

## Studies on Internal Bremsstrahlung Radiation Accompanying Second Forbidden $\beta$ -Decay of $^{36}\text{Cl}$ and $^{99}\text{Tc}$

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**Abstract:** The internal bremsstrahlung (IB) radiation accompanying the  $\beta$ -decay of the second forbidden  $\beta$ -transition of  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  sources are studied. The IB measured using a scintillation  $\gamma$ -spectrometer. It was analyzed by the variable width peeling-off-method, and then corrected applying the proper corrections. The analyzed and corrected IB spectrum was compared with those calculated according to Monte Carlo simulation and modified KUB calculations (M.KUB). It was found that the present experimental results have a better agreement with the theory than the previous work. Also it was found that the present experimental results are in a better agreement with M.KUB calculations than Monte Carlo simulation. This may be due to that the effect of Coulomb-charge (Z) on the IB process is treated in a more refined manner in M.KUB calculations than Monte Carlo simulation. The  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  sources have the same degree of forbiddenness but different values of Coulomb charge (Z) and different values of  $\beta$ -endpoint energy (709 KeV and 292 KeV respectively). Therefore the effects of Coulomb charge (Z) and values of  $\beta$ -endpoint energy on the IB-process are studied. It was found that these effects have a large contribution into IB-spectrum for the sources of larger Coulomb charge (Z) and lower  $\beta$ -endpoint energy, as in the case of  $^{99}\text{Tc}$ -source.

**Keywords:** Internal bremsstrahlung radiation, Radiative  $\beta$ -decay, Second forbidden  $\beta$ -transition and Radioactive sources  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$ .

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### I. Introduction:

The  $\beta$ -decay processes are accompanied by a weak electromagnetic radiation called internal bremsstrahlung (IB). IB has a continuous energy distribution. A review article by Persson [1] gives a detailed survey for the progress in the study of IB process. It appears that, unfortunately, in the majority of the cases studied, agreement between theory and experiment, as also among various experimental results has not yet been satisfactorily established.

Common to all these studies is the fact that the latter half of the IB spectrum deviated positively from the theories. The higher the energy, the higher was the magnitude of deviation. This disagreement is more significant at the forbidden  $\beta$ -transitions. It could not be explained until recently. Many more studies are needed to arrive at any definite conclusion. With this end in view, it was decided to reinvestigate the IB processes associated with the second forbidden  $\beta$ -decay of  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  sources.

In the literature the IB of  $^{36}\text{Cl}$  was measured by Venkaramiah [2] using the magnetic deflection technique. They compared their experimental results with the theoretical calculations of allowed and first forbidden  $\beta$ -transitions. Therefore, they suggested that it would be interesting to see the comparison of the experimental results with a theory developed for second forbidden  $\beta$ -transitions. Also one of the authors investigated before the IB of  $^{36}\text{Cl}$  Basha et al. [3] and compared the experimental results with the theoretical calculations of allowed and first forbidden  $\beta$ -transitions. Therefore it would be interesting to reinvestigate the IB of  $^{36}\text{Cl}$  and compare its experimental results with the theoretical calculations for second forbidden  $\beta$ -transitions.

The IB spectrum of  $^{99}\text{Tc}$  and other radioactive sources were measured by Keshava et al. [4] using the base line shift method. They compared their experimental results with the theoretical calculations of first forbidden  $\beta$ -transitions. They found an agreement between experimental results and theoretical calculations of Lewis and Ford [5]. We think that this agreement may be fortitious, because  $^{99}\text{Tc}$  is second forbidden  $\beta$ -transition and Lewis and Ford [5] is for first forbidden  $\beta$ -transition. Also Gundu Rao et al [6] studied the IB spectrum of  $^{99}\text{Tc}$  using the magnetic deflection method. They found no agreement between experimental results and theory. Also the IB of  $^{99}\text{Tc}$  is investigated by El-Konsol et al [7]. They compared their experimental results with the theoretical calculations of allowed and first forbidden  $\beta$ -transitions.

In this work the experimental results of  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  are compared with the theoretical calculations of modified KUB and Monte-Carlo simulation for second forbidden  $\beta$ -transitions. Also in this work we investigated the effect of Coulomb charge (Z) and  $\beta$ -end point energy values on the IB processes accompanying  $\beta$ -decay.

## II. Theoretical calculations:

### 2.1. $\beta^-$ -Particles energy spectra:

A quantum mechanical theory which satisfactorily gives the shape of  $\beta^-$ -particles energy spectra has been developed by Fermi. The Fermi theory can be found in textbooks (Evans, [8]; Konopinski[9]; Wu and Moskowski, [10]. The number of  $\beta^-$ -particles  $N(W) dW$  in the energy range  $W - W + dW$  can be approximately written as Cengiz[11].

$$N(W)dW \cong \frac{|P|^2}{\tau_0} 2\pi\alpha Z(W_0 - W)^2 W dW \quad (1)$$

with  $W = E/mc^2 + 1$  and  $W_0 = E_m/mc^2 + 1$  being the total energy and the maximum total energy of the  $\beta^-$  particle of kinetic energy  $E$  and the maximum kinetic energy  $E_m$  in units of electron rest mass energy,  $mc^2$ , where  $|P|^2$  is the squared modulus of the transition matrix element,  $\tau_0$  is a time constant, and  $\alpha$  is the fine structure constant.

For allowed transitions,  $|P|^2$  is independent of  $\beta^-$ -particle energy and is the order of unity. The allowed distribution of Eq. (1) must be multiplied with the shape correction factor in order to obtain the shape of the forbidden distribution. Many first-forbidden distributions and a few second-forbidden distributions can and do have the same shape as allowed distributions. Well-established examples include  $^{32}\text{P}$ ,  $^{186}\text{Re}$  and  $^{198}\text{Au}$  by Langer and Price [12]. So, the shape correction factor is ignored.

### 2.2. Bremsstrahlung cross section

For the nuclei of charge  $Ze$ , the differential electron bremsstrahlung cross-section (DEBCS) for the emission of a photon of energy  $k$  by an incident electron of kinetic energy  $E$  and total energy  $E_0 = E + mc^2$ , in the energy range  $\tau$  and  $\tau + d\tau$  ( $\tau = k/E_0$  is the photon energy in units of total electron energy), has been proposed

$$\frac{d\sigma_b}{d\tau} = 4\alpha r_e^2 Z(Z + \delta) \frac{d\tau}{\tau} \left[ 1 + (1 - \tau)^2 - \frac{2}{3}(1 - \tau) \right] \times \left[ \Phi(\Gamma, Z) + F_1(\beta', Z) + F_2(\beta, Z) - \frac{1}{3} \ln(Z) \right], \quad (2)$$

where  $r_e$  is the classical electron radius. The electron-electron contribution is approximated by replacing the  $Z^2$  Bethe-Heitler cross-section expression by  $Z(Z + \delta)$ , where  $\delta = 0.75$  is the experimentally determined value of Lanzl and Hanson [14]. The Bethe-Heitler screening parameter  $\Gamma$  may be written as

$$\Gamma = \frac{100mc^2}{E_0 Z^{1/3}} \frac{\tau}{1 - \tau}. \quad (3)$$

The correction functions  $F_1(\beta', Z)$  and  $F_2(\beta, Z)$  are given by

$$F_1(\beta', Z) = \alpha Z(1 - \beta'^5), \quad (4)$$

$$F_2(\beta, Z) = 8.5 \left( \frac{mc^2}{E_0} \frac{\alpha Z}{\beta} \right)^2, \quad (5)$$

where  $\beta$  and  $\beta'$  are the velocity of the incident electron before and after interaction in units of velocity of light, respectively. With the two correction functions  $F_1(\beta', Z)$  and  $F_2(\beta, Z)$  included in the DEBCS, the best fit to the available experimental cross-section data gives the screening function as

$$\Phi(\Gamma, Z) = 4.6 \left( 1 + \frac{1}{Z^2} \right) - \frac{1}{\beta} \ln(\Gamma + \beta - 0.3). \quad (6)$$

The forms of the correction functions  $F_1(\beta', Z)$  and  $F_2(\beta, Z)$  and the screening function  $\Phi(\Gamma, Z)$  have been empirically determined, their parameters having been chosen by minimizing the sum of the squares of the differences between the predictions of Eq. (2) and the experimental data by Al-Beteri and Raeside[13].

The radiative stopping power is proportional with the scaled bremsstrahlung cross section. Al-Beteri and Raeside[13] were compared the values of scaled bremsstrahlung cross section with experimental values and the values of Seltzer and Berger [15] which include the contribution from electron-electron bremsstrahlung (Figs 3-6 in Ref. [32]). The agreement between the values of Al-Beteri and Raeside [13], the experimental data and the values of Seltzer and Berger is in good. The ESTAR (Stopping Powers and Ranges for Electrons) database (<http://physics.nist.gov/PhysRefData/Star/Text/method.html>) is very useful for the electron transport. The radiative stopping powers are evaluated in ESTAR with a combination of theoretical bremsstrahlung cross

sections described by Seltzer and Berger[15]. The uncertainties of the radiative stopping powers are estimated to be 2 % above 50 MeV, 2 % to 5 % between 50 MeV and 2 MeV, and 5 % below 2 MeV by Berger et al. (<http://physics.nist.gov/PhysRefData/Star/Text/method.html>).

**2.2 Monte Carlo Simulation:**

In the beta decay, the end-point energies of  $\beta^-$  mode ( $E_m$ ), decay energies of electron capture (EC) mode ( $E_c$ ), branch ratios, ( $\eta$ ), and relative probabilities ( $P$ ) of the investigated radioisotopes (<http://atom.kaeri.re.kr>) are given in Table 1. In this study, the EC event was not included.

For the radioisotopes with two branches ( $\eta_1, \eta_2$ ) and two end-point energies, ( $E_{m1}, E_{m2}$ ) the end-point energy is sampled by generating a uniform random number  $\xi$  ( $0 < \xi < 1$ ) by means of following conditions:

$$\xi \leq \eta_1. \tag{7}$$

If the inequality in Eq. (7) holds, then the end-point energy is selected as  $E_m = E_{m1}$ , otherwise it is selected as  $E_m = E_{m2}$ .

The energy distribution of  $\beta^-$ -particles, Eq. (1) has been sampled using the acceptance-rejection method. The details of the simulation were given by Cengiz and Almaz[11]. To carry out the acceptance-rejection sampling with the usual rectangular rejection, the envelope rejection function is chosen as  $r(W) = 1$  and the energy distribution of  $\beta^-$ -particles, Eq. (1) is normalized in the form in which its maximum value will be equal to  $r(W)$ . Eq. (1) gives a maximum value at  $W = W_0/2$ . So, the normalized energy distribution of  $\beta^-$ -particles is obtained as

$$N(W)dW = W^2 \frac{(W_0 - W)^2}{(W_0 / 2)^4} dW \tag{8}$$

$W$  is sampled using a  $\xi$ -value from the direct (inversion) method as

$$W = W_c + \xi(W_0 - W_c). \tag{9} \quad \text{By}$$

generating a second  $\xi$ -value the inequality,

$$\xi \leq W^2 \frac{(W_0 - W)^2}{(W_0 / 2)^4} \tag{10}$$

is tested. If the inequality Eq. (10) holds, the sampled  $W$  is accepted; otherwise, the procedure Eqs.(9) and (10) is repeated with a new pair of uniform random numbers. The kinetic energy of  $\beta^-$  particles emitted in  $\beta^-$ -decay is calculated from  $E = (W-1)mc^2$ .

In  $\beta^-$ -decay, the  $\beta^-$ -particle emitted in  $\beta^-$ -decay, undergoes the internal bremsstrahlung (IB) event in the field of emitting nucleus. The IB process has a probability per electron emission of approximately  $\alpha$ , the fine-structure constant (Struzynski and Pollock [16], Evans [8] ). The existence of this case has been observed by Aston [17]. When the probability is taken into account, the shape of the IB distribution remains the same. Therefore, this probability is taken one.

The  $d\sigma_b(\tau)/d\tau$  formula has been sampled using the acceptance-rejection method. The details of the simulation were given by Cengiz and Almaz (2004). The envelope rejection function,  $h(\tau)$ , is chosen as

$$h(\tau) = \frac{f(\tau)}{f(\tau_{\min})}, \tag{11} \text{ where}$$

$$f(\tau) = \left[ 1 + (1 - \tau)^2 - \frac{2}{3}(1 - \tau) \right] \left[ \Phi(\Gamma, Z) + F_1(\beta', Z) + F_2(\beta, Z) - \frac{\ln Z}{3} \right]. \tag{12}$$

$\tau$  is sampled using a  $\xi$ -value from the

$$\tau = \tau_{\min} \left( \tau_{\max} / \tau_{\min} \right)^\xi. \tag{13} \quad \text{By}$$

generating a second  $\xi$ -value the inequality,

$$\xi \leq h(\tau). \tag{14}$$

is tested. If the inequality in Eq. (14) holds, the sampled  $\tau$  was accepted as a valid sample for the fractional bremsstrahlung photon energy ( $k = \tau E_0$ ); otherwise, the procedure with Eqs. (13) and (14) was repeated with a new pair of uniform random numbers. In this scheme, the energy of the bremsstrahlung photon,  $k$  and the remaining electron energy  $E - k$  are stored and a new electron is selected to be followed. The number of electron trajectories is chosen to be  $2 \cdot 10^7$

### 2.3 Modified KUB calculations (M.KUB):

The original theory of the IB was suggested by Knipp and Uhlenbeck[18] and by Bloch[19] (called KUB-Theory) for allowed  $\beta$ -transitions, neglecting the effect of Coulomb field (Z). Then Nilsson[20] as well as Lewis and Ford [5] calculated the IB-spectrum after taking into consideration the influence of the Coulomb field (Z) into IB-process. Despite these modifications for the original KUB-theory, the still absence of complete agreement between experiments and theories can be shown particularly for IB-spectrum accompanying forbidden  $\beta$ -transitions.

Therefore a new trail of theoretical calculations was carried out in the present work as applied for the first time by El-Konsol et al.[7].

In these calculations the shape correction factor for second forbidden  $\beta$ -transitions ( $C_2$ ) which was suggested by Konopiniski and Uhlenbeck[21] is used. This correction factor ( $C_2$ ) is calculated according to the following equation (15), for the sources under investigation ( $^{36}\text{Cl}$  and  $^{99}\text{Tc}$ ), and applied into Nilsson-theory[20] (where in Nilsson theory [20] the Coulomb correction factor was considered in a more refined manner than in the other theories). This is called modified KUB-calculations (M.KUB).

$$C_2 \cong p^4 + \frac{10}{3} p^2 q^2 + q^4 \cong (W^2 - 1)^2 + \frac{10}{3} (W^2 - 1)(W_0 - W)^2 + (W_0 - W)^4 \quad (15)$$

Where p and q are the moments of the associated electron and neutrinos respectively, W and  $W_0$  as defined before in equation (1).

## III. Experimental details:

### 3.1 $^{36}\text{Cl}$ :

The  $^{36}\text{Cl}$ -source was supplied by the radiochemical centre Amersham (Buckinghamshire, England) with an activity of approximately  $658.6 \times 10^4$  Bq.

This source decays to the ground state of  $^{36}\text{Ar}$  by  $\beta$ -emission with a branching ratio of 98.1% and an end point energy of 709 KeV. Positron emission with a 0.0017% branching ratio and an end point energy of 115 KeV was also observed. The  $\beta$ -transition was characterized by  $\Delta j=2$ ,  $\Delta \pi=0$  and  $\log ft=13.3$ , which is classified as non unique second forbidden  $\beta$ -transition (<http://atom.kaeri.re.kr>). It also decays to the ground state of  $^{36}\text{S}$  by an electron capture (Ec) with a branching ratio of 1.9% and an end point energy of 1135 KeV (25-27). For Ec transition one can find that  $\Delta j=2$ ,  $\Delta \pi=0$  and  $\log ft=13.58$  (<http://atom.kaeri.re.kr>).

The IB spectrum accompanying  $\beta$ -transition of  $^{36}\text{Cl}$  was measured with a single channel scintillation spectrometer utilizing a NaI (Tl) crystal of 2.54 Cm diameter and 1.91 Cm height optically attached to a 50 AVP photomultiplier. Details of the experimental arrangement and all necessary corrections which were applied for the measured IB can be found in the previous literature Khalil [22-24]. It was observed that the measured IB spectrum of  $^{36}\text{Cl}$  was accompanied with the appearance of a photo-peak at an energy 1135 KeV, which is due to the electron effect (Ec) as declared above. However this contribution of electron of electron capture was neglected because of its low intensity 1.9% (Basha et al.[25]).

### 3.2 $^{99}\text{Tc}$ :

$^{99}\text{Tc}$  is of a second forbidden beta transition type with a half-life  $\approx 2.19 \times 10^5$  yr and  $\beta$ -end point energy 292 KeV. Its decay scheme was given in (<http://atom.kaeri.re.kr>). The 89 KeV gamma line was not observed in the present measurement. It was observed only the contribution of the 141 KeV gamma line intensity to the IB spectrum of  $^{99}\text{Tc}$ -source. A certain Procedure similar to that suggested by Khalil [22,26] has been used. This procedure is based on choosing a mono-energetic source with gamma-line energy equal to that of the source under investigation. Measuring the spectrum of this mono energetic source under the same condition and geometry of the IB spectrum measurements. Then by simple subtraction one can easily obtain the pure IB spectrum.

The measured IB spectrum was then analyzed into its constituents by performing the peeling off procedure starting from the high energy tail of the spectrum. In this procedure, the complete pulse high spectra of various mono energetic gamma lines of energies covering the whole IB range studied (100 KeV-1200 KeV) were measured by the scintillation spectrometer at different high tension and at different sources to crystal distances. Dividing the full width at half maximum (FWHM) by the corresponding energy one can obtain the energy resolution of the spectrometer.

The measured IB intensity was then corrected for the solid angle, absorption, crystal efficiency, back scattering and external bremsstrahlung. The energy resolution of the whole gamma-spectrometer was determined to be 12%. All the details of the experimental processes and methods of analysis are given in the earlier work by Khalil [22-24] and Basha et al.[3].

#### **IV. Results and discussion:**

The measured IB spectrums of  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  sources are presented in figures (1) and (2) respectively. For each isotope the measured IB-spectrum was corrected according to the aforementioned corrections. For  $^{36}\text{Cl}$  the investigated energy region is from 305-648 KeV. Therefore the corrected experimental IB results of  $^{36}\text{Cl}$  are normalized into the theoretical calculations of Monte-Carlo simulation and M.KUB theory at 350 KeV as shown in figure (3). For  $^{99}\text{Tc}$  the investigated energy region is from 50-290 KeV. Then the experimental IB results of  $^{99}\text{Tc}$  are normalized into the theoretical calculations of Monte-Carlo simulation and M.KUB theory at 50 KeV as shown in figure (4).

In figure (3), one can observe that the present experimental IB results of  $^{36}\text{Cl}$  are in a better agreement with the theoretical calculations than in the previous work by Venkataramiah et al. [2]. Also, we observed in figure (4) that the present experimental IB results of  $^{99}\text{Tc}$  are in a better agreement with the theoretical calculations than in the previous work by Keshava et al. [6] and GunduRao et al. [4]. But we observed in figures (3) and (4), that still there is a positive deviation between experimental results and theoretical calculations close to  $\beta$ -end point energy. This may be due to detour effect on IB-process. This detour transition is accompanying with the forbidden  $\beta$ -transitions and was discussed early by Ford and Martin [27]. May be, if we take into account this detour effect into the theoretical calculations of M.KUB and Monte Carlo simulation we can get a better agreement between experiment and theory close to  $\beta$ -end point energy.

The  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  sources have the same degree of forbiddance of  $\beta$ -transition but different values of Coulomb charge ( $Z$ ) and different values of  $\beta$ -endpoint energy (709 KeV and 292 KeV respectively). Therefore the effects of Coulomb charge ( $Z$ ) and values of  $\beta$ -endpoint energy on the IB-process are studied. We represented in figure (5) the ratio values of experimental results into theoretical calculations of modified KUB  $R(\text{Exp./M.KUB})$  as a function of the ratio values of  $(K/E_{\beta\text{max}})$  for the sources under investigation  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$ . Also in figure (6) the ratio values of experimental results into theoretical calculations of Monte Carlo  $R(\text{Exp./Mont Carlo})$  for  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  are presented. From the figures (5) and (6) one can observe that the ratio values of  $^{99}\text{Tc}$  is larger than ratio values of  $^{36}\text{Cl}$  in the investigated energy region.  $^{99}\text{Tc}$  has a larger Coulomb charge ( $Z$ ) and lower value of  $\beta$ -endpoint energy in comparison with  $^{36}\text{Cl}$ . Then one can say that the effects of Coulomb charge ( $Z$ ) and value of  $\beta$ -end point energy on the IB-processes are large for the sources of larger Coulomb charge ( $Z$ ) and lower value of  $\beta$ -endpoint energy as mentioned before in Khalil [23-24].

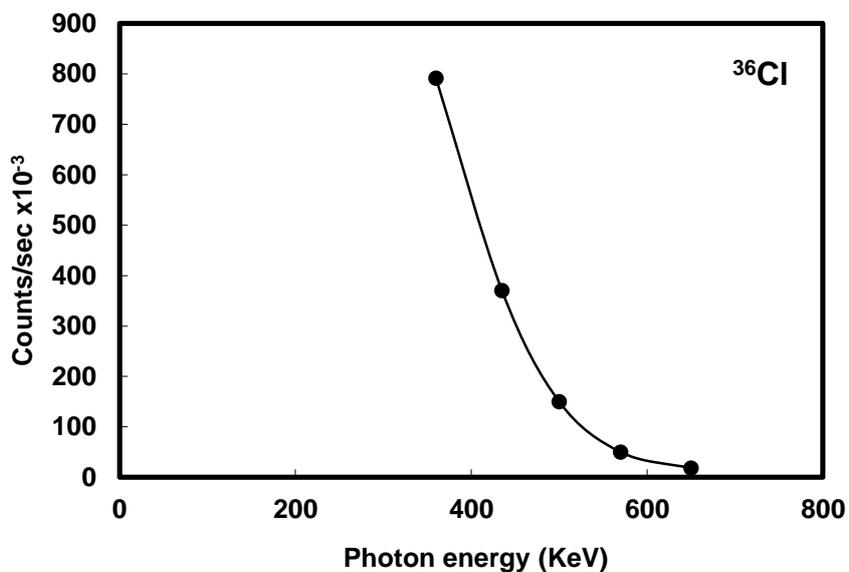
The above study may lead to the conclusion that, if the effects of Coulomb correction,  $\beta$ -endpoint energy, degree of forbiddance, and consequently detour contribution are considered in a more refined manner than those of the aforementioned theories, a good agreement can be expected between experiment and theory at the higher energy values, close to  $\beta$ -endpoint energy, as suggested before by Khalil [23-24].

#### **Acknowledgement:**

E.I.Khalil thanks Prof. S. El-Konsol and Dr. A.M. Basha for their interest and encouragement.

**Table 1.** In beta-decay, the end-point energies of  $\beta^-$  mode ( $E_m$ ), the energies of EC mode ( $E_\epsilon$ ), branch ratios ( $\eta$ ) and relative probabilities ( $P$ ) for the radioisotopes,  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  (<http://atom.kaeri.re.kr>).

Nucleus	Mode of decay		$\eta$		$P$	
	$\beta^-$ $E_m$ (keV)	EC $E_\epsilon$ (keV)	Beta	EC	$\beta^-$	EC
$^{36}_{17}\text{Cl}$	709.23	1142.3	0.9810	0.0190	1.00	not including
$^{99}_{43}\text{Tc}$	293.5	---	0.999984	--	1.00	---
	204.0		0.000016		0.00	



**Fig. (1):** Typical IB spectrum of  $^{36}\text{Cl}$

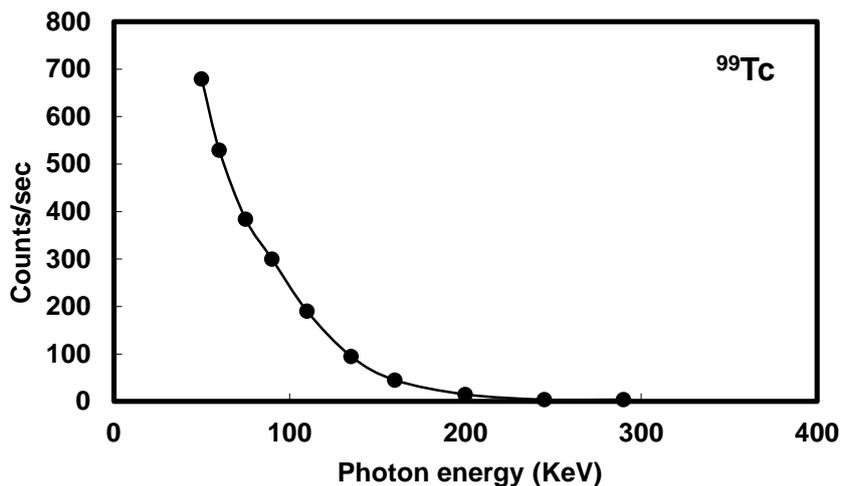


Fig. (2): Typical IB spectrum of  $^{99}\text{Tc}$

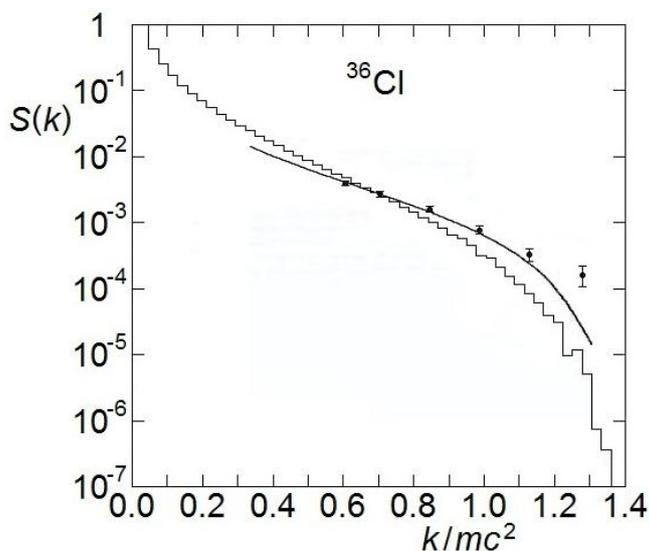


Fig.(3): The experimental points of Internal bremsstrahlung of  $^{36}\text{Cl}$  with the theoretical calculations of M.KUB (solid line) and Mont Carlo (zdzaj line).

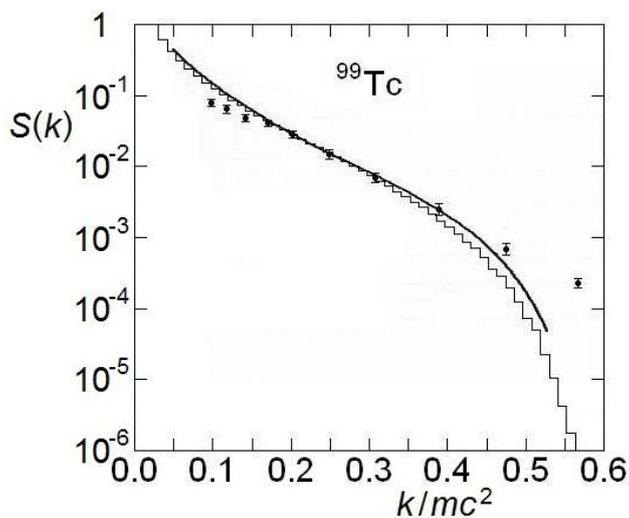


Fig.(4): The experimental points of Internal bremsstrahlung for  $^{99}\text{Tc}$  with the theoretical calculations of M.KUB (solid line) and Mont Carlo (zdzaj line).

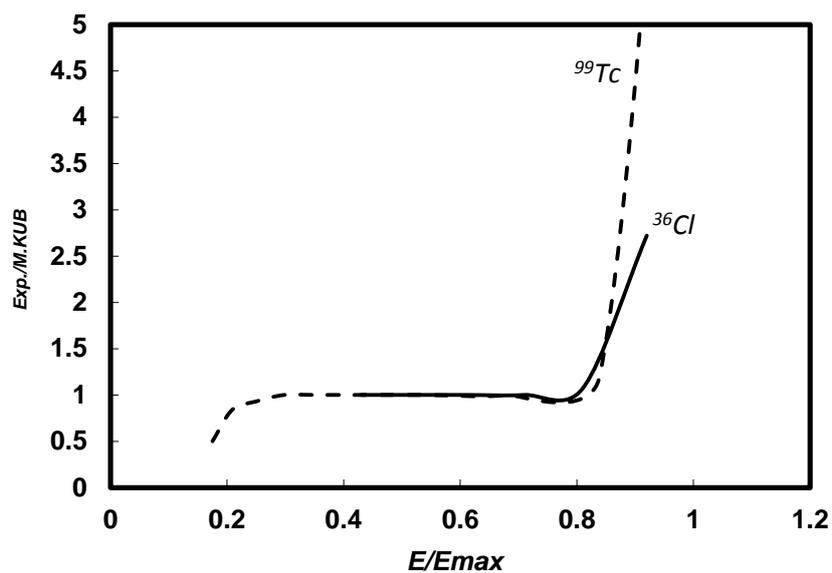


Fig. (5): The ratio values of ( Exp./M.KUB ) with  $(K/E_{\beta_{max}})$  for  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  Sources.

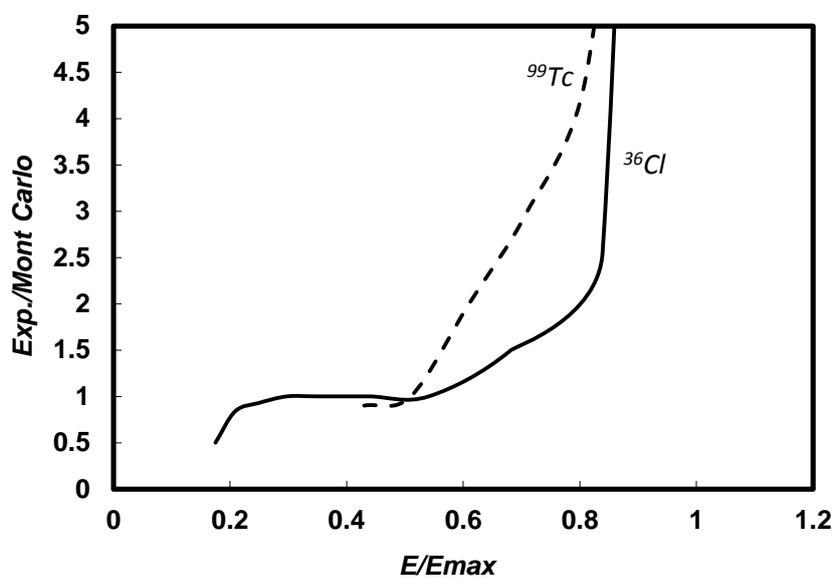


Fig. (6): The ratio values of ( Exp./Mont Carlo ) with  $(K/E_{\beta_{max}})$  for  $^{36}\text{Cl}$  and  $^{99}\text{Tc}$  Sources.

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