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حساب التنشيط الإشعاعي المستحث للكالسيوم (مكون أساسي في العظام) ولبعض آثار العناصر الأخرى باستخدام نيوترونات ذات طاقة 14 مليون الكترون فولت

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□ الملخّص □

يعتبر الكالسيوم مكونا أساسيا لعظام الإنسان وكذلك توجد نسب ضئيلة من بعض العناصر الأخرى تدخل في تركيب العظام.

عندما يتعرض الإنسان للإشعاع فإن هذه العناصر تنشط إشعاعيا ويصبح من المفروض أن نحسب كمية الإشعاع التي امتصتها هذه العناصر لمعرفة الجرعة المناسبة سواء في حالة التشخيص للمرض أو علاجه.

بعض نظائر هذه العناصر تكون نسبتها ضئيلة جدا (ولكنها موجودة) بحيث يصعب قياسها عمليا وفي هذه الحالة نلجأ إلى إجراء بعض الحسابات النظرية لحساب مقاطع التنشيط الإشعاعي لهذه التفاعلات وتكون النتائج مؤشر لقيمة هذا التنشيط.

في هذا البحث تم استخدام الكود EXIFON لحساب مقاطع التنشيط الإشعاعي وبالتالي تم استخدام هذه المقاطع لحساب قيمة الإشعاع المستحث داخل عظام الإنسان بمعلومية نسب تواجد العناصر الداخلة في تركيب العظام ، مثل الكالسيوم والحديد والنحاس والزنك ، والأسترونشيوم والباريوم والرصاص وذلك من خلال استخدام نيوترونات ذات طاقة 14 مليون الكترون فولت.

*أستاذة في شعبة طبيعة المفاعلات والنيوترونات مركز البحوث النووية–هيئة الطاقة الذرية–جمهورية مصر العربية

**مساعد باحث في شعبة طبيعة المفاعلات والنيوترونات مركز البحوث النووية–هيئة الطاقة الذرية–جمهورية مصر العربية

1. INTRODUCTION

At 14-MeV neutron laboratory energy, primary reactions like (n,p), (n, ń) and (n,α) are quite prominent. The (n,γ) reaction can be ignored when particle emission is possible. The data of (n,p) and (n, α) type reactions are important for making estimates of radiation damage (Qaim, 1971). A study of the induced activities per gm per neutron flux for calcium and Fe, Cu, Zn, Sr, Ba, and Pb trace elements in bone using the 14-MeV primary reaction cross-sections are described in this work.

The EXIFON code (Kalka, 1991) is based on an analytical model for statistical multistep direct and multistep compound reactions. It predicts emission spectra, angular distributions and activation cross-sections including equilibrium, preequilibrium as well as direct processes. The model is restricted to neutron, proton and α-induced reactions with neutrons, protons, alphas and photons in the outgoing channels. Multiple particle emissions are considered up to three decays of the compound system.

The estimation of the value of induced activity per unit neutron flux per gm of bone sample can be readily made for a reaction, which produces activity of appropriate half-life for laboratory measurements using these computed cross-sections. These estimates may be useful for the study of neutron activation analysis with conventional 14-MeV neutron generators, when experimental cross-sections are not available; either because the abundance of some of the isotopes is very low or the cross-section for the desired reaction is of the order of few millibarns which makes their experimental measurements difficult.

2. BRIEF DESCRIPTION OF THE CODE

2.1. Total Emission Spectra of the Process (a,b)

It can be expressed as:

$$d\sigma_{a,b}(E_a) / dE_b = d\sigma_{a,b}^{SMD}(E_a) / dE_b + d\sigma_{a,b}^{SMC}(E_a) / dE_b \qquad (2.1)$$

The first term denotes the statistical multistep direct part (SMD). Besides particle-hole excitation, collective phonon excitations are also considered. The second term symbolizes the statistical multistep compound (SMC) emission, which is based on a master equation. Both terms together (SMD+SMC) represent the so-called first-chance emission process. In that case the following abbreviations are used:

E : the excitation energy of the composite system
= $E_a + B_a$,
E_a : the kinetic energy of the ingoing particle,
B_a : the binding energy of the ingoing particle, and
U : the excitation energy of the residual system
= $E - E_b - B_b$
= $E_a + B_a - E_b - B_b$
E_b : the kinetic energy of the outgoing particle,
B_b : the binding energy of the outgoing particle,

2.1.1 SMD Cross-section

This cross-section is a sum over s step direct processes,

$$d\sigma_{a,b}^{SMD}(E_a) / dE_b = \Sigma\, d\sigma_{a,b}^{s}(E_a) / dE_b \qquad (2.2)$$

with

$$s = N_p = N_h$$

N_p and N_h are particle and hole number.
Detailed description of the equations describing the processes are given in the code report.

2.1.2 SMC Cross-section

This cross-section has the familiar form (b= n, p, a, g):

$$d\sigma_{a,b}^{SMC}(E_a) / dE_b = \sigma_a^{SMC}(E_a) \Sigma ((\tau_N(E)/h) \Sigma G_{N,b}^{(DN)}(E,E_b)) - \quad (2.3)$$

where

$\sigma_a^{SMC}(E_a)$: acts as a normalization constant and is given by

$$\sigma_a^{SMC}(E_a) = \sigma_a^{OM}(E_a) - \Sigma \sigma_{a,c}^{SMD}(E_a) \quad (2.4)$$

$\sigma_a^{OM}(E_a)$: the optical model cross-section

$\tau_N(E)$ satisfies the time-integrated master equation

$$-h d\tau_{NN0} = G_{N-2}^{(+)}(E)^{-}\tau_{N-2}(E) + G_{N+2}^{(-)}(E)^{-}\tau_{N+2}(E) - G_N(E)\tau_N(E) \quad (2.5)$$

N is the exciton number

$= N_p + N_h$ (particle and hole numbers),

N_0 = 2,3

N = Ö1.4gE

g : is the single particle state density,

G_N^{-} : is the damping width.

2.2 Total Emission Spectra of the Process(a,xb)

In that case a third term is added to the two terms in equation (2.1) and the equation becomes

$$d\sigma_{a,xb}(E_a) / dE_b = d\sigma_{a,b}^{SMD}(E_a) / dE_b + d\sigma_{a,b}^{SMC}(E_a) / dE_b + d\sigma_{a,xb}^{MPE}(E_a) / dE_b \quad (2.6)$$

The last term, the so-called multiple particle emission (MPE) includes the second-chance, third-chance emissions. It can be summarized as

$$d\sigma_{a,xb}^{MPE}(E_a) / dE_b = \Sigma d\sigma_{a,cb}(E_a) / dE_b + \Sigma d\sigma_{a,cdb}(E_a) / dE_b + \ldots \quad (2.7)$$

In which case, for example, for a second-chance process (a,cb) and c ¹ g the following master equation has to be solved:

$$d\sigma_{a,cb}(E_a) / dE_b = \grave{o}\, dE_c (d\sigma_{a,c}(E_a) / dE_c) \Sigma (\tau_{N-1}(E_1)/h) .(\Sigma G_{N-1,b}^{(DN)}(E_1,E_b) -) \quad (2.8)$$

Here the master equation has to be solved for each intermediate excitation energy E_1, given by

$E_1 = E - E_c - B_c$

$= E_a + B_a - E_c - B_c$

The escape widths in the last equation are calculated using the residual excitation energy U, given by

$U = E_1 - E_b - B_b$

$= E_a + B_a - E_b - B_b - E_c - B_c$

2.3. Activation Cross-sections

The following (model-independent) relations between the optical model (OM) reaction cross-sections are satisfied (at each incident energy E_a)

$$\left.\begin{aligned} \sigma'^{OM}_{a} &= \Sigma\, \sigma'_{a,b} \\ \sigma'_{a,b} &= \Sigma\, \sigma'_{a,bc} \\ \sigma'_{a,bc} &= \Sigma\, \sigma'_{a,bcd} \end{aligned}\right\} \qquad (2.9)$$

with the total first-chance emission is given by

$$\sigma'_{a,b} = \sigma'^{SMD}_{a,b} + \sigma'^{SMC}_{a,b}$$

In this code, the activation cross-sections are given by

$$\left.\begin{aligned} \sigma'_{a,b\gamma} &= \sigma'_{a,b} - S\,\sigma'_{a,bc,} \\ \sigma'_{a,cb\gamma} &= \sigma'_{a,cb} - S\,\sigma'_{a,cbd} \end{aligned}\right\} \qquad (2.10)$$

where $b,c,d \neq \gamma$

For example, the (n,p) activation cross-section has the form

$$\sigma'_{n,p\gamma} = \sigma'_{n,p} - \sigma'_{n,pn} - \sigma'_{n,2p} - \sigma'_{n,p\alpha} \qquad (2.11)$$

2.4. General Features

The physical units are : energy in MeV, Length in fm, cross-sections in mb. Incident energies are in LS, emission energies in CMS. The emission energy bin width ΔE_b depends on the incident energy : ΔE_b = 0.2 MeV (for E_a < 25 MeV) ; ΔE_b = 0.5 MeV (for 25 MeV $\geq E_a <$ 50 MeV) ; ΔE_b = 1.0 MeV (for $E_a \geq$ 50 MeV), Incident energy bin widths ΔE_a used for excitation functions are taken as any multiple of ΔE_b.

In a calculation the following parameters can be changed : strength of residual interaction, radius parameter, Fermi energy, phonon width and the global Optical Model parameter set for protons. In addition the pairing shift can be modified. The pairing energy has the most influence on description of emission spectra.

The calculated cross-sections at 14-MeV neutron energy are listed in tables 1 through 7 for the isotopes of Ca, Fe, Cu, Zn, Sr, Ba and Pb respectively, along with the available experimental data (Molla, 1977; Pepelnik, 1985; Csikai, 1993; Avrigeanu, 1988; Bostan, 1991; Tan,1995; Filatenkov, 1997; Baba, 1996; Gledenov, 1997; Vonach, 1991; Ikeda, 1991a; Yamada, 1990; Nakajima, 1991; Ikeda, 1991b; Bhuiyan, 1995; Kasugai, 1995; Nakajima, 1996; Nesaraja, 1997; Garuska, 1991 and Takao, 1998). The results are reasonable within experimental errors with only very few exceptions (^{44}Ca(np), ^{54,57}Fe(np), ^{63}Cu(np), Zn isotopes). In fact, the experimental cross-sections for ^{57}Fe are discrepant among themselves (55mb, 135mb). The very low abundance does not allow for precise measurements.

3. INDUCED ACTIVITIES

The induced saturated activity (Khanchi,1984, 1988 and Aggarwal, 1991) in micro-Curies per unit neutron flux per gm of the biological samples containing $^{A}_{Z}X_{N}$ for the experimental investigations is given by the relation:

(0.693 x no. of atoms of $^{A}_{Z}X_{N}$ x $\sigma_{(n,p) \text{ or } (n,\alpha)}$ (cm^2) x isotopic abundance x elemental concentration in bone) / 3.7 x 10^4 $T_{1/2}$(seconds)

where

$^{A}_{Z}X_{N}$: the isotope under consideration whose proton number is Z, neutron number is N with A = N + Z atomic mass unit (amu)

And 1 amu = 1.6605 x 10^{-24} gm

$T_{1/2}$ = phys. half life of the residual nuclei formed in the (n,p) or (n,α) reaction. Roberto (1983) reported average values for the trace elements in bone. These are listed in table 8.

The induced activities for these trace elements have been listed in table 9 for all the (n,p) and (n,α) reactions including just the isotopes with appropriate half-lives for laboratory measurements.

These computed activities for bone in micro-Curies/gm/neutron flux may provide useful estimates for carrying out the experimental investigations (Khanchi, 1989) with 14 MeV neutron generators having fluxes of $10^8 – 10^{10}$ neutrons per cm^2 per sec.

Also these computed activities can be very useful in designing the experiment for estimating the irradiation time of such samples by activation analysis, for trace elemental analysis.

Table 1. Calcium Isotopes

Isotope	Isotopic Abundance %	(n,p) (mb)				(n,α) (mb)		
		Molla (1977)	Pepelnik (1985)	Csikai (1994)	Present Results	Pepelnik (1985)	Csikai (1994)	Present results
^{40}Ca	96.97			470	474			180.5
^{42}Ca	.64		187±8.2		232.4		138.1→147.9	142
^{43}Ca	.145		150±8.2		-			-
^{44}Ca	2.06	46±5	44±7.5		74.1	29±1.2		23.5
^{46}Ca	.0033				16.7			2.6
^{48}Ca	.185				0.1			-

Table 2. Iron Isotopes

Isotope	Isotopic Abundance %	(n,p) (mb)							(n,α) (mb)				
		Molla (1977)	Csikai (1994)	Avirgeanu (1988)	Ercan (1991)	Tan (1996)	Filatenkov (1997)	Present Results	Csikai (1994)	Avirgeanu (1988)	Baba (1996)	Gledenov (1997)	Present results
^{54}Fe	5.84		315±10	380 or 390	340(30)			570.6	88.5±6	80→90		80→85	56.2
^{56}Fe	91.68	98±7		115	123(6)	108.6±2.2	112(4)	122.6			40→45		52.6
^{57}Fe	2.17	55±4			135(10)			28.4					31.9
^{58}Fe	.31	7±1.5						-					-

Table 3. Copper Isotopes

Isotope	Isotopic Abundance %	(n,p) (mb)							(n,α) (mb)			
		Molla (1977)	Csikai (1994)	Ercan (1991)	Tan (1996)	Filatenkov (1997)	Vonach (1991)	Present Results	Csikai (1994)	Filatenkov (1997)	Ikeda (1991a)	Present results
^{63}Cu	69.1	125±50	54 87.4				54±4	81.9	51,45.2 61.4→73	45.3(.8)	43.8±2.5 40.4±2.3	65.8
^{65}Cu	30.9	27±2.3		14(5)	29.3±1.5	21.3(.7)		19.1				18.3

Table 4. Zink Isotopes

Isotope	Isotopic Abundance %	(n,p) (mb)											(n,α) (mb)			
		Molla (1977)	Pepelnik (1985)	Ercan (1991)	Tan (1996)	Yamada (1990)	Nakajima (1991)	Ikeda (1991b)	Bhuiyan (1995)	Kasugai (1995)	Nakajima (1996)	Nesaraja (1997)	Present Results	Csikai (1994)	Ercan (1991)	Garuska (1991)	Present results
^{64}Zn	48.89	160±12		140(40)				185→190	210		200		377.1				145.2
^{66}Zn	27.81	72±8		69(10)	69±8	72±18	76			≈60	77.5		21.3	18.4→27.7			21.8
^{67}Zn	4.11			68(7)							-	≈28	-				-
^{68}Zn	18.56	9.8±1.4	9.1±.7										28.1		11(7)	6→8	25.4
^{70}Zn	.62		2.9±.3										-				-

Table 5. Strontium Isotopes

Isotope	Isotopic Abundance %	(n,p) (mb)	(n,α) (mb)
		Present results	
^{84}Sr	.56		
^{86}Sr	9.86		
^{87}Sr	7.02		
^{88}Sr	82.56	6.4	1.8

Table 6. Barium Isotopes

Isotope	Isotopic Abundance %	(n,p) (mb)				(n,α) (mb)
		Pepelnik (1985)	Csikai (1994)	Filatenkov (1997)	Present Results	Present Results
^{130}Ba	.101					
^{132}Ba	.097					
^{134}Ba	2.42		6±2			
^{135}Ba	6.59					
^{136}Ba	7.81					
^{137}Ba	11.32		5±1		10	5.1
^{138}Ba	71.66	2.1±.2		2.21(0.1)	.9	1.3

Table 7.Lead Isotopes

Isotope	Isotopic Abundance %	(n,p) (mb)	(n,α) (mb)			
		Presnt Results	Csikai (1994)	Filatenkov (1997)	Takao (1998)	Present results
^{204}Pb	1.4	3.7				-
^{206}Pb	25.1	2.3	.57±.04	.47(.026)		-
^{207}Pb	21.7	2.4				-
^{208}Pb	52.3	0.1				-
Pb(n,He)					1→2.5 mb	

Table 8. Elemental concentration in bone

Element	Conc. Range (µg / gram)
Ca	(100 – 180) x 10^3
Fe	5 – 500
Cu	1 – 20
Zn	50 – 120
Sr	45
Ba	5 – 25
Pb	10 - 40

Table 9. Evaluated Induced Activities

Isotope	Nuclear Reaction	$T_{1/2}$ residual nucleus	Induced activity µCi/gram/flux	Isotope	Nuclear Reaction	$T_{1/2}$ residual nucleus	Induced activity µCi/gram/flux
^{40}Ca	np	1.28 x 10^9 y	(3.21-5.77)x10^{-19}	^{57}Fe	np	1.6 m	(0.63-63.30)x10^{-11}
	nα	35 d	(0.16-0.29)x10^{-8}		nα	-	-
^{42}Ca	np	12.36 h	(0.89-1.60)x10^{-9}	^{58}Fe	np	65 s	(0.31-31.50)x10^{-12}
	nα	269 y	(0.28-0.51)x10^{-14}		nα	3.55 m	-
^{43}Ca	np	22.3 h	(0.71-1.27)x10^{-10}	^{63}Cu	np	100 y	(0.32 -6.40)x10^{-17}
	nα	-	-		nα	5.271 y 10.5 m	(0.48 -9.70)x10^{-16} (0.13 -2.58)x10^{-10}
^{44}Ca	np	22.1 m	(0.18-0.32)x10^{-7}	^{65}Cu	np	2.52 h	(0.11 -2.23)x10^{-12}
	nα	1.83 h	(0.19-0.34)x10^{-10}		nα	1.5 m 13.9 m	(0.11 -2.14)x10^{-10} (0.12 -2.32)x10^{-11}
^{46}Ca	np	115 s	(0.12-0.21)x10^{-9}	^{64}Zn	np	12.7 h	(0.19 -0.45)x10^{-10}
	nα	5.4 m	(0.65-1.16)x10^{-11}		nα	-	-
^{48}Ca	np	6.8 s	(0.006-0.01)x10^{-7}	^{66}Zn	np	5.1 m	(0.60 -1.44)x10^{-9}
^{54}Fe	np	312 d	(0.76-76.80)x10^{-15}		nα	100 y	(0.16 -0.39)x10^{-16}
	nα	27.7 d	(0.21-21.60)x10^{-16}	^{67}Zn	np	61.9 y	-
^{56}Fe	np	2.579 h	(0.12-12.10)x10^{-10}		nα	-	-
	nα	-	-	^{68}Zn	np	31 s	(0.13 -0.33)x10^{-8}

Table 9. Evaluated Induced Activities (continued)

Isotope	Nuclear Reaction	$T_{1/2}$ residual nucleus	Induced activity µCi/gram/flux	Isotope	Nuclear Reaction	$T_{1/2}$ residual nucleus	Induced activity µCi/gram/flux
^{68}Zn	np	3.8 m	$(0.19\text{-}0.45)\times10^{-9}$	^{137}Ba	np	30.17 y	$(0.50\text{-}2.45)\times10^{-18}$
	nα	2.52 h	$(0.43\text{-}1.03)\times10^{-11}$		nα	-	-
^{70}Zn	np	5 s 47 s	$(0.29\text{-}0.69)\times10^{-10}$ $(0.31\text{-}0.73)\times10^{-11}$	^{138}Ba	np	32.2 m 2.9 m	$(0.13\text{-}0.66)\times10^{-12}$ $(0.14\text{-}0.72)\times10^{-11}$
	nα	18 s	-		nα	9.1 h 15.6 m	$(1.11\text{-}5.60)\times10^{-14}$ $(0.39\text{-}1.95)\times10^{-12}$
^{88}Sr	np	17.8 m	2.85×10^{-11}	^{204}Pb	np	3.77 y	$(2.40\text{-}9.60)\times10^{-19}$
	nα	10.7 y 4.48 h	0.25×10^{-16} 0.53×10^{-12}	^{206}Pb	np	4.2 m	$(1.23\text{-}4.90)\times10^{-12}$
^{134}Ba	np	2.062 y	$(0.94\text{-}4.70)\times10^{-18}$	^{207}Pb	np	4.77 m	$(0.94\text{-}3.77)\times10^{-12}$
	nα	-	-	^{208}Pb	np	3.05 m	$(0.15\text{-}0.61)\times10^{-12}$

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