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A simple method to measure proton beam energy in a standard medical cyclotron*

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Abstract

A simple and rapid technique to measure the proton beam energy in the external beam line of a medical cyclotron has been examined. A stack of 0.1 mm thick high purity copper (Cu) foils was bombarded and the relative activity of ^{65}Zn produced in each foil was compared to a computational model that predicted activity, based on proton stopping power, reaction cross-sectional data, and beam energy. In the model, the beam energy was altered iteratively until the best match between computed and measured relative activities of the stack of disks was obtained. The main advantage of this method is that it does not require the comparison of the activities of different isotopes of zinc arising from (p, xn) reactions in the Cu, which would require the gamma photon detector being calibrated for different energy responses. Using this technique the proton beam energy of a nominally 18 MeV standard isochronous medical cyclotron was measured as 17.49 ± 0.04 (SD) MeV, with a precision of 0.2 % CV.

Key words cyclotron, beam energy, copper foils, ^{65}Zn, isotope

Introduction

In cyclotron use, it is sometimes important to measure the proton beam energy to ensure optimum yields for desired radioisotopes, and to ensure that production of unwanted radionuclide impurities is kept within tolerable limits[1].

Measuring proton energy using activation of a stack of natural copper (Cu) foils has been documented by other researchers but typically this involves measuring the ratios of activities of different Zn radioisotopes (usually ^{65}Zn and ^{62}Zn; see for example Kim et al.[1] and Kopecký [2]). One version of the method has utilised the measured activities in foils from the $^{63}Cu(p,n)^{63}Zn$ reaction, convoluted with the reaction cross section and proton stopping power, to determine the energy distribution of protons released during the high-power laser bombardment of solid targets[3]. One challenge of these methods is that if the detection efficiency of the device used to measure the gamma emissions of the radioisotopes is energy dependent, the photon count rate at different energies must be corrected. Although this is achievable with calibration standards, concentrating on a single isotope removes a possible source of error, especially if there is doubt about the accuracy of the energy correction. We determined the beam energy by constructing a computational model that calculates the relative activity of ^{65}Zn in each foil of the stack, as a function of beam energy. This computed distribution of activities is then compared with the measured activities and manipulated until a best fit is achieved, using incident proton beam energy as the adjustable parameter.

Materials and methods

The computational model

Stopping power data from the National Institute of Standards and Technology (NIST) (Berger et al.[4]) were used to calculate the stopping power for a proton in solid natural Cu (69.15% ^{63}Cu, 30.85% ^{65}Cu)[5] for any energy between 10^{-3} MeV and 5×10^{2} MeV. Curves were fitted to these data, allowing stopping power to be calculated as a continuous function of proton energy (Figure 1). A Weibull curve was fitted for energies up to 0.1 MeV, and two

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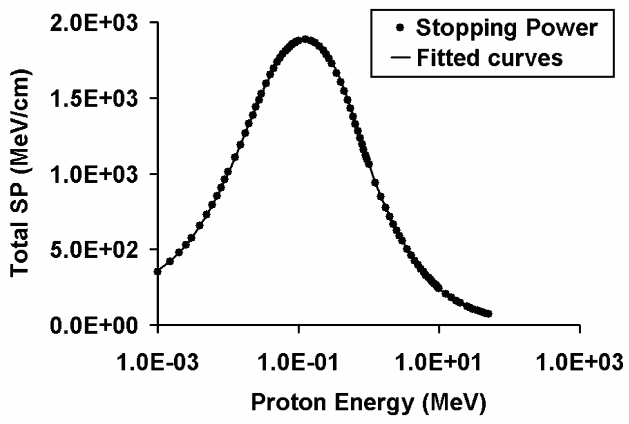

Figure 1. *Stopping power (SP) data plus fitted curves for protons in natural copper (from Ref. 4), as a function of proton energy.*

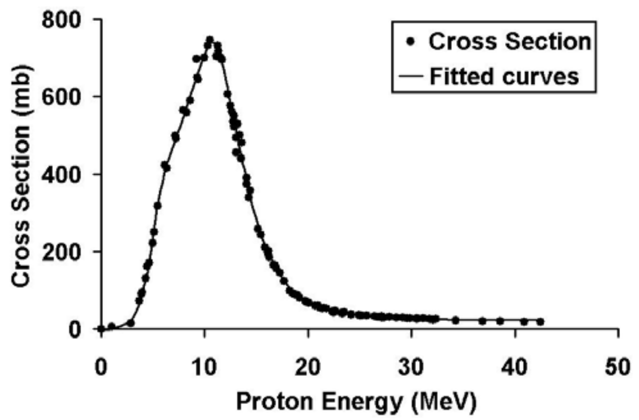

Figure 2. *Cross section data, plus fitted curves for the $^{65}Cu(p,n)^{65}Zn$ reaction (from Refs 2, 6, and 7), as a function of proton energy.*

Morgan-Mercer-Flodin (MMF) curves were used for the energy ranges 0.1 to 1 MeV, and 1 MeV to 50 MeV, respectively. The curve equations were obtained from the library of the curve fitting application CurveExpert (Curve Expert 1.37, Hyams, D.G., Starkville, USA).

For the ^{65}Cu(p,n)^{65}Zn reaction, exponential (0 to 5 MeV), 4th degree polynomial (5 to 11 MeV), and Weibull (15 MeV upwards) curves were fitted to combined reaction cross section data from papers by Collé, Kishore, and Cumming[6], Grüttner[7], and Kopecký[2]. The data and fitted curves are shown in Figure 2. This allowed the reaction cross section for ^{65}Cu(p,n)^{65}Zn to be calculated as a continuous function of proton energy.

The target consisting of eight stacked natural Cu foils of total thickness 0.8 mm was modelled as being divided into 0.001 mm distance increments (indexed as '*i*') along the beam path. By computing the activity of ^{65}Zn produced in each distance increment, and then summing these activities over the increments spanned by each foil in the stack, the total activity of ^{65}Zn produced in each foil can be calculated. A monoenergetic beam at normal incidence to the front face of the first foil, and a reasonable initial estimate of the primary beam energy were assumed. The proton energy at the entrance and exit of each distance increment was calculated using the fitted stopping power curve. The exit energy of one increment was used as the entrance energy of the next. The mean beam energy, E_i, assigned to the distance increment '*i*' was the mean of the entrance and exit energies.

However, as the proton beam penetrates the target the energy at any penetration depth '*x*' is not a unique value, but assumes a distribution. This phenomenon is known as 'energy straggling'. The energy spread is determined by the initial energy distribution (assumed monoenergetic in this experiment) at the entrance of the target; the elemental composition and density of the assumed homogeneous target material, and *x*. This distribution can be calculated easily using the SRIM-2008 Monte Carlo code for ion transport [12]. From the computed ion trajectories examined at selected depths '*x*', it was found that each energy distribution is closely Gaussian in shape. It is impractical to apply SRIM-2008 at each distance increment *i*. In order to minimise computations the code was applied at increments of 0.05 mm until the beam was completely absorbed by the target. A Gaussian curve $y=a*exp[-(E-\bar{E}_x)^2/(2c^2)]$ was fitted to each of the computed energy-spread distributions spanning the range of penetration depths, where E is proton energy, $\bar{E}_x$ is the mean energy of the protons at depth x, and a and c are constants to be fitted. The fitted parameters that describe the shape of the Gaussians (a, $\bar{E}_x$, and c) vary smoothly with x, allowing continuous functions to be derived that describe these parameters as a function of penetration depth. The area under each energy-straggling curve is proportional to the number of protons that penetrate to that depth. This area was assumed to be constant for all depths, based on the results of the SRIM simulations which showed that the loss of beam in the forward direction was less than 1%, down to particle energies low enough to be ignored in the computation. Thus, for computational purposes the energy distribution at distance increment i can be expressed as a histogram with elements $H(i,j)(j=1,n(i))$, where $H(i,j)$ is histogram element number j at distance increment i, and $n(i)$ is the number of histogram divisions. The element $H(i,j)$ represents the fraction of incident protons with energies between $E(i, j)$ and $E(i,j+1)$, where $E(i,j+1) - E(i,j) = \Delta E$. The energy interval ΔE is chosen to be sufficiently small that the histogram reasonably represents the bell-shape of the energy straggle distribution. The number of activated atoms, $P(i)$, of ^{65}Zn arising from proton beam transport through distance increment i, can be expressed as:

$$P(i) = \frac{k\rho y \Delta x T I \left(\sum_{j=1,n(i)} H(i,j) * \sigma[E(i,j)] \right)}{zM} \quad (1)$$

where

k is $(6.24 * 10^{21})$* Avogadro's number

ρ is the target material density (kg m^{-3})

y is the fraction by number of the ^{65}Cu atoms in the natural Cu

Δx is the distance travelled between adjacent increments (m)
T is the bombardment time (s)
I is the incident proton current, assumed to be constant (A)

$H(i,j)$ $(j=1,n(i))$ are the elements of the histogram for distance increment i
$E(i,j)$ is the proton energy (MeV) of the jth histogram element at penetration increment i
$\sigma[E(i,j)]$ is the reaction cross section for $^{65}Cu(p,n)^{65}Zn$ at energy $E(i,j)$ (m^2)
z is the charge in electron units of the incident particles
M is the atomic weight of the target material ($g\ mol^{-1}$);

and where:

$$\sum_{j=1,n(i)} H(i,j) = 1 \tag{2}$$

This assumes that the number of ^{65}Cu nuclei capable of being activated remains essentially constant during the bombardment and that the bombardment time $T << T_{1/2}$, where $T_{1/2}$ is the half life of the activated isotope. However, a significant proportion of the activated nuclei can decay during the bombardment time if the product has a short half life. This can be allowed for. If we drop the 'i' index and define R, the (constant) activation rate of target atoms (s^{-1}) as $R = P/T$ then we can calculate the total number of activated nuclei $N(T)$ present following a uniform bombardment for a time T as;

$$N(T) = \int_0^T R e^{-\lambda(T-t)} dt \tag{3}$$

where λ is the decay constant for the activated isotope.
Then;

$$N(t) = \frac{R}{\lambda}\left(1 - e^{-\lambda T}\right) \tag{4}$$

Since the activity $A = \lambda N$, we can write

$$A = R\left(1 - e^{-\lambda T}\right) \tag{5}$$

With irradiation to saturation ($T \to \infty$) we have for the saturation activity, $A_s = R$; the saturation yield, $A_s / I = R / I$, can then be easily calculated. The saturation yield occurs when the rate of production equals the rate of decay.

One of the input variables for the model is the reaction cross section. Unfortunately, published cross section data for the $^{65}Cu(p,n)^{65}Zn$ reaction are inconsistent. Four publications have reported these data. Those of Collé, Kishore, and Cumming[6], Grüttner[7], and Kopecký[2] are in general agreement. A curve fitted to these data gives the maximum reaction cross section as 727 mb at 10.5 MeV. However the later publication of Levkovskij[8] reported a maximum cross section of 912 mb for 9.5 MeV protons, an increase of 25%. Since the latter report lacks independent verification, the three earlier publications were collectively used for predicting ^{65}Zn yield.

Comparisons of model with published data

The predicted yield of ^{65}Zn from a Cu target was then compared to data published by Nickles[9]. In Nickles' paper, various thick, enriched targets were bombarded with an 11 MeV proton beam. The product yields from saturating bombardment using a 1 μA beam were determined with an accuracy of ± 20 %. For ^{65}Zn, the saturation yield was 8436 MBq μA^{-1}.

The model defined by Equation 1, together with the equation $A_s / I = P / IT$, predicted a saturation activity, under the conditions described by Nickles[9], of 6580 MBq μA^{-1}. The difference of 22% is in reasonable agreement.

The model was also compared to data presented by Dmitriev and Molin[10], who bombarded a thick, natural Cu target with 22 MeV protons. A saturation yield of 4980 ± 530 MBq μA^{-1} was reported, compared to the model's prediction of 4530 MBq μA^{-1}, a difference of 9% which is in good agreement.

Bombardment of copper foils

Circular Cu foils of diameter 25mm, purity >99.99% and 0.1 mm thickness were obtained from the manufacturer (Goodfellow Cambridge, Huntingdon, UK). Uniformity of foil thickness was checked by multiple micrometer measurements on each foil. A compact stack of eight foils was bombarded at normal incidence using an in-house constructed external beam line bolted to a target port of an IBA Cyclone 18/9 medical cyclotron (IBA Molecular, Louvain-la-Neuve, Belgium). The external beam line consisted of an evacuated tube ($\approx 10^{-6}$ torr) with a path length of 41.5 cm from the isolation valve of the cyclotron target port to the beamline target face. The beamline also incorporated beam collimation which allowed a circular beam of no more than 10 mm diameter to strike the centre of the target. Further, only that portion of the beam falling directly onto the target contributed to the measured cyclotron beam current. There were no electromagnetic beam-shaping optics connected to the beam line; beam alignment relative to the target centre could be altered by limited articulation of the beam tube. The energy of protons passing into the beam line through the isolation valve was nominally 18 MeV.

Beam alignment and beam collimation were checked directly after bombarding the Cu foils by placing the first of the bombarded foils on Gafchromic radiotherapy film (International Specialty Products, Wayne, USA) for 90 seconds, immediately post-bombardment.

Three separate bombardment runs were performed, each with a stack of eight foils. Bombardment time was 30 s, and beam current 10 μA.

Measuring foil activities

Because the determinants of activity of the activated isotope include primary beam energy and penetration depth, the energy can be estimated by measuring the activity of an appropriate (p,xn) produced radioisotope in each Cu foil of

Table 1. *Measured and predicted relative activities of ^{65}Zn produced in 0.1 mm-thick Cu foils from bombardment by a proton beam of nominal energy 18 MeV. Results are normalised to the highest measured activity. Predicted activities are derived from the best fit of the computational model to the experimental data using beam energy as the fitting variable. Foil "1" is the first to encounter the beam.*

Foil	1	2	3	4	5	6	7	8
Run 1								
Measured	0.197	0.339	0.655	1.000	0.847	0.305	0.005	0.000
Predicted	0.237	0.414	0.720	1.000	0.815	0.358	0.011	0.000
Run 2								
Measured	0.202	0.353	0.661	1.000	0.838	0.272	0.000	0.000
Predicted	0.239	0.417	0.725	1.000	0.809	0.347	0.010	0.000
Run 3								
Measured	0.193	0.349	0.668	1.000	0.847	0.287	0.000	0.000
Predicted	0.237	0.414	0.720	1.000	0.815	0.358	0.011	0.000
Mean (SD)								
Measured	0.198 (0.004)	0.347 (0.007)	0.661 (0.007)	1.000 (0.000)	0.844 (0.005)	0.288 (0.017)	0.002 (0.003)	
Predicted	0.238 (0.001)	0.415 (0.002)	0.722 (0.003)	1.000 (0.000)	0.813 (0.003)	0.355 (0.006)	0.011 (0.001)	

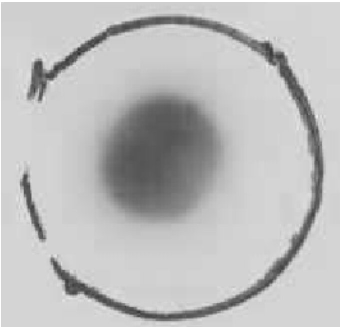

Figure 3. *Image formed by placing an irradiated foil on Gafchromic film. The larger circle indicates the outline of the foil while the dark spot represents the impact of the beam.*

the stack. A few days following bombardment with protons at a nominal energy of 18MeV, the predominant residual manifestation of the activation of natural Cu is ^{65}Zn ($T_{1/2}$ = 244d) arising from the $^{65}Cu(p,n)^{65}Zn$ reaction. Other products, ^{63}Zn ($T_{1/2}$ = 38 min) and ^{62}Zn ($T_{1/2}$ = 9.26 hr), arise from the reactions $^{63}Cu(p,n)^{63}Zn$ and $^{63}Cu(p,2n)^{62}Zn$, respectively. The main photopeak of ^{65}Zn is at 1115.5 keV (50.2% intensity) with the next highest intensity at 511 keV (2.8 %) (LNHB[11]). Hence it was decided to measure the activity of ^{65}Zn produced by measuring the area under the 1115.5 keV photopeak for each foil, and normalising the counts to those from the most active foil. Since the measured photopeaks from each foil are all at the same energy there is no need to allow for different detector efficiencies at different photon energies, a possible source of error. Activities were counted using a Canberra GammaSpect MCA system incorporating a Canberra GC1018 coaxial germanium detector. Software supplied with the unit (Genie 2000 Spectroscopy System, Canberra Industries, Meriden, USA) was used to identify photopeaks and establish the counts beneath each photopeak.

The relative activity of ^{65}Zn in each Cu foil was measured several days post-bombardment . The net peak area (NPA; NPA = total photopeak counts – background) under the 1115.5 keV photopeak was calculated. The Region of Interest, that is, the contiguous set of channels used in the NPA calculation, was checked visually to ensure that it exceeded the width of the photopeak, extending by two channels in each direction along the energy axis into the background. Background counts and correction for deadtime were automatically dealt with by the software.

Calculating beam energy on Cu target

To estimate the beam energy, the measured relative activity in each foil (activity / maximum activity seen across all foils) was compared to the predicted relative activity, derived from the computational model. The model predicted the activity on each foil that would be observed for a particular beam energy, using Equation 1. The energy resolution, ΔE, of all energy straggling histograms was set at 0.01 MeV. For each foil, the difference (residual) between the computed and the measured relative activities was calculated. The residuals were then squared and summed over the 8 foils. The primary beam energy used in the model was then adjusted until the residual sum of squares was minimised.

Results

Beam alignment

Validating assessments of beam alignment using Gafchromic film post-bombardment indicated no significant eccentricity in beam alignment (Figure 3).

Beam energy

Measured versus predicted relative activities are shown in Table 1. The beam energy calculated by minimising the differences between computed and measured relative activities is shown in Table 2, for each of the three runs. Results with and without the adjustment for energy

Table 2. *Primary proton beam energy determined from ^{65}Zn activities in a target stack of eight Cu foils of thickness 0.1 mm, in each of three runs. Results are shown with and without the adjustment for energy straggling. Energies are expressed in MeV.*

Run	Without straggling	With straggling
1	17.52	17.53
2	17.45	17.46
3	17.48	17.49
Mean (SD)	17.48 (0.03)	17.49 (0.04)*

*No significant difference with inclusion of straggling.

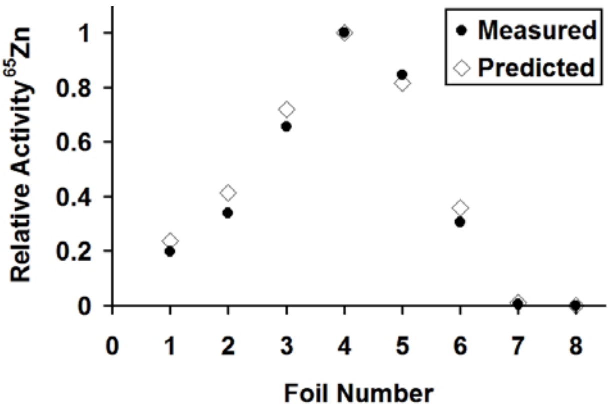

Figure 4. *Measured and predicted relative activities of ^{65}Zn produced in 0.1 mm-thick Cu foils from bombardment by a proton beam of nominal energy 18 MeV (Run 1). Both are normalised to 1 at peak activity.*

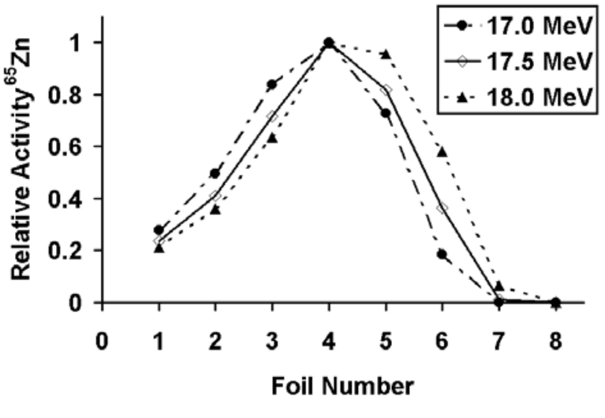

Figure 5. *Computed relative activities of ^{65}Zn produced in 0.1 mm-thick Cu foils following bombardment by proton beams of nominal energy 17.0 MeV, 17.5 MeV, and 18.0 MeV. All are normalised to 1 at peak activity. Joining lines are to aid visual interpretation.*

straggling are compared. The mean energy from the three runs did not change significantly with the inclusion of the straggling effect. The best estimate of the primary beam energy is 17.49 ± 0.04 (SD) MeV with a coefficient of variation of the measurement of 0.2%. A graphical representation of the measured and predicted activities (the latter corrected for energy straggling) is shown for a typical run (Run 1) in Figure 4.

For comparison purposes the computed relative activities in the copper foils are plotted for beam energies 17.0, 17.5, and 18.0 MeV in Figure 5. These give an indication of the ability of the method to discriminate energy differences at least of the order of 0.5 MeV.

Discussion

It is desirable to be able to determine reasonably precisely the beam energy of the cyclotron incident on a target. Though the energy degradation induced by a homogeneous foil of known uniform thickness and known atomic composition can be calculated easily, it is not possible to calculate accurately the primary energy of the beam incident on a target from the defining parameters and engineering of the cyclotron, such as dee geometry, magnetic pole shapes and magnetic field distribution. This is partly because of the complexity of the beam orbit arising from variabilities in the manufacturing and positioning of the magnet poles plus variability introduced by the beam extraction process[13].

Methods employing activation of copper foils for the estimation of proton beam energy have been widely applied over the range of energies (≈4 to 35 MeV) spanned by the excitation functions for the set of $^{nat}Cu(p,xn)^{62,63,65}Zn$ reactions. Kim et al. investigated monoenergetic beam energies by measuring the ratios of activities of ^{62}Zn to ^{65}Zn in target stacks of five 0.1 mm Cu foils, and reported reasonable agreement with theory[1]. Robson et al. measured the activity in Cu foils of ^{63}Zn using annihilation gamma rays and convoluted this with the reaction cross section and proton stopping powers to measure the polyenergetic energy distributions (≈5 to 40 MeV) of protons ejected in the forward and backward directions, arising from bombardment of an aluminium target with a high powered laser[3].

All these methods require measurement of absolute activities of the activation isotopes, accurate knowledge of the beam current, or both. In particular the gamma spectrometer must be calibrated accurately for efficiency at the energy of each relevant photopeak, which ideally requires an appropriate standard source for each peak.

However, if the requirement is for the "simple" measurement of the energy of a monoenergetic proton beam, then the applicable version of the stacked Cu foil technique does not require knowledge of the beam current, duration of bombardment, or calibration of the gamma spectrometer, only that the measurement of a single photopeak be stable over the duration of the experiment. The method described in this report requires only that the activities of all foils be compared at the photopeak 1115.5 keV for ^{65}Zn.

This study has shown that the proton beam energy of a "medium-energy" medical cyclotron can be measured with an acceptable precision (0.2% CV), at low beam current (10 μA), and with a short bombardment time (30 s).

Conclusions

This study describes a simple method for measuring the primary proton beam energy in a medical cyclotron. The method does not rely on any corrections for the energy – dependent sensitivity of the detector used to measure the activation of the target, and does not require an accurate measurement of the beam current or bombardment time. Irradiation of the target-foil stack takes less than one minute of cyclotron run-time. The results are in reasonable agreement with the expected value.

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