NUCLEAR DATA FOR CHARGED-PARTICLE MONITOR REACTIONS AND MEDICAL ISOTOPE PRODUCTION

EVALUATION OF REACTION CROSS SECTION DATA FOR A FEW MEDICALLY IMPORTANT RADIONUCLIDES

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Outline

- Introduction
- Scientific background
- Areas of investigation in CRP
- Positron emitters
- Research proposal
- Evaluation of ^{76}Br
- Conclusions

Introduction

Nuclear data needed for medical applications

Nuclear Structure Data

Nuclear Decay Data

Nuclear Reaction Data

Evaluated Nuclear Data

Purpose for the production of radionuclides

Diagnostics

Therapy

Theragnostics

Scientific Background

- **IAEA-TECDOC-1211**(Charged particle database for medical radioisotope production: diagnostic radioisotopes and monitor reactions)
- **IAEA-Technical report series -473** (Nuclear Data for the production of therapeutic radionuclides)
- **IAEA, INDC(NDS)-0591,0596** (Summary reports of meetings)

Nuclear Model Calculations

- EMPIRE
- TALYS
- STAPRE
- ALICE-IPPE

The nuclear model calculations is an integral part of evaluation

Scientific Background at GCU Lahore

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ELSEVIER

A comprehensive evaluation of charged-particle data for production of the therapeutic radionuclide ^{103}Pd

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^{103}Pd is an important radionuclide having a half-life of 16.99 d, which is suitable for internal radiation therapy, especially used for the treatment of prostate cancer. Its production in no-carrier-added form is done via charged-particle-induced reactions and the data are available in EXFOR library. We evaluated six charged-particle-induced reactions, namely ^{nat}Ag(p,x)^{103}Pd, ^{103}Rh(p,n)^{103}Pd, ^{103}Rh(d,2n)^{103}Pd, ^{100}Ru(α,n)^{103}Pd, ^{101}Ru(α,2n)^{103}Pd, and ^{102}Ru(^{3}He,2n)^{103}Pd process. In the first case, analysis was done up to about 100 MeV but in other cases only up to about 25 or 40 MeV. Furthermore, an evaluation of the data for the ^{nat}Ag(p,x)^{103}Ag process was also done since it may serve as a typical example for the ^{103}Ag→^{103}Pd precursor system. A statistical procedure supported by nuclear model calculations using the codes STAPRE, EMPIRE 2.19, and TALYS was used to validate and fit the experimental data. The recommended sets of data derived together with confidence limits are reported. The application of those data, particularly in the calculation of integral yields, is discussed. A comparison of the investigated routes from the viewpoint of practical applicability to the production of ^{103}Pd is given. Presently the ^{103}Rh(p,n)^{103}Pd reaction is the method of choice.

Scientific Background at GCU Lahore

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Evaluation of excitation functions of proton and deuteron induced reactions on enriched tellurium isotopes with special relevance to the production of iodine-124

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Cross-section data for the production of medically important radionuclide ^{124}I via five proton and deuteron induced reactions on enriched tellurium isotopes were evaluated. The nuclear model codes, STAPRE, EMPIRE and TALYS, were used for consistency checks of the experimental data. Recommended excitation functions were derived using a well-defined statistical procedure. Therefrom integral yields were calculated. The various production routes of ^{124}I were compared. Presently the ^{124}Te(p,n)^{124}I reaction is the method of choice; however, the ^{125}Te(p,2n)^{124}I reaction also appears to have great potential.

AREAS OF INVESTIGATION IN CRP

- Monitor reactions for charged particle beams
- Discrepancies and new directions in data for production of diagnostic gamma emitters
- Production of novel positron emitters (direct production and production via generator isotopes)
- Production of therapeutic isotopes (alpha emitters, electron and X-ray emitters)
- Decay data

Status of production data of positron emitters

Positron Emitter	Status
^{52}Fe, ^{55}Co,	Evaluation required
^{61}Cu, ^{66}Ga, ^{68}Ga	Evaluation required, re-measure and re-evaluate the positron intensity of ^{66}Ga
^{72}As, ^{73}Se, ^{76}Br	Evaluation required, re-evaluate the positron intensities
^{86}Y, ^{89}Zr	Evaluation required, re-evaluate the positron intensities
^{94m}Tc	Re-evaluate the positron intensity
^{110m}In	Include the production data
^{120}I	Evaluation required, re-evaluate the positron intensities

Selected positron emitters and nuclear reactions for their production

Positron Emitter	Nuclear Reaction
^{52}Fe	$^{55}Mn(p,4n)\,^{52}Fe$, $^{nat}Ni(p,x)^{52}Fe$, $^{52}Cr(^{3}He,3n)^{52}Fe$
^{55}Co	$^{58}Ni(p,a)\,^{55}Co$, $^{54}Fe(d,n)^{55}Co$, $^{56}Fe(p,2n)\,^{55}Co$
^{66}Ga	$^{66}Zn(p,n)\,^{66}Ga$, $^{63}Cu(a,n)^{66}Ga$
^{72}As	$^{nat}Ge(p,xn)\,^{72}As$
^{76}Br	$^{76}Se(p,n)^{76}Br$, $^{77}Se(p,2n)^{76}Br$, $^{75}As(a,3n)^{76}Br$
^{86}Y	$^{86}Sr(p,n)^{86}Y$, $^{85}Rb(a,3n)^{86}Y$, $^{88}Sr(p,3n)^{86}Y$
^{89}Zr	$^{89}Y(p,n)^{89}Zr$, $^{89}Yr(d,2n)^{89}Zr$
^{120}I	$^{120}Te(p,n)^{120}I$, $^{122}Te(p,3n)^{120}I$

Research proposal for the first year of CRP

1- $^{58}Ni(p,\alpha)^{55}Co$

2- $^{54}Fe(d,n)^{55}Co$

3- $^{56}Fe(p,2n)^{55}Co$

4- $^{66}Zn(p,n)^{66}Ga$

5- $^{63}Cu(\alpha,n)^{66}Ga$

6- $^{120}Te(p,n)^{120}I$

7- $^{122}Te(p,3n)^{120}I$

Evaluation Methodology of Charged Particle Data

- **Choice of charged particle induced reactions**
  (*target material, type and energy of charged particle, reaction cross section, competing reactions)*

- **Compilation of activation cross section data**
  (EXFOR, recent publications)

- **Scrutiny of data**
  (reliability, consistency, etc.)

- **Normalization of data**
  - extrapolation to 100 % enrichment of target material
  - adjustment of data to newly evaluated γ-ray intensities (NUDAT)
  - normalization of data to monitor cross section values given in IAEA-TECDOC-1211.

Evaluation Methodology (continued)

- Ratio of experimental to calculated data plotted as a function of energy, followed by a polynomial fitting (with errors as the weighting factor) to estimate the *f(E)*. Data beyond 3 σ limit were rejected. The 95 % confidence limits obtained for the *f(E)* were transformed back to cross section to estimate the uncertainty.

- Evaluation procedure repeated with all model calculations. Recommended data generated by averaging the normalized model calculations.

Evaluation Methodology (continued)

- Ratio of experimental to calculated data plotted as a function of energy, followed by a polynomial fitting (with errors as the weighting factor) to estimate the $f(E)$. Data beyond 3 σ limit were rejected. The 95 % confidence limits obtained for the $f(E)$ were transformed back to cross section to estimate the uncertainty.

- Evaluation procedure repeated with all model calculations. Recommended data generated by averaging the normalized model calculations.

Recently completed work related to CRP, ^{76}Br

Table 1
Investigated nuclear reactions for the production of ^{76}Br, Q-values and references.

Nuclear reaction	Q-value (MeV)	References
$^{76}Se(p,n)^{76}Br$	−5.74	Paans et al. (1980), Kovács et al. (1985), Levkovskij (1991), Hassan et al. (2004)
$^{77}Se(p,2n)^{76}Br$	−13.16	Janssen et al. (1980), Levkovskij (1991), Hassan et al. (2004), Spahn et al. (2009)
$^{78}Se(p,3n)^{76}Br$	−23.66	Levkovskij (1991), Spahn et al. (2009)
$^{75}As(^{3}He,2n)^{76}Br$	−3.95	Paans et al. (1980), Alfassi and Weinreich (1982), Misaelides et al. (1987)
$^{75}As(\alpha,3n)^{76}Br$	−24.5	Nozaki et al. (1979), Paans et al. (1980), Alfassi and Weinreich (1982), Hermanne et al. (1994)
$^{79}Br(p,4n)^{76}Kr\rightarrow^{76}Br$	−32.05	Dikšić et al. (1979)

Table 2
Nuclear reactions leading to the formation of different bromine impurities during the production of ^{76}Br.

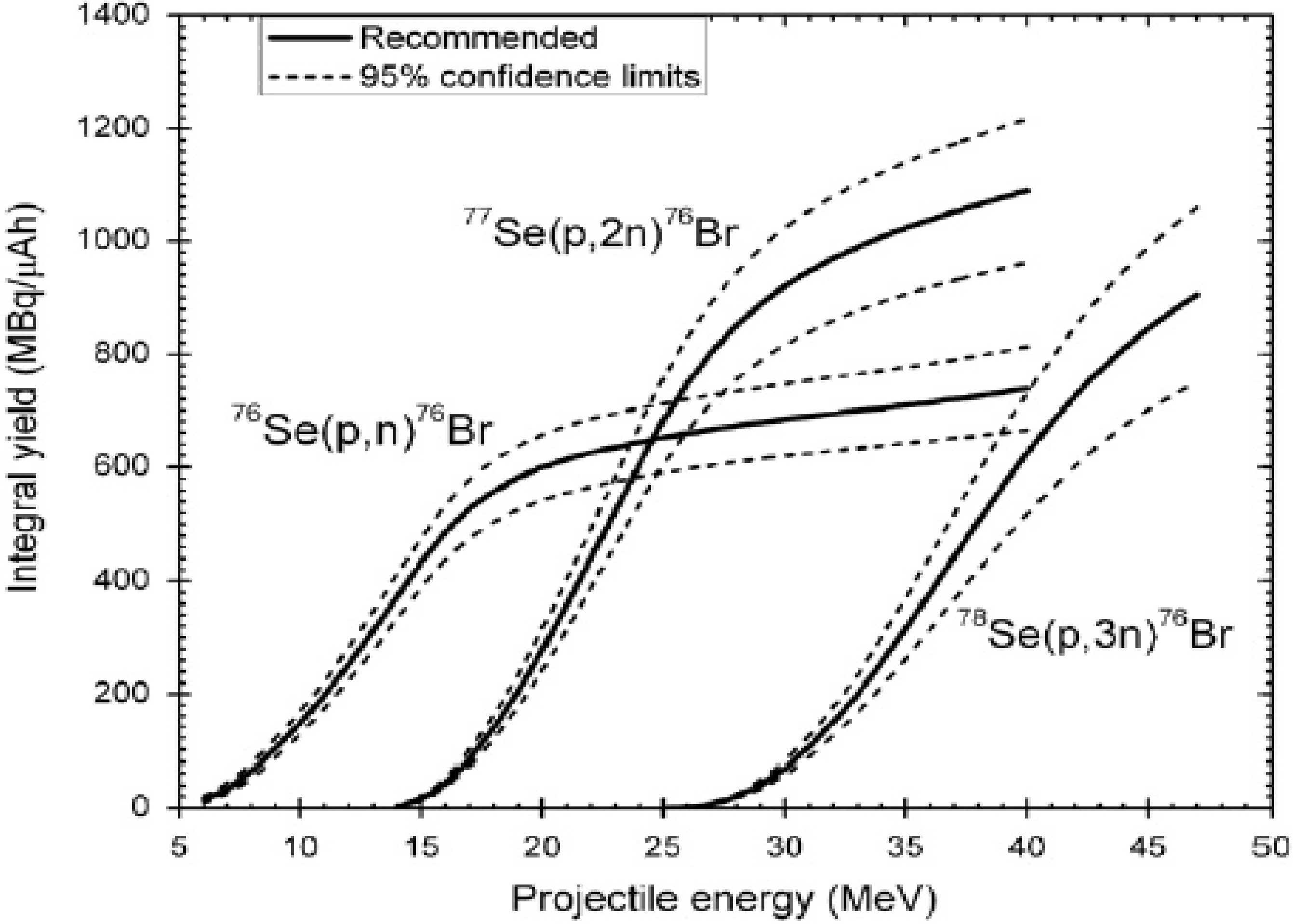
Nuclear reaction	Q-value (MeV)	References
$^{76}Se(p,2n)^{75}Br$	−14.97	Paans et al. (1980), Kovács et al. (1985), Levkovskij (1991), Hassan et al. (2004)
$^{77}Se(p,n)^{77}Br$	−2.15	Johnson et al. (1958), Fedorec et al. (1977), Janssen et al. (1980), Levkovskij (1991), Hassan et al. (2004), Spahn et al. (2010)
$^{77}Se(p,3n)^{75}Br$	−22.38	Levkovskij (1991), Spahn et al. (2009)
$^{78}Se(p,2n)^{77}Br$	−12.64	Janssen et al. (1980), Levkovskij (1991), Spahn et al. (2010)
$^{78}Se(p,4n)^{75}Br$	−32.88	Spahn et al. (2009)
$^{75}As(^{3}He,3n)^{75}Br$	−13.18	Paans et al. (1980), Alfassi and Weinreich (1982)
$^{75}As(\alpha,2n)^{77}Br$	−13.5	Waters et al. (1973), Nozaki et al. (1979), Alfassi and Weinreich (1982), Qaim et al. (1986), Hermanne et al. (1994)
$^{75}As(\alpha,4n)^{75}Br$	−33.7	Paans et al. (1980), Alfassi and Weinreich (1982), Hermanne et al. (1994)
$^{79}Br(p,3n)^{77}Kr\rightarrow^{77}Br$	−22.82	Dikšić et al. (1979), Levkovskij (1991)

Evaluated cross section data

Recommended cross sections for the $^{76}Se(p,n)^{76}Br$ reaction.

E (MeV)	σ (mb)	95% confidence limits		E (MeV)	σ (mb)	95% confidence limits	
		Upper	Lower			Upper	Lower
6.0	33	53	22	17.0	341	369	314
6.5	149	206	91	17.5	289	312	266
7.0	270	341	199	18.0	241	261	222
7.5	355	423	287	18.5	204	220	188
8.0	421	483	359	19.0	172	185	158
8.5	474	531	416	19.5	147	158	135
9.0	515	569	461	20	125	135	115
9.5	547	599	495	21	96	103	88
10.0	566	616	516	22	75	81	69
10.5	583	633	534	23	63	68	58
11.0	592	642	542	24	54	60	48
11.5	598	649	547	25	48	51	45
12.0	604	656	552	26	43	46	40
12.5	607	660	555	27	38	41	36
13.0	611	663	559	28	35	38	32
13.5	610	660	559	29	32	35	30
14.0	609	659	560	30	30	33	27
14.5	597	644	550	31	28	31	25
15.0	566	611	522	32	26	30	22
15.5	521	562	480	33	25	29	21
16.0	463	500	427	34	24	28	20
16.5	401	433	369	35	24	28	20

Calculated integral yields



Yield calculations of impurities

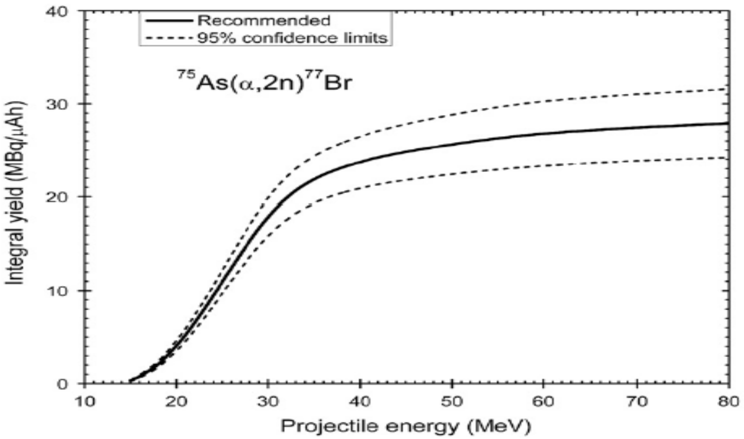


Fig. 25. Calculated integral yields for the ^{75}As(α,2n)^{77}Br reaction.

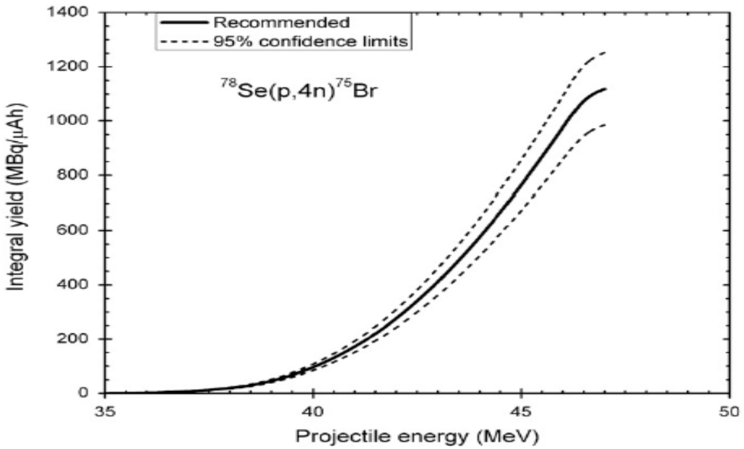


Fig. 27. Calculated integral yields for the ^{78}Se(p,4n)^{75}Br reaction.

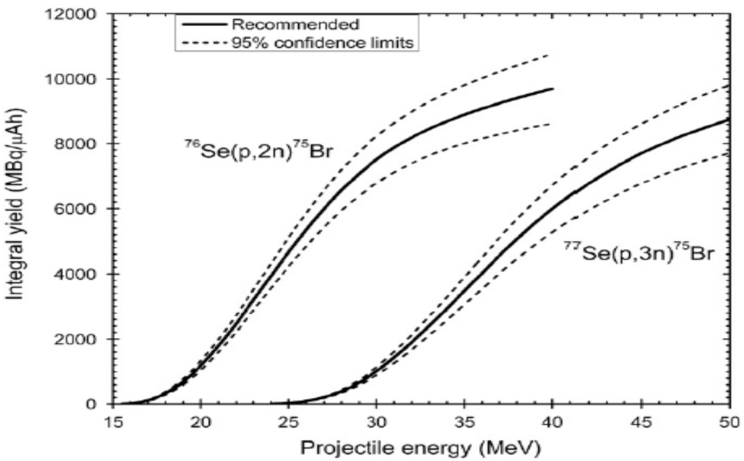


Fig. 26. Calculated integral yields for the ^{76}Se(p,2n)^{75}Br and ^{77}Se(p,3n)^{75}Br reactions.

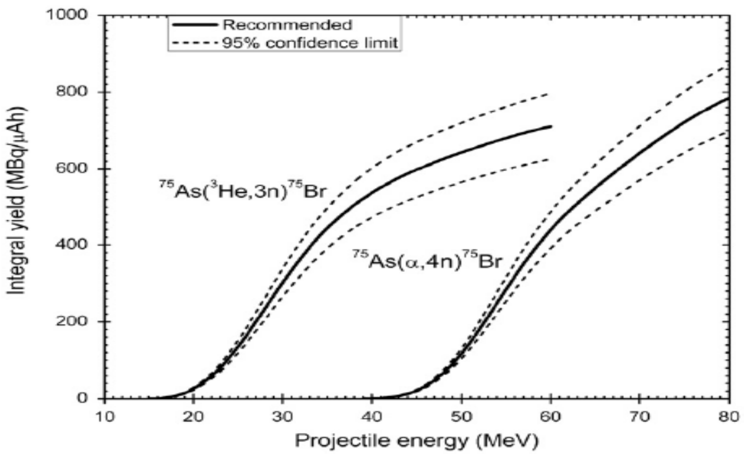


Fig. 28. Calculated integral yields for the ^{75}As(^{3}He,3n)^{75}Br and ^{75}As(α,4n)^{75}Br reactions.

Comparison of production routes

Comparison of production routes of ^{76}Br*.

Nuclear process	Energy range (MeV)	Thick target yield of ^{76}Br (MBq/μAh)			Radionuclidic impurities (%)	
		Suggested	Upper limit	Lower limit	^{77}Br	^{75}Br
^{76}Se(*p*,*n*)^{76}Br	15 → 7	402	437	366	< 0.1[a]	
^{77}Se(*p*,2*n*)^{76}Br	28 → 18	712	786	638	2.44	55
^{78}Se(*p*,3*n*)^{76}Br	42 → 32	577	675	479	8.46	47.7
^{75}As(^{3}He,2*n*)^{76}Br	20 → 11	12.8	14	11.7	1.2[b]	195
^{75}As(α,3*n*)^{76}Br	50 → 40	35.4	38.4	32.5	5.4	325
^{79}Br(*p*,4*n*)^{76}Kr → ^{76}Br	62 → 47	30[c]			< 0.1	

* All values calculated from the recommended excitation functions unless otherwise stated.

[a] Assuming the averaged (*p*,γ) reaction cross section of < 1 mb.

[b] Calculated from the measured cross sections of Alfassi and Weinreich (1982).

[c] Assuming a two step removal of radiobromine—first removal about 17 h after EOB, second removal 25 h thereafter. The yield of ^{76}Br and the impurity level correspond to the second fraction.

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Evaluation of excitation functions of proton, ^{3}He- and α-particle induced reactions for production of the medically interesting positron-emitter bromine-76

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ABSTRACT

Cross section data for production of the medically interesting radionuclide ^{76}Br ($T_{1/2}$=16.2 h) via the proton induced reactions on ^{76}Se, ^{77}Se, ^{78}Se and ^{79}Br, and ^{3}He- and α-particle induced reactions on ^{75}As were evaluated. The nuclear model codes STAPRE, EMPIRE and TALYS were used to check the consistency in the experimental data and a statistical procedure was applied to derive the recommended excitation functions. A comparison of various production routes of ^{76}Br (and of ^{75}Br) is presented.

Conclusions

- Methodology developed will be used to evaluate charged particle induced reaction cross section data over a wide energy range *(making use of a statistical procedure, supported by nuclear model calculations (EMPIRE, TALYS, ALICE-IPPE) to fit the data)*
- Method will be applied to evaluate several p, d, ^{3}He- and α-particle induced reactions used for the production of medically important radionuclides
- Recommended sets of data will be reported with 95 % confidence limits
- Data should be useful for optimization of production routes of the radionuclides at cyclotrons

List of few recent publications

- **M. Hussain, S. Sudár, M.N. Aslam, H.A. Shah, R. Ahmad, A.A. Malik, S.M. Qaim, A comprehensive evaluation of charged particle data for production of the therapeutic radionuclide ^{103}Pd. (Applied Radiation and Isotopes, 67, 1842-1854, 2009).**
- **M. Hussain, S. Sudár, M.N. Aslam, R. Ahmad, A.A. Malik, S.M. Qaim, Evaluation of charged particle induced reaction cross section data for production of the important therapeutic radionuclide ^{186}Re. (Radiochim Acta, 98, 385-395, 2010).**
- **M. Hussain, S. Sudar, M.N. Aslam, R. Ahmad, A.A.Malik, H.A. Shah, S.M. Qaim,Comprehensive evaluations of charged particle data for production of the therapeutic radionuclides ^{103}Pd, ^{186}Re and ^{67}Cu. (J. Korean Physical Society, 59, 1987-1990, 2011).**
- **M.N. Aslam, S. Sudár, M. Hussain, A.A. Malik, H.A. Shah, S.M. Qaim, Charged particle induced reaction cross section data for production of the emerging medically important positron emitter ^{64}Cu; A comprehensive evaluation. (Radiochim Acta, 97, 669-686. 2009).**
- **M.N. Aslam, S. Sudár, M. Hussain, A.A. Malik, H.A. Shah, S.M. Qaim, Evaluation of excitation functions of proton and deuteron induced reactions on enriched tellurium isotopes with special relevance to the production of iodine-124. (Applied Radiation and Isotopes, 68, 1760-1773, 2010).**
- **M.N. Aslam, S. Sudár, M. Hussain, A.A. Malik, S.M. Qaim, Evaluation of excitation functions of ^{3}He and α-particle induced reactions on antimony isotopes with special relevance to the production of iodine-124. (Applied Radiation and Isotopes, 69, 94-104, 2011).**
- **M.N. Aslam, S. Sudár, M. Hussain, A.A. Malik, S.M. Qaim, Evaluation of excitation functions of proton, 3He and α-particle induced reactions for production of the medically interesting positron emitter bromine-76. (Applied Radiation and Isotopes, 69, 1490-1505, 2011).**

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Evaluation Methodology of Charged Particle Data

- **Choice of charged particle induced reactions**
  (*target material, type and energy of charged particle, reaction cross section, competing reactions)*
- **Compilation of activation cross section data**
  (EXFOR, recent publications)
- **Scrutiny of data**
  (reliability, consistency, etc.)
- **Normalization of data**
  - extrapolation to 100 % enrichment of target material
  - adjustment of data to newly evaluated γ-ray intensities (NUDAT)
  - normalization of data to monitor cross section values given in IAEA-TECDOC-1211.

Evaluation Methodology (continued)

Comparison of normalized data with the results of nuclear model calculations (STAPRE, EMPIRE, TALYS) over a given energy range

(model parameters varied within recommended limits (RIPL-2) to get good description of experimental data)

Basic consideration $\sigma_{ev}(E) = f(E)\,\sigma_{model}(E)$

where $\sigma_{ev}(E)$, $\sigma_{model}(E)$ and $f(E)$ are the evaluated cross section, model calculated cross section and the energy dependent normalisation factor, respectively.

It appears a good assumption to approximate the $f(E)$ with a third order polynomial function. The ORIGIN code of the OriginLab Corporation was used for the fitting procedure.

Evaluation Methodology (continued)

- Ratio of experimental to calculated data plotted as a function of energy, followed by a polynomial fitting (with errors as the weighting factor) to estimate the *f(E)*. Data beyond 3 σ limit were rejected. The 95 % confidence limits obtained for the *f(E)* were transformed back to cross section to estimate the uncertainty.

- Evaluation procedure repeated with all model calculations. Recommended data generated by averaging the normalized model calculations.

- Thank you for your attention