | time [s] | fluence [cts] | [Bq] | |
| Al 2 mm U | 31200 | $6.54 \cdot 10^{6}$ | 121.0 ± 4.6 | |
| Al 2 mm C | 31200 | $6.54 \cdot 10^{6}$ | 160.0 ± 6.9 | |
| Al 2 mm D | 31200 | $6.54 \cdot 10^{6}$ | 164.0 ± 10.2 | |
| Al 2 mm B | 31200 | $6.54 \cdot 10^{6}$ | 1.18 ± 0.25 | |
| Al 0.5 mm U | 89100 | $2.17 \cdot 10^{6}$ | 9.05 ± 0.62 | |
| Al 0.5 mm C | 89100 | $2.17 \cdot 10^{6}$ | 11.20 ± 0.73 | |
| Al 0.5 mm D | 89100 | $2.17 \cdot 10^{6}$ | 10.50 ± 0.35 | |
| Al 0.5 mm B | 89100 | $2.17 \cdot 10^{6}$ | 0 | |
| Cu 0.25 mm U | 89100 | $2.17 \cdot 10^{6}$ | 2.60 ± 0.26 | |
| Cu 0.25 mm C | 89100 | $2.17 \cdot 10^{6}$ | 2.60 ± 0.13 | |
| Cu 0.25 mm D | 89100 | $2.17 \cdot 10^{6}$ | 2.59 ± 0.17 | |
| Cu 0.25 mm B | 89100 | $2.17 \cdot 10^{6}$ | 0 | |

7.2. Data analysis

The calibration factor of the IC is derived from Eq. (2). The IC output is constituted by TTL pulses (counts) and the calibration factor is then obtained in terms of number of beam particles per count.

The surface atomic density N_x , assuming that all atoms of the foils are Al atoms, is given by:

$$N_x = \frac{\rho_{Al}}{M} \cdot N_{AV} \cdot X_{Al} \tag{4}$$

where ρ_{Al} is the aluminum density, M the Al molar mass, N_{AV} the Avogadro's constant and X_{Al} the foil thickness.

The counting rate of the IC, the irradiation time t_{IRR} and the waiting time t_{WAIT} are read from a log-file, where data are stored every second.

7.2.1 Aluminum activation

The results show that the recoil process of the ²⁴Na spallation product has some importance. In fact the activities of the central and downstream foils are systematically higher than the activity of the upstream foil. From the results one can take as reference the average activity between the central and the downstream foils, i.e. (10.85 ± 0.39) Bq for the 0.5 mm foil and (162.0 ± 6.1) Bq for the 2 mm foil. The upstream foil is not considered due to the loss of ²⁴Na recoil nuclei escaping the foil. With these values Eq. (2) gives a calibration factor of 22093 particles/ct for the 0.5 mm foil.

7.2.2 Copper activation

Following the same procedure above, one obtains an average activity of (2.60 ± 0.11) Bq and a calibration factor of 16195 particles/ct.

7.2.3 Monte Carlo simulations

FLUKA simulations allowed the absolute activity to be scored in each Al foil. The simulations were conducted using a rectangular proton beam (2 cm x 2 cm) with a momentum of 280 GeV/c and flat particle distribution. The irradiation settings approximately reproduced the experimental conditions: irradiation time of 5 hours, cooling time of 10 minutes, beam intensity of $1.0 \cdot 10^7$ particles/s. The results are summarized in **Table 2**. All the reported values have been corrected by the factor of 1.77 for the ²⁷Al(p,x)²⁴Na reaction cross section as explained in Section 4. The simulations were also performed for a 1 mm thick foil.

Table 2. Induced activity and related statistical uncertainty obtained from the FLUKA simulations. For each foil the computed activation reactions are listed.

| Desition | Computed | Foil activity [Bq] | | Bq] |
|----------|--|--------------------|-------|--------|
| Position | reaction(s) | 0.5 mm | 1 mm | 2 mm |
| Up | 27 A 1(p x) ²⁴ No | 41.4 | 85.9 | 179.6 |
| | AI(p,x) Na | (0.5) | (0.9) | (1.4) |
| Central | ²⁷ Al(p,x) ²⁴ Na | 43.9 | 89.8 | 202.7 |
| | | (0.5) | (0.9) | (1.4) |
| Down | ²⁷ Al(p,x) ²⁴ Na | 44.1 | 94.6 | 218.7 |
| | | (0.7) | (1.2) | (1.75) |
| Up | $^{27}Al(p,x)^{24}Na +$ | 43.4 | 91.7 | 203.6 |
| | 27 Al(n, α) 24 Na | (0.7) | (1.1) | (2.0) |
| Central | $^{27}Al(p,x)^{24}Na +$ | 46.0 | 98.4 | 232.7 |
| | 27 Al(n, α) 24 Na | (0.9) | (1.3) | (2.1) |
| Down | $^{27}Al(p,x)^{24}Na +$ | 46.3 | 104.1 | 252.7 |
| | 27 Al(n, α) 24 Na | (0.7) | (1.5) | (2.0) |

8. Discussion

8.1. Al activation: evaluation of the contribution of competing reactions

The contribution of the neutron-induced reactions, calculated via the Monte Carlo simulations, to the production of ²⁴Na, as percent contribution to the activity induced by the primary particles and in terms of contribution per mass thickness is given in **Table 3**.

Table 3. Contribution of secondary neutrons to the foil activity (simulations).

| Foil and position | Induced activity [Bq] | | % contribution from secondary neutrons | |
|-------------------|-------------------------------------|-------|---|-------------------|
| | 27 Al(n, α) 24 Na | Total | On primary activity (*) | Per 100 mg/cm^2 |
| 0.5 mm C | 2.1 | 46.0 | 1 8% | 3 50/2 |
| 0.5 mm C | 2.1 | 40.0 | 4.870 | 5.570 |
| 0.5 mm D | 2.2 | 46.3 | 5.0% | 3.7% |
| 1.0 mm C | 8.6 | 98.4 | 9.6% | 3.6% |
| 1.0 mm D | 9.5 | 104.1 | 10.0% | 3.7% |
| 2.0 mm C | 30.0 | 232.7 | 14.8% | 2.7% |
| 2.0 mm D | 34.0 | 252.7 | 15.5% | 2.9% |

(*) (Neurons activity)/[(Total activity) – (Neutrons activity)]

According to the Monte Carlo simulations, the contribution of the competing reactions to the total activity lies in the range 2.7% - 3.7% per 100 mg/cm² of foil mass thickness.

From the experimental results, the correct calibration factor can be obtained by extrapolation to an infinitely thin (0 mm) foil. The calibration factor determined with a foil of given thickness can then be expressed as the sum of two terms. The first is due to the partial activity induced in the foil by the primary protons. The second is proportional to the foil mass thickness via a coefficient k (expressed in percent per 100 mg/cm²):

$$C_{x} = C_{\text{prim}} + k \cdot C_{\text{prim}} \cdot x \tag{5}$$

where C_x is the calibration factor obtained for a foil of mass thickness x, k is a constant, C_{prim} is the fraction of the calibration factor which is due to the partial activity induced by the protons, x is the foil mass thickness.

In this formula k refers to the partial activity due to the protons, even if in the literature the contribution always refers to the total activity. This is done for two reasons: first, to make Eq. (8) below more consistent; second, because the difference is of little importance.

Taking the calibration factors calculated from the Al activation experiments:

$$C_{0.5 \text{ mm}} = C_{\text{prim}} + k \cdot C_{\text{prim}} \cdot 135 \text{ mg/cm}^2$$
(6)

$$C_{2 mm} = C_{prim} + k \cdot C_{prim} \cdot 540 \text{ mg/cm}^2$$
(7)

From the two equations we obtain:

$$k = \frac{C_{0.5 mm} - C_{2 mm}}{(C_{2 mm} \cdot 135 mg/cm^2) - (C_{0.5 mm} \cdot 540 mg/cm^2)} =$$

= 0.00033/mg/cm² = 3.3%/(100 mg/cm²) (8)

The calibration factor extrapolated at 0 mm of foil thickness, C_{prim} , is (19240 ± 1654) particles/ct, where the uncertainty refers to the measured activity, the cross section value and the foil thickness. This value differs by more than 20% from the theoretical calibration factor calculated in Section 5. The investigation on this discrepancy is still ongoing and will be cleared by the next experiment which is already planned with more foil thicknesses. A reason may be the uncertainties on the theoretical calibration factor s explained in Section 5. A preliminary experiment carried out using an out-of-beam foil had already shown that the background contribution to the induced activity is negligible.

The reliability of the calibration factor has been confirmed by comparative measurements with another ionization chamber installed in the H4IRRAD line. From the results obtained by simultaneous measurements with the two monitors of the H4IRRAD beam the difference in the chambers readout was constantly below 4% over a broad range of beam intensities [18].

9. Conclusions

The three factors that influence the reliability of the activation foil technique via the ${}^{27}Al(p,3pn){}^{24}Na$ reaction were investigated.

The cross section value was derived from literature data and was employed to apply a correction factor to the results derived from the Monte Carlo simulations.

The contribution of the competing reactions on the calibration factor was estimated comparing the results of the Monte Carlo simulations with the experimental data. They are in good agreement and the contribution of the neutron-induced reactions to the foil activity can be estimated as $(3.3 \pm 0.5)\% / 100 \text{ mg/cm}^2$, where the percent relative contribution refers to the partial activity induced by the primary particles and the mass thickness refers to the foil.

The effect of the cross-contamination of the foil due to the recoil nuclei produced by the spallation process was verified. As expected, the induced activity of the upstream foil was systematically lower than the others due to the loss of ²⁴Na recoil nuclei escaping the foil.

Results from the activation experiment of the Cu foils give a calibration factor that is not consistent with what obtained via the Al activation. This can be due to two reasons. First, the activation experiment was limited to a single foil and thus it was not possible to assess the possible contribution of secondary particles to the total induced activity. Second, the induced activity was significantly lower than that in Al, which led to a poor statistics on the experimental data. The experiment has recently been repeated at H4IRRAD (operating since 2012 at momentum of 400 GeV/c) with better statistics and the analysis of the results is ongoing. A similar experiment has also been conducted at the CERF facility, where the beam is composed for 1/3 by protons and 2/3by pions. The data analysis, also ongoing, is in this case complicated by the fact that the high-energy cross sections for protons and pions are not the same. From the results of these experiments a better understanding of the reliability of this monitor reaction should be derived.

Acknowledgements

We wish to thank M. Magistris and Y. Donjoux for putting at our disposal the γ spectrometry lab and for useful discussion.

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