Re-estimation of absolute gamma ray intensities of ^{56}Mn using k_0-standardization

M. AHMAD, W. AHMAD, M. U. RAJPUT, A. QAYYUM

(Physics Research Division, Pakistan Institute of Nuclear Science &Technology, P.O. Nilore, Islamabad, Pakistan)

Abstract The thermal neutron capture gamma ray facility at Pakistan Research Reactor (PARR-1) is being used for the re-estimation of various properties like capture cross-sections, resonance integral, absolute gamma intensities, etc. of different isotopes. The data for gamma ray transitions from the capture of thermal neutrons by ^{55}Mn are not in good agreement specifically below 2 MeV. So there is a need to re-estimate its intensities with better accuracy. Analytical grade $MnCl_2$ powder and high purity Mn metal pieces were used in this study. Standard ^{152}Eu and ^{60}Co radioactive sources as well as thermal neutron capture γ-rays in chlorine were chosen for efficiency calibration. The k_0 standardization technique was applied for these measurements to eliminate systematic errors in efficiencies. Chlorine also acted as a comparator in k_0- factor calculations. The results have been tabulated for the main gamma rays from ^{56}Mn in the low as well as in the medium energy regions. The absolute intensities are in good agreement with most of the reported values.

Key words Absolute intensities, Prompt gamma rays, k_0-standardization, Manganese-56

CLC numbers O571.42+1, O657.4

1 Introduction

The nuclear properties like capture cross section, resonance integral, emission probabilities, etc. of various isotopes are of prime importance for their applications in specific areas of reactor physics. These fundamental properties have been determined since the beginning of nuclear physics but still there are large variations in the data. Reactor neutron activation analysis (NAA) is an important tool for the determination of material composition.[1,2] Similarly Prompt Gamma Neutron Activation Analysis (PGNAA) has proven its potential particularly for the elemental analyses of those elements which cannot be determined by conventional neutron activation techniques.[3,4] Although the resulting gamma ray spectra of the irradiated materials are usually very complex, the use of high-resolution germanium detectors (HPGe) has made the job easy for the spectroscopic purposes.[5] However, quantitative multi-elemental analysis is sometimes not worthwhile with NAA and PGNAA in their classical forms using relative methods because standards are required for each and every element to be determined.[6] This is not only time consuming but impossible in certain cases when unexpected elements are present. Non-availability of proper standards has always been a practical problem in these methods.

Besides elemental analyses, the properties like absolute emission probabilities can also be determined by a relative method. In this method, the sample and the standard are irradiated separately. A standard matching the sample composition, homogeneity and geometry for every element present in the material under investigation is selected and experiments are performed under identical beam conditions.[7] One of the problems is that the analytical results are seriously affected by the geometrical and matrix differences among the samples. So, to make this technique widely applicable, Molnar et al[8] proposed k_0 standardization for PGNAA. This method relies on the prompt k_0-factors for efficiency normalized analytical sensitivity ratios of elements of interest with respect to a suitable comparator. Chlorine is regarded as one of the

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most commonly used elements as comparator for evaluating prompt k_0-factors because it forms chlorides with most of the commonly used materials.[9] In earlier measurements of k_0-factors in NAA, pure gold was used as comparator.[10] Moreover the k_0 standardization can be applied directly to determine absolute elemental concentration if the exact composition of only one element present in the sample is known.[11]

The efficiency calibration of detection system over the desired energy range from a few keV to about 10 MeV is an important requirement for this technique.[12,13] The gamma rays from ^{35}Cl(n,γ) and ^{14}N(n,γ) are regarded as accurate standards for this purpose.[14] Sudarshan et al [15] have studied capture gamma rays from ^{60}Co to be used as multi γ-rays efficiency standard for PGNAA. They observed that the γ-rays from ^{59}Co(n,γ)^{60}Co can be a good supplement to chlorine and nitrogen due to the fact that capture cross section in ^{59}Co is much higher than in ^{14}N and comparable to ^{35}Cl resulting in higher γ-rays yield. Unlike other standards, which are only available in the form of compounds, cobalt can be used in metallic form. This feature results in the reduction of unwanted background in the γ-ray spectrum. Similar is the case of manganese. The thermal neutron cross sections of manganese are accurately known and it has γ-lines with good intensity over a wide energy range.

The data for gamma rays from the thermal neutron capture in ^{55}Mn are in poor shape, especially below 2 MeV.[16] Above 2 MeV, there are several fairly complete data sets. Below 2 MeV, there is a lot of variation in the existing data. So the intensities of prominent gamma lines up to 2 MeV will be a useful addition to the data. Moreover since k_0-factor can eliminate systematic errors in efficiencies, so manganese can also be a good choice for efficiency calibration in low as well as in medium energy region.

2 Experimental

The thermal neutron capture gamma ray setup at the tangential through tube of Pakistan Research Reactor-I (PARR-I) was used in this study. The details of indigenously designed and fabricated facility have been given elsewhere.[17,18] The size of neutron beam at the target position as determined by solid state nuclear track detectors (SSNTD) CR-39 was of about 3 cm in diameter.[19] The neutron flux was measured using gold foils with and without cadmium cover as well as with pure titanium metal. Its value at the target position was ~ 2×10^6 n/(cm^2•s)[20]. The detection system consisted of a 30% coaxial type Canberra HPGe detector kept at a distance of 40 cm from the sample position at an angle of 90^0 with respect to the neutron beam direction. The HPGe detector was placed between two annulus 8″×8″ NaI(Tl) detectors and the system was operated in Compton suppression mode. The whole assembly was surrounded by 4″ to 8″ thick lead shielding on all sides except towards the sample. Lithium carbonate plugs with total thickness of 8″ were placed between HPGe detector and the target to stop stray neutrons hitting the detector and thereby damaging the crystal. The detector system was coupled to a PC-based multi-channel analyzer with 8K-conversion gain. The detector had a resolution of about 2 keV at 1332 keV gamma line from ^{60}Co.

Before irradiating the samples, standard radioactive sources ^{60}Co and ^{152}Eu from IAEA and NH_4Cl from Merck, Germany (for thermal neutron capture in chlorine) were used in the same configuration to obtain absolute efficiencies.[21] The care was taken to maintain the sample size less than the beam diameter. Spec-pure $MnCl_2$•$4H_2O$ (1.5423 g) was used to determine prompt k_0-factors of manganese with respect to chlorine. The prompt γ-ray spectrum of manganese was recorded by irradiating 1.21 g of high purity manganese metal pieces crushed into powder form. The background spectra were also recorded at frequent intervals to take into account the fluctuations in the background and to access the contaminant γ-lines.

An important prerequisite for determining the absolute γ-ray emission probabilities is the measurement of full-energy photo-peak detection efficiency in the energy range of interest.[22] Since there is no single radioactive source available to cover such a wide energy range, the data both from standard radioactive sources and from thermal neutron capture gamma rays (n,γ) reactions were used. In case of γ-lines from (n,γ) reactions, absolute disintegration rates were not known. So only the relative efficiencies were worked out. Since accurate gamma ray detection efficiency is

essential in the k_0 standardization method, to get the absolute efficiency curve the data points were normalized with those from a calibrated radioactive source.

The prompt experimental k_0-factor of the element 'x' with respect to the comparator 'c' is given by Ref. [9]:

$$k_{0,x} = \frac{A_x/\varepsilon_c}{A_c/\varepsilon_x} \quad (1)$$

where ε_c and ε_x are the full energy photo-peak detection efficiencies of the comparator and the unknown sample respectively, and A_x and A_c are the analytical sensitivities (counts per second per unit mass of the element) of the sample and the comparator respectively.

3 Results and discussion

Prompt gamma spectra of manganese chloride and pure manganese are shown in Figs.1 and 2 respectively. The prominent γ-lines from chlorine are 517, 786, 1164, 1952 and 1959 keV. Due to much lower intensity, most of the gamma rays from manganese in the spectrum are not clearly visible. The 212 keV and 314 keV are prompt whereas 846 keV line is activation line from Mn. The tallest line in the spectra is 511 keV annihilation peak which is always present whenever the data are collected during reactor operation. The vertical scale in the figures is adjusted according to the height of peaks of interest. The data were obtained for time long enough to accumulate sufficient counts in the peak for analysis as well as to minimize the errors due to counting statistics. After irradiation, delayed γ-rays from radioactive ^{56}Mn were also obtained and 846 keV line was counted. This gamma line was used for neutron flux estimation to complement the values obtained with Ti. The background spectrum with reactor beam on is shown in Fig.3 for comparison. This background was recorded when the reactor was at full power (10 MW). The intensities of lines indicate good shielding for spurious gamma rays in the background.

In the present work, a fifth-order polynomial of the form

$$(\ln \varepsilon) = \sum_{i=0}^{5} a_i (\ln E)^i \quad (2)$$

has been used as a fitting function for the efficiencies "ε" where a_i's are fitting parameters and "E" is the gamma ray energy of interest. Absolute efficiencies in the energy range of 121–1408 keV were first determined by using ^{60}Co and ^{152}Eu data in Eq.(1).

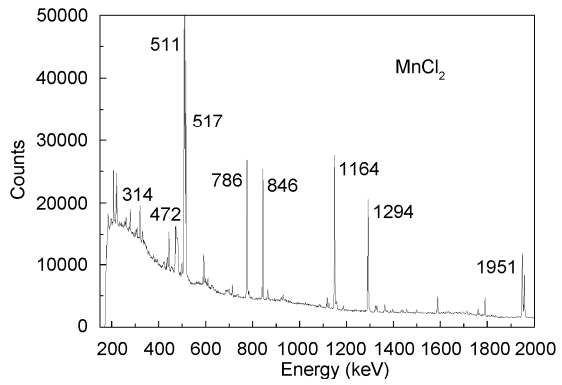

Fig.1 Prompt gamma ray energy spectrum of $MnCl_2$.

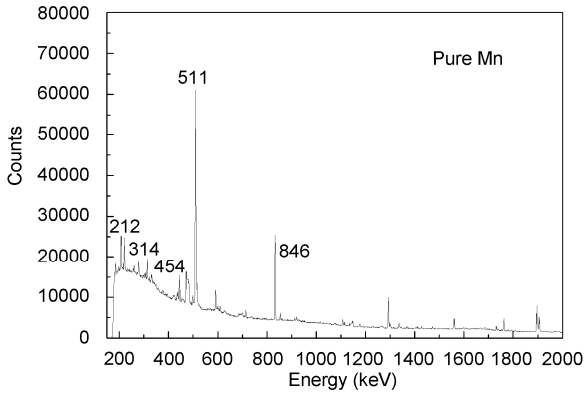

Fig.2 Prompt gamma ray energy spectrum of pure manganese.

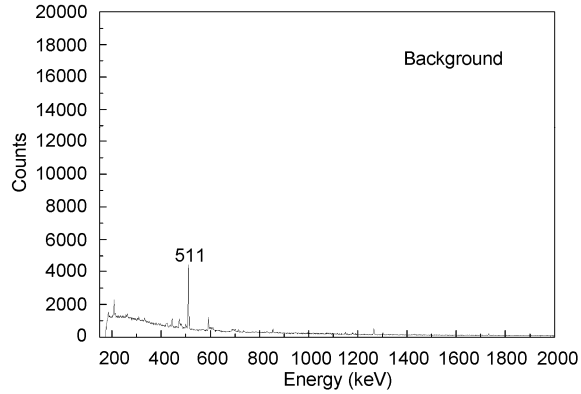

Fig.3 Background spectrum during reactor operation at full power of 10 MW.

An initial estimate was obtained by normalizing the relative efficiency of 1164 keV peak of ^{36}Cl and

342 keV of ^{49}Ti with the absolute efficiency of 1173 keV line from ^{60}Co and 344 keV line from ^{152}Eu. With these initial values, the entire set of peak areas from ^{36}Cl and ^{49}Ti were subjected to least-squares analysis using Eq. (2) to get a new set of a_i's. The absolute efficiency curve thus obtained is shown in Fig.4. The typical efficiency values are in the range of 10^{-5} and the errors on the fitted efficiency were in the order of 1%-3% in the lowest to highest energy range.

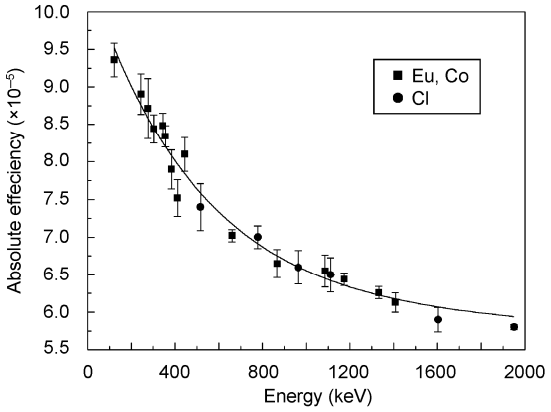

Fig.4 Absolute efficiency curve up to 2 MeV.

The k_0-factors for γ-rays from ^{56}Mn with respect to 1951 keV gamma line from ^{36}Cl were obtained using the data of manganese chloride. In literature we could only find the value of k_0-factor for 314 keV gamma ray of ^{56}Mn. We obtained a value of 0.149 whereas the values reported by other laboratories in NIST[23], JAERI[24] and Budapest[9] are 0.138, 0.152 and 0.148 respectively. The k_0-factors for the prominent gamma lines of manganese are tabulated in Table 1. As already mentioned that in the literature only the value of k_0-factor for 314 keV gamma line of ^{56}Mn is given, so the other values are being reported for the first time.

Table 1 Measured values of k_0-factor for ^{56}Mn

Energy (keV)	Efficiency ($\times10^{-5}$)	Experimental k_0-factor
188.51	8.890(4)	0.021(5)
212.14	8.793(8)	0.222(6)
271.38	8.548(3)	0.112(11)
314.44	8.382(2)	0.149(7)
375.69	8.157(9)	0.024(4)
454.75	7.890(3)	0.061(13)
1140.31	6.382(3)	0.036(3)
1403.39	6.059(2)	0.046±(5)
1511.00	5.952	0.041(4)

The values of k_0 can be calculated theoretically using the following relation give by Molnar et al:[9]

$$k_0 = \frac{(\sigma\theta\gamma / M)_x}{(\sigma\theta\gamma / M)_C} \quad (3)$$

where σ is thermal neutron absorption cross section, θ is isotopic abundance, γ is the absolute intensity of gamma rays of interest and M is atomic weight of the element. Here subscripts 'x' and 'c' refer to the unknown and comparator, which are Mn and Cl respectively. The values of σ, θ and M used in this study are given in Table 2, which were taken from Refs. [25], [26] and [27] respectively. The absolute intensities of gamma rays from ^{56}Mn were thus obtained by the following relation:

$$\gamma_x = \frac{\sigma_c\theta_c\gamma_c M_x}{\sigma_x\theta_x M_c} \times k_0 \quad (4)$$

The intensities worked out using the k_0-factor are presented in Table 3. The comparison with the data from other laboratories shows good agreement. Most of the values in the present study are quite close to those reported in Ref. [28] by Los Alamos Neutron Lab except at the lowest energy of 188 keV. When compared with the data compiled by Lone et al,[29] the

Table 2 Parameters used for calculations

Parameter	^{55}Mn	^{35}Cl	^{48}Ti
σ (barn)	13.3(2)	33.1(3)	6.09(13)
θ (%)	100	75.77(45)	73.720(22)
M	54.938049(9)	35.4527(9)	47.867(1)

Table 3 Measured values of absolute intensities

Energy (keV)	Absolute intensity				
	This study	Lone [29]	LANL [28]	Von Asseche [30]	ENSDF
188.51	1.307(5)	1.80	2.5	1.80	1.8
212.14	13.881(9)	8.8	13.0	10.6	11
271.38	7.029(2)	4.48	8.0	5.7	5.8
314.44	9.331(2)	6.76	9.4	9.4	9.5
375.69	1.496(3)	0.59	0.83	0.83	0.84
454.75	3.829(5)	2.22	2.9	2.9	2.9

1140.31	2.276(4)	0.66	3.0	N.A	N.A
1403.39	2.916(7)	0.88	3.5	N.A	N.A
1511	2.558(5)	N.A	2.7	N.A	N.A

value of absolute intensity for 188 keV is a little higher while all other values are lower than the values reported by other labs. Similar is the case for the values determined by Von Asseche et al.[30] In general the agreement is good enough but the present results may be regarded as more accurate due to application of k_0 standardization approach.

4 Conclusion

The k_0-standardization has been applied to prompt gamma neutron activation analysis of prominent gamma rays from ^{56}Mn up to 2 MeV. The only known values of k_0-factor for 314 keV photopeak agree well with the reported values in the literature, whereas other values of main gamma ray lines in manganese have been obtained for the first time. The measured absolute intensities of these peaks match with the theoretical values within the experimental errors. Due to the carefully measured gamma ray detection efficiencies using the standard radioactive sources from IAEA and the thermal neutron capture in Cl, it can be said that the values are more reliable. These data can also be useful for efficiency calibration in the low and medium energy regions.

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