Measurement of the $^{19}\text{F}(\text{n},2\text{n})^{18}\text{F}$ Cross Section from 18 to 27 MeV

C.L. Hartmann and P.M. DeLuca, Jr.

Department of Medical Physics

University of Wisconsin - Madison

Madison, WI 53706

Abstract

The $^{19}\text{F}(\text{n},2\text{n})^{18}\text{F}$ cross section was measured at neutron energies of 18, 21, 23, and 27 MeV. Nearly monoenergetic neutrons bombarded teflon (CF_2), Zr, and Au samples. $^{19}\text{F}(\text{n},2\text{n})^{18}\text{F}$ cross section values were determined relative to $^{nat}\text{Zr}(\text{n},\text{xn})^{89}\text{Zr}$ and $^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$ from measurements of the ^{18}F , ^{89}Zr , and ^{196}Au activities. Our results are in agreement with previous measurements below 20 MeV and extend the usefulness of this reaction to 27 MeV.

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INTRODUCTION

Investigations of the effects of fast neutrons upon matter often require accurate fluence determinations. Examples are measurements of helium production in metals from neutron bombardment and the kinetic energy transferred to charged particles, or kerma. The former is important for fusion reactor design[1], while the latter is essential for fast neutron radiation therapy of malignant tumors[2]. Recent measurements[3,4] of the kerma per unit neutron fluence, kerma factor, for several low-Z elements and 15-20 MeV neutrons revealed discrepancies with kerma factor values calculated from elemental cross section information. Extension of these kerma factor determinations to 20-30 MeV neutron energy relies upon the accurate measurement of neutron fluence.

^{18}F production by $^{19}\text{F}(\text{n},2\text{n})$ affords a convenient determination of neutron fluence. The anticipated 50-100 mb cross section and 110 min half-life match the required fluence and bombarding time for kerma measurements. Several measurements of this cross section are reported below 20 MeV[5,6,7,8,9,10,11,12], but only one at higher neutron energies[13]. Below 15 MeV and above 18 MeV, the results disagree by more than the stated uncertainties. The values obtained by Brill', *et al.* [13] for 14 to 34 MeV follow an energy dependence similar to other work, but are about 1.5 times greater in magnitude. Hence, there is uncertainty regarding the cross section above 20 MeV.

To improve the accuracy of this reaction cross section for determining absolute neutron fluence at higher energies, we measured the $^{19}\text{F}(\text{n},2\text{n})^{18}\text{F}$ cross section at 18, 21, 23, 25,

and 27 MeV. Monoenergetic neutrons bombarded Zr and Au metal samples as well as "teflon¹" samples. ^{18}F , ^{89}Zr , and ^{196}Au activities were corrected for variations in neutron flux, attenuation of neutrons during irradiation, self-absorption of sample gamma rays, and activity due to contaminating lower energy neutrons. $^{19}\text{F}(n,2n)^{18}\text{F}$ cross section values were then deduced by comparison with the $^{nat}\text{Zr}(n,xn)^{89}\text{Zr}$ and $^{197}\text{Au}(n,2n)^{196}\text{Au}$ activation cross sections.

EXPERIMENTAL PROCEDURE

Approximately monoenergetic neutrons from the $^3\text{H}(d,n)^4\text{He}$ reaction irradiated $(\text{C}_2\text{F}_4)_n$, Zr, and Au samples. Neutrons were produced by bombarding a tritium gas target with deuterons from the University of Wisconsin Tandem Accelerator. A 50.8 mm long and 6.4 mm inner diameter stainless steel cylinder contained the gas at about 0.6 atm pressure. After traversing a 2.5 μm molybdenum entrance foil and the target gas, the 0.8 μA deuteron beam stopped in a 0.25 mm thick gold disk. Deuterons lost a maximum of 0.2 MeV in the tritium gas.

During activation, samples were positioned along the deuteron beam axis, 150 mm from the midpoint of the tritium target. Activation samples were 25.4 mm-diameter cylindrical disks. The $(\text{C}_2\text{F}_4)_n$ and Zr samples were 6.35 mm thick and had masses of about 7 g and 20 g, respectively. The 2.5 g Au samples were 0.254 mm thick.

Samples were irradiated for different lengths of time. To avoid saturation of the ^{18}F

¹polytetrafluoromethane, $(\text{C}_2\text{F}_4)_n$

activity, $(C_2F_4)_n$ bombardment was limited to 2 h. At 18, 21, and 25 MeV, Au and Zr samples were irradiated for at least 8 h during which time additional Zr and $(C_2F_4)_n$ samples were included for 2 h. At 23 and 27 MeV, Zr and $(C_2F_4)_n$ samples were exposed for 2 h.

Recording the number of deuterons striking the target and the response of a NE-102 plastic scintillator monitored the relative neutron fluence. A high NE-102 bias avoided measurement of lower energy neutrons produced by $^2H(d,n)$ interactions with deuterium contaminating the tritium gas as well as breakup neutrons from deuteron interactions with the target components and gas. Recording of the deuteron beam current permitted correction for neutron flux variations.

After irradiation, a 42 mm-diameter cylindrical Ge(Li) spectrometer detected decay quanta from each of the samples. ^{196}Au activity was assessed from the number of full energy events associated with the 333 and 356 keV photons. Observation of events from the 909 keV decay photon served to measure ^{89}Zr activity. $^{18}F \beta^+$ activity was determined from a measurement of the annihilation quanta.

A set of standard photon sources, traceable to the National Institutes of Standards and Technology, served to calibrate the energy-dependent photopeak efficiency of the Ge(Li) detector. The stated uncertainties of the ^{137}Cs , ^{60}Co , ^{57}Co , ^{22}Na , source activities were 3.7%, 1.9%, 4.4%, and 3.7%, respectively. Normalization of a ^{152}Eu source's 1112 keV gamma-ray emission to the efficiency measured with the Cs, Co, and Na sources allowed

several additional ^{152}Eu emissions to complete the energy dependent photo-peak efficiency calibration. Branching ratios and half-lives for these sources were taken from Browne, *et al.* [14]

RESULTS

The $^{19}\text{F}(\text{n},2\text{n})^{18}\text{F}$ cross section was determined relative to that of the $^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$ and $^{\text{nat}}\text{Zr}(\text{n},\text{xn})^{89}\text{Zr}$ cross sections by comparing activities induced in F, Au, and Zr. To determine the $^{19}\text{F}(\text{n},2\text{n})^{18}\text{F}$ cross section, the $^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$ or $^{\text{nat}}\text{Zr}(\text{n},\text{xn})^{89}\text{Zr}$ cross section was scaled by the ratio of effective neutron flux during irradiation, $\bar{\phi}_{\text{Zr}}/\bar{\phi}_{\text{F}}$ or $\bar{\phi}_{\text{Au}}/\bar{\phi}_{\text{F}}$, and the ratio of activities at the end of bombardment, $A_{\text{o,F}}/A_{\text{o,Au}}$ or $A_{\text{o,F}}/A_{\text{o,Zr}}$. The effective neutron flux ratio accounts for flux variation and radioactive decay during irradiation. Additional corrections were made for neutron attenuation, varying sample position, and attenuation of activation gamma radiation.

Table 1 summarizes the $^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$ and $^{\text{nat}}\text{Zr}(\text{n},\text{xn})^{89}\text{Zr}$ cross section values used. These values were extracted from polynomial fits to measurements of Bayhurst, *et al.*[15], who compared activation in Au and $^{\text{nat}}\text{Zr}$ with ^{24}Na activity from $^{27}\text{Al}(\text{n},\alpha)$ as well as with the response of a proton recoil telescope. Bayhurst also measured the $^{90}\text{Zr}(\text{n},2\text{n})^{89}\text{Zr}$ cross section and used it as an additional monitor reaction. Their $^{90}\text{Zr}(\text{n},2\text{n})^{89}\text{Zr}$ cross section values agree with measurements by Prestwood, *et al.*[16] for neutron energies between 16 and 20 MeV. Bayhurst's $^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$ values are also in agreement with those of Veesser, *et al.*[17] and, to a lesser extent, with those of Prestwood, for neutron energies

Table 1: Assumed $^{197}\text{Au}(n,2n)^{196}\text{Au}$ and $^{nat}\text{Zr}(n,xn)^{89}\text{Zr}$ Cross Section Values[15] vs. Neutron Energy

Energy [MeV]	$\sigma^{197}\text{Au}(n,2n)$ [mb]	$\sigma^{nat}\text{Zr}(n,xn)^{89}\text{Zr}$ [mb]
18.0	1729 ± 86	581 ± 23
21.0	870 ± 45	613 ± 26
23.0	-	603 ± 26
25.0	352 ± 17	563 ± 24
27.0	-	480 ± 21

between 15 and 25 MeV.

We calculated the effects of neutron flux variation with time by dividing the irradiation period into several one hour segments and assuming that the flux was constant during each segment. The activity induced during each one hour period was corrected for radioactive decay during the segment and between the end of the segment and the end of bombardment.

An expression for the cross section in terms of measured counts and effective neutron flux is

$$\sigma = \frac{A_o}{N\bar{\phi}} = \frac{(\lambda C / \epsilon_p B) e^{\lambda \Delta t_w}}{(1 - e^{-\lambda \Delta t_c}) N \bar{\phi}}, \quad (1)$$

where A_o is the activity of the sample at the end of bombardment, $\bar{\phi}$ is the effective neutron flux, C is the net number of background corrected counts in the photopeak, Δt_w the time between the end of bombardment and start of counting, Δt_c the time of counting, ϵ_p the photopeak efficiency, and B is the decay branching ratio. Although $\bar{\phi}$ was not directly determined during the experiment, the ratios of effective flux for Au or Zr to

that for $(C_2F_4)_n$, $\bar{\phi}_{Zr}/\bar{\phi}_F$ and $\bar{\phi}_{Au}/\bar{\phi}_F$, were accurately measured and used to calculate the $^{19}F(n,2n)^{18}F$ cross section relative to Zr and Au. Half-lives and branching ratios were taken from Browne, *et al.*[14]

To obtain final $^{19}F(n,2n)^{18}F$ cross section values, we corrected for fluence variations due to differing sample distance and attenuation, self-absorption of the activation radiation, ^{11}C activation from $^{12}C(n,2n)$ in the $(C_2F_4)_n$ samples, and activation from contaminating $^2H(d,n)$ neutrons.

Distance corrections are based on the assumption that the tritium cell is a uniform line source of neutrons, and that the area of each activation sample is small compared with its distance from the neutron source. These corrections comprised 5% to 10% of final cross section values.

Neutron absorption corrections account for attenuation from intervening samples and within a sample. At 18 MeV, absorption cross sections were taken as the ENDF/B-V total cross section minus that for elastic and inelastic scattering. For energies above 20 MeV, the absorption cross section was assumed to be the same fraction of the total cross section as tabulated in ENDF/B-V at 20 MeV. Polynomial fits to the measurements of Peterson, *et al.*[18] provided total cross section values at each energy. The uncertainty in the neutron attenuation correction was assessed by assuming that absorption cross sections were uncertain by $\pm 20\%$. Absorption corrections accounted for less than 2% of final cross section values.

Gamma-ray attenuation corrections account for the attenuation of the 333, 355, 909, and 511 keV gamma rays within the Au, Zr and $(\text{C}_2\text{F}_4)_n$ samples, as well as that of the 511 keV gamma-ray attenuation in the 3 mm lucite jacket surrounding the $(\text{C}_2\text{F}_4)_n$ samples. We assumed that gamma rays started inside the activated sample and calculated gamma attenuation using interaction probabilities from the tabulations of Plechaty, *et al.*[19]. The resulting corrections were 1-6% of the final cross sections.

Above 22 MeV incident neutron energy, the $(\text{C}_2\text{F}_4)_n$ samples' β^+ activity was corrected for contributions from the $^{12}\text{C}(\text{n},2\text{n})^{11}\text{C}$ reaction. Activation of zirconium irradiated simultaneously with the $(\text{C}_2\text{F}_4)_n$ samples determined the average neutron flux and served to predict the resulting ^{11}C $(\text{C}_2\text{F}_4)_n$ activity. $^{12}\text{C}(\text{n},2\text{n})$ cross section values were deduced from a polynomial fit to the measurements of Ackerman, *et al.*[20]. To assess the accuracy of this correction, each $(\text{C}_2\text{F}_4)_n$ sample was counted immediately and 2 h after irradiation. As the ^{11}C half-life is 20.3 min compared with 109.77 min for ^{18}F [14], the ^{11}C activity correction is much smaller for the later counting than the earlier. Corrected fluorine activities from the initial and second counting agreed within $\pm 2\%$.

Because the tritium target gas contained 5% deuterium, we corrected zirconium and fluorine activities for contributions due to neutrons from the $^2\text{H}(\text{d},\text{n})$ reaction. Compared with reactions in the gas, reactions with deuterium driven into the beamstop were infrequent, since the target assembly was bombarded with a $0.8 \mu\text{A}$ beam for less than 80 h. Energies of the $^2\text{H}(\text{d},\text{n})^3\text{He}$ neutrons are below the F, Zr, and Au activation thresholds

except at 25 and 27 MeV. Neutrons from full energy $^2\text{H}(\text{d},\text{n})^3\text{He}$ reactions (not deeply inside the beamstop) had energies above the Au activation threshold but not above the F or Zr activation thresholds in the 25 MeV measurement, and were above the thresholds for all activation reactions at 27 MeV. In correcting the 27 MeV measurement for these neutrons, all $^2\text{H}(\text{d},\text{n})^3\text{He}$ reactions were assumed to occur between bombarding deuterons and deuterium contaminating the tritium target gas or not deeply inside the beamstop, and that the amount of contaminating deuterium was constant. Excess ^{196}Au activity at 25 MeV allowed an estimate of the amount of deuterium in the target. We calculated the expected Au activity at 25 MeV from the neutron fluence determined by Zr activation. The 3.5% correction to the 27 MeV measurement was added linearly to the uncertainty.

Table 2 summarizes the present measurements of the $^{19}\text{F}(\text{n},2\text{n})^{18}\text{F}$ cross section along with those from the evaluations of ENDF/B-VI[21] and Howerton[22], the latter being largely based upon the measurements by Ryves, *et al.*[12]. The last column indicates the reference material. Average measured values and those referred to Zr and Au activation are indicated. Stated uncertainties assumed uncertainties of 2-5% in A_o (this includes effects due to the efficiency calibration, positioning, carbon activation correction, and counting statistics), 1-2% in $\bar{\phi}_{(\text{Zr or Au})}/\bar{\phi}_F$, 4-5% in the Zr and Au activation cross sections, and 1-2% in the absorption correction factor (due to uncertainty in absorption cross sections).

The error in $\bar{\phi}_{(\text{Zr or Au})}/\bar{\phi}_F$ arose from uncertainty in the relative neutron flux measurement and from assuming a constant neutron flux during each time segment. The $\bar{\phi}_{\text{Zr}}/\bar{\phi}_F$

and $\bar{\phi}_{Au}/\bar{\phi}_F$ ratios calculated separately from scintillator response and deuteron charge agreed to $\pm 2\%$. In determining $\bar{\phi}_{Au}/\bar{\phi}_F$ and $\bar{\phi}_{Zr}/\bar{\phi}_F$, the neutron flux was assumed constant during each of the ≈ 1 -h segments. To test this assumption, $\bar{\phi}_{Zr}/\bar{\phi}_F$ was calculated with each segment divided into many smaller intervals based upon the variation in a chart-recorded history of deuteron current. For every energy, the resulting ratios were within $\pm 0.2\%$ of those determined for the constant segment flux assumption.

The neutron energy spread was deduced from a distance-weighted average of neutron energies in the gas cell. Slowing down of deuterons in the tritium gas as well as straggling in the Mo entrance foil and tritium gas yielded the values reported in Table 2.

DISCUSSION

By comparing the activity in $(C_2F_4)_n$ with the activity in Au and Zr following neutron irradiation, we measured the $^{19}F(n,2n)^{18}F$ cross section at 18, 21, 23, 25, and 27 MeV. Results are presented in Table 2 and compared with measurements by Menlove, *et al.* [9], Picard, *et al.* [10] and Ryves, *et al.* [12] in Figure 1.

Fluorine cross section values (not shown) determined by comparison of long and short Zr activation were in agreement, as were values deduced from Au activity based upon either the 333 or 356 keV ^{196}Au decay photons. For the 18- and 21-MeV neutron irradiations, the Zr based cross section values are somewhat less than those based upon Au activation. Average values reported in Table 2 are the weighted average of all determinations at each energy.

Table 2: $^{19}\text{F}(n,2n)^{18}\text{F}$ Cross Section Values In mb

Energy [MeV]	Present Work	ENDF/B VI [21]	LLNL [22]	Comments
16.5±0.3			69.2±2.2	
17		79.8		
17.4±0.2			76.0±2.5	
18.1±0.2	80.8±4.2			Zr activation
	85.5±4.4			Au activation
	83.0±3.0	86.2	81.5±2.5	Zr, Au average
19.0±0.2		96.8	82.8±4.0	
20.0		102.6		
21.0±0.1	82.6±4.5			Zr activation
	88.7±9.3			Au activation
	83.7±5.0			Zr, Au average
23.0±0.1	72.7±4.2			
25.0±0.1	57.2±2.9			
27.0±0.1	51.4±4.7			

As shown in Figure 1, the measured values below 21 MeV agree with the results of Ryves, Menlove, and Picard. Our 18 MeV value of 83.0 ± 3.0 mb agrees well with the ENDF/B-VI value of 86.2 mb and Howerton's value of 81.5 mb. At 21 MeV the measured value of 83.7 ± 5.0 is consistent with Howerton's value at 19 MeV, but is significantly less than the 20 MeV ENDF/B-VI evaluation. Not surprisingly the present measurements are systematically lower than Brill's [13]. Thus, we find that for neutron energies below 20 MeV our measurements are in good agreement with previous work and for energies above 20 MeV our values are substantially lower than the only previous measurement. The present work extends the usefulness of the $^{19}\text{F}(n,2n)^{18}\text{F}$ reaction to 27 MeV.

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Table and Figure Captions

Table I Assumed $^{197}\text{Au}(n,2n)^{196}\text{Au}$ and $^{nat}\text{Zr}(n,xn)^{89}\text{Zr}$ Cross Section Values[15] *vs.* Neutron Energy

Table II $^{19}\text{F}(n,2n)^{18}\text{F}$ Cross Section Values In mb (10^{-27} cm^2)

Figure 1 $^{19}\text{F}(n,2n)^{18}\text{F}$ cross section values plotted *vs* neutron energy. Besides the present work, $\bullet$, values reported by Menlove[9], $\circ$, Picard[10], $\triangle$, and Ryves[12], $\square$ are indicated.

Energy [MeV]	$\sigma_{^{197}\text{Au}(n,2n)}$ [mb]	$\sigma_{\text{nat Zr}(n,xn)^{89}\text{Zr}}$ [mb]
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