Measurement of the $^{nat}Lu(p,x)^{175}Hf$ excitation function

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1. Introduction

It is of great interest to study the chemical properties of the transactinide elements (Z > 103) due to the influence of relativistic effects on the chemical properties. These effects are expected to result in deviation of periodic group trends [2]. In order to assess this deviation of chemical behavior for the heaviest elements, their chemical behavior should be compared to that of their lighter homologs that reside in the same periodic group. However, chemically studying the heaviest elements presents several challenges which stem from the short half-lives and extremely low production rates [3]. In order to determine the suitability of a chemical system for transactinide chemistry, off-line experiments are first performed with the homologs and pseudo-homologs of the element of interest, at sub-tracer scale levels. In order to perform experiments with sub-tracer scale concentrations, radionuclides are used, some of which are not commercially available. Here we report the measurement of the $^{nat}Lu(p,x)$ excitation function for production of ^{175}Hf ($t_{1/2}$ = 70. d) for off-line chemical studies of Group IV homologs.

2. Experimental methods

Long-lived ^{175}Hf was produced using the $^{nat}Lu(p,x)$ reaction at the Single Event Effects (SEE) beam line of the K500 cyclotron at the Texas A&M University Cyclotron Institute. The stacked foil activation technique is commonly used to experimentally determine excitation functions [4,5,6] and has been used in this study. The extensively measured $^{nat}Ti(p,x)^{48}V$ reactions were used as monitor reactions [7] to assess the beam energy and intensity. A schematic of the target stack and target holder is shown in Fig. 1 a and b, respectively. A HD^+ beam was accelerated in the K500 cyclotron and underwent stripping via an Al stripper foil to H^+ before being delivered into the SEE beam line target chamber. A proton beam with an energy of 22.1 ± 0.05 MeV, one of the lowest achievable proton energies for the K500 cyclotron, was incident on the target stack. A stack of three ^{nat}Ta foils, each ~81 μm thick, was placed in front of the first Lu foil in order to degrade the beam to a lower, more appropriate energy, 16.7 MeV. Energy losses of the proton beam through each degrader and target were calculated using SRIM-2008 [9]. The average beam current was 1.85 μA. The geometry of the aluminum target holder ensured that nearly the whole beam passed through each target.

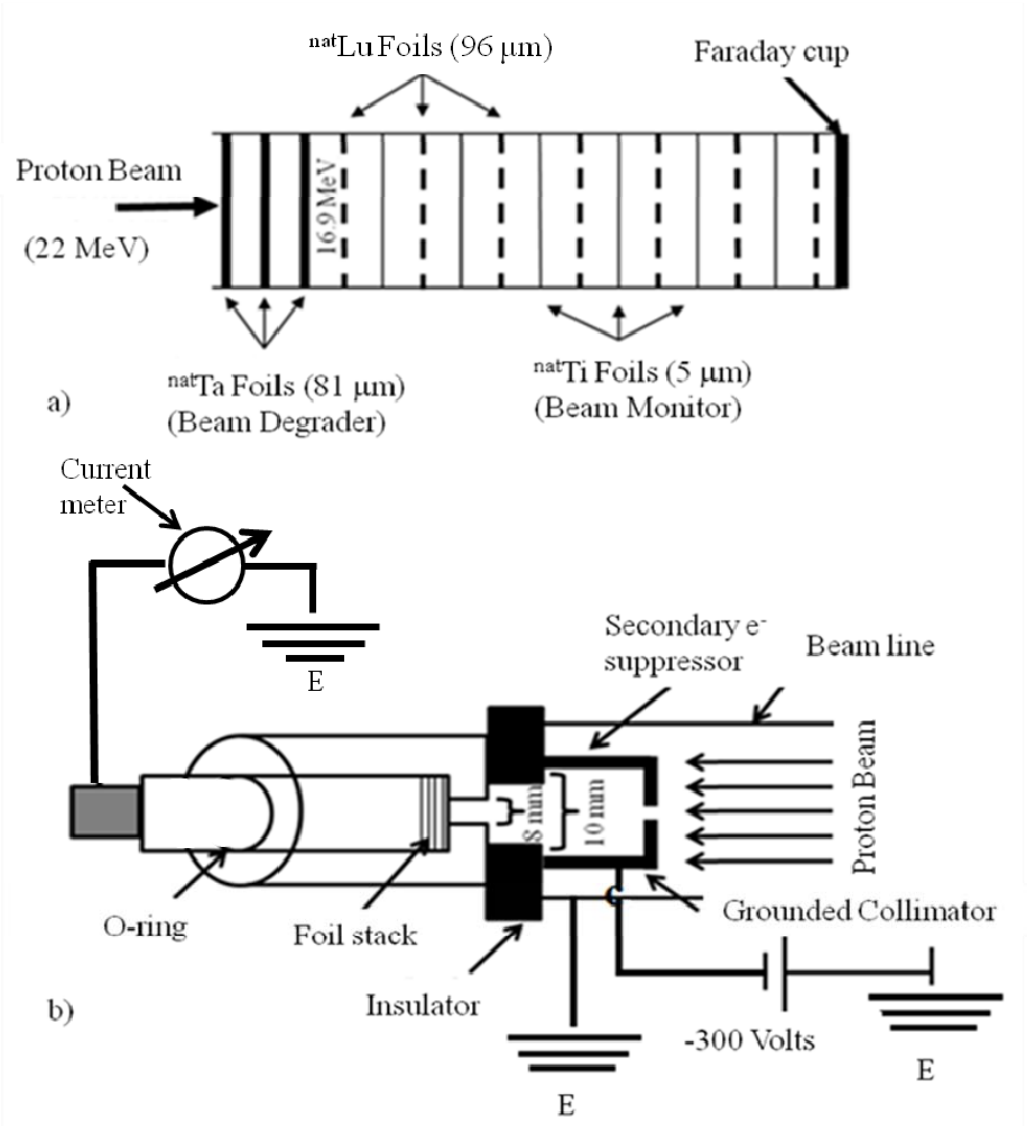

FIG. 1. Schematic drawings of the experimental setup. a) The foil degrader proceeding from left to right. b) The custom aluminum target holder mounted on the beam line [8].

The beam energy in the center of each target is shown in Table I. These values have an estimated 0.2 % error due to the error associated with the reported initial beam energy and an estimated 4% error associated with the energy loss calculations. The beam dose was determined using a current integrator connected to the back end of the target chamber, which also held the targets in place. The data from the current integrator was used to determine the reaction cross sections. At the end of irradiation, the target foils were allowed to cool for approximately 90 minutes to allow for the decay of short-lived products, transportation, and preparation of targets for γ-ray measurements. The foils were then carefully removed from the target holder in irradiation order. Each foil was then counted on a 70% HPGe γ-ray detector that

was energy calibrated using a National Institute of Standards and Technology (NIST) traceable ^{152}Eu point source and efficiency calibrated using a polynomial fit. The 343 and 983.5 keV lines were monitored for ^{175}Hf and ^{48}V, respectively. The resulting spectra were processed using the GF3 module within RadWare [10]. The cross sections were then calculated based on the obtained results.

Table I. Calculated beam energy on the center of each target (E_{cot}) with an estimated error of 0.2% and 4% from the initial beam energy and energy loss calculations, respectively.

Target	Thickness (μm)	E_{cot} (MeV)
Ta-1	80.8±1.5	21.3
Ta-2	80.8±1.5	19.8
Ta-3	80.8±1.5	18.1
Lu-1	96 ±7	16.7
Ti-1	4.9±0.2	16.0
Lu-2	96 ±7	15.3
Ti-2	4.9±0.2	14.6
Lu-3	96 ±7	13.8
Ti-3	4.9±0.2	13.1
Lu-4	96 ±7	12.2
Ti-4	4.9±0.2	11.5
Lu-5	96 ±7	10.6
Ti-5	4.9±0.2	9.7
Lu-6	96 ±7	8.7
Ti-6	4.9±0.2	6.9
Lu-7	96 ±7	5.2

3. Results and Discussion

3.1 γ-ray measurements of proton irradiated ^{nat}Ti and ^{nat}Lu target foils

The spectra collected from the peak of the excitation functions are shown in Fig. 2 a and b, respectively. All unlabeled peaks can be attributed to the minor products of the irradiation, minor gamma rays of ^{175}Hf, the natural decay series, background or electronic noise. The lack of γ-rays should be noted in the Lu foil spectrum. This indicates that ^{175}Hf is free of γ-ray interferences and can be produced for off-line chemical studies.

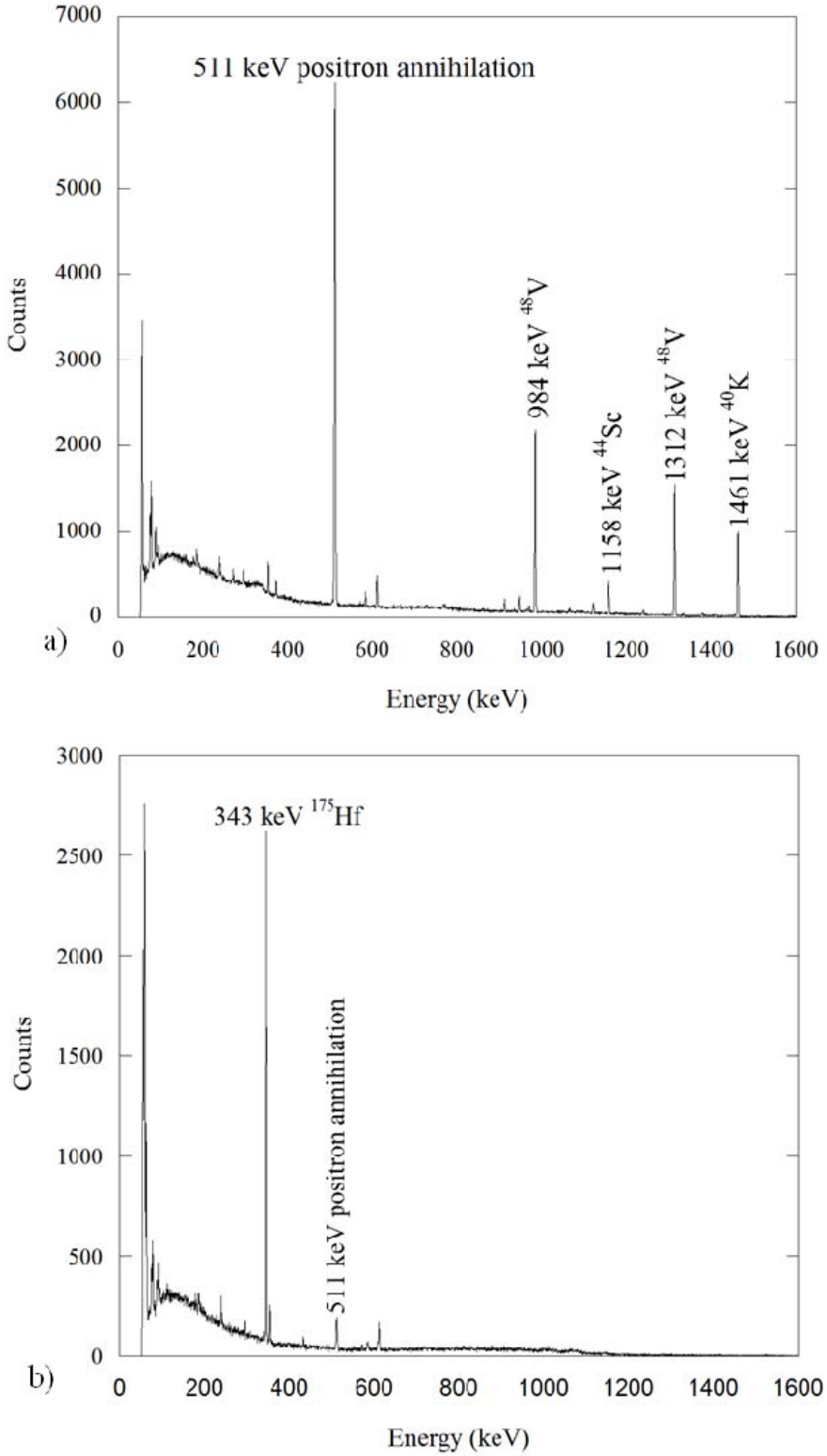

FIG. 2. The γ-ray spectra for proton irradiated foils at the peak of the excitation function. a) The spectrum from the ^{nat}Ti foil that was bombarded with 13.1 MeV protons in the center of the target. b) The spectrum from the ^{nat}Lu foil that was bombarded with 10.6 MeV protons in the center of the target

3.2 Excitation function for the ^{nat}Ti(p,x)^{48}V reaction

The measured excitation function for the ^{nat}Ti(p,x)^{48}V reaction is shown in Fig. 3 and the resulting cross sections and associated errors are listed in Table II. All cross sections were calculated based on the absolute detector efficiency, γ-ray intensity, decay corrections to the produced activity, beam intensity and the areal density of the target. For the ^{nat}Ti(p,x)^{48}V reaction the cross section was based upon the 983.5 keV γ-ray line which has an intensity of 99.98% [11]. All data points within the measured cross section were systematically high compared to the recommended data available through the International Atomic Energy Agency (IAEA) [7]. The cross sections were shifted down based on a χ^2 minimization fit. The correction lowered the cross sections by 27.32%. The error in the cross section is likely due to a systematically low output reading of the beam current integrator. The shape of the excitation function is in excellent agreement with the recommended data without further corrections, when the error bars of the reported data are considered. The maximum cross section was 368±26 mb and occurred at proton energy of 13.1 MeV.

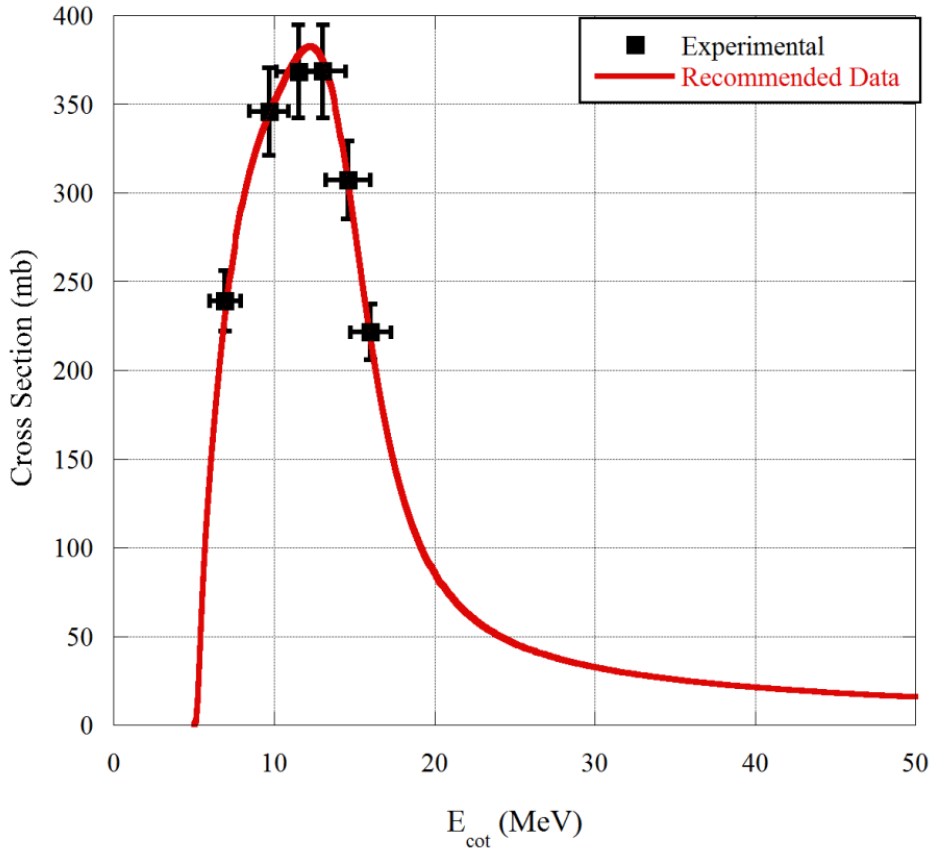

FIG. 3. The excitation function for the ^{nat}Ti(p,x)^{48}V reaction, systematically corrected and plotted as the squares, compared to the recommended data from the IAEA, plotted as a solid line [7].

Table II. Experimentally determined cross sections for the $^{nat}Ti(p,x)^{48}V$ reaction with their associated error. These data have been corrected with an experimentally determined correction factor of 27.32%; see the main text for a discussion.

Target	E_{cot} (MeV)	Cross section (mb)
Ti-6	6.9	239±17
Ti-5	9.7	346±25
Ti-4	11.5	368±26
Ti-3	13.1	368±26
Ti-2	14.6	307±22
Ti-1	16.0	221±16

3.3 Excitation function for the $^{nat}Lu(p,x)^{175}Hf$ reaction

The measured and predicted excitation functions for the $^{nat}Lu(p,x)^{175}Hf$ reaction are presented in Fig 4. The 27.32% systematic error correction that was applied to the $^{nat}Ti(p,x)^{48}V$ reaction was also applied to the $^{nat}Lu(p,x)^{175}Hf$ cross section data. The resulting cross sections and associated errors are

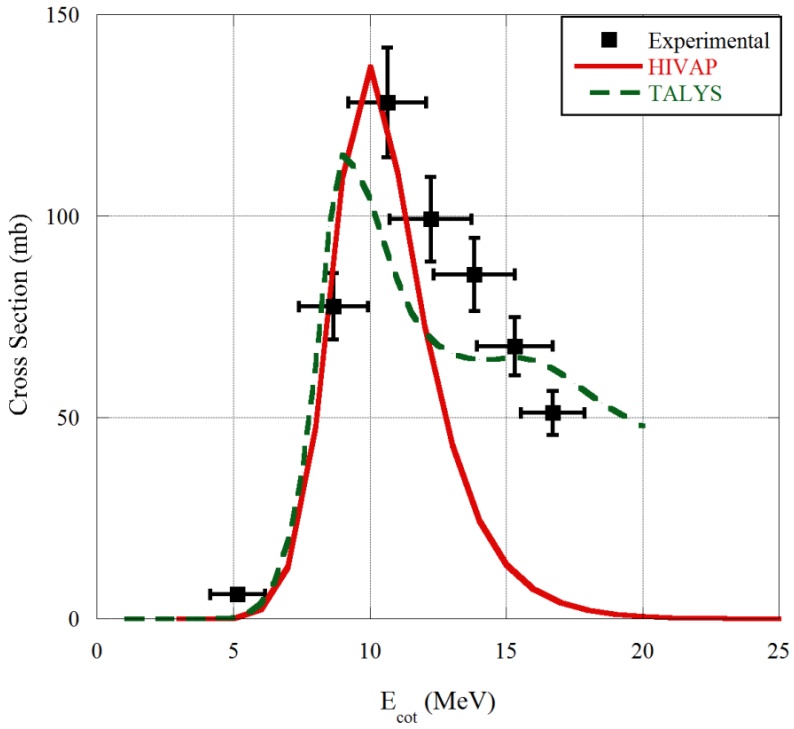

FIG. 4. The measured excitation function for the reaction $^{nat}Lu(p,x)^{175}Hf$ compared to that calculated with the HIVAP and TALYS codes. The reported error bars (shown in Table 1) are largely due to the error in the target thickness. The error at 5.2 MeV is smaller than the data point.

listed in Table III. The cross section was calculated based on the 343 keV γ-ray line which has an intensity of 84% [11]. Experimentally, the maximum cross section is 128±14 mb and is observed at a proton energy of 10.6 MeV.

Both the HIVAP and TALYS codes were used to model the excitation function for the $^{nat}Lu(p,x)^{175}Hf$ reaction [12,13]. Within the experimental error bars, both the HIVAP and TALYS codes predict the peak of the excitation function accurately. This indicates that both the HIVAP and TALYS codes predict particle evaporation from the compound nucleus accurately. The overall shape of the excitation function is poorly predicted by the HIVAP code, which can be explained by the fact that the HIVAP code does not account for the pre-equilibrium decay process (see [14] for a review). In this case, a localized region of high energy density is formed as the projectile interacts with the target nucleus, and particles can be emitted from this region before the dinuclear system can fully equilibrate to become a compound nucleus. Due to the high local temperature, these particles are emitted with significant kinetic energies; when the compound nucleus finally forms it has a relatively low excitation energy. In this way, reaction products can be formed at projectile energies greater than those normally expected from the traditional compound nucleus reaction mechanism, and the measured excitation function extends to higher projectile energies [14]. This can be seen both in the recommended excitation function for $^{nat}Ti(p,x)^{48}V$ in Fig. 3 and the experimental data for $^{nat}Lu(p,x)^{175}Hf$ in Fig. 4. [14]. The TALYS code predicts the shape of the ^{175}Hf excitation function much better than the HIVAP code, as the TALYS code accounts for the pre-equilibrium decay process, although a pronounced shoulder is not observed.

It will be necessary to use the K150 cyclotron to achieve higher beam intensities desired for the larger scale production of ^{175}Hf (not the K500, as used in this experiment) [15]. Based on the measured excitation function, a beam intensity of 10 μA and a target thickness of 300 μm, approximately 0.5 mCi of ^{175}Hf can be made in an 8 hour irradiation period using the K150 cyclotron.

Table III. Experimentally determined cross sections for the $^{nat}Lu(p,x)^{175}Hf$ reaction. These data have been corrected with an experimentally determined correction factor of 27.32%; see the main text for a discussion.

Target	E_{cot} (MeV)	Cross section (mb)
Lu-7	5.2	6.0±0.5
Lu-6	8.7	77±8
Lu-5	10.6	128±14
Lu-4	12.2	99±11
Lu-3	13.8	85±9
Lu-2	15.3	67±7
Lu-1	16.7	51±5

4. Conclusions

The ^{nat}Lu(p,x)^{175}Hf excitation function has been measured. During the γ-ray measurements, very little γ-ray contamination was seen, indicating that relatively clean ^{175}Hf can be produced at the Cyclotron Institute at Texas A&M University for use in off-line chemical studies of Rf homologs. The maximum cross section was determined to be 128±14 mb, observed at a proton beam energy of 10.6 MeV. A complete description of this work has been published in Ref [1].

[1] M. E. Bennett, D.A. Mayorov, K.D. Chapkin, M.A. Alfonso, T.A. Werke. Nucl. Instrum. Methods Phys. Res. **B276**, 62 (2012).

[2] V. Pershina, Chem. Rev. **96**, 1977 (1996).

[3] M.Schädel, Angew. Chem. Int. Ed. **45**, 368 (2006).

[4] F. Tárkányi, F. Szelecsényi, S. Takács, A. Hermanne, M. Sonck, A. Thielemans, M.G. Mustafa, Yu. Shubin, Z. Youxiang. Nucl. Instrum. Methods Phys. Res. **B168**, 144 (2000).

[5] M.A. Avila-Rodriguez, J.S. Wilson, M.J. Schueller, S.A. McQuarrie. Nucl. Instrum. Methods Phys. Res. **B266**, 3353 (2008).

[6] S. Takács, F. Tárkányi, A. Hermanne, R.A. Rebeles. Nucl. Instrum. Methods Phys. Res. **B269**, 2824 (2011).

[7] F. Tárkányi, S. Takács, K. Gul, G. Hermanne, M.G. Mustafa, M. Nortier, P. Obložinský, S.M. Qaim, B. Scholten, Yu.N. Shubin, Z. Youxiang, *Beam Monitor Reactions*, IAEA document number IAEA-TECDOC-1211, 49 (2001).

[8] A.A. Alharbi, M. McCleskey, G. Tabacaru, B. Roeder, A. Banu, A. Spiridon, E. Simmons, L. Trache, R.E. Tribble, *Progress in Research*, Cyclotron Institute, Texas A&M University (2008-2009), p.V-19.

[9] J.F. Ziegler, Computer Code SRIM-2008, www.srim.org (accessed June 2011).

[10] D.C. Radford, RadWare Software Package, http://radware.phy.ornl.gov (accessed June 2011)

[11] R.B. Firestone, L.P.Ekström, Table of Radioactive Isotopes, http://ie.lbl.gov/toi/index.asp (accessed June 2011).

[12] W. Reisdorf, Z. Phys. A **300**, 227 (1981).

[13] A.J. Koning, S. Hilaire and M.C. Duijvestijn, .TALYS-1.0., *Proceedings of the International Conference on Nuclear Data for Science and Technology*, April 22-27, 2007, Nice, France, editors O.Bersillon, F.Gunsing, E.Bauge, R.Jacqmin, and S.Leray, EDP Sciences, 2008, p. 211-214.

[14] M. Blann, Ann. Rev. Nucl. Sci. **8**, 886 (1975).

[15] Cyclotron Institute: Cyclotron Institute Proposed Upgrade, http://cyclotron.tamu.edu/facility_upgrade.pdf (accessed July 2011)