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### The ORNL Analysis Technique for Extracting \$\$-Delayed Multi-Neutron Branching Ratios with BRIKEN

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#### 35 Abstract

Many choices are available in order to evaluate large radioactive decay networks. There are many parameters that influence the calculated  $\beta$ -decay delayed single and multi-neutron emission branching fractions. We describe assumptions about the decay model, background, and other parameters and their influence on  $\beta$ -decay delayed multi-neutron emission analysis. An analysis technique, the ORNL BRIKEN analysis procedure, for determining  $\beta$ -delayed multi-neutron branching ratios in  $\beta$ -neutron precursors produced by means of heavy-ion fragmentation is presented. The technique is based on estimating the initial activities of zero, one, and two neutrons occurring in coincidence with an ion-implant and  $\beta$  trigger. The technique allows one to extract  $\beta$ -delayed multi-neutron decay branching ratios measured with the <sup>3</sup>He BRIKEN neutron counter. As an example, two analyses of the  $\beta$ -neutron emitter <sup>77</sup>Cu based on different *a priori* assumptions are presented along with comparisons to literature values.

#### 36 1. Introduction

Measuring single and multi-neutron emission after  $\beta$  decay of neutron-rich 37 nuclei is important in order to understand the evolution of nuclear structure and 38 its impact on  $\beta$ -decay properties far from stability. Multi-neutron emission after 39  $\beta$  decay of neutron-rich nuclei also impacts astrophysical r-process calculations 40 that estimate the abundance of various nuclei in the galaxy [1, 2]. Present and 41 future  $\beta$ -decay experiments with neutron-rich exotic nuclei created from the 42 fragmentation of heavy ions involve complex decay networks. It is important 43 to have a robust method to reliably extract the decay information associated 44 with each nucleus. The  $\beta$  delayed neutrons at RIKEN (BRIKEN) collaboration 45 measured the  $\beta$  decays of many neutron-rich nuclei that exhibit zero, single, 46 and multi-neutron emission probabilities,  $P_{xn}$  (where x = 0, 1, 2, ...) [3]. 47

<sup>48</sup> Techniques for evaluating single neutron branching ratios,  $P_{1n}$ , with <sup>3</sup>He <sup>49</sup> tubes [4, 5] must be extended to include the possibility of multi-neutron  $\beta$  decay. <sup>50</sup> So far, in heavy nuclei, only one case of a large  $\beta$ -delayed 2 neutron emitter, <sup>86</sup>Ga ( $P_{2n} = 20(10)\%$ ), has been reported [6]. The BRIKEN collaboration aims

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to extend current knowledge of two and more neutron emitters in medium and
heavy mass nuclei [3].

In this paper we present an analysis technique that may be applied to other 54 situations, though the discussion of the parameters is focused on the BRIKEN 55 experiment. The analysis technique is based upon measuring zero, one, and two 56 neutron activities detected in coincidence with an ion-implant and a  $\beta$  trigger, 57 but the technique may be applied to any decay activity in coincidence with an-58 other detector. The associated systematic and statistical uncertainties present 59 several challenges evaluating  $P_{xn}$ . This paper discusses these challenges and 60 presents one analysis procedure, the ORNL BRIKEN analysis procedure, used 61 to evaluate  $P_{xn}$ . Alternative analysis methods along with expanded experimen-62 tal detail will be published separately [8]. This manuscript also discusses the 63 analysis of BRIKEN data using as an example <sup>77</sup>Cu data. The analysis of <sup>77</sup>Cu 64 is chosen because it is a known  $\beta$ -delayed neutron emitter, with a known half 65 life of 468(2) ms [13] and a consistently measured single neutron decay fraction, 66  $P_{1n} = 31.0(38)\%$  [14] and  $P_{1n} = 30.3(22)\%$  [15]. The present paper does not 67 comment on the evaluation of the associated  $\gamma$ -ray detection, which will be pre-68 sented in a future publication. In addition to presenting the ORNL BRIKEN 69 analysis method, we offer comments on the inputs and parameters and their 70 influence on the errors in evaluating  $P_{xn}$ . 71

#### 72 2. Brief BRIKEN Detector Description

<sup>73</sup> The BRIKEN detector as used in the experiments at RIKEN consists of <sup>74</sup> 140 <sup>3</sup>He neutron detector tubes, a dual purpose ion-implant and  $\beta$  detector <sup>75</sup> (implant- $\beta$  detector), and two HPGe clovers and one of the experimental setups <sup>76</sup> is schematically shown in figure 1.

The BRIKEN detector was designed to maximize the neutron efficiency while 77 keeping the neutron efficiency as uniform as possible over a wide range of initial 78 neutron energies. The uniform neutron efficiency minimizes the contribution to 79 the neutron efficiency uncertainty from the initial neutron kinetic energy. This 80 effect and its impact on the BRIKEN design is discussed in [7]. From the analysis 81 presented in [7] and neutron source measurements, the average single neutron 82 efficiency of the BRIKEN detector is 62(2)% for neutrons with kinetic energies 83 ranging from thermal energies to 5 MeV. Further details of the BRIKEN setup 84 used in the commissioning experiments can be found in [7, 8]. 85

BRIKEN was placed on the zero degree beam line following BigRIPS at the RI Beam Factory (RIBF) of the RIKEN Nishina Center. The nuclei were identified per event by means of the BigRIPS separator [9].

<sup>89</sup> Several different implant- $\beta$  detectors were used in the various BRIKEN ex-<sup>90</sup> perimental runs at RIKEN. Two different silicon based implant- $\beta$  detectors <sup>91</sup> were used in separate runs, the AIDA detector [10] and the WAS3ABi detector <sup>92</sup> [11]. In conjunction with the WAS3ABi detector, a YSO scintillator [12] based <sup>93</sup> implant- $\beta$  detector was also used. All of the implant- $\beta$  detectors are segmented <sup>94</sup> in order to reduce ion-correlated background  $\beta$  triggers. Two HPGe clovers from the CLARION array of Oak Ridge National Laboratory were used to detect  $\gamma$ rays in coincidence with  $\beta$  and  $\beta$ -delayed neutron decays.

#### 97 3. Main Analysis Result

In this section, the fundamental equation used in the analysis is presented. A derivation of this fundamental equation is presented in Appendix A. The fundamental equation that contains only implant- $\beta$  time dependent terms can be written as

$$\begin{pmatrix} A_{0n}(t) \\ A_{1n}(t) \\ A_{2n}(t) \end{pmatrix} = A(t)\epsilon_I \varepsilon_\beta r_{0n} \mathbf{E} \begin{pmatrix} P_{0n} \\ P_{1n} \\ P_{2n} \end{pmatrix},$$
(1)

where  $A_{xn}(t)$  is the implant- $\beta$  activity with detecting x neutrons at time t (or summed over a range of times), A(t) is the overall activity at the same time,  $\epsilon_I$  is the implant efficiency,  $\varepsilon_\beta$  is the  $\beta$  efficiency for zero neutron decays,  $r_{0n}$  is the probability to detect no background neutrons in a given time window,  $P_{xn}$ is the branching probability for emitting x neutrons, and **E** is a matrix given by

$$\mathbf{E} = \begin{pmatrix} 1 & a_{1}\epsilon_{10n} & a_{2}\epsilon_{20n} \\ r_{1n}/r_{0n} & a_{1}\left(\epsilon_{11n} + \epsilon_{10n}r_{1n}/r_{0n}\right) & a_{2}\left(\epsilon_{21n} + \epsilon_{20n}r_{1n}/r_{0n}\right) \\ r_{2n}/r_{0n} & a_{1}\left(\epsilon_{11n}r_{1n}/r_{0n} + \epsilon_{10n}r_{2n}/r_{0n}\right) & a_{2}\left(\epsilon_{22n} + \epsilon_{21n}r_{1n}/r_{0n} + \epsilon_{20n}r_{2n}/r_{0n}\right) \\ \end{cases}$$

In the matrix  $\mathbf{E}$ ,  $a_x$  is the ratio of the x-neutron  $\beta$  efficiency  $(\varepsilon_{\beta x})$  to 0-neutron  $\beta$ efficiency  $(\varepsilon_{\beta})$ ,  $\epsilon_{xyn}$  is the probability to detect y neutrons given that x neutrons were emitted  $(x \ge y)$ , and  $r_{xn}$  is the probability that x background neutrons are detected within a given time window. By either considering the reasoning in Appendix A or merely extending the patterns in Equation 2, the matrix  $\mathbf{E}$  is easily extended to include three and four neutron terms  $(A_{3n}(t), A_{4n}(t), P_{3n}, P_{4n}, r_{3n}, r_{4n}, \epsilon_{33n}, etc...)$  if needed.

After solving equation 1 for the  $P_{xn}$  and taking the ratio of  $P_{xn}$  while requiring the sum to be 1.0, the dependence of the results on the variables A(t),  $\epsilon_I$ ,  $\varepsilon_\beta$ , and  $r_{0n}$  is removed.

Equations 1 and 2 are applicable to any situation where decay data can be separated into coincidence with a noisy secondary detector. In our case the secondary detector is the BRIKEN neutron detector. In most cases the  $a_x$  can be ignored by setting them equal to 1.0.

#### 121 4. Discussion of BRIKEN Specific Parameters

<sup>122</sup> Calculating  $P_{xn}$  involves evaluating the number of correlated implant trig-<sup>123</sup>gers with  $\beta$  triggers versus implant- $\beta$  times ( $\beta$  time minus implant time), here-<sup>124</sup>after referred to as implant- $\beta$  activities. Using the estimated initial activity (the <sup>125</sup>activity at the implant time) from the implant- $\beta$  activity gated in coincidence <sup>126</sup>on the neutron multiplicity gives a way to obtain the  $P_{xn}$ .

For each ion-implant signal all associated  $\beta$  signals within  $\pm 10$  sec within  $\pm 3$  pixels of the implant pixel of AIDA are correlated in software. Each pixel

in AIDA has a 0.58 mm pitch in both the x and y direction. The implant- $\beta$ 129 time correlation plot from a 60 hour BRIKEN run for BigRIPS selected <sup>77</sup>Cu 130 implanted ions is shown in figure 2. In addition to the implant- $\beta$  time correlation 131 activity plots, there are implant- $\beta$  time correlation activity plots gated on the 132 number of neutrons detected within the neutron thermalization time window, 133  $T_{th} = 200 \ \mu s$ , after each  $\beta$  signal (neutron-multiplicity implant- $\beta$  activities). 134 The activity gated on zero neutrons detected is shown in figure 3, the activity 135 gated on one neutron detected is shown in figure 4, and the activity gated 136 on two neutrons detected is shown in figure 5. Below we describe how the 137 estimated initial activity of the neutron-multiplicity implant- $\beta$  activities are 138 used to calculate the  $P_{xn}$ . 139

<sup>140</sup> Before discussing the connections between the initial activity of the neutron-<sup>141</sup> multiplicity implant- $\beta$  activities and the  $P_{xn}$ , a discussion of several required <sup>142</sup> parameters is presented. Some of these required parameters can be measured, <sup>143</sup> while others must be estimated. The evaluation and propagation of uncertainties <sup>144</sup> from measured and estimated parameters through the analysis is presented. A <sup>145</sup> discussion of the parameters considered in the BRIKEN  $P_{xn}$  evaluations is given <sup>146</sup> below.

#### 147 4.1. Implant- $\beta$ Background

Random  $\beta$  signals in coincidence with each implant contribute to the nearly 148 constant background in each implant- $\beta$  time correlation plot. These random 149  $\beta$  signals originate from other nearby implant  $\beta$  signals and implant  $\beta$  signals 150 that are not detected by the  $\beta$  trigger. The small slope of the background is 151 associated with short time drops (up to tens of seconds) in the rate of implanted 152 ions from an otherwise DC beam. When the beam drops before an implant, this 153 lowers the correlated  $\beta$  counts before the implant. Similarly, beam drops after 154 an implant lower the background counts after the implant. Because there are 155 relatively few beam drops, this is a small yet observable effect. 156

An accurate description of the background affects the fitting of the neutron-157 multiplicity implant- $\beta$  activities. Especially when the background models dif-158 fer on the order of the daughter and granddaughter activities. One way to 159 minimize the impact of the background modeling is to fit over a shorter time, 160 this minimizes the impact of variations of the background. For the  $^{77}$ Cu zero 161 neutron-multiplicity implant- $\beta$  activity, the background slope is on the order of 162 1.5 counts per second, while for the  $^{77}$ Cu one neutron-multiplicity implant- $\beta$ 163 activity, the background slope is on the order of 0.2 counts per second. While 164 this is small, it contributes a bias to the fit of the <sup>77</sup>Cu descendent activities. 165

The background is linearly modeled,  $C_0 + C_1 * t$ , before the implant and it is assumed that the background after the ion-implant time is linearly modeled as,  $C_0 - C_1 * t$ , with  $C_0$  and  $C_1$  calculated from the background before the implant. There is some uncertainty in this assumption and an approach is taken to minimize the impact of the background uncertainty on the estimation of the initial activity.

The ion-implants have very little background signal, due to the large unique signal of stopping a heavy ion with 100 - 200 MeV/u energy and the isotopic

identification plus coincident timing from the BigRIPS detectors [9], though the
 ion-implants do create background in the other detectors.

#### 176 4.2. <sup>3</sup>He Neutron Detector

The neutron-rich nuclei studied have roughly 100 - 200 MeV/u of kinetic 177 energy and their implantation creates background signals in all of the detectors, 178 including the silicon, scintillator,  $\gamma$ , and <sup>3</sup>He neutron detectors. The <sup>3</sup>He detec-179 tors see two types of background neutron counts. The first type of background 180 the <sup>3</sup>He counters see is an increase in neutron and  $\gamma$  counts associated with 181 the implanted energetic ion, referred to as the prompt flash. The second type 182 of neutron counter background is from the neutron room background in online 183 conditions, referred to as random neutron background. 184

The prompt flash neutron background associated with the stopping of energetic ions detected in the <sup>3</sup>He counters is removed by rejecting neutrons detected in the <sup>3</sup>He counters within one neutron thermalization time,  $T_{th}$ , after the implant time.

Random neutron backgrounds contribute to the implant- $\beta$  activities time 189 structure since they occur in coincidence with the  $\beta$  signal, and therefore these 190 need to be accounted for in the analysis. Random neutron background proba-191 bility coincidences that occur within one neutron thermalization time window 192 after the  $\beta$ -trigger time in the <sup>3</sup>He detectors are denoted by  $r_{0n}$  for the probabil-193 ity of zero background neutrons detected in coincidence,  $r_{1n}$  for the probability 194 of one random background neutron detected, and  $r_{2n}$  for the probability of two 195 random background neutrons detected within  $T_{th}$  of the  $\beta$ -signal time (written 196 generally as  $r_{xn}$  where x = 0, 1, 2, ...). 197

The magnitude of the background neutron coincidence probability,  $r_{xn}$ , can 198 be estimated by requiring decays that have no possible  $P_{2n}$  decay ( $Q_{\beta 2n} < 0.0$ ) 199 to have an average calculated  $P_{2n}$  consistent with zero. This requirement leads 200 to an estimation of the background neutron coincidence probabilities. Using 201 the analysis presented below, the predicted  $^{77}$ Cu  $P_{2n}$  versus the ratio of the 202 probability of detecting one neutron to detecting zero neutrons,  $r_{1n}/r_{0n}$ , with 203 an assumed small two neutron detection probability is shown in Figure 6. Be-204 cause it is energetically impossible for  $^{77}$ Cu to emit two neutrons, where the 205  $P_{2n}$  curve crosses zero gives the estimated  $r_{1n}/r_{0n}$  ratio. This technique gives 206 consistent results for  $r_{1n}/r_{0n}$  for other nuclei that have zero  $P_{2n}$  that were mea-207 sured with BRIKEN. The two neutron background coincidence rate is of order 208  $(r_{1n}/r_{0n})^2$  and therefore in general can be neglected compared to the one neu-209 tron coincidence rate, though in the equations below it is tracked for the sake 210 of completeness. 211

#### 212 4.3. Parent-Daughter $\beta$ Efficiencies

<sup>213</sup> The daughter nuclei may have a different  $\beta$ -trigger efficiency than the parent <sup>214</sup> decay. If the daughter nuclei decay has a different  $\beta$ -trigger efficiency than <sup>215</sup> the parent nuclei decay and it is not accounted for in the Bateman equation, <sup>216</sup> this will influences the fit of the parent activity. For many decays the parent and daughter nuclei have radically different  $\beta$ -decay energy windows,  $Q_{\beta}$  and they may have different low energy  $\gamma$  rays that have large conversion electron branches. Both of these factors can lead to different  $\beta$ -detector efficiencies for parent and daughter nuclei which depend strongly on the low energy threshold of the implant- $\beta$  detector. The Bateman equations need to be adapted in order to account for these effects and to minimize the influence of related uncertainties on  $P_{xn}$ .

#### 224 4.4. Neutron Multiplicity Dependent $\beta$ Efficiencies

Analogously to parent and daughter nuclei possibly having different  $\beta$ -detection 225 efficiencies, the different neutron multiplicity components of a single  $\beta$  decay can 226 have different  $\beta$  detection efficiencies. The component of the  $\beta$ -decay with no 227 neutrons emitted has in general a larger decay energy,  $Q_{\beta}$ , available for the 228  $\beta$  and  $\bar{\nu}_e$  to share, than for the one neutron component of the  $\beta$ -decay. This 229 impacts the  $\beta$ -detection efficiency of the  $\beta$  detector. Similarly, the component 230 of the  $\beta$ -decay with one neutron emitted generally has a larger decay energy, 231  $Q_{\beta n} = Q_{\beta} - S_n$ , available than two neutron component of the  $\beta$ -decay decay, 232  $Q_{\beta 2n} = Q_{\beta} - S_{2n}$ , which again can impact the  $\beta$ -detection efficiency. 233

Another effect that impacts the  $\beta$  efficiency is the final depth that the im-234 planted nuclei stops within the implant- $\beta$  detector. For nuclei stopped very near 235 the silicon surface approximately 50% of the emitted electrons leave no energy 236 deposit in the ion-implant pixel of the  $\beta$  detector. The implantation depth also 237 influences the number of detected minimally ionizing  $\beta$  particles, which to a 238 good approximation are  $\beta$  particles with energy above 1 MeV. Minimally ion-239 izing  $\beta$  particles deposit about 400 keV per mm of silicon. With a  $\beta$ -detection 240 threshold of 200 keV, it is possible for a high energy  $\beta$  to leave less than the 241 threshold energy in the implant- $\beta$  detector if it travels through less than 0.5 242 mm of silicon. To a first approximation to calculate the effect of the implanta-243 tion depth on the  $\beta$  efficiency one can assume ~ 55% of minimally ionizing  $\beta$ s 244 are detected. The number of minimally ionizing  $\beta$  particles can be estimated 245 by assuming a Gamow-Teller  $\beta$  emission spectrum with end-point  $Q_{\beta}, Q_{\beta n}$ , or 246  $Q_{\beta 2n}$ , as appropriate. Simulations and further discussion of this effect can be 247 found in [8]. 248

In this paper the  $\beta$  efficiency for  $\beta$  decays that emit no neutrons ( $P_{0n}$  decays) 249 is written as  $\varepsilon_{\beta}$ , while the  $\beta$  efficiencies for  $\beta$  decays that emit one ( $P_{1n}$  decays) 250 or two neutrons ( $P_{2n}$  decays) are given by  $\varepsilon_{\beta 1}$  and  $\varepsilon_{\beta 2}$ , respectively. For <sup>77</sup>Cu 251  $(Q_{\beta n} = 5.61 \text{ MeV and } Q_{\beta} = 10.17 \text{ MeV } [13])$ , an implant- $\beta$  detector threshold 252 of 200 keV and assuming a Gamow-Teller  $\beta$  distribution leads to a ~ 1% relative 253 difference in the number of  $\beta$ s detected. And, still assuming a Gamow-Teller  $\beta$ 254 distribution, up to a  $\sim 10\%$  relative difference in the number of high energy  $\beta$ 255 particles detected if the ion-implant position in the silicon detector is taken into 256 257 account. To account for possible additional effects, a 15% uncertainty in the ratio of the one neutron emission  $\beta$  efficiency to the zero neutron  $\beta$  efficiency is 258 assumed for <sup>77</sup>Cu to be  $\varepsilon_{\beta 1}/\varepsilon_{\beta} = 1.00(15)$ . 259

#### <sup>260</sup> 4.5. Energy Dependence of Neutron Efficiency

As emphasized in [4], the overall neutron efficiency depends on the energy of the emitted neutron. The energy of neutrons emitted in  $P_{(x+1)n}$  events in general will have lower energy compared with  $P_{xn}$  events, though how much lower is challenging to estimate. By using  $Q_{\beta}$  and the neutron separation energy,  $S_n$ , values, estimates of the absolute upper emitted neutron energies can be made.

#### <sup>267</sup> 5. Extracting Activities with the Bateman Equation

#### <sup>268</sup> 5.1. Impact on Bateman Equations

The impact of differing parent-daughter  $\beta$  efficiencies is not included in the 269 original Bateman equation solution [16]. In order to properly fit the full Bate-270 man equation, the  $P_{xn}$  need to be known, and for unmeasured  $\beta$ -delayed neutron 271 emitting nuclei this is not the case. In addition, the parent and daughter  $\beta$  ef-272 ficiencies need to be known. The modification to the Bateman equation for 273 differing parent-daughter  $\beta$  efficiencies is similar to the correction due to the 274  $P_{xn}$  daughter-neutron daughter factor, and disentangling these two values is 275 not well defined from the fit of the adapted Bateman equation to the data. 276

The Bateman equation solutions for zero, one, and two neutron ion-implant 277  $\beta$  activities depend on the  $P_{xn}$  values, the parent and daughter  $\beta$  efficiencies, 278 and on the neutron efficiency in a more intricate way than the full ion-implant 279  $\beta$ -decay time activity does. Effectively, these parameters are not uniquely iden-280 tifiable from the fit. Fortunately, precise knowledge of these parameters is not 281 required to estimate the  $P_{xn}$ . Even with ambiguity in the parameter values, the 282 estimated initial activities from the neutron-multiplicity ion-implant- $\beta$  activities 283 can be used to calculate the  $P_{xn}$ . 284

In order to minimize the influence of the relative daughter  $\beta$  efficiencies and 285 the unknown  $P_{xn}$  values on the Bateman fits, the estimated initial activity of the 286 zero, one, and two coincident neutron implant- $\beta$  activity curves  $(A_{0n}, A_{1n}, A_{2n})$ 287 can be extracted instead of the full number of counts obtained from a original 288 Bateman equation fit. The initial activity precision is affected by the statistics, 289 but is mainly influenced by the parent half-life uncertainty. It is worth noting 290 that the full statistics are used to estimate the initial activity. The influence 291 of unknown daughter  $\beta$  efficiencies and of the initially unknown  $P_{xn}$  dominate 292 the errors. The impact of these uncertainties are minimized by looking at the 293 estimated initial activity, see figures 3, 4, 5. Finally, it is worth noting that the 294 initial activity at the implant time can be read directly from the decay curve in 295 order to make online estimates of the  $P_{xn}$ . 296

#### <sup>297</sup> 5.2. Bateman Fitting Ranges

The time range used for fitting the adapted Bateman equations is an important factor. For the BRIKEN implant- $\beta$  detectors there was electronic noise in AIDA for the first 30 ms immediately after the ion-implant time, so this early time data is not included in the fit. This noise has been corrected after the first experimental runs and the initial cutoff time has been reduced to around 10 ms. This electronic noise is much longer than, and therefore dominates, the ion-implant exclusion time,  $T_{th}$ , mentioned previously. In the <sup>77</sup>Cu data we do not use the first 40 ms of data, which does not impact the calculations due to the much longer <sup>77</sup>Cu half life of 468(2) ms [13]. For much shorter half lives this becomes a limiting factor.

Choosing the higher time cutoff depends on several factors. First is the 308 limitation of the background being modeled as linear, as discussed previously. 309 The second limitation is the accuracy of the modified Bateman equation and 310 what is actually being fit as the maximum time is increased. There is effectively 311 no more direct information about the parent decay after six parent half lives, so 312 fitting beyond that only gains information on the daughter and grand daughter 313 decays. But the daughter decays are not the primary information we are after, 314 we are after the parent decay information. For all of the adapted Bateman 315 equation fits, the endpoint of each fit is varied from 6 to 10 times the parent 316 half life. 317

#### 318 5.3. Initial Activity Contamination by Daughter Activities

The early ion-implant- $\beta$  activities for the  $A_{xn}(t)$  have small quantifiable contributions from the daughter decays. By looking at early times, times much smaller than the daughter half life just after the ion-implant time, the amount of daughter activity at time t is given approximately by

$$A_D(t) \sim (\lambda_D t) A_{P0},\tag{3}$$

where  $A_D(t)$  is the daughter activity at time t,  $\lambda_D$  is the daughter decay rate, and  $A_{P0}$  is the initial activity of the parent. This approximation is valid as long as  $\lambda_D t \ll 1$  and that there are enough  $A_{P0}$  counts at early times. In the <sup>77</sup>Cu example, the number of daughter decays at time t = 10 ms amounts to  $\sim 0.2\%$ of the initial activity of <sup>77</sup>Cu.

#### 328 5.4. Influence of Daughter Parameters on Initial Activities

All of the parameters related to the daughter decays,  $P_{xn}$  values, daughter 329  $\beta$  efficiencies, and daughter half lives, minimally influence the initial activity 330 deduced from the fit. This is because all of the parameters in the modified 331 Bateman equation at early times are proportional to terms shown in equation 332 3. And therefore as time goes to zero, the direct influence of the parameter 333 uncertainties on the initial activity fit also goes to zero. The daughter parame-334 ters still influence the estimation of the parent half life, but as we demonstrate 335 below this error has reduced influence on the  $P_{xn}$ . 336

This line of argument is only true for experiments with no directly implanted daughter nuclei in the same pixel within the analysis time window. For experiments with a nonzero initial daughter activity equation 3 does not apply and hence the propagation of errors in the daughter nuclei parameters do not necessarily reduce to zero as in equation 3.

#### <sup>342</sup> 5.5. Influence of Half Life on the Initial Activities

The parent half life uncertainty influences the  $P_{xn}$  uncertainty, but the impact on the calculated  $P_{xn}$  is mitigated by the linear nature of the solution of equations 1 and 2. Since the parent half life is the same for all three decay components, the impact on the  $P_{xn}$  errors of the half life uncertainty is minimized.

In figure 7, the assumed <sup>77</sup>Cu half life is varied by  $\pm 50\%$  and the impact on the calculated <sup>77</sup>Cu  $P_{1n}$  is (+2, -16)%. If the <sup>77</sup>Cu half life is assumed unknown by  $\pm 10\%$ , the impact on the calculated <sup>77</sup>Cu  $P_{1n}$  is  $\pm 2\%$ . In the case of the literature value of <sup>77</sup>Cu, 468(2) ms [13, 14, 15], the resulting uncertainty of  $P_{1n}$ is  $\pm 0.2\%$ . This is a negligible number when compared with the other sources of uncertainty.

One way to evaluate the half life error is to use the one neutron implant- $\beta$ 354 activity to estimate the half life, because the uncertainty in the zero neutron 355 implant- $\beta$  activity is usually larger. The one neutron implant- $\beta$  activity half 356 life is then used in the zero neutron implant- $\beta$  activity to calculate the  $P_{xn}$ . 357 We demonstrate this for the <sup>77</sup>Cu below. For more neutron rich nuclei, the 358 challenge of extracting a half life due to daughter contamination will be present 359 in the one and even the two neutron implant- $\beta$  activities and therefore it may 360 be more challenging to obtain a precise half life. But due to the linear nature 361 of the ORNL BRIKEN analysis technique, the impact of the half-life error on 362 the  $P_{xn}$  is reduced. 363

#### <sup>364</sup> 6. Statistical and Systematic Uncertainties Summary

Knowledge of the parent half life has an impact on the estimated errors of  $P_{xn}$ . In many cases, knowledge of the half life is available from previous experiments, but for many of the exotic neutron-rich nuclei measured with BRIKEN, the half lives are currently unknown or have extremely large uncertainties.

In  $\beta$ -neutron decays, up until recently it has been possible to use the one 369 neutron decay activity to get a good half-life measurement, because it is a clean 370 spectrum with little to no contamination from the daughter decays. For exotic 371 neutron-rich nuclei this may no longer be the case because the daughter nuclei 372 decays may also have a significant  $\beta$ -delayed neutron decay channel, and ex-373 tracting the half-life from one, and even two, neutron implant- $\beta$  activity curves 374 may not be a precise measure of the  $\beta$ -decay half life. Another effective way 375 to measure a more precise half life is to measure an associated  $\gamma$  ray and its 376 half life gating on the  $\gamma$  energy in the HPGe detectors. But this is not always 377 possible, such as in cases where there are no detected  $\gamma$  rays associated with the 378 particular decay, whether from low statistics or from no  $\gamma$  rays being emitted. 379 In each case the single best possible estimate of the half life should be used to 380 fit all of the x-neutron activity decay curves, though what is considered best 381 will depend on the specifics of each nuclei and its daughters. 382

#### <sup>383</sup> 7. Example - <sup>77</sup>Cu

The decay of <sup>77</sup>Cu is presented to demonstrate the analysis procedure de-384 scribed in this manuscript. For  $^{77}$ Cu the half life is well known, 468(2) ms 385 [13, 14, 15], but as an exercise, the evaluation is also presented as if the half life 386 is unknown and the half lives for the zero, one and two neutron decay activities 387 are treated as independent. This means the half lives are (slightly) different 388 for each x (x = 0, 1, 2) neutron implant- $\beta$  activity, which in turn leads to large 389 uncertainties in the calculated  $P_{xn}$  values. In the analysis of nuclei measured 390 with BRIKEN, the same half life is used for zero, one, and two neutron decay 391 activity curves. 392

<sup>393</sup> By varying the initial activities,  $A_{xn}$ , with the uncertainties from the adapted <sup>394</sup> Bateman equation fit and propagating the results through equation 1 the statis-<sup>395</sup> tical errors in the  $P_{xn}$  can be calculated. To calculate the systematic errors, one <sup>396</sup> can vary the parameters ( $\epsilon_{11n}$ ,  $a_x$ ,  $r_{xn}/r_{0n}$ , etc..) in equation 1 and equation 2 <sup>397</sup> by their respective uncertainties independently or correlated, as is appropriate, <sup>398</sup> while evaluating the  $P_{xn}$  repeatedly.

The decay of <sup>77</sup>Cu is well characterized,  $[Q_{\beta} = 10.17(15) \text{ MeV}, Q_{\beta n} =$ 399 5.61(15) MeV,  $Q_{\beta 2n} = -2.21(15)$  MeV] [13]. The negative  $Q_{\beta 2n}$  for <sup>77</sup>Cu means 400 that two neutron decay is not possible. In figures 3, 4, and 5 the implant- $\beta$ 401 activities with zero, one, and two neutron multiplicity as a function of time, 402  $A_{xn}(t)$ , for <sup>77</sup>Cu are shown. Approximate initial activities,  $A_{xn}$ , can be read off 403 the histograms, though associating a precise uncertainty for the read off initial 404 activity poses challenges. The initial activities and uncertainties from the fits 405 with the adapted Bateman equation without using information on the <sup>77</sup>Cu half 406 life and not requiring the zero, one, and two neutron implant-decay curve half 407 lives to be the same are  $A_{0n} = 914(106)$ ,  $A_{1n} = 209(15)$ , and  $A_{2n} = 2.5(7)$ . 408

The initial activities and uncertainties from the fits with the adapted Bateman equation assuming the known half life,  $T_{1/2} = 468$  ms, are  $A_{0n} = 908(11)$ ,  $A_{1n} = 212(3)$ , and  $A_{2n} = 2.6(4)$ . Notice the uncertainties are much smaller than in the unknown and independently varied half-life case. The resulting <sup>77</sup>Cu half life from the one neutron decay activity fit is  $T_{1/2} = 471(25)$  ms and if half life is used in the analysis of all three decay activity curves it gives identical results as using the known half life of 468(2) ms.

Since there are two neutron counts with a decay detected, one might naively 416 think there is possibly a small two neutron decay branch. But if one compares 417 the initial two neutron activity to the initial one neutron activity, the ratio is a 418 little over 0.01, which is just the relative probability to detect a single random 419 background neutron in the <sup>3</sup>He detectors in our thermalization time window, 420  $r_{1n}/r_{0n} = 0.012$ . Using the same argument, about 10 of the one neutron activity 421 counts,  $A_{1n} = 212(3)$ , are actually zero neutron events in coincidence with a 422 background neutron. In this case it is a small correction,  $\sim 5\%$  relative error, 423 but in other cases with different relative  $P_{xn}$  values this can be a much larger 424 correction. For example, a large  $P_{0n}$  and a small  $P_{1n}$ , on the order of a percent 425 or two, will have a large component of random coincidences in the one neutron 426 decay curve. This observation holds similarly for a large  $P_{1n}$  and a small  $P_{2n}$ . 427

Using these initial activities and assuming a single neutron efficiency of 62%428 [7], a relative daughter  $\beta$  efficiency,  $a_1 = 1.0$ , and estimating the noise by 429 requiring the  $P_{2n}$  is zero which gives  $r_{1n}/r_{0n} = 0.012$ , as shown in figure 6. For 430 the case where the <sup>77</sup>Cu half life is fixed to the known value and varying the  $A_{xn}$ 431 by their uncertainties 100,000 times while inputing these values into equation 1, 432 a fit of the resulting distribution is shown in figure 8 with a Gaussian function 433 and reporting the  $\overline{P}$  and  $\sigma_P$ , one obtains  $P_{0n} = 71.2(5)\%$ ,  $P_{1n} = 28.8(5)\%$ , and 434  $P_{2n} = 0.000(1)\%$ . For the case with an unconstrained <sup>77</sup>Cu half life and the 435 same neutron efficiency one obtains  $P_{0n} = 71.1(33)\%$ ,  $P_{1n} = 28.9(33)\%$ , and 436  $P_{2n} = 0.000(2)\%$ , the results are shown in figure 10. 437

If in addition to the statistical uncertainties, the single neutron efficiency is varied as 62(2)% [7], and the relative neutron-multiplicity as  $\beta$  efficiency as  $a_1 = 1.00(15)$  (motivated previously), the calculated  $P_{xn}$  distributions are shown in figures 9 and 11. Fitting each distribution with a Gaussian function, one obtains  $P_{0n} = 70.8(30)\%$ ,  $P_{1n} = 29.2(30)\%$ , and  $P_{2n} = 0.000(1)\%$  using the known half life and leaving the half life unconstrained one obtains  $P_{0n} =$ 70.7(44)%,  $P_{1n} = 29.3(44)\%$ , and  $P_{2n} = 0.000(2)\%$ .

Since the <sup>77</sup>Cu half life is well known, our reported one neutron branching fraction,  $P_{1n} = 29.2(30)\%$ , is in 1  $\sigma$  agreement with the literature values of  $P_{1n} = 31.0(38)\%$  [14] and  $P_{1n} = 30.3(22)\%$  [15]. The two literature values were obtained using two different techniques, providing confidence in the value.

#### 449 8. Summary

We have presented the fundamentals of the BRIKEN analysis and shown 450 two evaluations of  $^{77}$ Cu  $\beta$ -neutron precursor decay properties and the associated 451 statistical and systematic uncertainties as examples. We present a general result 452 that simplifies calculation and propagation of uncertainties. We also present 453 a discussion of extracting zero, one, and two neutron activities appropriate 454 for the BRIKEN setup. This discussion is applicable to other experiments if 455 daughter implants are spatially and temporally distinguishable from the nuclei 456 of interest implants. If this is not an appropriate description of a particular 457 other experiment, the conversion of activities to  $P_{xn}$  in equations 1 and 2 is 458 still valid. For <sup>77</sup>Cu the BRIKEN result for the one neutron branching fraction, 459  $P_{1n} = 29.2(30)\%$  agrees with previous measurements of  $P_{1n}$  in the literature. 460 This agreement increases our confidence in the evaluation procedure presented 461 in this paper. 462

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#### <sup>561</sup> Appendix A. Derivation of Equations 1 and 2

In this appendix we describe the derivation of equation 1 and 2. For the 562 derivation we only consider up to a two neutron emitting nucleus. The extension 563 of the analysis to three and four neutron decays is straight forward. The basis 564 of the derivation is to consider all of the possible ways to detect y neutrons 565  $(0 \le y \le x)$  given that x neutrons  $(0 \le x \le 2)$  are emitted. For clarity, in the 566 567 first part of the derivation we ignore the dependence of the relative  $\beta$  efficiency on the number of neutrons emitted, that modification is shown following the 568 basic derivation. 569

The possible ways to detect no neutrons for various decay events are listed 570 here. There are only three possible ways. The first possibility is a decay with 571 zero neutrons emitted and no background neutrons detected. The second possi-572 bility is a decay with one neutron emitted but that neutron is not detected and 573 no background neutrons are detected. The third possibility is a decay with two 574 neutrons emitted but neither neutron is detected and no background neutrons 575 are detected. Using the notation used in equations 1 and 2, the ways to detect 576 zero neutrons can be written as 577

$$A_{0n}(t) = A(t)\epsilon_I \epsilon_\beta r_{0n} \left( P_{0n} + \epsilon_{10n} P_{1n} + \epsilon_{20n} P_{2n} \right).$$
(A.1)

Next is the list of possible ways to detect one neutron from various decay 578 events. There are five possible ways. The first possibility is a decay with zero 579 neutrons emitted and one background neutron detected. The second possibil-580 ity is a decay with one neutron emitted and that neutron is detected and no 581 background neutrons are detected. The third possibility is a decay with one 582 neutron emitted but that neutron is not detected and one background neutron 583 is detected. The fourth possibility is a decay with two neutrons emitted and 584 only one of those neutrons are detected and no background neutrons are de-585 586 tected. The fifth possibility is a decay with two neutrons emitted and neither of those neutrons are detected but one background neutron is detected. Using 587 the notation used in equations 1 and 2, the ways to detect one neutron can be 588

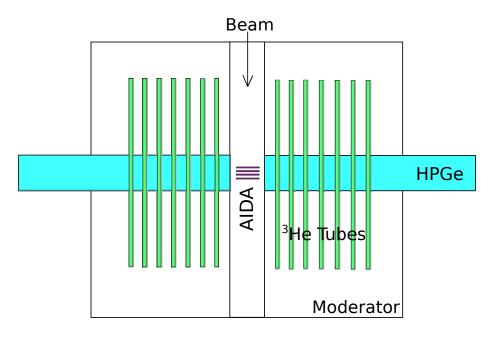


Figure 1: (Color online) Schematic top view of the BRIKEN detector. The AIDA silicon detectors (purple) are referred to as implant- $\beta$  detectors, because the nuclei of interest are first implanted into these detectors and then the  $\beta$  particles emitted in subsequent  $\beta$  decays are also observed in the same detectors. For the analysis described in the text, only coincident information from the <sup>3</sup>He tubes and one of the implant- $\beta$  detectors is required.

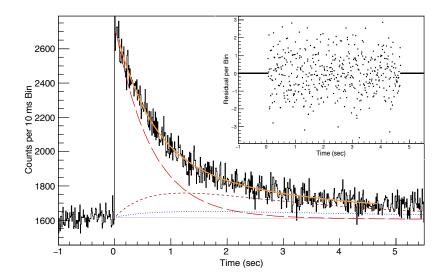


Figure 2: (Color online) Fit of adapted Bateman equation to <sup>77</sup>Cu data with an implant- $\beta$  trigger correlation and no information on the number of neutrons from the <sup>3</sup>He tubes. The residual of the  $i^{th}$  bin is defined as  $R_i = (data_i - fit) / \sqrt{n_i}$ , where  $n_i$  is the number of counts in the  $i^{th}$  bin. Shown in the plot are the total fit (orange - solid), <sup>77</sup>Cu (red - long dashed), <sup>77</sup>Zn (dark red - short dashed), <sup>76</sup>Zn (blue - dotted), background (light gray - solid), and the data (black - solid). All decay curves are offset by the background. The granddaughter decays are not shown to preserve clarity.

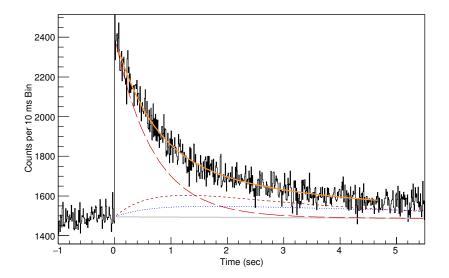


Figure 3: (Color online) Fit of adapted Bateman equation to  $^{77}\mathrm{Cu}$  data with an implant- $\beta$  trigger correlation and zero neutrons detected in the <sup>3</sup>He tubes. Colors and comments are as in figure 2.

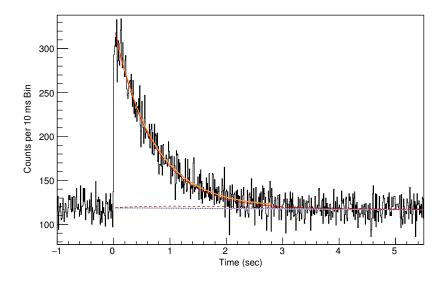


Figure 4: (Color online) Fit of the adapted Bateman equation to  $^{77}$ Cu data with an implant- $\beta$  trigger correlation and one neutron detected in the <sup>3</sup>He tubes. Colors and comments are as in figure 2, though the total and the  $^{77}$ Cu decay are almost indistinguishable.

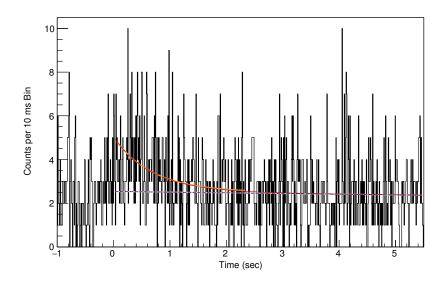


Figure 5: (Color online) Fit of the adapted Bateman equation to  $^{77}$ Cu data with an implant- $\beta$  trigger correlation and two neutrons detected in the <sup>3</sup>He tubes. Colors and comments are as in figure 2, though the total and the  $^{77}$ Cu decay are almost indistinguishable.

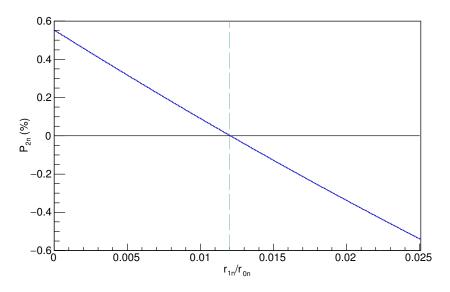


Figure 6: The variation of the calculated  $^{77}$ Cu  $P_{2n}$  with statistical uncertainties versus the ratio of one neutron background coincidence probability to zero neutron background coincidence probability. The vertical dashed line at 0.012 is the zero crossing point.

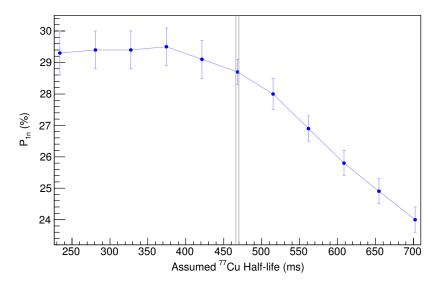


Figure 7: The variation of the calculated  $P_{1n}$  versus input <sup>77</sup>Cu half life. This demonstrates the technique's level of stability to uncertainties in the half life. The experimental <sup>77</sup>Cu half life is bounded by the two gray lines [13]. The solid blue line is drawn to guide the eye.

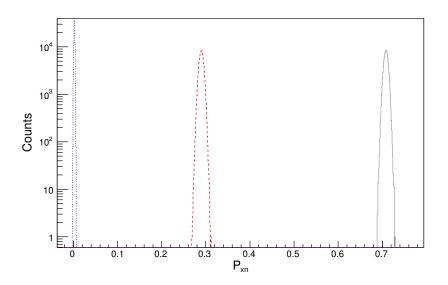


Figure 8: (Color online) Statistical variation of <sup>77</sup>Cu initial activities and the impact on the  $P_{xn}$  assuming the known <sup>77</sup>Cu half life,  $T_{1/2} = 468(2)$ ms.  $P_{0n}$  is shown as a solid gray line,  $P_{1n}$  is shown as a dashed red line, and  $P_{2n}$  is shown as a dotted blue line.

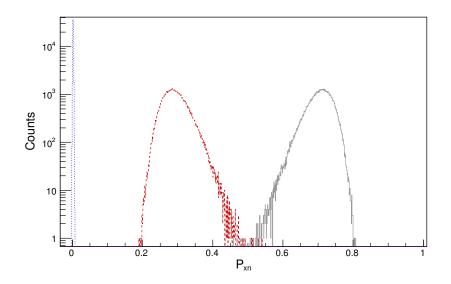


Figure 9: (Color online) Statistical and systematic errors after variation of <sup>77</sup>Cu initial activities and the other parameters described in the text and their impact on the  $P_{xn}$  assuming the known <sup>77</sup>Cu half life,  $T_{1/2} = 468(2)$ ms. Colors and line styles are as in Figure 8.

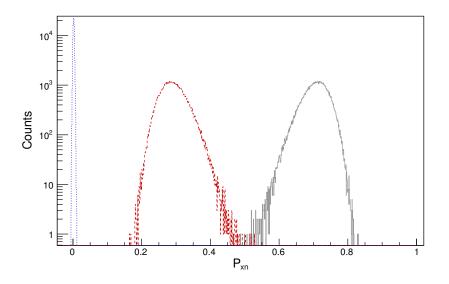


Figure 10: (Color online) Statistical variation of  $^{77}$ Cu initial activities and the impact on the  $P_{xn}$  with non-fixed  $^{77}$ Cu half life. Colors and line styles are as in Figure 8.

589 written as

$$A_{1n}(t) = A(t)\epsilon_I \varepsilon_\beta \left( P_{0n}r_{1n} + \epsilon_{11n}r_{0n}P_{1n} + \epsilon_{10n}r_{1n}P_{1n} + \epsilon_{21n}r_{0n}P_{2n} + \epsilon_{20n}r_{1n}P_{2n} \right),$$
(A.2)

The last enumeration of possibilities considered is the list of possible ways to 590 detect two neutrons from various decay events. There are six possible ways. The 591 first possibility is a decay with zero neutrons emitted and two background neu-592 tron detected. The second possibility is a decay with one neutron emitted and 593 that neutron is detected in coincidence with one background neutron detected. 594 The third possibility is a decay with one neutron emitted but that neutron is not 595 detected but two background neutrons are detected. The fourth possibility is a 596 decay with two neutrons emitted and both emitted neutrons are detected along 597 with no background neutrons detected. The fifth possibility is a decay with two 598 neutrons emitted and only one of the emitted neutrons is detected along with 599 one background neutron detected. Lastly, the sixth possibility is a decay with 600 two neutrons emitted and neither of the emitted neutrons is detected but two 601 background neutrons are detected. Using the notation for equations 1 and 2, 602 the ways to detect two neutrons can be written as 603

$$A_{2n}(t) = A(t)\epsilon_I \epsilon_\beta \left( P_{0n}r_{2n} + \epsilon_{11n}r_{1n}P_{1n} + \epsilon_{10n}r_{2n}P_{1n} + \epsilon_{22n}r_{0n}P_{2n} + \epsilon_{21n}r_{1n}P_{2n} + \epsilon_{20n}r_{2n}P_{2n} \right).$$
(A.3)

<sup>604</sup> Equations A.1, A.2, and A.3 are not quite equations 1 and 2, one additional set <sup>605</sup> of parameters remains to be inserted.

<sup>606</sup> Due to the possible large difference between  $Q_{\beta}$ ,  $Q_{\beta n}$ , and  $Q_{\beta 2n}$  (decay <sup>607</sup> energy for zero, one, and two neutron decays) the associated  $\beta$  efficiencies ( $\varepsilon_{\beta}$ , <sup>608</sup>  $\varepsilon_{\beta 1}, \varepsilon_{\beta 2}$ ) may not be the same. Adding these parameters to the equations, the <sup>609</sup> zero neutron equation becomes

$$A_{0n}(t) = A(t)\epsilon_I r_{0n} \left(\varepsilon_\beta P_{0n} + \varepsilon_{\beta 1}\epsilon_{10n} P_{1n} + \varepsilon_{\beta 2}\epsilon_{20n} P_{2n}\right), \qquad (A.4)$$

<sup>610</sup> with similar changes to the one and two neutron equations.

<sup>611</sup> After factoring out  $\varepsilon_{\beta}$ ,  $r_{0n}$ , and group the  $A_{xn}(t)$  and the  $P_{xn}$  into vectors, <sup>612</sup> the remaining components are the matrix **E**, we arrive at the equations 1 and <sup>613</sup> 2, the basis of the ORNL BRIKEN analysis technique.

<sup>614</sup> The extension of this analysis to three and larger neutron emission is straight <sup>615</sup> forward, with the additional modification that the random probability of three <sup>616</sup> and four background neutrons should be included and that the  $\beta$  efficiencies <sup>617</sup> and neutron efficiencies for three and four neutron decays should be included.

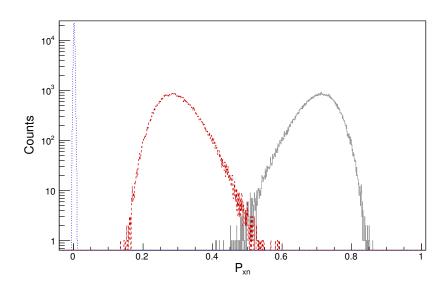


Figure 11: (Color online) Systematic and statistical variation of <sup>77</sup>Cu initial activities and the other parameters described in the text and their impact on the  $P_{xn}$  with a non-fixed <sup>77</sup>Cu half life. Colors and line styles are as in Figure 8.