Measuring the absolute decay probability of ^{82}Sr by ion implantation

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Abstract

We have developed a method of implanted ion counting in order to determine the absolute branching ratio of the 776.5 keV γ-ray transition in the decay sequence of ^{82}Sr$\rightarrow^{82}$Rb$\rightarrow^{82}$Kr. A 215 MeV beam of ^{82}Sr was produced at the Holifield Radioactive Ion Beam Facility and passed through an ionization chamber that counted and identified the ions before they were implanted into thin aluminum foils. Subsequent offline measurements using a Ge detector deduced the probability per decay of ^{82}Rb for the 776.5 keV γ-ray in ^{82}Kr to be 0.1493(38) in agreement with the accepted average value of 0.1508(16). This new technique measures directly the number of decaying nuclei in a given sample and significantly reduces the dependence on knowledge of the complete decay level scheme.

Keywords:
Nuclear physics, ion implantation, absolute decay probability, PET

1. Introduction

The radionuclide ^{82}Rb has been used for many years in Positron Emission Tomography (PET) to image the heart muscle. In fact, a special issue [1] of

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this journal was devoted to the subject and the nuclear physics of the ^{82}Sr-^{82}Rb generator upon which it depends. Suppliers of ^{82}Sr typically use the 776.5 keV γ-ray transition in ^{82}Kr as a measure of how many ^{82}Sr nuclei are in the generator. This transition is isolated in the decay spectrum and its intensity can be easily determined with modern Ge detectors. In 1987, two groups reported the probability per decay of ^{82}Rb to emit a 776.5 keV γ ray following β decay. Their results, 0.1512(18) [2] and 0.149(4) [3], differed significantly from the previously accepted value of 0.134(5) [4] obtained using low resolution NaI(Tl) detectors. The currently accepted value [5], 0.1508(16), is the weighted average of the two newer measurements.

The two 1987 measurements were made under similar conditions:

- Determined relative γ-ray emission rates by using high resolution Ge or Ge(Li) detectors
- Measured positron emission rates by detecting annihilation radiation
- Used calculated electron-capture-to-positron ratios to determine the probability of annihilation radiation and X-ray emission per decay

Thus, the systematic errors of both measurements are similar and dependent upon knowledge of a complete decay scheme for ^{82}Rb in order to correctly apply the theoretical electron-capture-to-positron ratio. We have undertaken the present study to minimize this systematic uncertainty and to measure directly the decay probability of the 776.5 keV transition. Our technique has the added benefit of removing much of the chemistry and thus, significant impurities from other isotopes. In addition, it is applicable to most radioactive species including extremely short-lived isotopes [6, 7] .

2. Experimental technique

Our technique is to produce a beam of ^{82}Sr and accelerate it above 2.5 MeV per nucleon at the Holifield Radioactive Ion Beam Facility [8] at Oak Ridge National Laboratory. The beam is delivered to an ionization chamber that counts individual beam particles and measures their energy loss in the gas. Since the acceleration process selects mass and the energy loss is large and element specific at these energies, almost all contaminants are removed and/or identified sufficiently well to distinguish them from the isotope of interest. The ions are implanted into thin foils placed in the gas immediately

after the last anode of the ion chamber. These foils can then be removed from the vacuum system and the decay spectrum measured offline with a well-calibrated γ-ray analysis system. This technique counts every ion that gets implanted and hence the decay probability can be measured directly without relying on theoretical calculations or knowledge of the feeding to higher energy levels[2].

2.1. Radioactive ion beam production

The 370 MBq ^{82}Sr sample was purchased and shipped in a vial with a few drops of weak HCl. This solution was dropped into metal powder in a copper cup, capped with additional powder, pressed, and heated in air to convert the $SrCl_2$ to SrO. The cup was then placed in the copper cathode holder of the HRIBF multi-sample sputter source [9] where Cs ions bombarded the powder, ejecting negative ions of SrO from the cathode. The SrO^- molecule was extracted from the source, formed into a beam, mass analyzed, and injected into the 25 MV tandem accelerator. The molecule was dissociated at the high voltage terminal of the tandem and $^{82}Sr^+$ was further accelerated to 215 MeV. A magnet selected energy and a mono-energetic beam of ^{82}Sr was delivered to the experimental end station.

Two production runs were used as the first test indicated that improvements could be made. A significant problem was the purity of the beam when the powder used to hold the SrO was copper. Despite the techniques used, the Sr beam was only 10% of the total beam delivered to the implantation station. The impurity is believed to be copper which resulted from negative ions of $^{63}Cu^{35}Cl$ which has the same mass as $^{82}Sr^{16}O$. In fact, the negative ion beam current up to the terminal of the tandem was several nanoamperes which could only be from a stable contaminant. Even though the beam passed through a 180° magnet and the energy analyzing 90° magnet, a contaminant was delivered to the implantation station on the order of 13000 ions/s representing a reduction in beam intensity relative to the terminal of over seven orders of magnitude. The intensity and composition of the impurity was strongly affected by small energy changes suggesting it was not the only possible ^{82}Sr isobar, ^{82}Se. In this initial attempt, the ^{82}Sr intensity was

[2]To do a full correction for γ summing, knowledge of the preceding γ-transitions is needed. However, this correction and uncertainty is typically much smaller than other uncertainties.

1300-2000 ions/s and would have been adequate for our measurements if not for the high impurity of the beam.

Figure 1: Photograph of the ion source cathode assembly for the second production run. Four cathodes are filled beginning clockwise from the upper right position: ^{nat}Sr in Ag, ^{nat}Sr in Cu, ^{82}Sr in Ag, and ^{nat}SrO in Cu. The cathodes with Ag are in Ta cups and the others are in Cu holders. The first three cathodes were produced using the same technique. The fourth was produced by mixing SrO powder with Cu powder before filling and pressing. Color online.

As a result, our next attempt used silver powder instead of copper and replaced copper cups with tantalum cups. Previous experience in developing Be beams [10], indicated that Ag should give comparable results but would significantly reduce the copper that can be sputtered. Indeed, our ^{82}Sr beam intensity remained unchanged or slightly lower (1600 ions/sec) while reducing the copper contamination by a further order of magnitude (800 ions/sec). The overall reduction in total beam intensity proved beneficial for ion count-

ing. Obvious future improvements to this ion source would be to replace the copper cathode holder with a different material such as tantalum. The ion source cathode assembly used in the second production run is shown in Fig. 1.

2.2. Beam counting and implantation

The ^{82}Sr beam was tuned to a focus on a red alumina phosphor upstream from the entrance window to the ionization chamber [11]. The ionization chamber is comprised of six 9-mm anodes separated by 1 mm with a common cathode of approximately 60 mm. The typical configuration [11] uses entrance and exit windows that are 16 mm in diameter with metal support wires for the 0.9 μm mylar windows. The chamber is filled with CF_4 gas and can be operated as high as 270 mbar. In the present experiment 150 mbar was found to provide good identification of ions while ensuring the ions had enough energy to be implanted into the foils. The signals were processed through standard analog electronics and an event signal to start the data acquisition and provide a means for live time corrections was taken from the second anode. A pulser signal was injected onto the second anode to monitor electronic gain shifts and to possibly provide a further check on live time corrections. In addition, a 100 Hz pulser from the data acquisition was also used to monitor computer live time. The ^{82}Sr ions were implanted into thin kapton or aluminum foils of 19 mm diameter. In order to be counted as a probable implantation event, all six anodes of the ionization chamber must register a signal.

During the first run several problems were identified. The most significant problem was the high data rate and the pile-up that resulted. Pile-up occurred when a second ion (or more) entered the ion chamber during the time it takes to process the signals from the first ion. If the second ion occurred after the ADC gate, it was accounted for in the analysis by the live time correction. If it occurred before this, it appeared in the spectra as an event with higher than normal energy loss extending up to twice (or more) the full energy of a non-pileup event. The closer together the ions occur, the closer to twice the full energy loss was recorded. This can be seen in the energy loss spectrum shown in Fig. 2. Although many of the pileup events can be identified and counted, those close to the energy loss of a single ion cannot be disentangled and may result in significant uncertainty in the number of atoms. In addition, the high counting rate from the detector resulted in excessive dead time in the acquisition system. This dead time was reduced

by scaling down by a factor of 10 the rate of events processed. The second production run with its much lower total beam intensity allowed the removal of the scale-down module. A "fast CAMAC readout" module[3] developed for the HRIBF data acquisition was also added and cut the data processing time by almost half. In addition, a pile-up circuit was added to identify pile-up events which occur after the data acquisition inhibit was initiated.

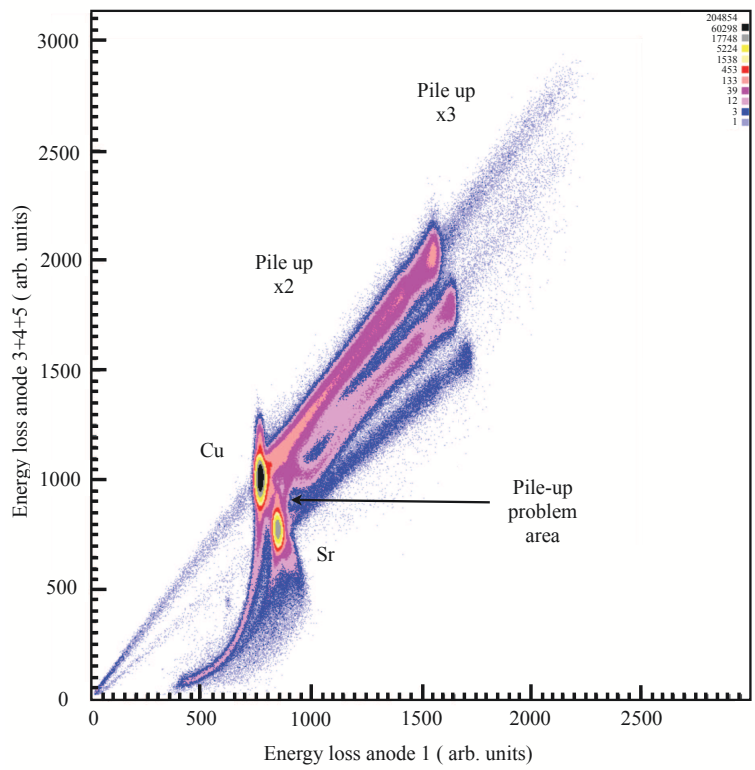

Figure 2: Energy loss spectrum from the first production run. Labels identify the features of data. The pile-up circuit discussed in the text was later used to identify events indicated by the arrow. See Figs. 5, 6, 7 for the improved data from the second production run. Color online.

Another potential problem that was identified was the presence of the exit window support wires. Although a small fraction of the total area of the window, any ion that is recorded by the ion chamber that hits the wires

[3]An auxiliary controller that performs 16-bit transfers with minimum overhead

would be lost in the implantation. Therefore, this window was removed and the foil position was changed so that the resulting path did not increase the amount of gas the ions must traverse. This had an advantage in that a silicon detector could be mounted temporarily in place of the foil and the implantation efficiency was measured to be better than 99.5% in our calibration runs. In fact, the only events in the ionization chamber that were missing from the silicon detector coincidence corresponded to low-energy events in all six anodes. Therefore, two-dimensional gates, that effectively contain both low and high energy thresholds, render a separate correction for implantation efficiency unnecessary under normal operating conditions. Kapton foils were intended to be used for the implantation since our γ-calibration sources were delivered on kapton foils. But it was thought that perhaps the implanted ^{82}Sr might migrate out of such a material so 0.127 mm aluminum foils were used instead.

2.3. Offline analysis

A standard n-type Ge detector with 15% intrinsic efficiency was used to measure the decay of the implanted ions. Energy resolution was better than 2 keV at 1.33 MeV and was calibrated for efficiency using several standard sources [12] mounted on thin kapton foils. The sources contained transitions from ^{241}Am, ^{109}Cd, ^{133}Ba, ^{152}Eu, ^{139}Ce, ^{57}Co, ^{85}Sr, ^{54}Mn, ^{88}Y, and ^{60}Co. This source combination had the advantage that ^{152}Eu has a 778.9 keV γ-ray that is less than 3 keV from the line of interest, includes a potential contaminant ^{85}Sr, and covers the energy range from 61 keV to over 1800 keV. The γ-ray analysis station was calibrated for 15 and 30 cm geometries and with and without thick aluminum positron annihilators surrounding the sources. Ultimately, the 15 cm geometry was used with positron annihilators in place as is shown in Fig. 3. The Ge detector was shielded by an annular cylinder with layers of steel, lead, tin, copper, and aluminum.

The absolute efficiency for 776.5 keV γ-rays was fit globally (down to 61 keV) and locally (above 340 keV) using two polynomial least squares formalization [13]. The center of the range of four resulting efficiencies was taken as the efficiency value with the range limits giving the uncertainty.

3. Results

Following the experience in the first production run the following parameters were chosen for the second implantation run:

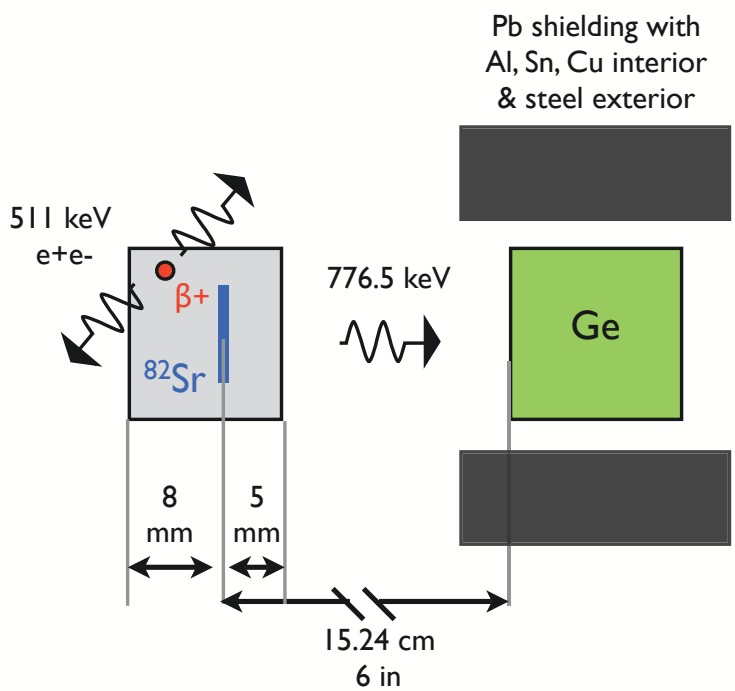

Figure 3: Sketch illustrating the setup for the γ-ray measurements. Color online.

- ^{82}Sr beam produced using pressed silver powder cathodes in tantalum cups
- ^{82}Sr beam energy at 215 MeV
- Ion chamber CF_4 gas at 150 mbar
- Pileup circuit in place with fast CAMAC data readout and no scale-down module active
- No exit window on the ionization chamber
- Aluminum implantation foils placed approximately 3 mm from the edge of the last anode
- 100 Hz pulser and event rate taken for live time corrections
- γ-ray analysis used the 15 cm geometry and aluminum annihilators

Beam was delivered over a period of 7 days with total beam intensity ranging from 1800 to 2500 ions/s. Four implantation samples were produced and the data sorted into bins of 30 s. The profile of the ^{82}Sr component in the beam is shown in Fig. 4.

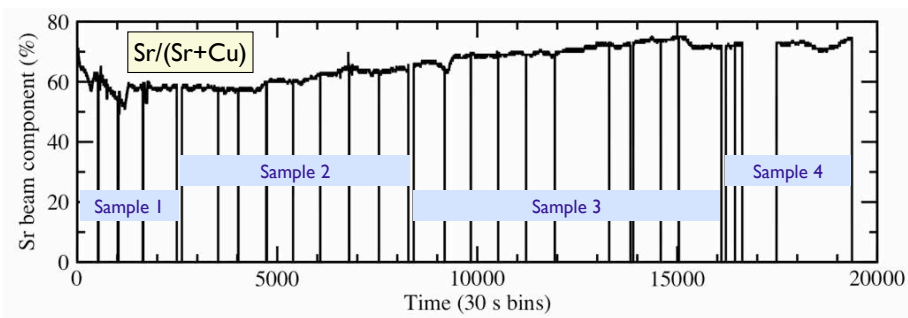

Figure 4: Concentration profile of ^{82}Sr in the beam throughout the experiment. Data are sorted into 30 s time bins and based on gates set in the two-dimensional energy loss spectra, an example of which is shown in Fig. 5. The time region corresponding to each sample implantation is indicated on the figure. Color online.

In order to be counted as an implantation event, the ion must have produced an energy loss signal in all anodes except the second because it has the

added pulser which may distort the data. However, the second anode would have been present as the event signal given to start the data acquisition is derived from this anode. Two-dimensional gates were set on the fourth and fifth anode spectrum which provided the best separation of the beam components as a result of the addition of the pile-up identifier. A representative example of the data is shown in Fig. 5. These single-ion events accounted for approximately 97% of all events.

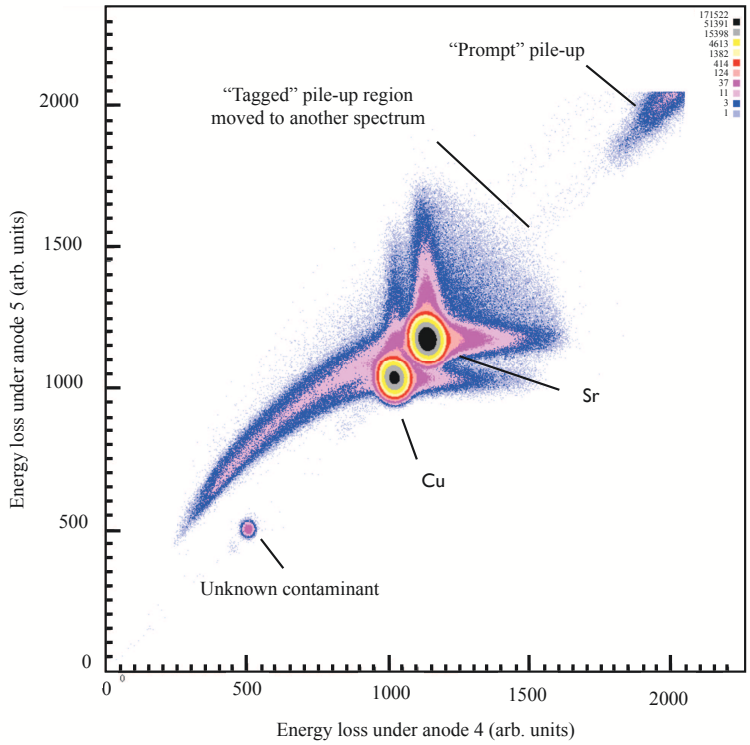

Figure 5: Energy loss spectrum from the second production run not tagged as pile-up. Pile-up data exists beyond the region shown (see Fig. 7). Labels identify the features of data. A two-dimensional gate enclosed the events labeled Sr and Cu and included the wings to the right and above each intense group. Narrow gates, discussed in the text, did not include the wings and were drawn tightly around the circular intense regions. The pile-up circuit discussed in text resulted in the lack of data in the region indicated. The data corresponding to this region are shown in Fig. 6. Color online.

The remaining 3% of events were identified as pile-up in either the pile-up identification tag or as events of nearly twice the energy-loss of single ion

events. Representative examples of the spectra used to count these events are shown in Figs. 6 and 7. All events in Fig. 6 and the high energy events in Fig. 7 represent two beam particles processed as a single event. Their location in the spectrum provides sufficient identification for each particle with the exception of those events in the same area as the full energy single events in Fig. 6. These events correspond to those that occur in the time near the close of the ADC gate where one event has been completely processed and identified while the other has not. These unidentified events can still be counted as their probability of being ^{82}Sr is given by the percentage of ^{82}Sr in the beam. Thus, the single full-energy ^{82}Sr events counts as two ^{82}Sr ions while the single full-energy contaminant ions count as two contaminant ions.

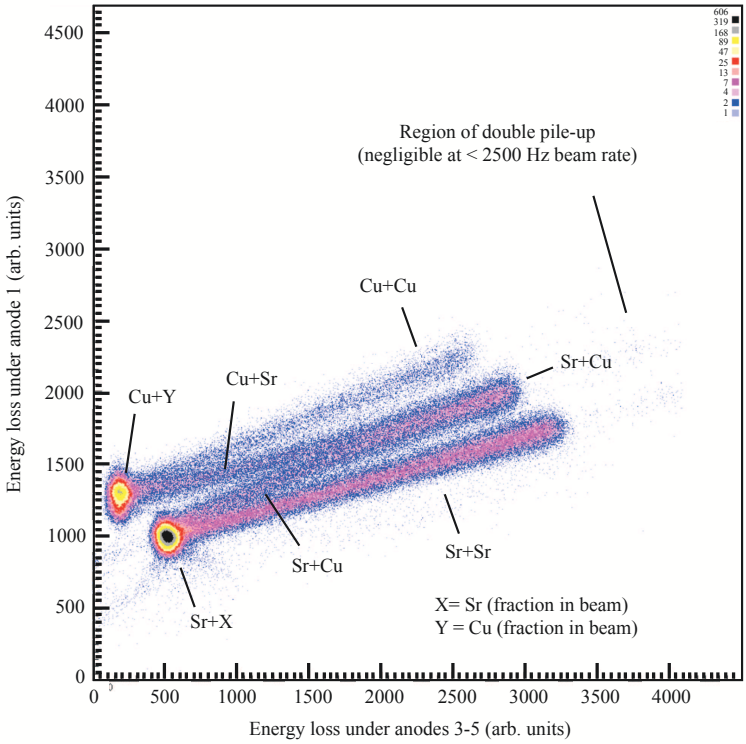

Figure 6: Energy loss spectrum from the second production run tagged by the pile-up circuit. See text concerning the full energy single events labeled with X and Y. Labels identify the features of data. Color online.

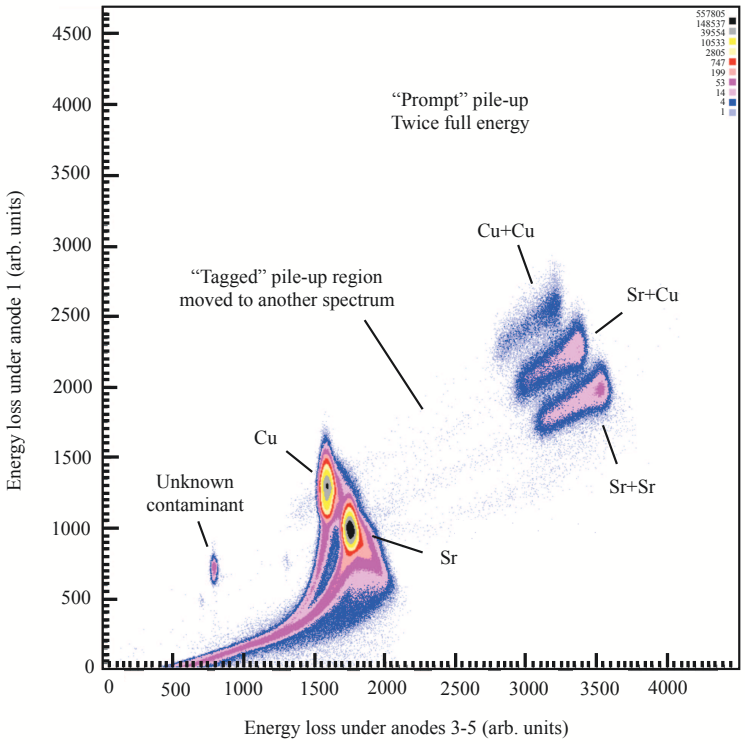

Figure 7: Energy loss spectrum from the second production run not tagged by the pile-up circuit. The high energy events correspond to pile-up events that occurred before the pile-up circuit could be initiated. Labels identify the features of data. Color online.

Two methods were used to determine the number of ^{82}Sr ions implanted into the foils. One method (A) involved summing two-dimensional gates around the appropriate groups in the spectra shown in Figs. 5, 6, and 7. The other method (B) determined the fraction of ^{82}Sr (see Fig. 4) in the beam as given by the ratio of the two-dimensional gates around the full-energy single event spectrum shown in Fig. 5. This fraction was then multiplied by the total number of events recorded adjusted for pile-up events (tagged and untagged). The number of untagged events was determined by the number of high energy events above the pile-up tagged region as shown in Fig. 7. In addition, a low-energy threshold was used to account for the low-energy events that were not implanted as revealed in the silicon detector implantation efficiency tests. Method B was checked using the same large gates of the method A as well as extremely tight circular gates drawn around the full-energy peaks. Both methods (and the narrow-gate check) yielded the same value within less than 0.1% of each other.

The four samples were measured together for γ-emission. By correcting each 30 s time bin for the half-life [3], [5], [14] of ^{82}Sr, 25.35(3) d, the total number of ^{82}Sr atoms remaining in the four samples at the start of the γ-measurement was 7.264(27)x10^8 atoms (average of the two methods). The general size of the corrections and their uncertainties included in the number of atoms remaining includes:

- Implantation efficiency - 1.000(3) and 0.995(3) - applied globally
- Data acquisition live time - $\sim$0.910(2) - adjusted per sample
- Statistical error - applied on each 30 s time bin
- Fractional purity (applicable to method B only)- applied on each 30 s time bin
- Half-life of 25.35(3) d - applied on each 30 s time bin
- Clock differences between implantation and γ measurement data acquisitions - 5 min - applied on each 30 s time bin

The largest contributions to the uncertainty in the number of atoms remaining were implantation efficiency (65%) and the data acquisition live time (35%). All others contributed at the 10^{-4} level or less. The implantation efficiency has two values because a magnetic steerer on the beam line was off

during the implantation of half of sample 2 and all of sample 3. This caused the beam to be shifted. The ring of material surrounding sample 3 was measured for 776.5 keV γ-rays in an attempt to quantify if any ^{82}Sr missed the implantation foil. There was indeed slight activity on the ring whereas other rings indicated no activity. The relative yield of ring 3 versus sample 3 indicated that 0.5(3)% of the sample's activity was on the ring. Therefore, for sample 3 and the latter half of sample 2, an additional correction to the energy threshold (see Sec. 2.2) was applied to account for the shutdown of the steerer. An analysis of the effects of the steerer and the geometry of the experiment indicated that the 6 mm ring should have all implants which missed the aluminum foil.

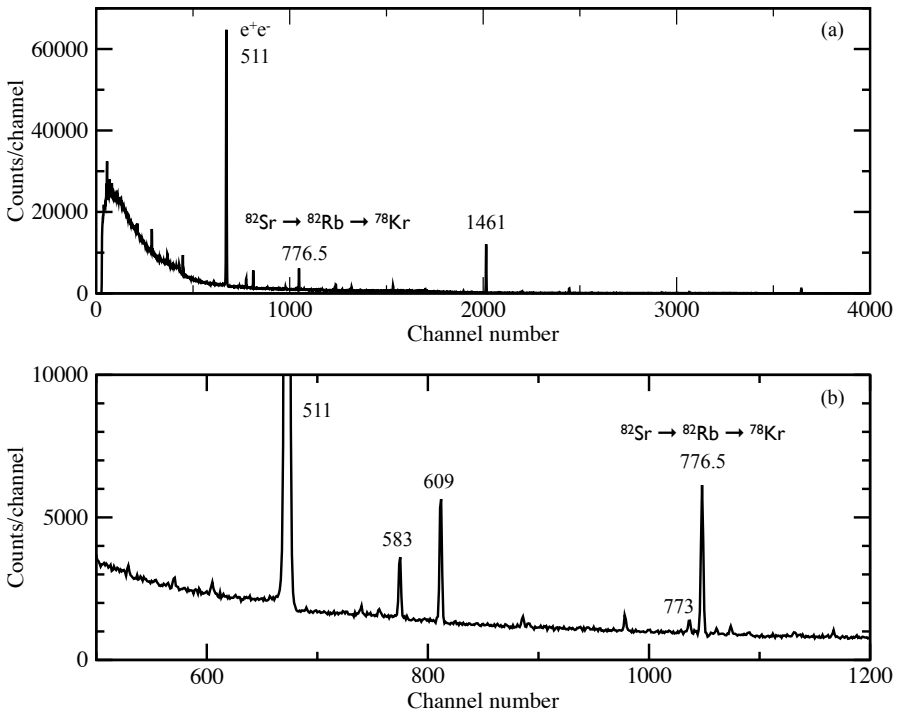

Figure 8: *(a)* Spectrum of γ-rays taken from four samples surrounded by an annihilator. *(b)* The same spectrum showing only the region near the 776.5 keV transition which is isolated and resolved from the 773 keV γ-ray from the background.

The γ-ray spectrum taken over a 20-day measurement is shown in Fig. 8. The 776.5 keV γ-ray yield was measured to be 13660(160) from a total of

1.895(34)x10^8 decays. The absolute photopeak efficiency for the 776.5 keV γ-ray was 0.0004872(58). The data was further corrected by 1.0088(2) for summing with the 511 keV annihilation radiation. Taking these quantities into account, the number of 776.5 keV γ-rays emitted per ^{82}Rb decay following decay of ^{82}Sr is 0.1493(37).

4. Conclusion

The decay of the ^{82}Sr-^{82}Rb generator, used in PET imaging of the heart muscle, was investigated using ion implantation techniques. A beam of radioactive ^{82}Sr was developed and accelerated above 2.5 MeV per nucleon. This beam was detected by an ionization chamber, individual ions were identified by their energy loss, and implanted in thin foils. Two methods of determining the number of ^{82}Sr atoms implanted were averaged and the subsequent decay of the atoms to ^{82}Rb and ^{82}Kr was measured by a Ge detector. The number of 776.5 keV transitions per decay of ^{82}Rb following the decay of ^{82}Sr was determined to be 0.1493(38) in agreement with the previously accepted value of 0.1508(16). This technique measures directly the number of atoms in the implanted sample and hence, only the absolute efficiency of the Ge analysis station needs to be calibrated. Any isotope with suitable half-life which can be made into a beam and accelerated can be used with this technique which offers different systematic errors than other techniques that rely on chemical separation coupled with relative measurements. Those other techniques often rely on detailed knowledge of a complete decay scheme and may be augmented by theoretical calculations.

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6. References

[1] Appl. Radiat. Isot. **38**, 171-239 (1987).

[2] S. M. Judge *et al.*, Appl. Radiat. Isot. **38**, 185 (1987).

[3] D. D. Hoppes *et al.*, Appl. Radiat. Isot. **38**, 195 (1987).

[4] H.-W. Muller, Nuclear Data Sheets, **50**, 1 (1987).

[5] J. K. Tuli, Nuclear Data Sheets, **98**, 209 (2003).

[6] J. A. Winger *et al.*, Phys. Rev. Lett. **102**, 142502 (2009).

[7] S. V. Ilyushkin *et al.*, Phys. Rev. C **80**, 054304 (2009).

[8] J. R. Beene *et al.*, J. Phys. G, in press.

[9] Y. Liu *et al.*, in *Proc. 17th International Conference on the Applications of Accelerators in Research and Industry*, AIP Conf. Proc. **680**, (American Institute of Physics, Woodbury NY, 2003), p 1017.

[10] D. W. Stracener *et al.*, Nucl. Instrum. and Meth. in Phys. Res. A **521**, 126 (2004).

[11] C. J. Gross *et al.*, Eur. Phys. J. A **25 S01**, 115 (2005).

[12] Sources from Eckert and Ziegler Isotope Products, URL: www.isotopeproducts.com.

[13] Philip R. Bevington, *Data Reduction and Error Analysis for the Physical Sciences*, (McGraw-Hill, New York, 1969); Microsoft Program EXCEL 2007 Windows Edition, Microsoft corporation.

[14] S. M. Judge *et al.*, Appl. Radiat. Isot. **38**, 193 (1987).