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Calculation of L-shell ionisation and x-ray production cross sections for some trans-uranium elements induced by protons

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ABSTRACT

L-shell ionisation and x-ray production cross sections of trans-uranium elements: Np, Pu and Am, induced by protons of energies 5.00–20.00 MeV, have been calculated. The cross sections for the targets were obtained using computer codes based on the ECPSSR theory. Due to the lack of experimental data of the targets considered, in view of their radioactive, toxic and unstable nature, the calculated cross sections were subsequently validated. This was achieved using the computer codes to evaluate the ionisation and x-ray production cross sections of Uranium induced by protons of energies in the 1.00–20.00 MeV range. This study also showed that the relative x-ray line intensities associated with the ionisation and x-ray productions of the targets considered are dependent on protons energies.

1. Introduction

Inner-shell ionisation of atoms by charged particles has been an area of interest because of its major application in several fields especially in atomic and nuclear physics; and in the particle induced x-ray emission (PIXE) spectroscopy [1]. A vast knowledge of the x-ray production cross sections is required for quantitative analysis of samples in PIXE technique [2]. The study of the ionisation process has increased rapidly over the years resulting in comparisons between experimental and calculated ioniation and x-ray production cross sections. Consequently, there exists a large amount of data on ionisation and x-ray productiond cross sections of stable elements induced by protons and alpha particles with few or no data for the unstable ones [2–4]. Application of the ionisation process in fundamental and applied studies require adequate knowledge of several fundamental parameters in order to obtain accurate quantitative results. This include the emission rates, fluorescence yields, Coster-Kronig transition probabilities (atomic parameters), ionisation cross sections and x-ray production cross sections [5,6].

Several models have been used in the description of the shell x-ray emission and ionisation process by protons. This include the Plane Wave Born Approximation (PWBA) theory [7], the Semi-Classical Approximation (SCA) theory [8], Binary Encounter Approximation (BEA) theory [9] and the Energy Loss Coulomb Repulsion Perturbed Stationary State Relativistic (ECPSSR) theory [10]. So far, the ECPSSR theory has been successful in describing the ionisation process for the K- and L-shells with close agreement between calculated and experimental cross sections values. The ECPSSR theory, though based on the PWBA model, takes into account the energy loss (E) during collisions and Coulomb deflection (C) of the projectile; and the perturbed stationary state (PSS) and relativistic (R) nature of the inner-shell electron. The ECPSSR ionisation cross section is formulated in terms of the PWBA cross section as [10]:

$$\sigma_s^{ECPSSR} = C_s\left(\frac{2\pi d q_{0s}\zeta_s}{z_s(1+z_s)}\right) f_s(z_s)\sigma_s^{PWBA}(\xi_s^R/\zeta_s,\ \zeta_s\theta_s) \tag{1}$$

where $\sigma_s^{PWBA}\left(\frac{\xi_s^R}{\zeta_s},\ \zeta_s\theta_s\right)$ represents the PWBA cross section, $d \equiv Z_1Z_2/Mv_1^2$ is the half distance of the closest approach and q_{0s} represents the approximate minimum momentum transfer U_{2s}/v_1. U_{2s} is the observed binding energy of the ejected target electron and v_1 represents the projectile velocity. C_s is the Coulomb deflection factor with s = K, L_1, L_2 or L_3. Z_1 and Z_2 represent the projectile's (ion) and target's atomic numbers, respectively; and θ_s is the reduced binding energy. η_s is the reduced incident ion energy, v_1 is the projectile velocity and U_s is the binding energy of the ejected electron. The dimensionless paramter ξ_s^R represents the relativistically corrected reduced ion velocity. The projectile energy loss during the collision process is represented by the correction factor $f_s(z_s)$ where, z_s is an argument parameter. An important parameter that play a major role in the calculation of the ionisation cross section of a target is the electron form factor. The electron form factor, $F_{Ws}(Q)$ is calculated in the PWBA cross sections as:

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$$\sigma_s^{PWBA} = (2j_2+1)4\pi a_0^2 \frac{Z_1^2}{(Z_{2s})^4 \eta_s} \int_{W_{\min}}^{W_{\max}} dW \int_{Q_{\min}}^{Q_{\max}} \frac{dQ}{Q^2} |F_{Ws}(Q)|^2 \quad (2)$$

where $Z_{2s} = Z_2 - S$, where S is the screening parameter (S= 0.3 for the K-shell and S = 4.15 for the L-shell). j_2 is the total angular momentum of the target nucleus. W_{min} and W_{max} are the minimum and maximum energy transfered to the target atom while Q_{min} and Q_{max} are the minimum and maximum momentum transferred to the target atom.

Over the years, several tabulations [11,12] have evaluated the ionisation cross sections for various targets but this procedure yielded results that are not in good agreement with experiment. This has led to the use of computer codes to calculate the inner shell cross sections employing available models in order to minimize these discrepancies. A few open source codes used in calculating the ECPSSR inner shell cross sections (Eq. 1) are found in literature [12–14]. These codes have been used to evaluate the ionisation and x-ray production cross sections for the K-shell resulting in excellent agreement with experimental data. However, as a result of the complex nature of the L-shell, calculated cross sections using computer codes are not in good agreement with experimental ones. This has led to several updates in available computer codes [15,16] employed in the calculation of inner shell ionisation cross sections. For example, the Inner-Shell Ionisation Cross sections (ISICSoo) code [15] is a C language program that computes ionisation and x-ray production cross sections for K-, L-, and M-shells. The code is in the ECPSSR and PWBA theoretical frame-works, using exact integration limits for calculating the form factors. The Electron Removal Cross Section, (ERCS08) program [16], which is implemented in FORTRAN, calculates cross section for K-, L- and M-shell ionisation according to various options of the ECPSSR theory. The Smit's code [17], implemented in Pascal, calculates the ionisation cross sections for the K- and L-shells using correct limits of integration of transfer momentum, which is given as:

$$Q_{\substack{\min \\ \max}} = \frac{\zeta_s^2 \theta_s^2}{\eta_s^R n_2^4} \left(1 \pm \sqrt{1 - \frac{m \zeta_s \theta_s}{M \eta_s n_2^2}} \right)^{-2} \quad (3)$$

where M is the projectile mass, n_2 represents the principal quantum number of the L-shell, η_s^R and m_s^R are the relativistically corrected reduced incident ion energy and electron rest mass, respectively. This parameter, $Q_{\max}{}^{\min}$ is erroneously evaluated in the ERCS08 and ISICSoo codes as reported in [17]. The Smit's code is a refinement of an earlier version [14] in order to obtain ionisation cross sections that yield better prediction of the original ECPSSR theory [10].

The wide discrepancies that exist between experimental and calculated x-ray production cross sections are also attributed to high level of uncertainties in the atomic parameters associated with inner shell transition [18,19]. In order to minimize these discrepancies, several values of the atomic parameters have been calculated using various theories such as the semi-empirical formula [5,20], Dirac Hartree Slater (DHS) model [18,21] and Dirac–Fock Model [19,22,23]. Of all the reported values of the atomic parameters, those of [19,22,23], based on the Dirac–Fock (DF) theory, were found to have the lowest uncertainties.

In this work, we determined the L-shell ionisation cross sections of the targets, $Z_2 = 93$, 94 and 95 induced by protons in the 5–20 MeV energy range using the Smit's code that is based on ECPSSR theory. The obtained ionisation cross sections were converted to x-ray production cross sections using a new computer code named Feat-code that is implemented in pascal. The targets considered are trans-uranium elements: Neptunium, Plutonium and Americium that have useful applications in nuclear technology. Neptunium is used in detectors of high energy neutrons, Plutonium serves as a heat source in radioisotope thermo-electric generators which are used to power some spacecraft and Americium is used in the ionisation chamber of modern smoke detectors. These elements (Z_2 = 93, 94 and 95) have limited cross sections reported in literature [4]. This is because they are unstable, radioactive and toxic. Also, the dependence of the targets L-subshells cross sections on projectile energies will be evaluated. In addition, L-subshell ionisation cross sections as well as x-ray production cross sections shall be calculated for $_{92}U$ in the same proton energy range of 5.00–20.00 MeV as it is done for the three trans-uranium targets.

Table 1
Atomic Parameters: Fluorescence and Coster-Kronig Yields of $_{92}U$, $_{93}Np$, $_{94}Pu$ and $_{95}Am$ based on [22], the Total Atomic and Fractional Radiative Widths of $_{92}U$, $_{93}Np$ and $_{94}Pu$ based on [19] and those of $_{95}Am$ based on [21].

Parameters		Targets			
		$_{92}U$	$_{93}Np$	$_{94}Pu$	$_{95}Am$
ω_i	ω_1	0.190	0.200	0.210	0.220
	ω_2	0.506	0.519	0.473	0.487
	ω_3	0.444	0.454	0.463	0.473
f_{ij}	f_{12}	0.035	0.030	0.030	0.030
	f_{13}	0.670	0.670	0.680	0.680
	f_{23}	0.140	0.136	0.230	0.222
Γ_i	Γ_1	2.353	2.482	2.619	2.886
	Γ_2	4.340	4.588	4.837	5.044
	Γ_3	3.621	3.810	4.005	4.170
F_{ip}	F_{3l}	0.046	0.047	0.047	0.048
	$F_{3\alpha}$	0.744	0.742	0.740	0.743
	$F_{3\beta}$	0.209	0.211	0.212	0.205
	$F_{2\beta}$	0.763	0.755	0.753	0.757
	$F_{2\gamma}$	0.221	0.223	0.225	0.218
	$F_{1\beta}$	0.679	0.720	0.717	0.682
	$F_{1\gamma}$	0.243	0.260	0.260	0.242

Table 2
Calculated Ionisation Cross Sections of $_{93}$Np, $_{94}$Pu and $_{95}$Am for the L_1, L_2 and L_3 subshells.

Proton	Ionisation Cross sections for the Targets								
Energy	$_{93}Np$			$_{94}Pu$			$_{95}Am$		
(MeV)	σ_{L1}	σ_{L2}	σ_{L3}	σ_{L1}	σ_{L2}	σ_{L3}	σ_{L1}	σ_{L2}	σ_{L3}
5.00	12.86	21.31	94.01	11.06	18.96	86.02	9.45	16.87	78.81
6.00	22.43	29.93	125.8	19.56	26.76	115.5	16.95	23.92	106.1
7.00	33.62	39.05	157.5	29.64	35.05	145.0	25.98	31.46	133.6
8.00	45.60	48.37	188.3	40.56	43.57	173.8	35.90	39.24	160.6
9.00	57.73	66.82	245.4	51.73	52.13	201.5	46.14	47.10	186.6
10.00	69.57	66.82	245.4	62.73	60.57	227.6	56.32	54.88	211.3
11.00	80.84	75.67	271.3	73.27	68.77	252.1	66.15	62.48	234.4
12.00	91.36	84.13	295.2	83.18	76.66	274.8	75.45	69.82	256.0
13.00	101.1	92.17	317.2	92.37	84.18	295.7	84.13	76.85	276.0
14.00	109.9	99.74	337.4	100.8	91.29	315.0	92.13	83.53	294.4
15.00	117.9	106.8	355.8	108.4	97.98	332.7	99.44	89.33	311.3
16.00	125.0	113.4	372.5	115.3	104.2	348.8	106.1	95.76	326.8
17.00	131.4	119.6	387.7	121.5	110.1	363.5	112.0	101.3	341.0
18.00	137.1	125.3	401.4	127.0	115.5	376.8	117.4	106.5	353.9
19.00	142.0	130.5	413.8	131.9	120.5	388.8	122.2	111.3	365.6
20.00	146.4	135.0	425.0	136.2	125.2	399.7	126.4	115.7	376.2

2. Methodology

In calculating the L-subshells ionisation cross sections, we used protons of 5–20 MeV energy range. This is because unstable elements like trans-uranium elements have a very weak spectroscopic resolution. This makes it extremely difficult to carry out measurements of their cross sections at lower energies of the incoming projectiles (in this case, protons). So, measurements can only be possible at large cross sections via ionisation by protons above the intermediate 1–4 MeV energy range. The ionisation cross sections for the trans-uranium targets were