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DETERMINATION OF γ-RAYS RELATIVE INTENSITIES FROM THE ^{35}Cl$(n,\gamma)^{36}$Cl REACTION ON FILTERED THERMAL NEUTRON BEAM

TRAN TUAN ANH AND PHAM NGOC SON
Nuclear Research Institute, Dalat
VUONG HUU TAN
Vietnam Agency for Radiation and Nuclear Safety, Hanoi, Vietnam

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E-mail: ttanhfr@yahoo.com

Abstract. *The relative intensities of prompt γ-rays from the ^{35}Cl(n, γ)^{36}Cl reation with thermal neutron have been used as secondary γ-ray intensity standards for the prompt gamma neutron activation analysis (PGNAA) and for nuclear data measurements due to a high capture cross section. The filter neutron technique was applied for producing a thermal neutron beam at the neutron channel N_o. 4 of the Dalat nuclear research reactor. The neutron flux and Cd-ratio are 8.72×10^6 n.cm^{-2}.s^{-1} and 134, respectively, determined by the gold foil activation method. A new PGNAA system with a HPGe detector of 58% relative efficiency and a digital spectrometer was used to detect prompt gamma rays from the ^{35}Cl(n, γ)^{36}Cl reaction. In this work, relative intensities of 23 prompt γ-rays have been determined on the filtered thermal neutron beam. The present results are in good agreement with literature values and data from previous measurements.*

Keywords: relative intensity, filtered thermal neutron beam, Dalat research reactor.

I. INTRODUCTION

The energies and emmission probabilities (intensities) of prompt γ-rays in the high energy region are very important for spectrometer calibration and for nuclear data measurements due to the experiments are only carried out by (n, γ) reaction. The ^{14}N(n, γ)^{15}N reaction is usually a primary γ-ray source for high energy and efficiency calibrations of detectors in PGAA [1–4]. However, this reaction has a small cross section (75 mbarns), hence it is difficult to detect prompt γ-rays of ^{15}N in a low thermal neutron flux $< 10^7$ n.cm^{-2}.s^{-1}. In this case, the ^{35}Cl(n, γ)^{36}Cl reaction is chosen as a secondary standard because of a large cross section (43.6 barns) and emitting γ-rays up to 9 MeV. Furthermore, the prompt γ-ray peak at 1951.1 keV of ^{36}Cl can be used as an internal mono-standard Cl in the k0-PGAA method [5–8]. Therefore, it is necessary to accurately determine the emission probabilities of prompt γ-rays from ^{35}Cl(n, γ)^{36}Cl reaction.

The neutron filter technique has been applied for producing quasi-monoenergetic neutrons of 55 keV and 144 keV as well as thermal neutrons at the horizontal channel No. 4 at the Dalat Research Reactor (DRR). The beams were used for nuclear data measurements, PGNAA and other applications [9]. In the present work, the relative intensities of prompt γ-rays in range of 0.5 – 8 MeV from the ^{35}Cl(n, γ)^{36}Cl reaction have been determined on the filtered thermal neutron beam by using a high quality gamma spectrometer and a recently upgrade PGAA facility at the DRR.

II. EXPERIMENTAL PROCEDURE

This experiment was carried out on the filtered thermal neutron beam with 98cm Si + 01cm Ti + 35g/cm^2 S at the horizontal channel N$_{0.}$ 4 of the DRR. The thermal neutron flux at the irradiation position and the Cd(Au) ratio were 8.72×10^6 n.cm^{-2}.s^{-1} and 134, respectively.

A ^{35}Cl target prepared from pure compound of NH$_4$Cl was wrapped in fluorinated ethylenepropylen resin (FEP) tape of 25 μm thickness. A blank sample was also prepared from the FEP for measurement of the background spectrum. The sample was placed at an angle of 45° with respect to the beam direction, into the sample box of polytetrafluoroethylene (PTFE) with sample-detector distance of 31 cm. The PGNAA system consists of a 58% coaxial horizontal HPGe detector with an energy resolution of 2.1 keV at 1333 keV and a digital spectrometer to measure prompt γ-rays from ^{35}Cl(n, γ)^{36}Cl reaction. The sample was simuirradiated/measured for sufficiently long time (48 hrs) to achieve good counting statistics. The gamma spectrum was observed by using GammaVision 3.2 software. The experimental arrangement and the gamma spectrum of ^{36}Cl were shown in Fig. 1 and Fig. 2, respectively.

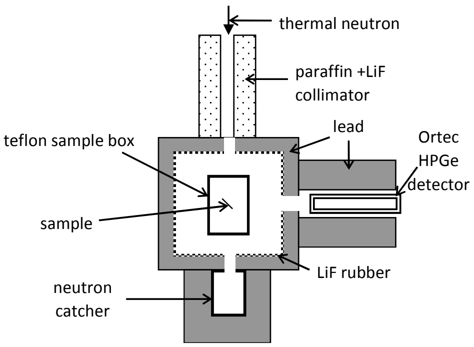

Fig. 1. Experimental arrangement in PGNAA facility

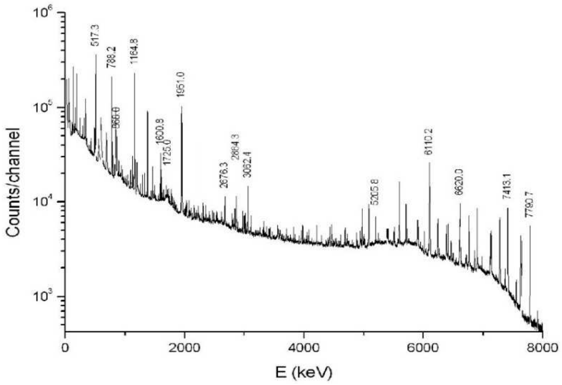

Fig. 2. Prompt γ-ray spectrum of ^{36}Cl

III. DATA ANALYSIS

The absolute efficiency curve of a HPGe detector can be determined by measuring a ^{152}Eu standard radioactive source for the energy range below 1408.0 keV. The relative efficiencies measured from prompt γ-rays of ^{58}Ni(n, γ)^{59}Ni reaction were then normalized to absolute efficiencies in order to extend the curve up to 9 MeV [10]. The response function of the HPGe detector was also simulated by using the MCNP5 code to calculate absolute efficiencies in the energy range of 50 keV – 10 MeV with uncertainties less than to 1% [11]. The relative standard deviations

Table 1. Comparison of experimental and MCNP efficiencies

Nuclide	**E** (keV)	**I$_\gamma$** (%)	ε_{MCNP}	$\Delta\varepsilon_{MCNP}$ (%)	$\varepsilon_{Exp.}$	$\Delta\varepsilon_{Exp}$ (%)	$\frac{\varepsilon_{MCNP}}{\varepsilon_{Exp.}}$
^{152}Eu	121.8	28.41	2.10E-03	0.14	1.91E-03	3.1	1.10
	244.7	7.55	1.72E-03	0.30	1.69E-03	3.2	1.02
	344.3	26.58	1.40E-03	0.18	1.50E-03	3.1	0.93
	411.1	2.237	1.24E-03	0.72	1.37E-03	4.2	0.91
	444.0	3.125	1.18E-03	0.61	1.31E-03	3.5	0.90
	778.9	12.96	8.06E-04	0.35	8.55E-04	3.1	0.94
	867.4	4.241	7.51E-04	0.61	7.81E-04	3.5	0.96
	964.1	14.62	7.00E-04	0.35	7.16E-04	3.2	0.98
	1085.9	10.13	6.47E-04	0.44	6.53E-04	3.2	0.99
	1089.7	1.731	6.45E-04	0.96	6.51E-04	7.4	0.99
	1112.1	13.4	6.36E-04	0.39	6.42E-04	3.1	0.99
	1213.0	1.415	6.01E-04	1.10	6.04E-04	4.3	0.99
	1299.1	1.632	5.74E-04	1.20	5.78E-04	4.8	0.99
	1408.0	20.85	5.43E-04	0.33	5.53E-04	3.1	0.98
						Aver.	**0.98**
						Std. dev. (%)	**5.1**
^{58}Ni(n,γ)^{59}Ni	878.0	5.29	7.45E-04	0.19	7.22E-04	1.5	1.03
	1188.8	1.253	6.09E-04	0.43	6.19E-04	2.3	0.98
	1301.4	1.17	5.73E-04	0.45	5.98E-04	6.1	0.96
	1949.9	1.067	4.32E-04	0.55	4.45E-04	2.7	0.97
	5312.7	1.2	1.82E-04	0.77	1.94E-04	4.7	0.93
	6105.2	1.58	1.57E-04	0.73	1.68E-04	4.2	0.94
	6583.8	1.86	1.45E-04	0.72	1.41E-04	4.2	1.03
	8120.6	2.98	1.14E-04	0.60	1.12E-04	3.6	1.02
	8533.5	16.2	1.08E-04	0.28	1.03E-04	2.1	1.05
	8998.4	33.4	1.02E-04	0.21	9.85E-05	2.1	1.03
						Aver.	**0.99**
						Std. dev. (%)	**4.3**

(%RSD) of calculated and experimental efficiency ratios were found 5.1% for ^{152}Eu and 4.3% for ^{59}Ni, respectively (see Table 1).

The full energy peak efficiency curve of HPGe detector in the energy range of 50 keV – 10 MeV was shown in Fig. 3.

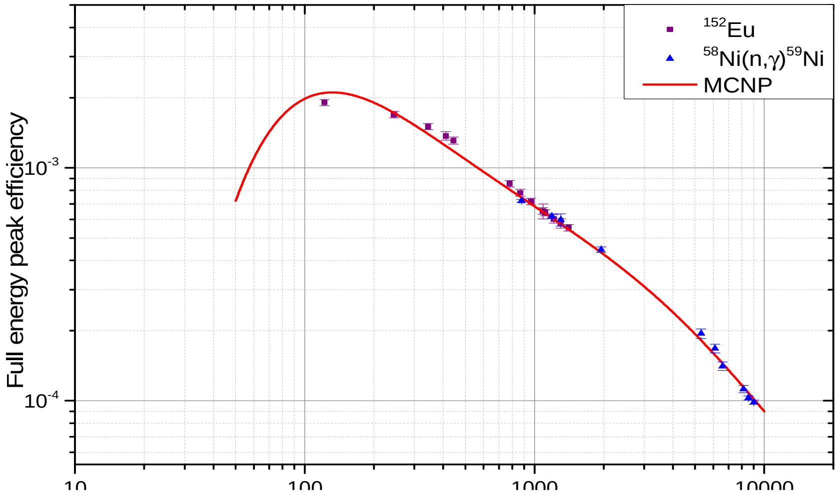

Fig. 3. Comparison of the measured and calculated full energy peak efficiencies

The peak areas of prompt γ-rays from ^{35}Cl(n, γ)^{36}Cl reaction were analyzed by the FitzPeaks Gamma Analysis Software [12]. In this work, overlapping peaks of 786.3 + 788.4 keV and 1951.1 + 1959.3 keV were deconvoluted and then resulted the net area for each peak.

The relative intensities normalized by the 1951.1 keV γ-ray can be written as:

$$I_k = \frac{\dfrac{\left(\frac{S_{\gamma,k}}{t_c} - \frac{S_{\gamma,b,k}}{t_{c,b}}\right)}{\varepsilon_{\gamma,k}}}{\dfrac{\left(\frac{S_{\gamma,s}}{t_c} - \frac{S_{\gamma,b,s}}{t_{c,b}}\right)}{\varepsilon_{\gamma,s}}} \cdot 100\%$$

where $S_{\gamma,k}, S_{\gamma,s}, S_{\gamma,b,k}, S_{\gamma,b,s}$ are net areas of gamma peak k^{th} and normalized peak and backgrounds, respectively. $t_c, t_{c,b}$ are counting times of sample and background. $\varepsilon_{\gamma,k}$, $\varepsilon_{\gamma,s}$ are efficiency of peak k^{th} and normalized peak. In this experiment, the background is very small and can be neglected.

The relative intensity uncertainties follow the propagation of error law:

$$\Delta I_k = I_k \sqrt{\left(\frac{\partial I_k}{\partial S_{\gamma,k}}\right)^2 \Delta S_{\gamma,k}^2 + \left(\frac{\partial I_s}{\partial S_{\gamma,s}}\right)^2 \Delta S_{\gamma,s}^2 + \left(\frac{\partial I_s}{\partial \varepsilon_{\gamma,s}}\right)^2 \Delta\varepsilon_{\gamma,s}^2 + \left(\frac{\partial I_s}{\partial \varepsilon_{\gamma,s}}\right)^2 \Delta\varepsilon_{\gamma,s}^2}$$

where $\Delta S_{\gamma,k}, \Delta S_{\gamma,s}$, $\Delta\varepsilon_{\gamma,k}$, $\Delta\varepsilon_{\gamma,s}$ are net area and efficiency uncertainties of gamma peak k^{th} and normalized peak, respectively.

IV. RESULTS AND DISCUSSION

The relative intensities of 23 prompt γ-rays normalized by the 1951.1 keV γ-ray from ^{35}Cl(n, γ)^{36}Cl reaction have been determined on the filtered thermal neutron beam at the Dalat research reactor. The present results were compared with literature values from the ENSDF library [10] and data from previous measurements [13–15] as shown in Table 2.

Table 2. Relative intensities of prompt γ-rays from ^{35}Cl(n, γ)^{36}Cl reaction normalized by 1951.1 keV

E (keV)	**Coceva** [13] $I_k(\pm\%)$	**Raman** [15] $I_k(\pm\%)$	**Molnár** [14] $I_k(\pm\%)$	**ENSDF** [10] $I_k(\pm\%)$	**Present(a)** $I_k(\pm\%)$	**Present(b)** $I_k(\pm\%)$
517.1	125.32(5.8)	117.82(2.1)	119.83(0.7)	119.71(0.7)	118.95(4.29)	120.97(0.7)
786.3	54.25(3.3)	51.49(2.9)	54.03(0.9)	53.83(0.9)	56.32(4.37)	56.14(1)
788.4	84.17(2.2)	81.19(2.4)	85.63(0.9)	84.9(1.2)	85.86(6.33)	85.57(4.7)
1131.2	9.86(2.9)	-	9.9(0.5)	9.9(0.5)	9.78(4.65)	9.65(1.9)
1164.9	140.28(2.6)	134.65(1.8)	140.86(0.4)	140.5(0.7)	152.24(6.29)	150.33(4.6)
1601.1	17.97(2.6)	19.01(1.6)	19.13(0.6)	19.06(0.9)	19.04(4.56)	18.89(1.7)
1951.1	**100(0.3)**	**100(0.2)**	**100(0.6)**	**100(0.6)**	**100(6.12)**	**100(4.4)**
1959.3	64.78(2.2)	64.36(2.3)	64.76(0.7)	64.84(0.7)	64.24(4.63)	64.25(1.9)
2676.3	8.11(2.5)	-	8.42(0.7)	8.37(1.2)	7.99(7.8)	8.14(6.6)
2863.8	29.76(1.9)	27.92(1.6)	28.74(0.6)	29.09(2.6)	28.48(4.78)	29.16(2.2)
2975.3	5.39(2.4)	-	5.95(1.2)	5.78(3.1)	5.13(6.42)	5.26(4.8)
3061.8	18.16(1.9)	17.08(1.8)	17.81(0.6)	17.79(0.9)	16.62(4.53)	17.08(1.6)
4440.4	5.4(2.2)	-	5.95(1)	5.7(4.4)	5.02(6.35)	5.24(4.7)
4979.7	18.65(2.7)	-	19.47(0.8)	19.29(1.2)	17.62(4.88)	18.39(2.4)
5517.2	8.71(2.5)	-	8.84(0.8)	8.79(0.9)	8.37(7.2)	8.7(5.8)
5715.2	27.39(2.8)	26.39(1.9)	28.74(0.9)	27.88(3.1)	26.17(4.75)	27.13(2.1)
5902.7	5.69(2.8)	-	5.87(1.2)	5.84(1.2)	5.43(4.78)	5.61(2.2)
6110.8	106.14(3.2)	102.97(1.9)	104.22(0.9)	103.66(0.9)	102.83(4.31)	105.97(0.8)
6619.6	40.38(2.1)	37.57(2)	39.98(0.9)	39.71(1.3)	38.97(4.61)	39.76(1.8)
6627.8	24.19(2.4)	21.98(2)	23.17(1.1)	23.2(2.1)	24.03(4.61)	24.51(1.8)
6977.8	11.81(2.8)	11.09(2.3)	11.71(1.4)	11.5(1.4)	10.97(5.56)	11.09(3.6)
7413.9	54.25(2.3)	49.5(2.5)	52(1.4)	51.8(1.5)	56.8(4.5)	56.7(1.5)
7790.3	42.86(2.3)	40.84(2.7)	42.01(1.2)	41.83(1.2)	39.9(4.6)	39.41(1.8)
8578.5	14.13(2.1)	13.51(2.6)	13.95(1.5)	13.89(1.5)	-	-
$\sum I_k \vert \bar{\sigma}_{I_k}$	**1017.7(2.5)**	**917.4(2.0)**	**1010.9(0.9)**	**1006.9(1.5)**	**1000.7(5.2)**	**1008.0(2.7)**

$\sum I_k$ is sum of relative intensities and $\bar{\sigma}_{I_k}$ is average of relative intensity uncertainties;
Present(a) - using experimental efficiencies; Present(b) - using calculated efficiencies.

In Table 2, 24 strong gamma lines from 0.5 – 8.5 MeV were chosen to determine relative intensities. The authors Coceva [13], Raman [15] and Molnar [14] have measured prompt gamma rays of ^{36}Cl on the high thermal and cold neutron beams by using a Compton-suppressed spectrometer. The detector efficiencies in a high energy region were determined by measuring ^{14}N(n, γ)^{15}N reaction while the present experiment only used a HPGe spectrometer in a single mode. In our work, the sum of intensities is in good agreement with the literature value. However, the average of relative intensity uncertainties in this case is quite high (5.2%) compared with the others. Main errors due to peak area uncertainties (1–5%) and detector efficiency uncertainties (2–7%). Increasing counting time to reduce statistical errors of counts is not recommended because the background is also increased while measuring the gamma spectrum and could not solve in a single mode system using a HPGe detector. Hence, the precise calculation of detector efficiencies with about 1% or better accuracy by MCNP code is significant figures important in the reduction of experimental uncertainties. The results in the last column of Table 2 showed that, relative intensities calculated with the MCNP efficiency function are in very good agreement with ENSDF data. The average of relative intensity uncertainties reduced to 2.7%. The differences between experimental and evaluated data were shown in Fig. 4.

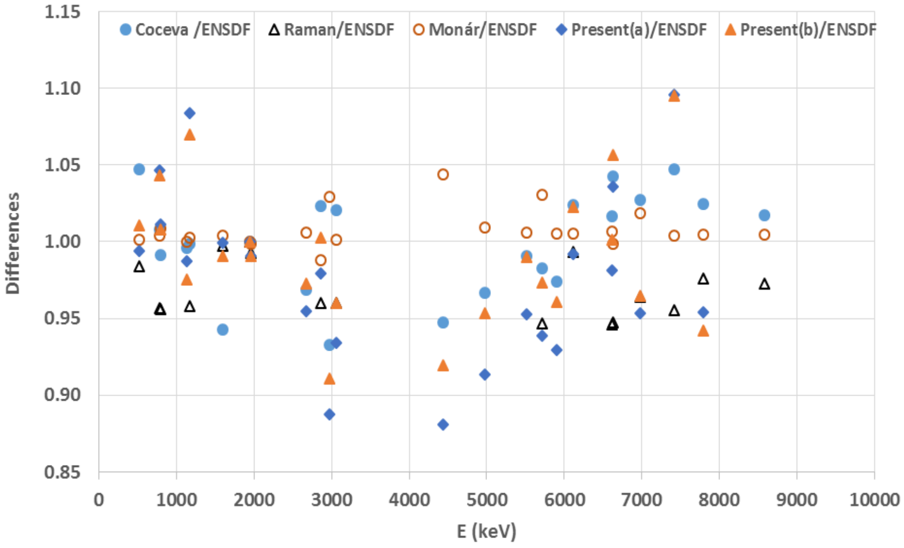

Fig. 4. Differences between experimental and evaluated data

V. CONCLUSION

Experimental relative intensities of prompt γ-rays from ^{35}Cl(n, γ)^{36}Cl reaction have been determined on the filtered thermal neutron beam at the Dalat research reactor. The full energy peak efficiencies were measured by a ^{152}Eu gamma source and prompt gamma rays from ^{58}Ni(n, γ)^{59}Ni reaction to obtain an efficiency curve from 0.1 – 9 MeV with 3.6% average relative uncertainty. The HPGe detector efficiencies in the energy range of 50 keV – 10 MeV were also calculated by a Monte Carlo MCNP code with 0.5% or better accuracy. Both efficiency functions have been used to determine relative intensities of ^{36}Cl prompt γ-rays normalized by 1951.1 keV. The obtained results are in good agreement with literature values and with data from previous measurements.

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