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Irradiated Radium-Beryllium Neutron Source Buildup

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ABSTRACT

Large increases in neutron source strength as a result of irradiation by neutrons were first reported in 1958. Later experimental work irradiating sources in the Materials Test Reactor indicated that resonance absorption effects could be safely ignored. Consideration of the differences in neutron energy spectrum between the MTR and usual power reactors suggested that resonant absorptions could be significant in power reactors. Calculations over a range of neutron spectra show the source build-up factor to be several times as large for spectra typical of power reactors as that for purely thermal reactors.

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I. INTRODUCTION

The possibility of orders of magnitude increases in neutron source strength for irradiated radium-beryllium sources were first reported by Pearlstein in 1958(1). Considering only thermal flux, he showed that transmutation of radium 226 by successive neutron captures and beta decay could lead to increases in source strength relative to the initial strength of close to 100.

Based on the work of Dudziak and Freeman in 1960(2), it appeared that any resonance effects could be safely ignored. An extensive experimental program irradiating sources to buildup factors as high as 17 was performed in the Materials Test Reactor. Calculations were performed using subcadmium fluxes measured by cobalt activation, and neglecting the shielding effect of the stainless steel encapsulation. It was recognized that this procedure introduced errors which tended to cancel one another. Comparison of calculated to measured results revealed no systematic deviations which could be attributed to this procedure, as shown in Slide 1. The maximum deviation was 30%. An uncertainty of this order can easily be accomodated in the design of shipping casks or in designing the optimum method for increasing a source strength by irradiation.

Although not stated in their article, the Dudziak and Freeman samples were irradiated in position L47 of the MTR[3]. This position has a cobalt cadmium ratio between eight and nine, the hardest neutron spectrum available for irradiation of samples in the MTR[4]. This cobalt cadmium ratio corresponds to a uranium 235 cadmium ratio of about 65, a much more thermal spectrum than encountered normally in power reactors. U-235 cadmium ratios for power reactors are often in the range of 10 to 5.

Consideration of the differences in neutron spectra between power reactors and the MTR plus the resonance cross-section data now available for radium 226 and actinium 227 suggests that buildup factors for radium-beryllium sources irradiated in power reactors could be substantially larger than previsouly indicated. Analyses of radium-beryllium neutron source buildup factors as a function of the spectrum hardness were therefore undertaken taking into account known resonance cross-sections.

II. NUCLEAR EVENTS

A Ra:Be source produces neutrons by the reaction $Be^9(\alpha,n)C^{12}$ where the alpha particles are emitted by Ra^{226} and its daughter products in an unirradiated source. Under neutron irradiation, the transmutation of Ra^{226} produces alpha emitters with higher specific activities than Ra^{226} itself. The isotopic coupling which describes the behavior of this source under irradiation can be represented as shown in Slide 2. The short lived isotopes with the exception of Ac-228 are not shown explicitly but are represented only by the total number of alpha particles involved in the transitions. Analytically assuming that all half lives not shown as well as that of Ac-228 are zero leads to no significant error.

III. CALCULATIONAL METHODS

The differential equations governing the number densities, N, of nuclei involved in chains coupled by radioactive decay and/or neutron absorption are of the form

$$\frac{d\,Ni}{dt} = \sum_{j \neq i} Nj\, Cj_i - \Lambda i\, Ni \qquad \text{(Slide 3)}$$

where C_{ji} is the coupling from nucleus j to nucleus i by neutron absorption or radioactive decay and Λ_i is the removal of nucleus i by the same processes. The form of C_{ji} is either λ_j, or $\phi_t \left(\sigma_{tj} + \frac{\phi_e \sigma_{ej}}{\phi_t}\right)$ where ϕ_t is the thermal flux, σ_{tj} is the thermal cross section at the effective temperature of the thermal flux, and $\phi_e \sigma_{ej}$ is the epithermal reaction rate of nucleus j integrated over all energies above some thermal cut off energy. Λ_i has the form

$$\left[\lambda_i + \phi_t \left(\sigma_{ti} + \frac{\phi_e \sigma_{ei}}{\phi_t}\right)\right]$$

where λ_i is the decay constant for nucleus i. These equations have been programmed at KAPL in FORTRAN IV for isotopic coupling of the order required here.

Solution of the above equations yields number densities for the various isotopes. The number densities can be converted to alpha yields per second by weighting with the alpha yields and appropriate isotopic decay constants. However, the neutron yield per alpha decay is strongly energy dependent. Runnals and Boucher[5] have fitted an empirical expression shown in Slide 4,

$$n_{max} = .152\ E^{3.65} \times 10^{-6}\ \text{neutrons/alpha}$$

relating the thick target neutron yield, n_{max}, to the alpha particle energy in Mev for beryllium targets. They used this expression to calculate thick target neutron yields for the Ra and Ac chains. The results were found to agree with the best available experimental data within experimental uncertainties. Dudziak and Freeman[2] extended the neutron yield calculation to the Th chain. The results summarized for the three chains are shown in Slide 4. The population weighting factor in the last column

$$W = \frac{1620\ \text{yr}}{t\ 1/2} \times \frac{Y}{485}$$

is required to normalize the final neutron source yield of an isotope to the fresh unirradiated radium-beryllium source strength. In this expression $t_{1/2}$ is the dominant half live and Y is the neutron yield per 10^6 decays including the alpha decays of short lived isotopes in equilibrium with the dominant half life. The contribution of a dominant controlling population, N, to the neutron source buildup factor is the ratio of the weighting factor product with N to the initial population of Ra-226.

$$\frac{N \times W}{N_o \ (Ra\text{-}226)}$$

The total buildup factor is obtained by summing the individual contributions.

It will be noted in this table that Ac-227 begins both the Ac chain and the Th chain. The Ac chain represents the sequence of events if there were no neutron capture in Ac-227. The chain labeled Th is the sequence of events with neutron capture. Development of the Th chain is clearly preferable for enhancement of the buildup factor.

Neutron absorption rates are needed to complete the analysis of the nuclear chains. The nuclear constants used are given in Slide 5. It will be noted that no resonance data are available for the isotopes involved other than Ra-226, Ac-227 and Pb-210. Since freshly prepared radium sources are assumed in our calculations, the Pb-210 data is ignored. Absorption rates are dependent upon the product $\sigma\phi$. If the .0253ev cross section, σ_o , is given for a $1/v$ absorber, the value at any temp T is given by

$$\sigma_T = \sigma_o \sqrt{\frac{T_o}{T}} \; ,$$

where temperatures are on the absolute scale and T_o is the temperature equivalent to .0253ev. Reactor thermal neutron flux distributions can be approximated by a hardened Maxwellian distribution. The average value of a $1/v$ cross section in such a distribution is the product of $\sqrt{\pi/2}$ and the value of the cross section at the effective temperature for the distribution. However the thermal cross sections given in Slide 5 were mostly measured in pile neutrons[7, 8, 9] at temperatures near room temperature. Thus, these cross sections are not point energy values, but are values for thermal neutron distributions. That is, the distribution factors (approximately $\sqrt{\pi/2}$) are included in the reported "thermal" cross section.

Since it is only the product $\sigma\phi$, and not its factors, which determine reaction rates, the hardened Maxwellian flux can be reduced to an effective 68F neutron flux,

$$\phi_{\text{eff}}^{68F} = \sqrt{\frac{T_o}{T_h}}\ \phi$$

where $T_o = 68+459$ on the Rankin absolute temperature scale, and T_h is the absolute value of the effective temperature for the harden Maxwellian flux, for use with the thermal cross sections in Slide 5.

In addition to the thermal cross sections, some of the isotopes of Slide 5 have important resonance cross sections. The most important cross sections in our analysis are for Ra-226 and Ac-227. Ra-226 has a single low energy resonance at .537ev. At this energy both the high energy Maxwellian tail of the thermal flux and the low energy end of the slowing down neutron flux contribute significantly to the resonance captures in Ra-226. The energy dependent flux over this resonance region was represented by the form

$$\phi(E) = \phi^{(3)} N \frac{E}{(k\ T_h)^2} e^{-\frac{E}{k\ T_h}} + \frac{b}{E} \qquad \text{Slide 6}$$

where $\phi^{(3)}$ is the thermal flux integral from 0 to .625ev, E is neutron energy, k is the Boltzmann constant, T_h is the effective temperature of the Maxwellian distribution and b is a constant for the slowing down flux. In the thermal range b was tapered to zero as the Maxwellian distribution became dominant for low energy. N is a normalization factor defined by the requirement

$$\phi^{(3)} = \int_0^{.625} \phi\ (E)\ dE. \qquad \text{Slide 6}$$

The Ac-227 resonances are at high energies unaffected by thermal neutrons. Furthermore, the low population of Ac-227 allows the direct use of the infinitely dilute resonance integral, I, to obtain the resonance cross section.

IV. CALCULATIONAL RESULTS

Since a thermal flux of 6×10^{13} neutrons per second per centimeter squared is readily obtainable for source buildup applications, we have illustrated the effect of neutron spectrum with this thermal flux, assumed to correspond to 68F. A convenient calculational tool for characterizing the reactor neutron energy spectrum is the Wigner-Wilkins parameter $\Delta^{(6)} \approx 4 \frac{\Sigma_a (k\ T)}{\xi\ \Sigma_s}$.

We have chosen to consider the span for Δ of 0 to 1 which encompasses uranium 235 cadmium ratios down to between 3 and 4. Slide 7 illustrates the radium-beryllium source buildup factor as a function of the integrated thermal neutron flux and of the spectrum parameter Δ. The curve for $\Delta = 0$, corresponds to assuming no resonance flux contribution to the buildup factor. It is readily apparent that buildup factors are much larger with hard thermal spectra compared to the buildup factors when resonance effects are neglected. Slide 8 shows the effect of thermal flux amplitude on the source buildup factor neglecting resonance effects. It can be seen that the buildup factor is not very sensitive to flux amplitude until the integrated thermal flux is greater than 10^{21} cm^{-2}. A similar result is obtained when resonance effects are considered. Thus, costs of procuring radium-beryllium sources irradiated to a desired buildup factor can potentially be sharply reduced by selecting the exposure location with the lowest cadmium ratio.

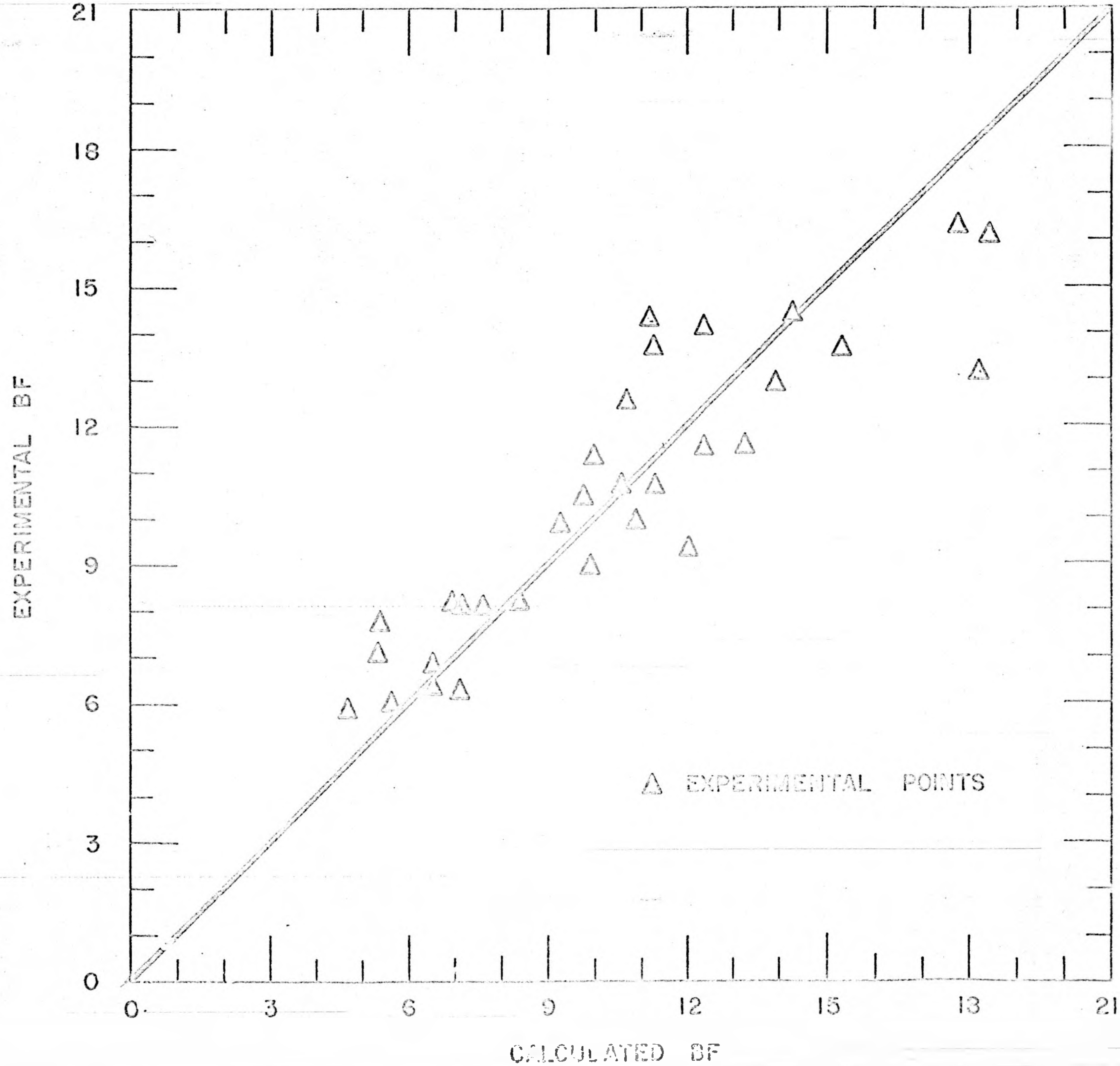
Another area in which these resonance effects should be considered is the design of transportation casks for irradiated sources. Neutron sources for reactor startup are commonly left in core to be built up in intensity by irradiation for subsequent startups. At the time of source removal, a reasonably reliable estimate of the source intensity is required as the basis for radiation protection design. If one calculates a buildup factor as low as five for a radium-beryllium source with the assumption of no resonant effects, one could easily be undercalculating by a factor of two or more. As the buildup factor and/or the spectrum parameter Δ get larger, neglect of the known resonant effects leads to underpredictions of up to factors of four. Non-conservative errors on the order of two or more are unacceptable in radiation protection design.

V. CONCLUSIONS

It must be emphasized that the results we are reporting are based upon very meager resonance cross section data for the isotopes concerned. Since the available data is for the isotopes of greatest importance, any new resonant cross section data should change our results to a much lesser extent than observed when including the available data relative to assuming no resonance effects. Until such time as experiments can be performed establishing the adequacy of our calculational approach including the significance of the unknown resonance effects in the isotopes involved in the radium-beryllium neutron source intensity buildup, calculations of the buildup factor should include not only the effects of the neutron spectrum and the available resonance cross section data but also an allowance for the potential presence of significant unknown cross sections.

CALCULATED vs. EXPERIMENTAL BUILDUP FACTOR

Slide 1



$$
\begin{tikzcd}
\text{Ra-226} \arrow[r, "1620\text{ yr.}", "(4\alpha)"'] \arrow[d, "n{,}\gamma"] & \text{Pb-210} \arrow[r, "22\text{ yr.}"] & \text{Po-210} \arrow[r, "138.4\text{ d}", "(1\alpha)"'] & {} \\
\text{Ac-227} \arrow[r, "21.2\text{ yr.}"] \arrow[d, "n{,}\gamma"] & \text{Th-227} \arrow[r, "18.17\text{ d}", "(1\alpha)"'] \arrow[d, "n{,}\text{ fission}"] & \text{Ra-223} \arrow[r, "11.4\text{ d}", "(4\alpha)"'] \arrow[d, "n{,}\gamma"] & {} \\
\text{(Ac-228} \arrow[r, "6.1\text{ hr.}"] & \text{) Th-228} \arrow[r, "1.91\text{ yr.}", "(1\gamma)"'] \arrow[d, "n{,}\gamma"] & \text{Ra-224} \arrow[r, "3.6\text{ d}", "(4\alpha)"'] \arrow[d, "n{,}\gamma"] & {} \\
 & {} & {} &
\end{tikzcd}
$$

Slide 2

$$\frac{dN_i}{dt} = \sum_{j \neq i} N_j C_{ji} - \Lambda_i N_i$$

$$C_{ji} = \lambda_j \quad \text{or} \quad \varphi_t \left(\sigma_{t_j} + \frac{\varphi_e \sigma_{ej}}{\varphi_t} \right)$$

$$\Lambda_i = \lambda_i + \varphi_t \left(\sigma_{tj} + \frac{\varphi_e \sigma_{ej}}{\varphi_t} \right)$$

Slide 3

Neutron Yields

Chain	Source or Ancestor	Dominant 1/2 Life	Alpha Part. per Decay	Neutrons per 10^6 Decays*	Population Weighting for Buildup Factor
Ra	Ra-226	1620 yr	4	485	1.
	Pb-210	22 yr	0	0	.0
	Po-210	138.4 d	1	67	591.
Ac	Ac-227	21.2 yr	0	0	.0
	Th-227	18.17 d	1	99	6647.
	Ra-223	11.4 d	4	610	65281.
Th	Ac-227	21.2 yr	0	0	.0
	Th-228	1.91 yr	1	71.6	125.
	Ra-224	3.64 d	4	692.9	232240.

* $n_{max} = .152E^{3.65} \times 10^{-6}$ neutrons/alpha

SLIDE 4

Neutron Crosssection Data

Thermal Crosssections (barns)

Ra-226	Ac-227	Th-227	Th-228	Ra-227	Ra-224
20	830	1500	123	130	12

Resonance Cross sections

Ra-226	E_0	Γ	Γ_n
	.537 ev	29 mv	.021 mv

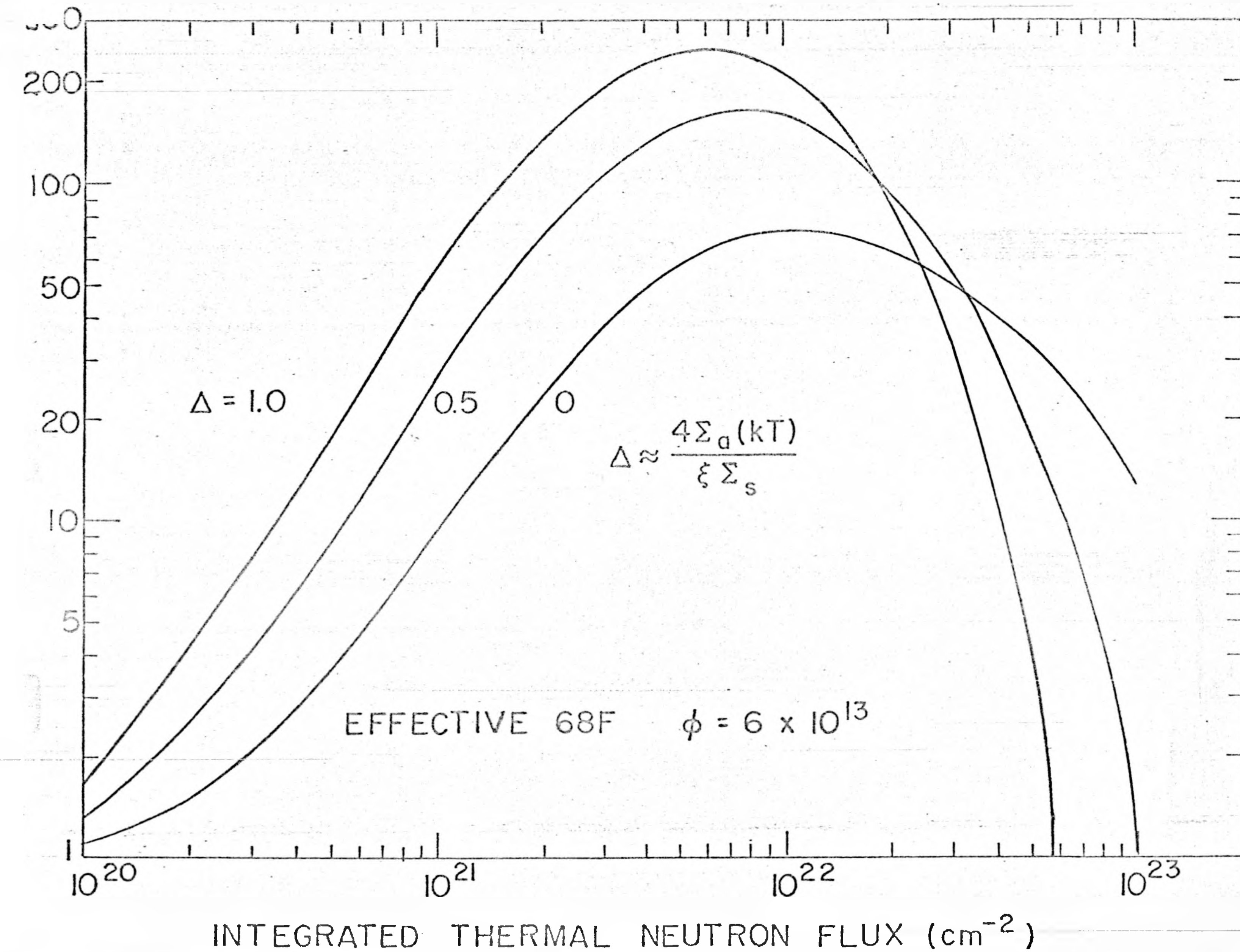
Ac-227 Resonance Integral = 1177 barns

Slide 5

$$\phi(E) = \phi^{(3)} N \left(\frac{E}{(kT_n)^2} e^{-\frac{E}{kT_n}} + \frac{b}{E} \right)$$

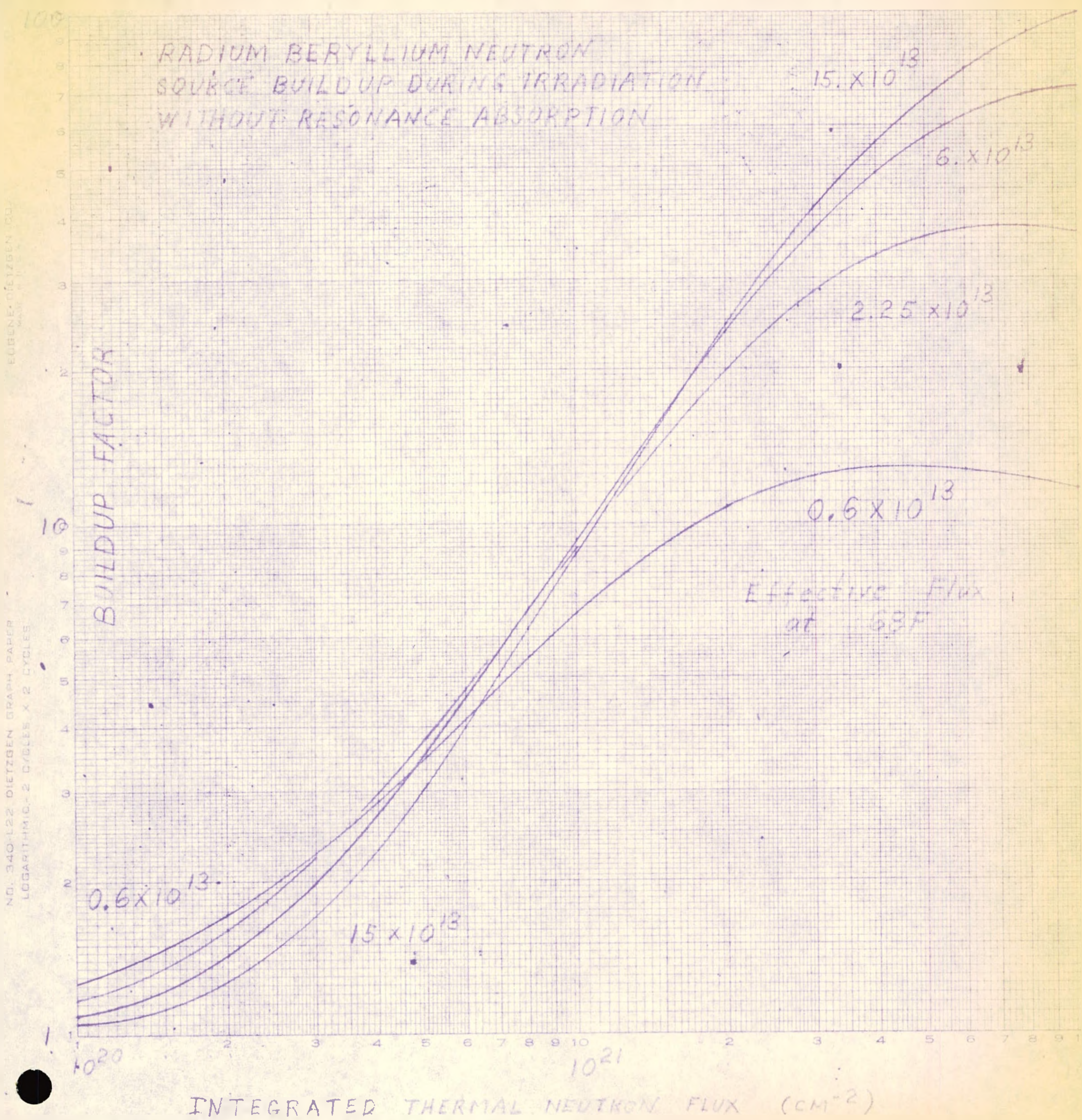
$$\phi^{(3)} = \int_0^{.625} \phi(E)\, dE$$

Slide 6



RADIUM BERYLLIUM NEUTRON SOURCE BUILDUP
DURING IRRADIATION

Slide 7



RADIUM BERYLLIUM NEUTRON SOURCE BUILDUP DURING IRRADIATION WITHOUT RESONANCE ABSORPTION
BUILDUP FACTOR
15. X 10^13
6. X 10^13
2.25 X 10^13
0.6 X 10^13
0.6 X 10^13
15 X 10^13
Effective Flux
100
10
1
10^20
10^21
INTEGRATED THERMAL NEUTRON FLUX (CM^-2)

Slide 8

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