Ion Beam Analysis of Targets Used in Controlatron Neutron Generators*

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Abstract

Controlatron neutron generators are used in neutron detection systems at Sandia National Laboratories. To provide for increased tube lifetime for the moderate neutron flux output of these Controlatron generators, metal hydride (ZrT_2) target fabrication processes have been developed. Also, to provide for manufacturing quality control of these targets, ion beam analysis techniques are used to determine film composition. The load ratios of ZrT_2 Controlatron neutron generator targets have been successfully measured by simultaneously acquiring RBS and ERD data using a He^{++} beam energy of 10 MeV. Several targets were measured and the film thicknesses obtained from RBS measurements agreed within ±2% with Dektak profilometer measurements. The target fabrication process and ion beam analysis techniques will be presented.

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Introduction

Pulsed neutron generators are used in several elemental analysis applications using prompt inelastic scattering (n,n′γ) and radiative capture (n,γ) reaction techniques. These applications range from bulk materials analysis to well logging and the assay of transuranic materials in hazardous waste [1]. Neutron generators can also be used for testing neutron detection systems. The required neutron flux used in these applications can range from ~10^6 to 10^{10} neutrons/pulse into 4π steradians. Therefore, these generators rely on the T(d,n)^{4}He inelastic reaction to create the desired high neutron flux. To provide reasonable generator lifetimes, high quality targets and sources must be used.

Controlatron neutron generators are used in neutron detection systems at Sandia National Laboratories. These tubes use a Penning ion source to create an ion beam consisting of mostly D_2^+, with small amounts of D^+ and D_3^+. This beam is pulsed on for ~10 μsec and upon hitting the tritiated target creates a neutron flux of ~10^6 neutrons into 4π steradians as measured by a lead probe neutron detector. An overview of Pb probe detector technology is found in [2]. A metal hydride (ZrT_2) target fabrication process has been developed to provide for increased tube lifetime, typically ≥10,000 shots at this neutron flux output. Also, to provide for manufacturing quality control of these targets, non-destructive ion beam analysis (IBA) techniques are used to determine film composition. While Sandia manufactures the targets used in the Controlatron neutron generator, the rest of the tube is manufactured and assembled by Thermo Scientific [3].

Experimental Method and Results

The Controlatron target substrate is cut from a molybdenum sheet using a molybdenum wire electrical discharge machining (EDM) process. The target has a diameter of 25.4mm, is 0.254mm thick, and has four equidistant protruding tabs on the circumference for mounting. In

preparation for thin film evaporation, the substrate is degreased, etched using a nitric/sulfuric acid solution, and then hydrogen fired at 800ºC. This process causes a surface roughening that provides for better film adhesion. Once cleaned, the Mo substrates are handled in Class 100 or Class 1000 clean rooms. The zirconium film is evaporated onto the target substrate in an ultra high vacuum system equipped with an electron beam deposition gun. Quartz witness coupons are also mounted in the evaporation chamber and are later used for confirming film thickness (using a Veeco Dektak Model 3ST profilometer). The substrates are positioned onto a dome shaped fixture which is rotated during deposition. The substrates and the vacuum system are baked out at temperatures up to 550ºC. Once the pressure is stabilized to 5 x 10^{-7} Torr or less, the zirconium film is evaporated onto the substrates at 425 – 475ºC at deposition rates of up to 100Å per second. The final film is transparent on the substrate and has a thickness of ~1.5 microns.

The zirconium film is reacted with tritium in another ultra high vacuum chamber located within a glovebox. The Controlatron targets are loaded into the chamber and heated at elevated temperatures to break up the zirconium oxide surface layer, a process called film activation. After the film is activated, tritium gas is introduced into the process chamber and allowed to react with the zirconium film to form a di-tritide (ZrT_2). The process tritium used is stored in a depleted uranium storage bed. The exact temperature during this process step is considered proprietary. However, one can see the range of temperatures available to load the film to form a di-tritide from the phase diagram [4] shown in Fig. 1. Finally, the gas is pumped away and the chamber allowed to cool to ambient temperature before removing them from the chamber. Again, the ZrT_2 film is transparent and so it is not possible to visually verify if the zirconium film reacted well with the tritium gas.

A 6 MV EN tandem Van de Graaff-Pelletron accelerator is used to non-destructively measure the composition of the ZrT_2 film using a 10.0 MeV He^{++} beam with the target tilted at a 70º angle. Both an RBS and ERD spectrum are acquired simultaneously using a multi-parameter multichannel analyzer (FAST ComTec MPA-3) to obtain the areal densities of the Zr and tritium. A few percent residual hydrogen and deuterium are also incorporated in the film during the loading process. The experimental geometry has been previously discussed in [5]. The beam energy was calibrated using resonances as described in ref. 5 and has an energy spread of ±1 keV. The backscattering detector used for RBS was an Ortec (BB-016-050-400), having a ~16 keV energy resolution and 400μm depletion depth. The backscattering angle θ and forward scattering angle φ were respectively, 165º and 30º, with an estimated deviation of ±0.2º. The detector solid angle Ω was 6.888 msr for RBS and 1.287 msr for ERD, as measured using a NIST traceable ^{238}Pu α source that had a certified activity of ±1.3%. Before an analysis run the number of incident ions collected into a Faraday cup and the backscattered yield from a rotating Au/Al paddle that chopped the beam at 1.25 Hz upstream from the sample was measured and used to calibrate the counts per μC. The beam was defined by two sets of tantalum slits into a rectangle 1.0 mm H x 0.36 mm W, with the projected beam dimensions onto a film at the 70º tilt angle being ~1.0 mm^2. The total number of incident ions on a sample for subsequent runs could then be measured using the relationship $Q = (Q_{cal}/2e)(Y/Y_{cal})$, where Q_{cal} is the integrated charge collected into the Faraday cup during the calibration run, e is the fundamental electronic charge, Y_{cal} is the Au yield from the paddle during the calibration run, and Y is the Au yield from a data acquisition run. The uncertainty in Q is ±2.0%.

The analysis program SIMNRA [6] was used to determine the Zr areal density. A typical RBS spectrum is shown in Fig. 2. A ~10nA beam current and a chamber vacuum of ~2 x 10^{-8}

Torr are typical operating conditions. Since the target substrate was purposely roughened to enhance film adhesion, the film has the same roughness. Because the target is also tilted to 70°, plural scattering (multiple large angle scattering of the beam because of surface roughness) becomes a problem [7]. Although a fit to the spectrum and an areal density may be obtained using the roughness simulation available in SIMNRA, see Fig. 3, the areal density is inaccurate because the shape of the spectrum is distorted due to plural scattering. However, it was determined from normal RBS spectra simulation and placing the Zr and Mo peak intersection at the half height of the slope between the two, as shown in Fig. 3, that the RBS thickness was the same as that obtained (within ~2%) from Dektak thickness measurements of quartz witness coupons inserted into the film deposition chamber. The RBS thickness was determined by using a mass density for Zr hydride of 5.47 g/cm^3.

The areal densities of the three isotopes of hydrogen (hydrogen, deuterium, and tritium or H, D, and T) were obtained using ERD. A particle telescope is used to separate the ERD spectra of the three isotopes. A 109 μm thick aluminized Mylar foil is placed in front of a curved aperture, and is used to range out the He^{++} ion beam forward scattered off the target, but allow recoiled H, D, and T through. The curved aperture allows for better energy resolution. A thin (65.2 μm) silicon surface barrier detector (Ortec TD-040-300-75-S) with an active region of 300 mm^2 is behind the aperture, immediately followed by a thick silicon surface barrier detector (Ortec BU-018-300-300), which also has a 300 mm^2 active region. These four components complete the particle telescope. H, D, and T recoiled from the target with enough energy to pass through both the Mylar foil and the thin detector create a signal ΔE which is used to start a timing window with the signal E created when the particle stops in the thick detector. The signals ΔE and E, as shown Fig. 4, are processed in the multi-parameter multichannel analyzer

to quantify the amount of each hydrogen isotope in the target. These signals can be projected onto the energy axis, see Fig. 4 inset, and integrated to provide the isotope yield Y. The areal density can then be calculated from the equation, $\eta = Y \cdot \cos\theta / Q\Omega\sigma(E,\varphi)$, using Sandia measured cross sections σ at beam energy E, taking into account the analysis beam mean energy loss in the film as discussed in [5]. Other terms of the equation were previously discussed.

Conclusions

The load ratios of ZrT_2 Controlatron neutron generator targets have been successfully measured by simultaneously acquiring RBS and ERD data using a He^{++} beam energy of 10 MeV. Several targets were measured and the film thicknesses obtained from RBS measurements agreed within ±2% with Dektak profilometer measurements. From previous work [5] involving load ratio measurements of $Er(HDT)_2$ targets it was determined that the overall measurement error was 8.3%, 3σ. As we have excellent agreement between RBS and profilometry measurements of thickness, the same error has been assigned to the measurement of the Controlatron targets. IBA measurements have confirmed di-tritide loading of the robust Zr target films fabricated by Sandia and the targets are now incorporated into Controlatron neutron tubes for use in neutron detection systems.

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Figure Captions

Figure 1. Phase diagram [4] showing H/Zr atomic ratio, pressure, and temperature regimes for hydriding Zr films.

Figure 2. Typical RBS spectrum for Controlatron target ZrT_2 films using a 10 MeV He^{++} beam energy and target tilt angle of 70°. An enhanced cross section for O, ratio-to-Rutherford of 250 at this energy, shows a thin surface oxide on the film.

Figure 3. The analysis of a typical RBS spectrum using SIMNRA software [6] is shown. Although the simulation using roughness parameters to fit the experimental data shows good agreement, comparison of the RBS thickness with Dektak profilometry measurement shows a ~15% discrepancy. Plural scattering that distorts the experimental profile because of the substrate and film roughness coupled with a target tilt angle of 70° is the likely cause. Simulation without roughness parameters while lining up the intersection of the Zr and Mo peaks at the half height of the step height between the two shows thickness agreement of ±2%.

Figure 4. Multiparameter ERD data acquired using a particle detector with a multiparameter multichannel analyzer allows for the separation of the hydrogen isotopes in the hydrided Controlatron targets. Regions of interest can be drawn around each isotope and the yield data projected onto the energy axis, such as is shown in the inset for tritium.

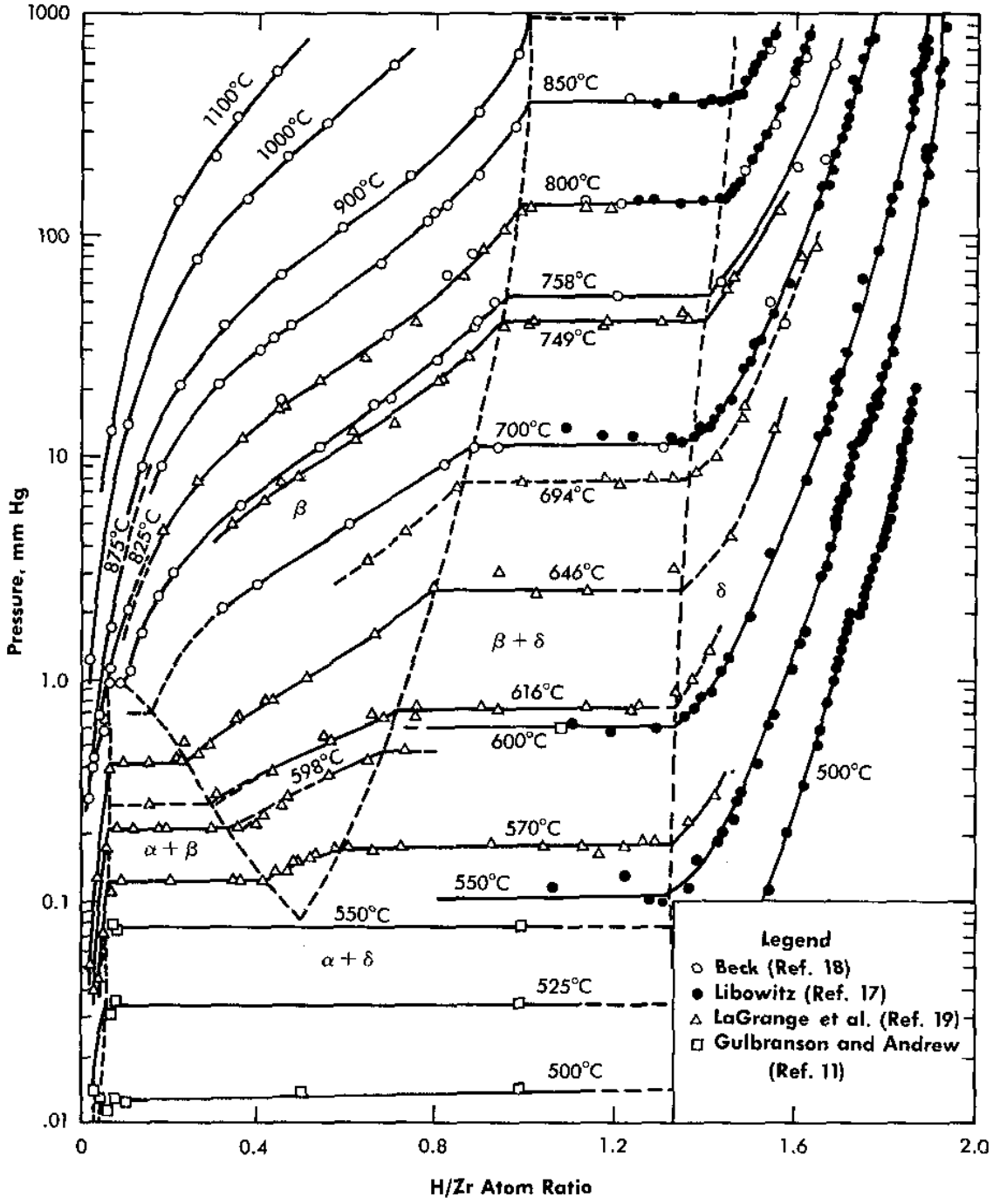

1000
100
10
1.0
0.1
.01
Pressure, mm Hg
0
0.4
0.8
1.2
1.6
2.0
H/Zr Atom Ratio
1100°C
1000°C
900°C
875°C
825°C
850°C
800°C
758°C
749°C
700°C
694°C
646°C
616°C
600°C
598°C
570°C
550°C
550°C
525°C
500°C
500°C
β
β + δ
δ
α + β
α + δ
Legend
Beck (Ref. 18)
Libowitz (Ref. 17)
LaGrange et al. (Ref. 19)
Gulbranson and Andrew (Ref. 11)

Fig. 1

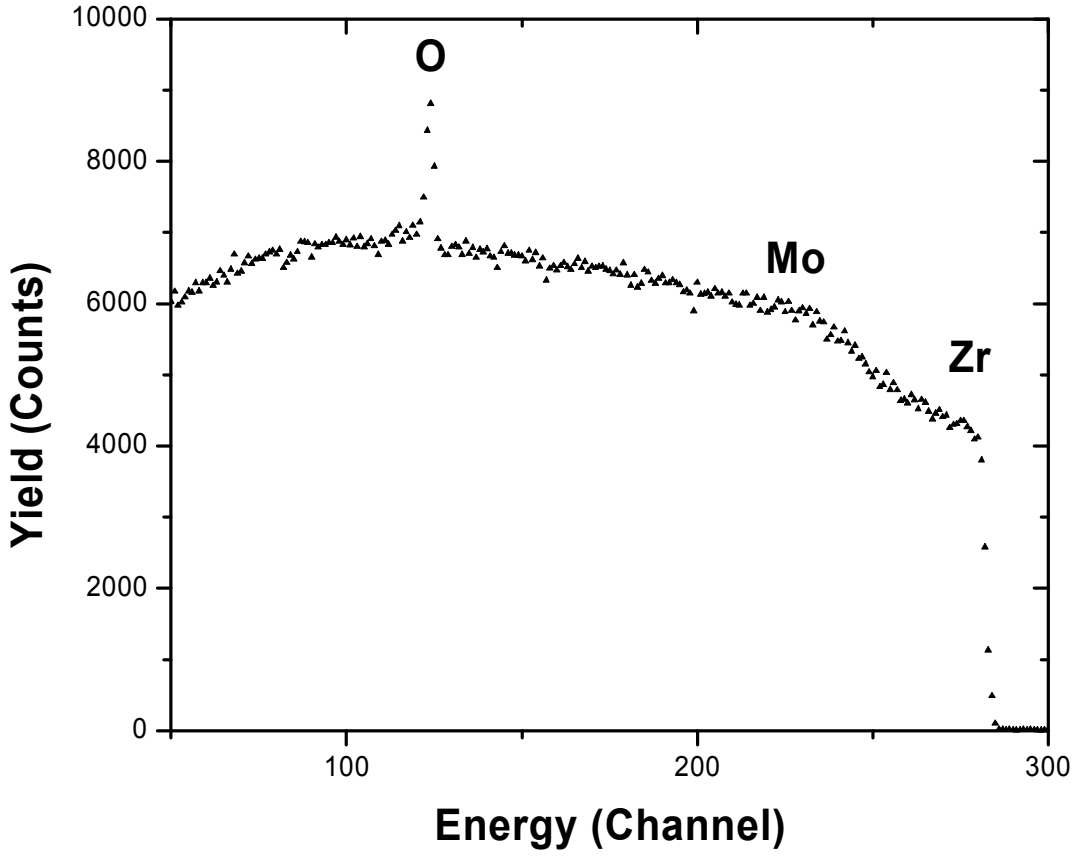
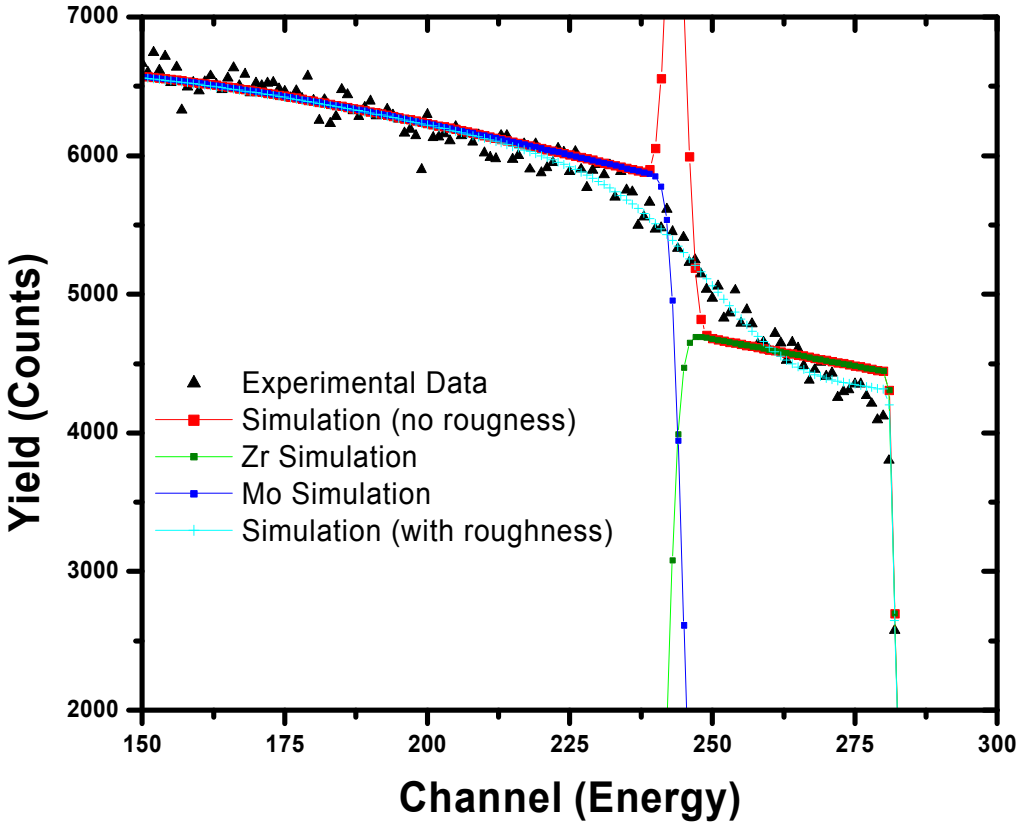

10000
8000
6000
4000
2000
0
Yield (Counts)
O
Mo
Zr
100
200
300
Energy (Channel)

Fig. 2

1


Yield (Counts)
Channel (Energy)
Experimental Data
Simulation (no rougness)
Zr Simulation
Mo Simulation
Simulation (with roughness)
7000
6000
5000
4000
3000
2000
150
175
200
225
250
275
300

2

3 **Fig. 3**

4

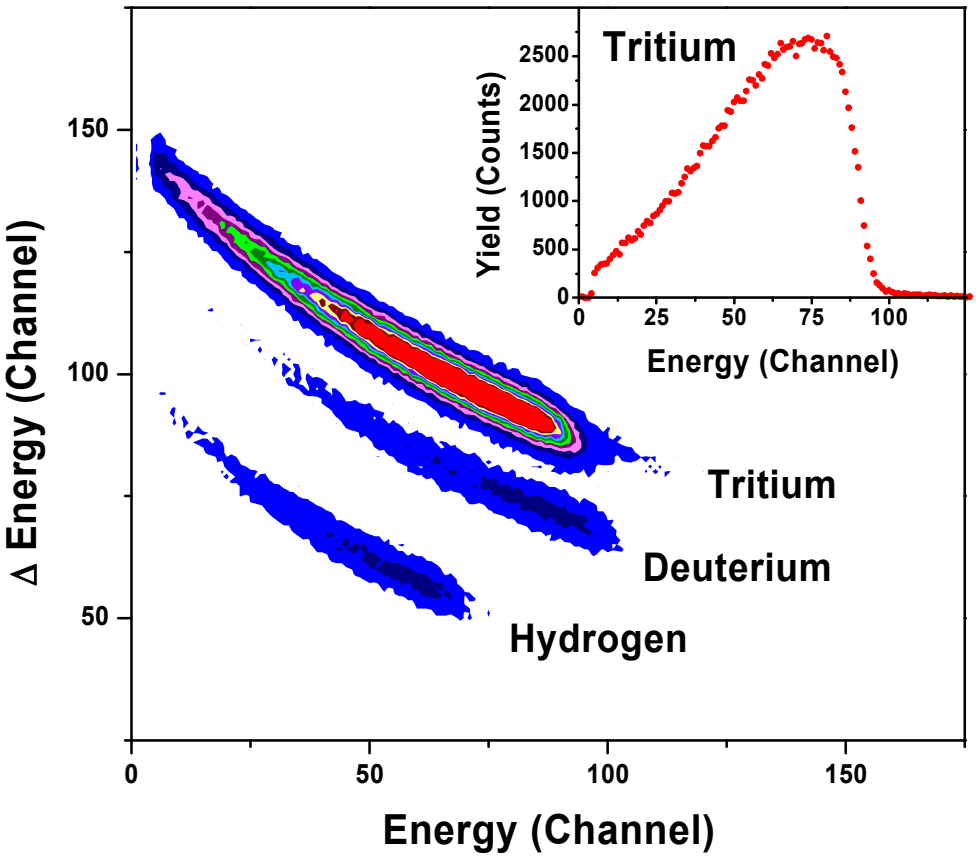

Tritium
Yield (Counts)
2500
2000
1500
1000
500
0
0
25
50
75
100
Energy (Channel)
Tritium
Deuterium
Hydrogen
Δ Energy (Channel)
150
100
50
0
50
100
150
Energy (Channel)

Fig. 4