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Charged Particles Emission in Fast Neutrons Processes on Mo Isotopes

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ABSTRACT

The Molybdenum nucleus, (protons numbers Z = 42 and mass A = 83-115) has 33 isotopes of which 7 natural (A = 92, 94, 95, 96, 98, 100) and four isomers. The first 6 natural isotopes are stable but the nucleus with A = 100 is unstable with the time of life of 7.8 x 10¹⁸ y. The isotopes with A = 100 is a fission product and it is used in medicine. Nuclear reactions induced by fast neutrons are of great interest for fundamental and applicative researches. For fundamental investigations fast neutron reactions are a source of new data on nuclear reaction mechanisms and structure of nuclei. For applications these reactions provide precise nuclear data for reactors technology (fission and fusion), processing of long lived nuclear waste, reprocessing of U and Th for transmutation and energy projects, accelerated driven systems (ADS), etc. Fast neutron cross sections data for charged particles emission are of interest also, because the accumulation of Hydrogen and Helium in the walls and vessels of nuclear facilities lead to the modification of their physical properties.

The following reactions ${}^{94}Mo(n,p){}^{94}Nb$ and ${}^{95}Mo(n,np){}^{94}Nb$ induced by fast neutrons were analyzed. Cross sections, isomers ratios, parameters of nuclear optical potentials were evaluated. The ${}^{94}Nb$ isotope can be found in the radioactive wastes. This nucleus is unstable, has a very large time of life ($T_{1/2} = 20300 \text{ y}$) and contributes to the low level geological activity of the environment due to the buried wastes.

OUTLINE

- **1. INTRODUCTION**
- 2. THEORETICAL BACKGROUND
- **3. COMPUTER CODES**
- 4. RESULTS
- **5. DISCUSSION**
- 6. CONCLUSIONS

1. INTRODUCTION - FAST NEUTRON ACTIVATION

GENERAL

Precise nuclear data for:

- Nuclear energy existing fission reactors ; future fusion reactors
- Long lived radioactive waste
- Reprocessing if U and Th (Transmutation and Energy)
- Accelerated Driven Systems (ADS)

Fast Neutrons with Emission of Charged Particles

- Neutrons Data for Structural Material needed to estimate gas production by (n,p) and (n,α) reactions

Tagged Neutrons

- Neutron Induced Processes (n,n') on Ca, Fe, CI, P, F, S

FAST NEUTRON REACTIONS – Mo Processes:

- ⁹⁴Mo(n,p)⁹⁴Nb, ⁹⁵Mo(n,np)⁹⁴Nb

- **1. INTRODUCTION FAST NEUTRONS**
- ACTIVATION
- COMPLEMENTARY TO ACTIVATION WITH SLOW NEUTRONS
- POSSIBILITY TO CREATE BETTER GAMMA EMITTERS COMPARED WITH SLOW NEUTRONS
- POSSIBILITY TO INVESTIGATE LARGE SAMPLE DUE TO:
 - PENETRATION DEPTH OF NEUTRONS
 - ESCAPE FROM SAMPLE OF EMITTED GAMMAS

PROCESSES

- (n,n'), (n,n' γ), (n,2n), (n,p), (n,np), (n, α), (n,n α) etc

DIFICULTIES

- HIGH BACKGROUND

METHODS OF BACKGROUND REDUCTION - NEUTRONS TAGGING

2. THEORETICAL BACKGROUNDS

- **Cross Sections Evaluations**
- -CS calculated with Talys
- Incident energy from threshold up to 25 MeV with the contribution of:
- Direct Processes -> DWBA
- Compound Processes >Hauser Feshbach Formalism
- Pre equilibrium >Two Component Exciton Model

Nuclear Potential (implemented in Talys) – Wood – Saxon (WS) with Real and Imaginary Type

- WS Components: Volume (V), Surface (S), Spin Orbit (SO)
- Potential Parameters: Obtained from Nuclear Data Processing
- Local with Real and Imaginary Part
- Global with Parameters by Koning Delaroche

Levels Density - Constant Temperature Fermi Gas Model

2. THEORETICAL BACKGROUNDS – HAUSER FESHBACH APPROACH

Cross section



Historically first HF expression

Hauser - Feshbach

 $W_{\alpha\beta}$ = Widths Fluctuation Factor (WFC)

WFC

- Indicates a correlation between the ingoing channel (incident) and outgoing channels
- At low energies (<1 MeV) WFC=1 no correlation between *in* and *out* channels
- Decreases slowly with the energy
- It is calculated by complicate procedures (ex Moldauer expression)

2. THEORETICAL BACKGROUNDS - ISOMER RATIO

 $R = \frac{Y^m}{Y^g}$ = Experimentally measured isomeric ratio

 Y_m, Y_g = Yields of isomeric and unstable ground states

General Expression

$$R = \frac{Y_m}{Y_g} = \frac{\int\limits_{E_{th}}^{E_m} N_0 \phi(E) \sigma_m(E) dE}{\int\limits_{E_{th}}^{E_m} N_0 \phi(E) \sigma_g(E) dE}$$

 E_{th} = Threshold energy of nuclear reaction

 $E_m =$ Maximal energy of incident gamma quanta

2. THEORETICAL BACKGROUNDS - ACTIVITY

$$A_{obs} = \frac{N\sigma\varphi\alpha\varepsilon}{\lambda} [1 - Exp(-\lambda t)] Exp(-\lambda T) [1 - Exp(-\lambda\Delta T)]$$

Where:

N = Number of atoms of the isotope of the element

- σ = cross section
- $a = \gamma$ -ray abundance
- φ = Neutron Flux
- **ε** = Detector efficiency
- λ = Decay constant
- t = Irradiation time
- T = Cooling time
- **ΔT = Counting time**

Yields and Activities – features not implemented yet in Talys

3. THEORETICAL BACKGROUNDS – TALYS

TALYS – Freeware soft working under LINUX – dedicated to nuclear reactions, fission and nuclear structure calculation

Possibility - to calculate inclusive and exclusive cross sections

Nuclear Reaction (binary) – X(x,y)Y

Inclusive cross section – including y particle from other open channels like $(x,ny), (x,2ny), \cdots$

Exclusive cross section – taking into account the y particle only from X(x,y)Y reaction

4. RESULTS.

FAST NEUTRON REACTIONS WITH CHARGED PARTICLES EMISSION CASE OF ${}^{94}Mo(n,p){}^{94}Nb$ Reaction ; $Q_{np} = -1.26$ MeV

Importance – Obtaining of ⁹⁴Nb Nucleus Natural Molybdenum: Mo Z = 42 Natural Isotopes of Mo with their abundance (%): - ${}^{92}Mo(14.65), {}^{94}Mo(9.19), {}^{95}Mo(15.87), {}^{96}Mo(16.67), {}^{97}Mo(9.58), {}^{98}Mo(24.29)$ - stables - ${}^{100}Mo(9.74)$ – Decay $\beta^{-}\beta^{-}$ to ${}^{100}Ru$

Density of Natural Mo:10.28 g/cm³ : Spin and Parity of ⁹⁴Mo: 0⁺

Isomers in ⁹⁴Mo(n,p)^{94m,g}Nb Reaction

Results on:

- Inclusive and Exclusive Cross Sections

 Production of isomer and ground states of ⁹⁴Nb (m and g states)

- Parameters of Nuclear Potentials in incident and exit channels

- Activities and other Concurrent Processes



4. RESULTS – ⁹⁴Mo(n,p)⁹⁴Nb





Processes

- Compound mechanism is dominant

-With the increasing of incident energy – multistep compound processes are enabled

-Direct processes can be neglected

States

- At low energies discrete states of residual nuclei can be important

- At higher energies – continuum states gives the main contribution to the cross sections (XS)

-Curve 4 from upper figure is the same with curve 3 from lower figure

4. RESULTS – ⁹⁴Mo(n,p)⁹⁴Nb – Comparison with Experimental Data



From literature

2 sets of experimental dataPoints (1), (2)

-Talys evaluation (3)

- Good agreement with Exp Data (1)

- Agreement obtained by variation parameters of optical potential

 Difference between experimental data – explained by the existence of many open channels with participation of protons

Good description of XS Data

- allow to evaluate angular distributions, isomer ratios and other physical values

4. RESULTS – ⁹⁴Mo(n,p)⁹⁴Nb – Angular Distributions



5 MeV

- Direct component has very low values

- Compound processes – from discrete and continuum states

14.1 MeV

- Compound processes are dominant – after 10 – 12 MeV they are coming from mutistep compound processes and from continuum states

- Still low direct component

- Angular correlations – necessary in the evaluation of different asymmetry effects observed in fast neutrons reactions

Chosen energies – possibility to measure in FLNP 14

4. RESULTS – ⁹⁴Mo(n,p)^{94 m, g}Nb – Isomer Ratio (IR)





IR

- Important for spin distribution investigation
- Isotopes production
- m isomer state; g ground state
- In our case 94 m, gNb
- m (Spin/Parity/HalfT/ Energy 3+ / 6.263 min / 41 keV
- g (Spin/Parity/HalfT 0+ / 2.03E03

IR calculation - most simple case

R

- point like target (no proton loss in target)

Formula

$$=\frac{Y_m}{Y_g}=\frac{\int\limits_{E_m}^{E_m}N_0\phi(E)\sigma_m(E)dE}{\int\limits_{E_m}^{E_m}N_0\phi(E)\sigma_g(E)dE}$$

Results – Protons energy – threshold -> 25 MeV Unit Flux – 1 IR = 3.27 ± 0.15 ¹⁵ 4. RESULTS – ⁹⁴Mo(n,p)^{94 m, g}Nb – IR Computer Modeling – 14.1 MeV



0

Real Target – finite thickness L00 – g – ground; L01 – m isomer

Angular Distribution

 $\frac{d\sigma}{d\Omega}(\theta) = p_1 + p_2 \cos^2(\theta) + p_3 \cos^4(\theta) + p_4 \cos^6(\theta)$

Angular Distribution

-Generation - Direct method - Solved Numerically

$$\frac{2\pi}{\sigma_{np}}\int_{0}^{\theta_{c}}\frac{d\sigma}{d\Omega}\sin\left(\theta\right)d\theta = r \Longrightarrow \theta_{c}, r \in [0,1), \theta \in [0,\pi)$$

	P ₁	p ₂	p ₃	P ₄	
L00	13•10 ⁻⁴ ±0.51 •10 ⁻⁸	-6•10 ⁻⁵ ±8.06 •10 ⁻⁷	2•10 ⁻⁵ ±1.97 •10 ⁻⁶	6.1•10 ⁻⁵ ±1.29 •10 ⁻⁶	
L01	12•10 ⁻⁵ ±1.27 •10 ⁻⁷	5•10 ⁻⁵ ±1.34 •10 ⁻⁶	5•10 ⁻⁵ ±1.30 •10 ⁻⁶	7•10 ⁻⁵ ±2.16 •10 ⁻⁶	

4. RESULTS – ⁹⁴Mo(n,p)^{94 m, g}Nb – IR Modeling – 14.1 MeV – Protons Spectra







Input Data and Simulated Results

- -100 000 events $E_{neutrons} = 14.1 \text{ MeV}$
- Flux = 1
- Maximal protons path (gp)- 1.168 g/cm²
- -Target thickness (gt) gt = 0.1 gp
- IR[teor] = 2.39
- IR[sim] = 2.37
- NL00 = 28577; NL01 = 67805
- Lost protons in target 3618

4. RESULTS – ⁹⁴Mo(n,p)^{94 m, g}Nb – IR Modeling – 14.1 MeV – Protons Spectra







Input Data and Simulated Results

- -100 000 events $E_{neutrons} = 14.1 \text{ MeV}$
- Flux = 1
- Maximal protons path (gp)- 1.168 g/cm²
- -Target thickness (gt) gt = gp
- IR[teor] = 2.39
- IR[sim] = 2.41
- NL00 = 18295; NL01 = 44067
- Lost protons in target 37638

4. RESULTS. CONCURENT PROCESSES

In the ⁹⁴Mo(n,p)⁹⁴Nb Reactions for Incident Neutrons from Threshold up to 20 MeV many channels are open

- 2 channels: ⁹⁴Mo(n,np)⁹³Nb (Q=-8.49 MeV) and ⁹⁴Mo(n,2n)⁹³Mo (Q=-9.67 MeV)



4. RESULTS. EXCLUSIVE REACTIONS. SECOND WAY OF ⁹⁴Nb PRODUCTION

⁹⁴Nb isotopes – obtained by ⁹⁵Mo(n,np)⁹⁴Nb reaction (Q= - 8.63 MeV) Realized an analyze of Inclusive and Exclusive Reactions Presented only the main Results on Exclusive Reactions



Concurrence between Direct and Compound Processes of Pre – Equilibrium origin mainly

CS close to ⁹⁴Mo(n,p)

High Energy part dominated by Direct Processes -> Increasing of High Energy Region

Isomer Ratio - Neutron Flux =1 R = $0.927 \div 0.019$ - Neutron Flux ~ $1/E^{0.9}$ R = $0.923 \div 0.018$

4. RESULTS.⁹⁴Nb PRODUCTION. CONCURENT PROCESSES

Concurrent Processes:

 ${}^{95}Mo(n,p){}^{95}Nb (Q = -0.1432 MeV);$ ${}^{95}Mo(n,2np){}^{93}Nb (Q = -15.857 MeV)$ Exclusive Processes for ${}^{95}Mo(n,p){}^{95}Nb$



Production of ⁹⁵Nb

Standard Talys Input – Acceptable Description of Experimental Data

Isomer Ratio

Neutron Flux =1 R = 0.4024 \oplus 0.025 Gam. Tr. E_y = 0.2356 MeV

(n,2np)-importance at 17-20 MeV



4. RESULTS. EXCLUSIVE REACTIONS. TALYS POTENTIAL PARAMETERS Cross Sections Evaluation Realized with Standard Input of Talys

Wood – Saxon Potential Parameters – ⁹⁴Mo(n,p)⁹⁴Nb n + ⁹⁴Mo channel

V [MeV]	r _v [fm]	a _v [fm ⁻¹]	W [MeV	r _w [fm]	a _w [fm ⁻¹]	V _{so} [MeV]	r _{vso} [fm]	a _{vso} [fm ⁻¹]
50.99	1.22	0.658	0.16	1.22	0.658	5.99	1.05	0.58

p + ⁹⁴Nb channel

V	r _v	a _v	W	r _w	a _w	V _{so}	r _{vso}	a _{vso}
[MeV]	[fm]	[fm ⁻¹]	[MeV	[fm]	[fm ⁻¹]	[MeV	[fm]	[fm ⁻¹]
61.94	1.215	0.664	0.13	1.215	0.664	6.03	1.043	0.59

4. RESULTS. EXCLUSIVE REACTIONS. TALYS POTENTIAL PARAMETERS Cross Sections Evaluation Realized with Standard Input of Talys

Wood – Saxon Potential Parameters – ⁹⁴Mo(n,p)⁹⁴Nb n + ⁹⁵Mo channel

V [MeV]	r _v [fm]	a _v [fm ⁻¹]	W [MeV	r _w [fm]	a _w [fm ⁻¹]	V _{so} [MeV]	r _{vso} [fm]	a _{vso} [fm ⁻¹]
51.27	1.215	0.664	0.16	1.215	0.664	5.99	1.044	0.59

p + ⁹⁵Nb channel

V	r _v	a _v	W	r _w	a _w	V _{so}	r _{vso}	a _{vso}
[MeV]	[fm]	[fm ⁻¹]	[MeV	[fm]	[fm ⁻¹]	[MeV	[fm]	[fm ⁻¹]
62.10	1.215	0.664	0.13	1.215	0.664	6.03	1.044	0.59

4. RESULTS. ACTIVITIES. ISOMER RATIO MEASUREMENTS

MODEL - REAL TARGET OF 1 CM X 1 CM X 0.1 CM DESINSITY – 10.28 g/cm³

Goal – Activity obtained in Experiment at Different Energies

⁹⁵ Mo(n,np) ^{94m} Nb		⁹⁵ Mo(n,np) ^{94g} Nb		⁹⁵ Mo	(n,p) ^{95m} Nb	⁹⁵ Mo(n,p) ^{95g} Nb		
E _n [MeV]	σ [mb	A _{obs} [decay]	σ [mb	A _{obs} [decay]	σ [mb	A _{obs} [decay]	σ [mb]	A _{obs} [decay]
14.1	5.76	1.86 · 10 ⁶	3.09	1.7 · 10 ⁻³	8.56	9818	6.95	7
20	61.8	2.00 · 10 ⁷	31.1	1.7 · 10 ⁻²	9.67	1109	48.5	47

 $\tau_{\rm m} = 6.263 {\rm m}; \ \tau_{\rm g} = 2.03 \cdot 10^6 {\rm y}$ $\tau_{\rm m} = 86.6 {\rm h}; \ \tau_{\rm g} = 34.975 {\rm d}$

Irradiation time = 3 min; Cooling time = 3 min; Counting time = 5 min

4. RESULTS. ACTIVITIES. ISOMER RATIO MEASUREMENTS

MODEL - REAL TARGET OF 1 CM X 1 CM X 0.1 CM DESINSITY – 10.28 g/cm³

Goal – Activity obtained in Experiment at Different Energies

⁹⁴ Mo(n,p) ^{94m} Nb		⁹⁴ Mo(n,p) ^{94g} Nb		⁹⁴ Mo(I	n,np) ^{93m} Nb	⁹⁴ Mo(n,np) ^{93g} Nb		
E _n [MeV]	σ [mb	A _{obs} [decay]	σ [mb	A _{obs} [decay]	σ [mb	A _{obs} [decay]	σ [mb]	A _{obs} [decay]
14.1	27.1	8.75 · 10 ⁶	9.48	6.4 · 10 ⁻³	6.95	0	6.95	0
20	22.2	7.17 · 10 ⁵	7.83	4.3 · 10 ⁻³	9.67	0	48.5	0

 $\tau_{\rm m} = 6.263 {\rm m}; \ \tau_{\rm g} = 2.03 \cdot 10^6 {\rm y}$ $\tau_{\rm m} = 16.13 {\rm y}; \ \tau_{\rm g} = {\rm stable}$

Irradiation time = 3 min; Cooling time = 3 min; Counting time = 5 min

Talys – for gamma and neutron induced reactions activities and yields calculations on sample target are not implemented Activities are evaluated by us – simple model of isomer ratios measurements

Results suggest the possibility of the isomer ratio measurements

5. DISCUSSIONS

Analyzed Nuclear Reactions Induced by Fast Neutrons for:

Fast Neutron Activation and Isomer Ratio Evaluation Reactions: ⁹⁴Mo(n,p)⁹⁴Nb and ⁹⁵Mo(n,np)⁹⁴Nb

For mentioned Processes were Evaluated with Talys:

- Cross Sections (Inclusive and Exclusive)
- Isomer Ratios
- Activities and Yields on Simple Modeled Measurements

Comparison with Existing Experimental Data

- using a Standard Talys Input the CS Evaluation are in well and / or acceptable agreement with Existing Experimental Data
- For many Evaluations like CS productions of isomer and isotopes and Isomer Ratios no experimental data
- Obtained New Nuclear Data on Potential and Level Densities Parameters

Cross Sections – by Talys

6. CONCLUSIONS

Talys – Rapid and Efficient Codes for Evaluation of Nuclear Data (Reaction Models ->Cross Section and Nuclear Structure)

Necessary a Better Description of Experimental Data in Analyzed Processes

Fast Neutrons – Obtained New Theoretical Evaluations on Isomer Production and Isomer Ratios - > New Measurements and Experiments

Improvement of Experimental and Theoretical Data for Tagged Neutrons Method Necessary to test New Elements

Necessary to use Computer Codes for Experiment Simulations – GEANT4 - Activities, Yields and other

Present Evaluations

- Experiment Proposal at JINR Dubna Facilities
- LNF JINR Dubna Facilities IREN, IBR-2
- LNR JINR Dubna Microtron MT-25

THANK YOU VERY MUCH

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