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Photoneutron and Photonuclear Cross Sections According to Packed Cluster Model

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خلاصة

حسبت قطاعات البث النيوتروني بأشعة جاما للنظائر ثوريوم 232، ثيتونيوم 237، يلوتونيوم 239، يورانيوم 233، 234، 235، 237، من الحد الأدنى لطاقة التفاعل حتى 20 مليون أليكترون فولت بواسطة تجمع مكونات النواه فى حزم الكم الذاتى لموجة مكونات النواه. وقد حققت خواص حزم تجمع مكونات النواه عدم وجود انبعاث نيوتروني مزدوج من أشعة جاما فىْ نظيرى اليورانيوم 233، 234، وكذلك عدم وجود انبعاث نيوتروني انشطارى لهما. كما فسرت حزم تجمع مكونات النواه اسبقية البث النيوتروني والبث النيوتروني الانشطارى فى حالة يورانيوم 235 كما ظهر توافق واضح بين المنحنيات المحسوبة لتفاعلات البث النيوتروني والقيم التجريبية لقطاعات هذه التفاعلات.

Abstract

Photonuclear Gross sections have been estimated for ^{232}Th, ^{237}Np, ^{239}Pu, ^{233}U, ^{234}U, ^{235}U, ^{238}U in the energy range from threshold up to 20 MeV, by perturbation balance in Packed Cluster. The Packed Cluster (γ, f) and (γ, n) cross sections require complete absence of any (γ, 2n) or (γ, nf) cross sections for ^{233}U and ^{234}U as in experiment. It also explains the early (γ, n) and (γ, nf) reactions in ^{235}U.

Introduction

Experimental data shows that actinide isotopes capable of photofission have also photoneutron properties. Near photofission threshold, (γ, n) photoneutron emission starts for all photofission isotopes. This level is a level for subcluster disturbance of Packed Cluster "P.C.". For some photofission isotopes (γ, 2n) Cross section[4] occurs at collective order of P.C.. The present work hopes to explain the characteristics and location of different gamma reactions, as well as the (γ, n), (γ, 2n) photoneutron emission cross sections for photofission isotopes and why there is no (γ, 2n) for ^{233}U and ^{234}U from P.C. data.

Wave Equation of Packed Cluster

When a photon establishes P.C. quantum levels, the remaining e.m. energy from photon perturbs the two clusters of P.C.. The Hamiltonian can be put to describe the motion of the c.g. and relative to it.

$$H = ((-h^2/2\mu)\nabla_r^2 + V(r)) + ((-h^2/2(m_1+m_2)\nabla_R^2 + (e/c)((Z_1A_1+Z_2A_2/(m_1+m_2))\frac{h}{i}\nabla_R)+(e/c)((z_1/m_1)A_1 - (z_2/m_2)A_2)(\frac{h}{i})\nabla_r \quad (1)$$

where z_1e, z_2e and m_1,m_2 are charges and masses of both subclusters, c is light speed. The solution of unperturbed cluster relative motion in (1) is

$$\psi_i = R(r)\ Y^m(\theta', \emptyset') \quad (2)$$

where θ', $\emptyset'$ are the directions of "r" relative to arbitary coordinate system; and R(r) satisfy eq(3) where h= palnk const. ÷ 2π

$$(h^2/2\mu)(d^2/dr^2)+(2/r)-(s(s+1)/r^2))R(r) + E.R(r)= (-)\ V(r)\ xR(r) \quad (3)$$

The potential energy of spherical Harmonic oscillator of P.C. is(5)

$$V(r)= {}^1/_2\ \mu\ \omega_o^2\ r^2 \quad (4)$$

$$R(r)= N_s \emptyset^s_n\ Exp.\ (-\alpha_o\ r^2) \quad (5)$$

where, $\emptyset^s_m$ (r) is a polynomial with r^s as lowest degree.

$$N_s = (2(\mu w/h)^{3/2}/\Gamma(s+3/2))^{1/2}(\mu w/h)^{1/2s} \quad (6)$$

$$\alpha_o= \mu w/2h \quad (7)$$

For e.m. ray propagation along Ø=O, magnetic vector potential along θ = O and ψi of initial system is inclined at (α, β) relative to ray direction.

$$Y^m_s(\theta', \emptyset') =((4\pi/(2s+1))\sum_{m=-s}^{s} Y^m_s(\theta, \emptyset)Y^{m*}_s(\alpha, \beta) \quad (8)$$

Hence the solution relative to e.m. ray is :

$$\psi_i = ((4\pi/(2s+1))\, N_s\, r_s\, (\mathrm{Exp.}\,(-\alpha_o\, r_s^2))\, \psi_u \qquad (9)$$

$$\psi_u = \sum_{m=1}^{s} (2(N_s^m)\, (\cos m(\emptyset-\beta))\, f_s^m(\alpha)\, f_s^m(\theta)) + (N_s^o)\, f_s^o(\alpha)\, f_s^o(\theta) \qquad (10)$$

Perturbation by E.M. Wave

The Matrix defining perturbation for quantum change in space

$$H_{ki} = \int \psi_k^*\, H'\, \psi_i\, dT \qquad (11)$$

where ψ_k is normalised final function of free motion

$$\psi_k = k^{3/2} \sum(i)^{s_f}\, Y_{s_f}^m(k)\, Y_{s_f}^m(r)\, (J_{s_f}(\rho))/\rho^{1/2} \qquad (12)$$

$\rho = kr$, $\hat{k}$ has direction $(\pi/2, 0)$, $\hat{r}$ has the direction $(\theta, \emptyset)$

Time Change for Probability of State

The changes of states occurs by time varying coefficient. The probability from time varying coefficient is time rated to yield cross section for density of final energy states.

$$\rho(E)\, dE = n^2\, dn\, d\Omega \qquad (13)$$

$$E = h^2k^2 / 2\mu' = h\omega - E_{th} \qquad (14)$$

$$\text{or } \rho(E) = (\mu' n^3/h^2 k^2)\, d\Omega \qquad (15)$$

The Packed Cluster Levels

The lowest nucleon wave division state makes the coupling state for P.C.

$$V(r) = (1/2)\, \mu\, (E_w/6 \times 90\, h)^2\, (3\mu ch/E_w)^2\, m_n$$

$$= 1/2\mu\, m_n c^2\, (\pi/2x90)^2 \qquad (16)$$

The outer level s=3 of P.C. is assumed to carry neutrons only and hence it may separate during perturbation causing μ= 18. For μ= 18, the reaction levels s=2,3 are (V(r))= 5.1 MeV, 7.65 MeV. For μ=21.6 (unseparated neutron shell) (V(r)) = 6.12 MeV, 9.18 MeV.

The (γ, n) reaction occurs at unseparated neutron shell at threshold (γ,n)=6.12 MeV and the fission threshold occurs at (γ,f)=5.1 MeV for separated neutron shell. Both (γ, n) and (γ,f) occur at s=2 as reaction threshold.

The two products of (γ, nf) and (γ,2n) occur at collective order resolution of P.C.. The threshold of (γ,nf) is hence for neutron emission from unseparated shell and for final function order 4, the (γ,nf) threshold = 12.18 MeV from eq.(4) at s = 4. As neutron emission occurs from the of 5 levels with 18 nucleons. Hence the unit state for P.C. is $(1/2E_w)/ 3x90 = 1.72$ MeV= hw_0. The P.C. has a reduced mass μ =(2x3/5)18=21.6 unit. The separation of its clusters is $r_c = \pi/k = \pi c (h \times 3/E_w)$. Hence from eq(4), the perturbation level of P.C. is separated neutron shell, the collective order for (γ,2n) is at $s= (5x4)^{1/2}$ at separated neutron shell yielding (γ,2n) threshold= 11.4 MeV; from eq.(4) at separated shell.

Coordination of Packed Cluster Order and Strates

The Packed Cluster yields [6] all thermal neutron resonance levels in fissionable nuclei with thermal neutron cross sections. As for ^{233}U, the (γ,n) reaction threshold and the (γ,f) reaction threshold are equal, it follows that (γ,n) starts from one cluster and (γ,f) starts from the other cluster. Hence it is not possible at one threshold to have two neutrons emitted in collective manner. Hence (γ,2n) is absent in ^{233}U. For ^{234}U, the number of states for (γ,f) from subclusters 2,3 are equal to those for (γ,n) from subclusters 2,3 as in tables 1,2. Hence (γ,n) and (γ,f) should occur alternatively, one type from one subcluster and the other from the other cluster. Hence there is no (γ,n) simultaneously from both subclusters or there is no (γ,2n) is ^{234}U.

Also when neutron emission is not resloved at collective order of subclusters there is no (γ,nf) as well in ^{233}U and ^{234}U.

For ^{235}U the final state s= 4 at (γ,2n) and the number n of final probable states is n=2. Hence the (γ,nf) occurs from two separate cluster of order s=2 or from separated shell clusters at (γ,nf) threshold =11.4 MeV. It follows that (γ,n) occurs at s=2 with separated shell at threshold=5.1 MeV. For (γ,2n) two different states for two neutrons occur at unseparated neutron shell with threshold 12.18 MeV. As (γ,n) occurs from subcluster order s=2, the (γ,f) occurs from subcluster order s=2, at mass states before separation.

Photoneutron Cross Sections

The cross section for (γ,n) is for neutron final free motion from s=2. As perturbation occurs from charged part of subclusters, it can occur from photon resolution of cluster order s=2 or 3. But the perturbation cross cross section is for charged part of order s=2. For E of photon in MeV

$$\sigma(\gamma,n) = \mu ((z_1/m_1 - z_2/m_2)^2 + (4z_1z_2/m_1/m_2) \sin^2 (\pi E/2E_\Delta)) \quad (1.08726)$$

$$((n_2 u_2^3 \exp.(-u_2^{3.5}) + n_3 u_3^3 \exp.(-u_3^{3.5}))/E \text{ mb} \quad (17)$$

Photoneutron Pair Cross Section

$$\sigma(\gamma, 2n)= \mu (271.87 n^3 ((1-u/3)^2/ 1.5 + u^2 (4.6285 (1-2u/21)^2 + 0.174191 u^4 (1-3u/26)^2 + 0.001595 u^6) u^{1/2} \exp.(-u))/E \text{ mb} \quad (18)$$

The above cross section is for collective order of P.C. at final state of free motion of neutrons. The photon energy is E in MeV, μ =18 but μ= 21.6 for ^{235}U as explained for reaction thresholds.

Results and Discussion

The value of subcluster charges z_1 and z_2 are the same for (γ,f), (γ, n), (γ, 2n) and (γ, nf). The thresholds for these reaction are given in table 1 as calculated from eq.(16). Table 2 gives the (γ, f) data[4] for comparison of states for 233U and 234U.

Table 1: (γ, n) and (γ, 2n) photoneutron thresholds MeV and states

Isotope	n_2	$E_{th,2}$(γ, n)	n_3	$E_{th,3}$ (γ, n)	E_{th} (γ, 2n)	E_{th} (γ, nf)	n(γ, 2n)
^{233}U	8	5.1MeV	16	8.6MeV	----	-----	----
^{234}U	11	6.12	20	9.18	----	-----	----
^{235}U	11	5.1	20	7.65	12.18	11.4	2
^{236}U	14	6.12	21	9.18	11.4	12.18	2.5
^{237}Np	10	6.12	18	9.18	11.4	----	2.5
^{238}U	16	6.12	25	9.18	11.4	12.18	3.5
^{232}Th	22	6.12	35	9.18	11.4	12.18	5

Table 2: Photofission barriers in MeV and number of fission states[4]

Isotope	n_2	$E_{th,2}$	n_3	$E_{th,3}$	z_1	z_2
^{233}U	11	5.1	22	8.6	17	22
^{234}U	11	5.4	20	8.4	19	25
^{235}U	12	5.3	20	8.6	17	22
^{236}U	11	5.3	18	8.2	16	21
^{237}Np	12	5.2	20	7.9	17	22
^{238}U	12	5.3	18	8	15	20
^{232}Th	12	5.4	15	7.5	9	12

The equal states of ^{234}U in n_2 $(\gamma,n)= n_2(\gamma,f)$ and n_3 $(\gamma,n)= n_3(\gamma,f)$ at $s_f=1$ causes (γ,n) peak at 1 MeV deviation from threshold [4] in (γ,n) for ^{234}U.

In figures, the P.C. cross sections are given by-.- lines. It may be concluded that the P.C. can also adequately describe the (γ,n) and $(\gamma,2n)$ reaction cross sections. The Packed Cluster explains the absence of $(\gamma,2n)$ and (γ,nf) in ^{233}U and ^{234}U. It can also explain the changes of the relative location of (γ,f) and (γ,n) as well as the changes of relative location of $(\gamma,2n)$ relative to (γ,nf).

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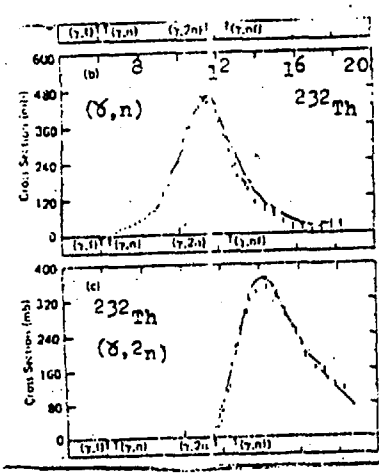
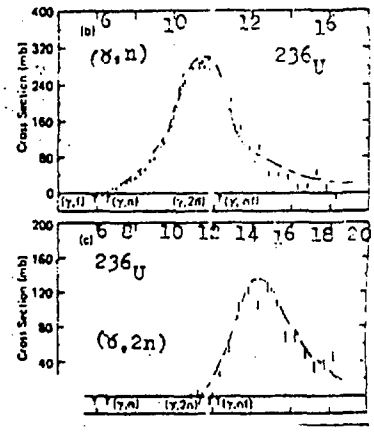

(b)
(γ,n)
232Th
(c)
232Th
(γ,2n)
Cross Section (mb)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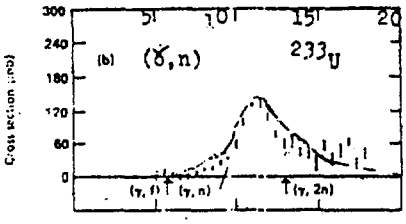
236U
(c)
236U
(γ,2n)
Cross Section (mb)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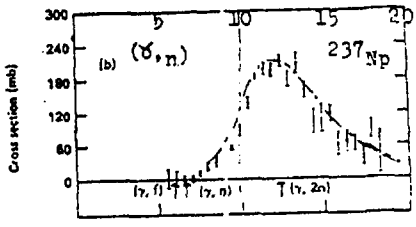
233U
Cross section (mb)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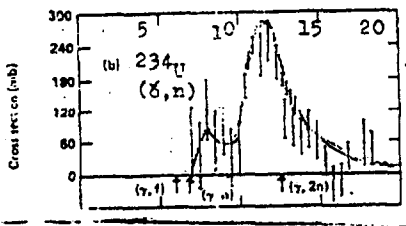
(γ,n)
237Np
Cross section (mb)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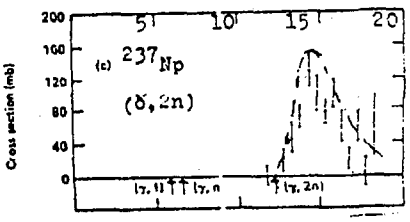
234U
(γ,n)
Cross section (mb)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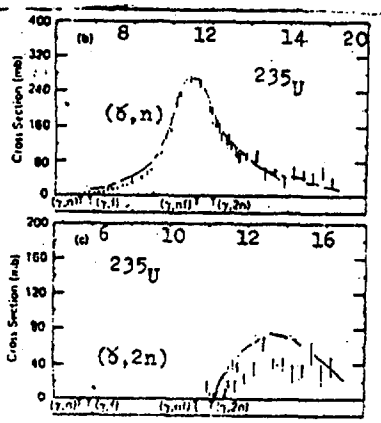
(γ,2n)
Cross section (mb)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235U
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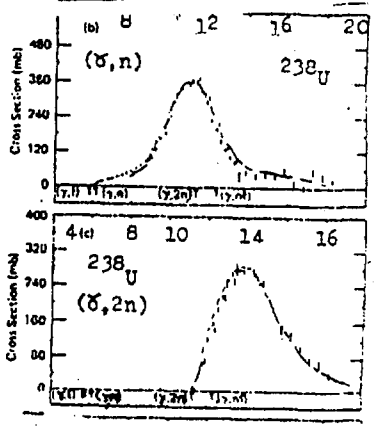
235U
(γ,2n)
Cross Section (mb)


(b)
(γ,n)
238U
(c)
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(γ,2n)
Cross Section (mb)

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The Application of Cerenkov Radiation in 14 MeV Neutron Activation for Determination of Chromium, Manganese, Iron and Cobalt

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خلاصة

تم استخدام اشعاع جرنكوف المتولد في خلية من الزجاج العضوي في التنشيط النيوتروني لقياس تراكيز العناصر ومنها الكروم ، المنغنيز ، الحديد والكوبلت . وذلك باستخدام مولد نيتروني وبطاقة تقرب من 14 م . أ . ف نوع Kaman 1003 موجود في قسم الكيمياء النووية ، جامعة لفبرا التكنولوجيه ، لفبرا ، المملكة المتحدة .

تم حساب العد لاشعاع جرنكوف للنويدات الباعثة لاشعة بيتا وكاما باستخدام قناة التريتيوم لجهاز العداد الوميضي السائل .

الاختلاف في عمر -نصف للنويدات المتولدة أثناء التنشيط النيوتروني وكذلك اختلاف الطاقات ، جعل من الممكن قياس تراكيز هذه العناصر في مزيجها باستخدام طريقة الامتصاص او طريقة الانحلال المزدوج .

اثبتت الطريقه زيادة في الحساسية وحدود الكشف مقارنة مع طريقة اطياف كاما التقليدية .

Abstrat

Cerenkov radiation produced in a perspex cell has been employed in 14 MeV neutron activation analysis of chromium, Manganese, Iron and cobalt, using a Kaman 1003 neutron generator fitted with a rotating target and capable of producing up to 10^{11} n.S^{-1} [located at the Nuclear Chemistry Department, Loughborough University of Technology, Loughborough, United Kingdom]. Cerenkov counting of β and γ -emitting radionuclide can be measured by counting in the tritium channel of liquid scintillation spectrometer.

Variations in β and γ -energies and half-lifes of the radionuclides, means that the determination of isotopes in mixtures is frequently possible either

by absorption techniques or on the basis of half-life using a double decay method. The method shows an increased sensitivity and detection limits for these elements compared with results obtained from conventional gamma spectral analysis.

Introduction

Cerenkov radiation produced when a charged particle passes through a medium with avelocity (V) greater than that of light, local polarization of the medium is produced along its path. The polarized molecules return to their quiescent state after passage of the charged particle with emission of light. It was shown theorictically by Frank and Tamm (1937)that this light is emitted in a cone of half- angle O in a forward direction to that of the direction of the travel of the particle when O is given by the equation:

$$O = Cos^{-1} (1/Bn)$$

n = refractive index of the medium

B = ratio of V/C where C is the velocity of light in vacuo.

It is clear that B must be greater than 1/n and there is therefore a threshold energy for the emission of Cerenkov radiation in a particular medium depending upon its refractive index.

Liquid scintillation counters are a suitable instrument to assay a β-emitter radio-nuclide such ^{14}C and tritium. The scintillation counting of tritium yields on an averange of 28 photons per disinteqration extending to a maximium of 90 whilst Cerenkov radiation for a β-emitter such ^{32}P (E max. 1.71 MeV) gives a mean value of 40 photons per disintegration. Therefore the tritium channel of a scintillation spectrometer is quite suit able for counting Cerenkov radiation many radioactive nuclides produced by first neutron irradiation are either β or β emitters are generally assessed by measurement of 0.51 meV gamma-radiation and β-emitters by measurement of associated gamma-radiation since pure β-emitters are relatively rare. Quantitative measurement of gamma radiation is not usually a chieved with high efficiency and measurement of Cerenkov radiation provided that the β-energy is suitably high can afford a much higher efficiency. β-emitter and electron released by the interaction of γ-radiation with medium are both capable of producing cerenkov radiation provided that the particle energy exceeds the threshold energy for the refractive index of the particular medium.

The 14 MeV neutron activation of chromium,manganese, iron and cobalt produces measurable amounts of radio isotopes by the nuclear reactions shown in Table (1).

Table (1) : Relevant nuclear reactions of chromunium, manganese, iron cobalt and the nuclear properties of the products

Nuclear Reaction	Radiation	Energy (MeV)	Half-life
$^{52}Cr(n,p)^{52}V$	β^-	2.73	3.76m
$^{55}Mn(n,a)^{52}V$		1.45	
$^{55}Mn(n,\gamma)^{56}Mn$	β^-	0.33	2.56h
		0.75	
		1.05	
		2.86	
$^{56}Fe(n,p)^{56}Mn$		0.845	
		1.81	
		2.12	
		2.52	
		2.66	
		3.39	
$^{56}Co(n,\gamma)^{m60}Co$	β^-	1.54	10.5m
		0.058	
	IT	1.332	

Experimental

A neutron generator fitted with a rotating target and capable of producing up to 10^{11} n.S^{-1} was employed for activation in this work.

All irradiated samples were counted using liquid scintillation spectrometer (tritium channel). The compounds irradiated were chromium oxide (CrO_3), manganese acetate $(CH_3\ COO)_2\ Mn.\ .4H_2O$, Ferric oxide Fe_2O_3, and cobalt nitrate $Co(NO_3)_2.\ 6H_2O$. Asuitable weight of each being dissolved in water and irrdiated in a small polythene vial (38 mm long, 8mm diameter) the volume of which was 0.8 ml. A neutron flux of the order $2X10^8$ n.S was used for all irradiation, the exact value being determined by the irradiation of a copper foil and subsequent measurement of the activity by the method described else where (1).

After irradiation the vial was mounted centrally between two cylinders of perspex (each 30 mm long, 26.5 mm diameter) bored with a central cylindrical hole (20 mm long, 10 mm diameter). The end of one cylinder had an annular hole 20 mm diameter and 2.5 mm deep bored in one end, so that the two fitted together to make a cylinder the same size as a standard scintillation vial. Samples were irradiated for 3 min. allowed to decay for 2 mins and then counted. The detection limit for each element defined as three times the standard deviation of the background count was determined.

Solutions of manganese & iron having the same density were then made up, irradiated as before and counted using a double counting technique. The first count was taken in the perspex cylinder but for the second count the small vial was surrounded by black paper to eliminate Cevenkov radiation occuring in the aqueous solution. The ratios of measured activities by the two methods was obtained for both manganese and iron.

Mixtures containing known and diferent amount of manganese & iron were irradiated with fast neutron and counted using double counting technique. In all cases counts were corrected to a 2 minute decay & 5 minute irradiation with neutron flux of $2X10^8$ $n.S^{-1}$. The amount of manganese & iron present have calculated from the simultaneous equations:

$$ax + by = c$$
$$dx + ey = f$$

where :

a = CPM/mg manganese (unscreened)
b = CPM/mg iron (unscreened)
c = CPM of manganese/iron mixture (nuscreened)
d = CPM/mg manganese (screened)
e = CPM/mg iron (screened)
f = CCPM of manganese/iron (screened)

x and y are the amounts (mg) of manganese and iron respectively in the mixtures.

Results and Discussion

Table 2 shows the detection limits obtained for chromium, iron, manganese and cobalt compared with the detection limits given by CUYPERS and CUYPERS (2) using classical gamma- spectral analysis from which it can be seen that there is a marked increase in sensitivity for chromium and manganese whilst the sensitivity for iron and cobalt is

slightly increased. The major activity produced by irradiation of manganese is due to the isotope ^{52}V (t 1/2=3.76 m) while irradiation of iron produced Mn (t 1/2 = 2.56 h). Analysis of a mixture of manganese and iron in the composition range of 19-83% mn showed a precision of 0.2-1.6% for manganese and 1.5-5.3% for iron.

Table (2) : Comparison of sensitivities of the technique used with those in literature for chromium, manganese, iron and cobalt

Isotope	Cts.obtained mg-1	Detection limit mg -counting (36)	Detection limit, mg Present work
Cr	1400	0.21	0.009
Mn	4226	0.49	0.003
Fe	84.8	1.3	0.142
Co	42	4.3	0.285

Table (3) shows the results of analysis mixtures of manganese and iron from which it can be ssen that the analysis of mixures can be achieved with reasonable accuracy.

Table (3) : Ratios of manganese and iron activities using a double decay technique under a flux of 2.01 x 108 n.cm-2. s-1 and irradiation time of 5 min

Element	CPM (mg-1) T_d = 2m	CPM (mg-1) T_d =20m	(CPM) T_d=20m / (CPM) T_d =2m
Mn	749.65	69.1	0.092
Fe	27.71	24.9	0.898

Conclusion

The method of Cerenkov counting in 14 MeV neutron activation analysis shows an increased and detection limits for chromium, iron, manganese and cobalt compared with results obtained from conventional gamma spectral analysis.

The extension of the use of Cerenkov radiation for irradiated samples previously reported (3), makes it possible to analyzed mixtures by simple variation in the technique. Provided that the isotopes produced during irradiation have significantly different radiation energies.

Refferences

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