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ICTP-IAEA Joint Workshop on Nuclear Data for Science and Technology: Medical Applications

30 September - 4 October, 2013

α, β+, γ and Auger-electron Decay Data in Nuclear Medicine – Experimental Determination, Status and Deficiencies Part II

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α, β⁺, γ and Auger-electron Decay Data in Nuclear Medicine – Experimental Determination, Status and Deficiencies Part II

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Workshop on Nuclear Data for Science and Technology: Medical Applications ICTP, Trieste, Italy, 30 September – 4 October 2013

Nuclear Data for Medical Applications

Rationale: Cancer therapy

Diagnosis



Beneficiaries: medical physicists, radioisotope producers, researchers ... **Objectives:** improve data for medical radioisotope production and patient dose delivery calculations in radiotherapy

ATOMIC AND NUCLEAR DATA SUPPORT FOR MEDICAL APPLICATIONS



- Production of radioisotopes for medical diagnosis and therapy
- Radiotherapy: photons, electrons, protons and heavy ions
- Brachytherapy combines radiotherapy with monoclonal antibodies
- Dosimetry and radiation safety
- Radiation transport data

Atomic and Nuclear Data

Nuclear structure and decay data

→ nuclear energy levels, half-lives and radioactive decay (α, β, γ) ENSDF MIRD DDEP

Atomic decay data

→ X-rays, Auger electrons

β^- decay of ¹³⁷Cs



β^- and β^+ decay

- β^{-} and β^{+} spectra:
- β emissions are NOT discrete even reasonably simple β spectra can be difficult if not impossible to analyse so as to quantify individual β transitions
- therefore, proposed decay schemes are commonly derived from measured γ and conversion-electron energies and emission probabilities
- ➤ balance population-depopulation of nuclear levels by total (γ + ce) transitions to provide the means of calculating β⁻ emission probabilities and (EC + β⁺) transition probabilities by difference

β^{-} and β^{+} decay

- β^{-} and β^{+} spectra:
- $(EC + \beta^{+}) \text{ transitions manipulated further to determine}$ $EC and <math>\beta^{+}$ components of their total (EC + β^{+}) transition probability to each specific nuclear level
- EC-to-positron ratios adopted from relevant tabulations, e.g.

N.B. Gove, M.J. Martin, Nuclear Data Tables, 10 (1971) 205-317

➢ would benefit from direct (and accurate) $P_{β+}$ measurements, despite inherent difficulties

Auger emitters and related ND needs

Auger, Coster-Kronig and super-Coster-Kronig
✓ low-energy (10 eV–10 keV) electrons – short range (few nm to 1 μm)
✓ commonly emitted by radionuclides that decay by EC, CE or IT

INVITED COMMENTARY

Cancer Therapy with Auger Electrons: Are We Almost There? Amin I. Kassis, PhD Harvard Medical School Boston, Massachusetts J. Nucl. Med. 44 (2003)

- high toxicity highly localized energy deposition in a small volume
- availability of many radionuclides with a wide range of physical halflives
- emission of gamma rays useful for imaging more than 100 Auger emitters most popular: ⁹⁹Tc^m, ¹¹¹In, ^{123,125}I, ⁶⁷Ga, ¹⁹³Pt^m new and emerging: ^{117,119}Sb, ¹⁶⁵Er

Medical applications - Auger electrons

Regaud and Lacassagne (1927) "The ideal agent for cancer therapy would consist of heavy elements capable of emitting radiations of molecular dimensions, which could be administered to the organism and selectively fixed in the protoplasm of cells one seeks to destroy."



Claudius Regaud (1870-1940)



Antoine Lacassagne (1884-1971)



Medical applications - Auger electrons

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Medical applications - Auger electrons

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"The ideal agent for cancer therapy would consist of heavy elements capable of emitting radiations of molecular dimensions, which could be administered to the organism and selectively fixed in the protoplasm of cells one seeks to destroy."

22-26 August 2011

Technical Meeting on Intermediate-term Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data

A.L. Nichols, et al., IAEA report INDC(NDS)-0596, September 2011

<u>Auger emitters:</u> ⁶⁷Ga , ⁷¹Ge, ⁷⁷Br, ^{99m}Tc, ¹⁰³Pd, ¹¹¹In, ¹²³I, ¹²⁵I, ¹⁴⁰Nd, ¹⁷⁸Ta, ¹⁹³Pt, ^{195m}Pt, ¹⁹⁷Hg



Atomic radiations - Basic concepts





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Atomic radiations - Basic concepts



Coster-Kronig electron





Initial vacancy



$$E_{L1L2M1} = E_{L1} - E_{L2} - E_{M1}^{L2}$$

L1 L2 M1 Coster-Kronig transition 2 new secondary vacancies

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Atomic relaxation and vacancy transfer

Vacancy cascade in Xe



- Full relaxation of an initial inner shell vacancy creates vacancy cascade involving X-ray (<u>radiative</u>) and Auger as well as Coster-Kronig (<u>non-radiative</u>) transitions
- Many possible cascades for a single initial vacancy
- Typical relaxation time ~10⁻¹⁵ seconds
 - Many vacancy cascades following a single ionisation event

Atomic radiations - Basic concepts

Vacancy cascade in Xe



Vacancies on the inner-shell can be produced by

- electron impact
- photo ionization
- ion-atom collision
- internal conversion
- electron capture
- secondary processes accompanying β-decay or electron capture

- X-rays and Auger electrons are an integral part of the radiations emitted in nuclear decay
- Atomic radiations are important for radioisotope applications (medical physics, nuclear astrophysics, nuclear engineering)
- ENSDF: atomic radiations are not included in ENSDF

Existing calculations Auger electron yield per nuclear decay

	RADAR	DDEP	Eckerman & Endo (2007)	Howell (1992)	Stepanek (2000)	Pomplun (2012)
⁹⁹ Tc ^m (6.007 h)	0.122	0.13	4.363	4.0		2.5
¹¹¹ In (2.805 d)	1.136	1.16	7.215	14.7	6.05	
¹²³ I (13.22 h)	1.064	1.08	13.71	14.9		6.4
¹²⁵ I (59.4 d)	1.77	1.78	23.0	24.9	15.3	12.2
²⁰¹ Tl (3.04 d)	0.773	0.614	20.9	36.9		
Vacancy propagation	Deterministic	Deterministic	Deterministic (+++)	Monte Carlo with charge neutralization	Monte Carlo	Monte Carlo

Existing programs

Common problems/limitations:

- Neutral atom binding energies are sometimes used for atoms with vacancies, i.e. for ions
- Single initial vacancy is considered. Secondary vacancies are ignored
- Atomic radiations <u>only</u> from primary vacancies on the <u>K and L shell</u>
- Limited information on <u>sub-shell</u> rates
- Auger electrons below ~ 1 keV are often omitted

BrlccEmis – Monte Carlo approach for vacancy creation and propagation

- Initial state: neutral isolated atom
- Nuclear structure data: from nuclear decay scheme evaluation (e.g. ENSDF)
- Electron capture (EC) rates: Schönfeld (1998Sc28)
- Internal conversion coefficients (ICC): Brlcc (2008Ki07)
- > Auger and X-ray transition rates: EADL (1991 Perkins)

Calculated for single vacancies!

Auger and X-ray transition energies: RAINE (2002Ba85)

Calculated for actual electronic configuration!

> Vacancy creation and relaxation from EC and IC: treated independently

- > <u>Ab initio treatment of the vacancy propagation:</u>
 - Transition energies and rates evaluated on the spot
 - Propagation terminated once the vacancy reaches the valence shell

BrlccEmis

- Reads the ENSDF file, evaluates absolute decay intensities of EC, GAMMA, CE and PAIR transitions
- Simulates a large number events: 100k-10M radioactive decays followed by atomic relaxation
- Electron configurations and binding energies stored in memory (and saved on disk). <u>New configurations only calculated if needed</u>

(⁵⁵Fe: 15 k, ²⁰¹Tl: 1300 k)

- Emitted atomic radiations stored on disk (Gb files)
- Separate files for X-rays and Auger electrons
- Smaller programs to sort/project energy spectra, and produce detailed reports

¹¹¹In EC decay – vacancy propagation



⁹⁹Tc^m atomic radiations



BrIccS v2.3 (9-Dec-2011)						
Z=43 (Tc, Technetium)						
γ-energy	: 2.1726 k	æV				
Data Sets: BrIccFO						
Shell	E(ce)	E3				
Tot		1.370E10 (20)				
M1	1.63	2.26E6 (4)				
M2	1.73	3.37E9 (5)				
M3	1.75	5.98E9 (9)				
M4	1.92	1.100E9 (16)				
M5	1.92	1.655E9 (24)				
M-tot	1.78	1.211E10 (17)				
N1	2.10	5.00E5 (7)				
N2	2.13	4.92E8 (7)				
N3	2.14	8.77E8 (13)				
N4	2.17	9.11E7 (13)				
N5	2.17	1.350E8 (19)				
N-tot	2.14	1.596E9 (23)				
01	2.17	3.49E4 (5)				
O-tot	2.17	3.49E4 (5)				

⁹⁹Tc^m atomic radiations – X-rays

	DDEP	BrIccEmis
Kα ₁	18.3672 4.21E-2	18.421 4.05E-2
Kα ₂	18.251 2.22E-2	18.302 2.13E-2
κβ	20.677 1.30E-2	20.729 1.18E-2
L	[2.134 - 3.002] 4.82E-3	2.466 4.72E-3
Μ		0.263 7.83E-4
Ν		0.047 8.73E-1

⁹⁹Tc^m atomic radiations – Auger electrons

	DDEP	BrIccEmis
KLL	[14.86:15.58] 1.49E-2	15.37 1.48E-2
KLX	[17.43:18.33] 2.79E-3	17.85 5.58E-3
КХҮ	[19.93:21.00] 2.8E-4	20.27 5.07E-4
K-total	2.15E-2	16.15 2.08E-2
CK LLM		2.08E-2 0.054
CK LLX		0.144 9.48E-3
LMM		2.016 9.02E-2
LMX		2.328 1.41E-2
LXY		2.654 6.07E-4
L-total	[1.6:2.9] 1.089E-1	1.765 1.24E-1

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⁹⁹Tc^m atomic radiations – Auger electrons

	DDEP	BrIccEmis
СК ММХ		0.104 7.10E-1
MXY		0.170 1.10E+0
Super CK NNN		0.014 5.36E-1
CK NNX		95% 0.012 8.45E-1
Total yield Auger electron per nuclear decay	0.13	w 500 eV 3.37

KLL-Auger spectrum of ¹¹¹Cd from EC decay of ¹¹¹In



KMM-, KMN- and KNN-Auger spectrum of ¹¹¹Cd from EC decay of ¹¹¹In



¹¹¹In EC decay – experiment vs calculation



E.A. Yakushev, et al., Appl. Radiat. Isot. 62 (2005) 451

- ESCA; FWHM = 4 eV
- Calculations normalized to the strongest experimental line

New model: ⁵⁵Fe EC decay

- primary vacancies from BrIcc (for CE) and EC (for electron capture)
- Binding energies calculated using RAINE code for electron configurations of a neutral atom or an ion with single or multiple vacancies
- Auger and CK transition rates calculated using non-relativistic perturbation theory with screened hydrogen wave functions (simple general formalism for all shells and transitions)
- Partially Defrosted Orbitals: calculations for all electronic configurations (complete treatment), but with singlevacancy transition rates





Slide courtesy of Kalman Robertson

Medical Internal Radiation Dose (MIRD)

- "The Program RADLST" documentation of the calculational methods and auxiliary data to quantify dose rates: T.W. Burrows, Brookhaven National Laboratory Report BNL-NCS-52142, February 1988
- 2. Access software at:

www.nndc.bnl.gov/mird/ www-nds.iaea.org/medportal/

Medical Internal Radiation Dose (MIRD)

MIRD	dose rate	data for	¹⁸ F as	determined	from	ENSDF	decay data
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Half-life of 109.77	min		November 1996			
β^+/EC decay mode		¹⁸ O daughter is stable				
Radiation	P _i (Bq-s) ⁻¹	Energy (MeV)	P _i ×E _i			
β_1^+	9.67×10 ⁻⁰¹	2.498×10 ^{-01*}	2.42×10 ⁻⁰¹			
γ^{\pm}	1.93	5.110×10 ⁻⁰¹	9.89×10 ⁻⁰¹			
K X-ray	1.80×10 ⁻⁰⁴	5.249×10 ⁻⁰⁴ *	9.42×10 ⁻⁰⁸			
K X-ray	4.74×10 ⁻¹²	5.000×10 ⁻⁰⁴ *	2.37×10 ⁻¹⁵			
Auger-K	3.07×10 ⁻⁰²	5.200×10 ⁻⁰⁴ *	1.60×10 ⁻⁰⁵			
Listed X, γ and γ^{\pm} rates	adiations		9.89×10 ⁻⁰¹			
Listed β , ce and Auger radiations			2.42×10 ⁻⁰¹			
Listed radiations			1.23			

* Average energy (MeV).

Medical Internal Radiation Dose (MIRD)

Half-life of 45.59 min	209 Tl daughter is radioactive α Novem		
α and β^- decay modes	²¹³ Pc	o daughter is radioactive	β ⁻ April 2007
Radiation	P _i (Bq-s) ⁻¹	Energy (MeV)	P _i ×E _i
α_1	1.55×10 ⁻⁰³	5.549	8.58×10 ⁻⁰³
α recoil	1.55×10 ⁻⁰³	1.053×10 ⁻⁰¹	1.63×10 ⁻⁰⁴
α_2	1.94×10 ⁻⁰²	5.869	1.14×10 ⁻⁰¹
α recoil	1.94×10 ⁻⁰²	1.114×10 ⁻⁰¹	2.16 ×10 ⁻⁰³
γ_1	1.27×10 ⁻⁰⁴	8.680×10 ⁻⁰¹	1.11×10 ⁻⁰⁴
β_3^-	5.86×10 ⁻⁰³	9.080×10 ^{-02*}	5.32×10 ⁻⁰⁴
β_{8}^{-}	3.07×10 ⁻⁰¹	3.204×10 ^{-01*}	9.85×10 ⁻⁰²
β-9	2.29×10 ⁻⁰³	3.768×10 ^{-01*}	8.62×10 ⁻⁰⁴
β^{-}_{10}	6.58×10 ⁻⁰¹	4.922×10 ^{-01*}	3.24×10 ⁻⁰¹
γ_1	4.29×10 ⁻⁰³	2.928×10 ⁻⁰¹	1.26×10 ⁻⁰³
Υ ₁₇	5.20×10 ⁻⁰⁴	1.119	5.82×10 ⁻⁰⁴
$K_{\alpha 1}$ X-ray	1.83×10^{-02}	7.929×10 ⁻⁰²	1.45×10 ⁻⁰³
$K_{\alpha 2}$ X-ray	1.10×10 ⁻⁰²	7.686×10 ⁻⁰²	8.45×10 ⁻⁰⁴
K_{β} X-ray	8.27×10 ⁻⁰³	8.980×10 ^{-02*}	7.43×10 ⁻⁰⁴
L X-ray	1.55×10 ⁻⁰²	1.110×10 ^{-02*}	1.72×10 ⁻⁰⁴
Listed X, γ and γ^{\pm} radiations			1.22×10 ⁻⁰²
Omitted X. γ , and γ^{\pm} radiations ^{**}			1.48×10^{-05}
Listed β , ce and Auger radiations			4.24×10 ⁻⁰¹
Omitted β , ce, and Auger radiations ^{**}			7.74×10 ⁻⁰⁴
Listed α radiations			1.22×10 ⁻⁰¹
Listed radiations			5.58×10 ⁻⁰¹
Omitted radiations**			7.89×10 ⁻⁰⁴

MIRD dose rate data for ²¹³Bi as determined from ENSDF decay data

* Average energy (MeV).

Alan Nichöl**E** ach omitted transition contributes < 0.100% to $\Sigma(P_i \times E_i)$.

Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data

Relevant recent past:

- Cross sections IAEA-NDS CRP 1995–2000: Charged Particle Cross-Section Database for Medical Radioisotope Production: Diagnostic Radioisotopes and Monitor Reactions, IAEA-TECDOC-1211, May 2001
- Cross sections IAEA-NDS CRP 2003–2010: Nuclear Data for the Production of Therapeutic Radionuclides, IAEA Technical Reports Series No. 473, IAEA, Vienna, Austria, December 2011
- 3. Decay data IAEA-NDS CRP 1998-2005: Update of X Ray and Gamma Ray Decay Data Standards for Detector Calibration and Other Applications, IAEA-STI/PUB/1287, published as 2 volumes, May 2007
- 4. Decay data IAEA-NDS CRP 2005-2010: **Updated Decay Data Library for Actinides**, prepared in draft, IAEA, Vienna, Austria, to be published

Immediate past and future:

Cross sections and decay data – one further IAEA-NDS CRP launched in late 2012 on the basis of

- **High-Precision Beta-Intensity Measurements and Evaluations for Specific PET Radioisotopes** (see IAEA report INDC(NDS)-0535, 2008)
- Improvements in Charged-Particle Monitor Reactions and Nuclear Data for Medical Isotope Production (see IAEA report INDC(NDS)-0591, 2011)

Consultants' Meeting, IAEA Headquarters, Vienna, Austria 3-5 September 2008, IAEA report INDC(NDS)-0535

Tadashi Nozaki	ex-RIKEN, Japan			
Syed Qaim	Forschungszentrum Jülich, Germany [Chairman			
Deon Steyn	iThemba Laboratory, South Africa			
Stephen Waters	ex-Cyclotron Unit, Hammer	smith Hospital, UK		
Roberto Capote	IAEA Nuclear Data Section	[Scientific Secretary]		
Alan Nichols	IAEA Nuclear Data Section	[Rapporteur]		

Consultants' Meeting, IAEA Headquarters, Vienna, Austria 3-5 September 2008, IAEA report INDC(NDS)-0535

Radionuclides – standard β^+ emitters	Requirements				
	t _{1/2}	<i>P</i> _β +	P _X	Ργ	evaluate
¹ C, ¹³ N, ¹⁵ O, ¹⁸ F none - well-defined decay data		ata			
⁶⁸ Ge/ ⁶⁸ Ga, ⁸² Sr/ ⁸² Rb	none - wel	ll-defined	⁶⁸ Ga an	d ⁸² Rb d	decay data
Radionuclides – hadron therapy					
¹⁰ C, ¹⁴ O, ¹⁷ F, ¹⁸ Ne, ¹⁹ Ne	nc	one – ade	quate d	ecay da	ta

Consultants' Meeting, IAEA Headquarters, Vienna, Austria

3-5 September 2008, IAEA report INDC(NDS)-0535

Radionuclides – non-standard $oldsymbol{eta}^+$ emitters		Requirements				
	<i>t</i> _{1/2}	P_{β^+}	P _X	Pγ	evaluate	
⁵⁷ Ni			\checkmark		\checkmark	
⁶⁶ Ga, ⁷² As, ⁷³ Se, ⁸⁶ Y, ⁹⁴ Tc ^m					\checkmark	
⁷⁵ Br, ⁷⁷ Kr					(√)	
⁶⁴ Cu				√ 1345.8-keVγ	(√)	
⁷⁶ Br, ¹²⁰ I					\checkmark	
⁸¹ Rb, ⁸² Rb ^m , ⁸³ Sr	inac	curately	defined d	ecay data	\checkmark	
²² Na, ³⁰ P, ³⁴ Cl ^m , ³⁸ K, ⁴⁵ Ti, ⁴⁸ V, ⁴⁹ Cr, ⁵¹ Mn, ⁵² Mn, ⁵² Mn ^m , ⁵² Fe, ⁵⁵ Co, ⁶¹ Cu, ⁹⁰ Nb, ¹¹⁰ In ^m , ¹²⁴ I, ¹⁵² Tb, ⁴⁴ Ti/ ⁴⁴ Sc, ⁶² Zn/ ⁶² Cu, ¹⁴⁰ Nd/ ¹⁴⁰ Pr		reasona	bly well-d	efined decay d	ata	

most important non-standard radionuclides for PET (2008).

increasingly important for PET (2008).

merit further work for potential PET applications (2008).

NUCLEAR DATA FOR CHARGED-PARTICLE MONITOR REACTIONS AND MEDICAL ISOTOPE PRODUCTION First Research Coordination Meeting, 3-7 December 2012 IAEA Report INDC(NDS)-0630

Marie-Martine Bé Brett Carlson Mazhar Hussain Anatoly Ignatyuk Guinyun Kim Filip Kondev Ondrej Lebeda Aurelian Luca Yasuki Nagai Haladhara Naik Alan Nichols Meiring Nortier Ingo Spahn Ferenc Tarkányi Laboratoire National Henri Becquere Instituto Tecnologico de Aeronautica, Government College University, Laho Institute for Physics and Power Engin Kyungpook National University, Repu Argonne National Laboratory, USA Nuclear Physics Institiute, Academy o National Institute of Physics and Nuc Osaka University, Japan Bhabha Atomic Research Centre, Indi University of Surrey, UK Los Alamos National Laboratory, USA Forschungszentrum Jülich, Germany Hungarian Academy of Sciences, Hun

Roberto Capote

IAEA Nuclear Data Section



Cross sections	Decay data	Additional comments
monitor reactions		
²⁷ Al(p,x) ^{22,24} Na	_	re-evaluate up to 800 MeV
$^{27}Al(d,x)^{22,24}Na$		re-evaluate
²⁷ Al(³ He,x) ^{22,24} Na		re-evaluate
$^{27}Al(\alpha,x)^{22,24}Na$		measure and re-evaluate
^{nat} Ti(d,x) ⁴⁶ Sc	_	re-evaluate
^{nat} Ti(³ He,x) ⁴⁸ V		measure and re-evaluate up to 46 MeV
$^{nat}Ni(d,x)^{56,58}Co$	_	measure and re-evaluate
^{nat} Cu(p,x) ⁵⁸ Co	_	measure and evaluate up to 100 MeV
$^{nat}Cu(p,x)^{62,63,65}Zn$	^{62,63} Zn	inconsistencies – measure and re-evaluate; evaluate
		^{62,63} Zn decay schemes
$^{nat}Cu(d,x)^{62,63,65}Zn$		measure and evaluate
$^{nat}Cu(\alpha,x)^{66,67}Ga, ^{65}Zn$		measure and re-evaluate
$^{nat}Mo(p,x)^{96}Tc^{g+m}$	_	evaluate
_	⁶¹ Cu	evaluate ⁶¹ Cu decay scheme

Cross sections	Decay data	Additional comments
<u>diagnostic γ emitters</u>		
90 Zr(n,p) 90 Y ^{g+m}	_	re-evaluate
¹⁰⁰ Mo(n,2n) ⁹⁹ Mo	⁹⁹ Tc ^m	evaluate; decay data - focus on Auger electrons
100 Mo(p,2n) 99 Tc ^{g+m}		measure and evaluate
¹⁰⁰ Mo(p,pn) ⁹⁹ Mo		measure and evaluate
100 Mo(d,3n) 99 Tc ^{g+m}		evaluate
¹⁰⁰ Mo(d,p2n) ⁹⁹ Mo		evaluate
¹⁰⁰ Mo(γ,n) ⁹⁹ Mo		measure
¹⁰⁰ Mo(γ,f) ⁹⁹ Mo		measure
⁶⁴ Zn(n,p) ⁶⁷ Cu	⁶⁷ Cu	measure and evaluate; evaluate ⁶⁷ Cu decay scheme
⁶⁷ Zn(n,p) ⁶⁴ Cu		measure and evaluate; ⁶⁴ Cu impurity from ^{nat} Zn(n,p)
		reaction
${}^{68}Zn(n,x){}^{67}Cu$		measure and evaluate
68 Zn(γ ,p) 67 Cu		measure
$^{112}Cd(p,2n)^{111}In$	¹¹¹ In	re-evaluate; decay data - focus on Auger electrons

Cross sections	Decay data	Additional comments
<u>diagnostic γ emitters</u> (cont.)		
124 Xe(p,2n) 123 Cs	_	re-evaluate – ¹²³ I production
¹²⁴ Xe(p,pn) ¹²³ Xe		re-evaluate – ¹²³ I production
124 Xe(p,x) 121 I		evaluate – ¹²³ I production: ¹²¹ I impurity from ¹²⁴ Xe(p,x) reaction
⁵¹ V(p,n) ⁵¹ Cr	_	evaluate all noteworthy production routes of ⁵¹ Cr
^{nat} Fe(p,x) ⁵¹ Cr		measure and evaluate
²⁰³ Tl(p,2n) ²⁰² Pb ^m (EC) ²⁰² Tl	_	re-evaluate
²⁰³ Tl(p,3n) ²⁰¹ Pb(EC) ²⁰¹ Tl		re-evaluate
²⁰³ Tl(p,4n) ²⁰⁰ Pb(EC) ²⁰⁰ Tl		re-evaluate
$^{nat}W(\alpha, x)^{186,188}Re$	_	measure

Cross sections	Decay data	Additional comments
<u>β+ emitters</u>		
$^{55}Mn(p,4n)^{52}Fe$	⁵² Fe	evaluate; evaluate ⁵² Fe decay scheme
^{nat} Ni(p,x) ⁵² Fe		evaluate
⁵² Cr(³ He,3n) ⁵² Fe		evaluate
⁵⁸ Ni(p,α) ⁵⁵ Co	_	evaluate
⁵⁴ Fe(d,n) ⁵⁵ Co		evaluate
⁵⁶ Fe(p,2n) ⁵⁵ Co		evaluate
^{nat} Fe(p,x) ⁵⁵ Co		measure and evaluate
⁶¹ Ni(p,n) ⁶¹ Cu	_	measure and evaluate
64 Zn(p, α) 61 Cu		measure and evaluate
⁶⁶ Zn(p,n) ⁶⁶ Ga	⁶⁶ Ga	evaluate; decay data - positron intensities:
		measurements, and ⁶⁶ Ga evaluate decay scheme
63 Cu(α ,n) 66 Ga		evaluate
⁶⁸ Zn(p,n) ⁶⁸ Ga	—	measure and evaluate
65 Cu(α ,n) 68 Ga		evaluate

Cross sections	Decay data	Additional comments
<u>β+ emitters</u> (cont.)		
^{nat} Ge(p,xn) ⁷² As	⁷² As	measure and evaluate; decay data - positron intensities:
		measurements, and evaluate ⁷² As decay scheme
75 As(p,3n) 73 Se	⁷³ Se	measure and evaluate; decay data - positron intensities:
72 (2) 72 9		measurements, and evaluate ⁷³ Se decay scheme
$^{72}\text{Ge}(\alpha,3n)^{73}\text{Se}$		evaluate
76 Se(p,n) 76 Br	⁷⁶ Br	measure and evaluate; decay data - positron intensities:
		measurements, and evaluate "Br decay scheme
$77Se(p,2n)^{76}Br$		measure and evaluate
75 As(α ,3n) 76 Br		evaluate
⁸⁶ Sr(p,n) ⁸⁶ Y	⁸⁶ Y	re-evaluate; decay data - positron intensities:
		measurements, and evaluate ⁸⁶ Y decay scheme
⁸⁸ Sr(p,3n) ⁸⁶ Y		evaluate
${}^{85}\text{Rb}(\alpha, 3n){}^{86}\text{Y}$		evaluate
⁸⁹ Y(p,n) ⁸⁹ Zr	⁸⁹ Zr	evaluate; measure and evaluate ⁸⁹ Zr decay scheme
89 Y(d,2n) 89 Zr		measure and evaluate
$^{nat}Y(\alpha, x)^{89}Zr$		measure and evaluate

Cross sections	Decay data	Additional comments
<u>β⁺ emitters</u> (cont.)		
⁹³ Nb(p,x) ⁹⁰ Nb	-	measure and evaluate
89 Y(α ,x) 90 Nb		measure and evaluate
⁹⁴ Mo(p,n) ⁹⁴ Tc ^m	⁹⁴ Tc ^m	evaluate; measure and evaluate ⁹⁴ Tc ^m decay scheme
92 Mo(α ,x) 94 Tc ^m		evaluate
$^{111}Cd(p,2n)^{110}In^{m}$	—	evaluate
120 Te(p,n) 120 I	120 I	evaluate; measure and evaluate ¹²⁰ I decay scheme
122 Te(p,3n) 120 I		
	⁶⁴ Cu	evaluate decay scheme – discrepant weak gamma
		emission

Cross sections	Decay data	Additional comments	
β⁺ emitters: generators			
⁶² Zn/ ⁶² Cu generator:		PET analogue of therapeutic ⁶⁷ Cu	
⁶³ Cu(p,2n) ⁶² Zn	_	measure and evaluate	
⁶⁸ Ge/ ⁶⁸ Ga generator:		PET analogue of new/proposed therapeutic ⁶⁷ Ga	
^{nat} Ga(p,xn) ⁶⁸ Ge	—	measure and evaluate	
⁶⁹ Ga(p,2n) ⁶⁸ Ge		measure and evaluate	
⁷¹ Ga(p,4n) ⁶⁸ Ge		measure and evaluate	
⁷² Se/ ⁷² As generator:			
75 As(p,4n) 72 Se	_	evaluate	
$^{nat}Br(p,x)^{72}Se$		evaluate	
⁸² Sr/ ⁸² Rb generator:			
^{nat} Rb(p,xn) ⁸² Sr	_	evaluate	

Cross sections	Decay data	Additional comments
<u>β⁺ emitters: generators</u>		
⁴⁴ Ti/ ⁴⁴ Sc	⁴⁴ Ti t _{1/2}	evaluate ⁴⁴ Ti half-life
⁵² Fe/ ⁵² Mn ^m	-	evaluate
$^{110}Sn/^{110}In^{m}$	_	evaluate; PET analogue of therapeutic ¹¹¹ In and ¹¹⁴ In ^m
¹¹⁸ Te/ ¹¹⁸ Sb	_	evaluate; PET analogue of new/proposed therapeutic ¹¹⁷ Sb and ¹¹⁹ Sb
¹²² Xe/ ¹²² I	_	evaluate; PET analogue of therapeutic ¹²³ I, ¹²⁵ I and ¹³¹ I
¹²⁸ Ba/ ¹²⁸ Cs	_	evaluate; PET analogue of new/proposed therapeutic ¹³¹ Cs
¹⁴⁰ Nd/ ¹⁴⁰ Pr	_	evaluate

Cross sections	Decay data	Additional comments
<u>a emitters</u>		
229 Th(α) 225 Ra(β ⁻) 225 Ac(α)	-	
decay chain to ²¹³ Bi:		
²³² Th(p,x) ²²⁵ Ra		measurements up to 200 MeV, and evaluate
²³² Th(p,x) ²²⁵ Ac		measurements up to 200 MeV, and evaluate
²²⁶ Ra(p,2n) ²²⁵ Ac		measure and re-evaluate
²³² Th(p,x) ²²⁷ Ac		evaluate; 227 Ac (t _{1/2} 21.8 y) long-lived
		contaminant of ²²⁵ Ac ($t_{\frac{1}{2}}$ 10.0 d)
230 U(α) 226 Th(α) decay chain:	²³⁰ U decay	evaluate all decay schemes in decay chain:
	chain	230 U(α) 226 Th(α) 222 Ra(α) 218 Rn(α) 214 Po(α) 210 Pb(β^{-})
		²¹⁰ Bi(β^{-}) ²¹⁰ Po(α) ²⁰⁶ Pb(stable)
231 Pa(d,3n) 230 U		evaluate
231 Pa(p,2n) 230 U		evaluate
232 Th(p,3n) 230 Pa(β^{-}) 230 U		²³⁰ Pa β^- branch of only 7.8% – evaluate
227 Th(α) 223 Ra(α) decay chain:	_	
232 Th(p,x) 227Th		

Cross sections	Decay data	Additional comments
electron and X-ray emitters		
130 Ba(n, γ) 131 Ba(EC) 131 Cs	_	evaluate
131 Xe(p,n) 131 Cs		evaluate
¹³³ Cs(p,3n) ¹³¹ Ba(EC) ¹³¹ Cs		evaluate
176 Hf(α ,2n) 178 W(EC) 178 Ta	¹⁷⁸ Ta	measure and evaluate; decay data - focus on Auger
		electrons
^{nat} Hf(p,x) ¹⁷⁸ Ta		measure and evaluate
_	¹⁰³ Pd	evaluate ¹⁰³ Pd decay scheme (including Auger
		electrons)

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Nuclear Medicine: Nuclear Data Considerations

Future applications in nuclear medicine?

• diagnostic

new developments over next 20 years?

• therapeutic

new developments over next 20 years?

If we answer the above question for nuclear medicine, we define our needs for nuclear data measurements and evaluations over both the intermediate- and longer-term timescales

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Radionuclides:

Diagnostic γ-ray emitters

β⁺ emitters

Therapeutic β^- , X-ray and γ -ray emitters

Therapeutic Auger-electron emitters

Therapeutic α emitters

Proton and heavy-ion beam therapy

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Nuclear Data:

Cross-section production data

Decay data

Modelling?

Intermediate Term:

5 to 15 years \rightarrow up to 2025

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Immediate past and future:

22-26 August 2011, IAEA Headquarters, Vienna, Austria

Intermediate-term Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data A.L. Nichols, S.M. Qaim and R. Capote Noy

IAEA report INDC(NDS)-0596, September 2011

Technical Meeting IAEA Headquarters, Vienna, Austria 22-26 August 2011

Diagnostic γ-ray emitters

Radionuclide	Requirements	Comments
99Tc ^m	¹⁰⁰ Mo(p,xn), (p,α), (d,xn); (γ,n), (γ,f); nuclear decay data evaluated in previous CRP (IAEA-STI/PUB/1287); Auger electrons	Accelerator production; highly-enriched ¹⁰⁰ Mo (> 99%) should be investigated; Auger-electron studies planned in CRP (2012)
⁹⁷ Ru	³ He and ⁴ He on Mo	Limited application
123	See IAEA-TECDOC-1211 and IAEA- STI/PUB/1287; Auger electrons	Several production reactions and discrepancies to be studied in planned CRP
¹⁴⁷ Gd	⁴ He on Sm; proton on Eu	Special application in MRI + SPECT
²⁰³ Pb		Special application in tracer studies
Alan Nichols		ICTP, Trieste, Italy, 30 September – 4 October 2013

β^+ emitters

Radionuclide	Requirements	Comments
¹¹ C, ¹³ N, ^{14,15} O, ³⁰ P, ³⁸ K	Activation cross sections for proton-induced reactions with energies up to 250 MeV	Cross sections well defined for $E_p < 20$ MeV \rightarrow higher energies of interest up to 250 MeV for proton therapy
³⁴ Cl ^m	Cross-section measurements and evaluations	Low priority
⁴³ Sc	Cross-section measurements and evaluations	Good positron-decay characteristics, but difficult to produce
⁴⁵ Ti, ⁴⁸ V, ⁴⁹ Cr, ⁹⁰ Nb	Cross-section measurements and evaluations	Potentially important for radioimmunotherapy
^{51,52} Mn	Cross-section measurements and evaluations	Special application in MRI + PET
⁵² Fe, ⁵⁵ Co, ⁶¹ Cu, ¹¹⁰ In ^m	Cross-section evaluations	Several novel applications; ⁵² Fe and ⁶¹ Cu decay-data evaluations planned in CRP (2012)
⁵⁷ Ni, ⁷² As, ⁷³ Se, ⁹⁴ Tc ^m	Cross-section measurements and evaluations; β ⁺ and X-ray emission probabilities	⁷² As, ⁷³ Se and ⁹⁴ Tc ^m decay-data evaluations planned in CRP (2012)

β^+ emitters (continued)

Radionuclide	Requirements	Comments
⁶⁴ Cu	Cross sections - see previous CRP (IAEA Technical Reports Series No. 473)	Important β ⁺ emitter, especially for radioimmunotherapy; decay-data evaluation planned in CRP (2012)
⁶⁶ Ga	Cross-section measurements and evaluations; β ⁺ and X-ray emission probabilities	Decay-data evaluation planned in CRP (2012)
⁶⁸ Ga	Cross-section measurements and evaluations	Direct production, as well as ⁶⁸ Ge/ ⁶⁸ Ga generator route
⁷⁵ Br, ⁷⁷ Kr	Cross-section measurements and evaluations; β ⁺ and X-ray emission probabilities	Limited application
⁷⁶ Br, ⁸⁹ Zr	Cross-section measurements and evaluations; β ⁺ and X-ray emission probabilities	Decay-data evaluations planned in CRP (2012)
⁸¹ Rb, ⁸² Rb ^m , ⁸³ Sr,	Cross-section measurements and evaluations; β ⁺ and X-ray emission probabilities	Limited application
⁸⁶ Y	Cross-section evaluations; β ⁺ and X- ray emission probabilities	Important positron emitter for quantification of dosimetry calculations; decay-data evaluation planned in CRP (2012)

β^+ emitters (continued)

Radionuclide	Requirements	Comments
⁹⁵ Ru	³ He and ⁴ He beam cross-section measurements and evaluations	Limited application; many gamma rays, together with ~ 14% β^+ emission
¹²⁰	Cross-section evaluations; β ⁺ and X- ray emission probabilities	Decay-data evaluation planned in CRP (2012)
¹²¹	Cross-section measurements and evaluations	Borderline – longer-term consideration (easier to produce than ¹²⁰ I); many gamma rays, together with ~ 11% β^+ emission
¹²⁴	Cross sections - see previous CRP (IAEA Technical Reports Series No. 473)	Important positron emitter for quantification of dosimetry calculations
¹⁵² Tb	Cross-section measurements and evaluations	Potentially useful as lanthanide-based positron emitter

β^+ emitters (continued): generators

Radionuclide	Requirements	Comments
⁴⁴ Ti/ ⁴⁴ Sc	Cross-section measurements and evaluations; evaluation of parent T _{1/2}	Long-lived parent ($T_{1/2}$ of 60 y); difficult to produce; evaluation of ⁴⁴ Ti $T_{1/2}$ planned in CRP (2012)
⁵² Fe/ ⁵² Mn ^m	Cross-section and decay-data measurements and evaluations	Special application in MRI + PET
⁶² Zn/ ⁶² Cu	Cross-section measurements and evaluations; β ⁺ and X-ray emission probabilities	⁶² Zn decay-data evaluation planned in CRP (2012)
⁶⁸ Ge/ ⁶⁸ Ga, ⁸² Sr/ ⁸² Rb	Cross-section measurements and evaluations	Well-established systems, but inadequate databases
⁷² Se/ ⁷² As	Cross-section measurements and evaluations; β ⁺ and X-ray emission probabilities	⁷² As decay-data evaluation planned in CRP (2012)
¹⁴⁰ Nd/ ¹⁴⁰ Pr	Cross-section measurements and evaluations; Auger-electron and other low-energy electron data for ¹⁴⁰ Nd microdosimetry	Radiotherapy + PET; parent ¹⁴⁰ Nd(EC) to operate as therapeutic radionuclide, while ¹⁴⁰ Pr is positron emitter (<i>in-vivo</i> generator)

Therapeutic β^- , X-ray and γ -ray emitters

Radionuclide	Requirements	Comments
⁴⁷ Sc	Cross-section measurements and evaluations	Low-energy β [−] emitter
⁶⁷ Cu	Cross sections - see previous CRP (IAEA Technical Reports Series No. 473); decay-data measurements and evaluation, particularly g.s. to g.s. transition	Important radionuclide – emission of low-energy β ⁻ particles, and preparation of organometallic complexes; decay-data evaluation planned in CRP (2012)
¹⁰³ Pd	Cross sections - see previous CRP (IAEA Technical Reports Series No. 473); decay-data discrepancies – measurements and evaluation; Auger electrons	Decay-data evaluation and Auger- electron studies planned in CRP (2012)
¹³¹ Cs	Cross-section measurements and evaluations	X-ray emitter
¹³¹ Ba	Cross-section measurements and evaluations; decay-data evaluation	X-ray emitter
¹⁶¹ Tb	¹⁶⁰ Gd(n,γ) ¹⁶¹ Gd(β ⁻) ¹⁶¹ Tb; decay-data measurements and evaluation	Low-energy β [−] emitter

Therapeutic β^- , X-ray and γ -ray emitters (continued)

Radionuclide	Requirements	Comments
¹⁶⁶ Ho	Cross sections and decay data – see previous CRP (IAEA Technical Reports Series No. 473 and IAEA-STI/PUB/1287); require cross-section measurements and evaluation for	High-flux reactor required for double-neutron capture
¹⁶⁹ Er	¹⁶⁴ Dy(2n,γ) ¹⁶⁶ Dy(β ⁻) ¹⁶⁶ Ho Cross-section measurements and evaluations, including spallation beam cross sections; decay-data measurements and evaluation	Low-energy β [−] emitter
¹⁷⁵ Yb	Cross-section measurements and evaluations for charged-particle reactions; decay-data measurements and evaluation	Low-energy β [−] emitter
¹⁹¹ Os / ¹⁹¹ Ir ^m	Cross-section measurements and evaluations	Low-energy β ⁻ emitter for radiotherapy + SPECT; potential <i>in-</i> <i>vivo</i> generator
¹⁹¹ Pt / ¹⁹¹ lr ^m Alan Nichols	Cross-section and decay-data measurements and evaluations	X-ray emitter; potential <i>in-vivo</i> generator ICTP, Trieste, Italy, 30 September – 4 October 2013

Therapeutic Auger-electron emitters

Radionuclide	Requirements	Comments
⁶⁷ Ga, ¹¹¹ In	Cross sections evaluated in two previous CRPS (IAEA-TECDOC-1211 (⁶⁷ Ga and ¹¹¹ In), and IAEA Technical Reports Series No. 473 (⁶⁷ Ga)); Auger electrons may become an issue	⁶⁷ Ga and ¹¹¹ In of increased application in internal radiotherapy; ¹¹¹ In Auger- electron studies planned in CRP (2012)
⁷¹ Ge	Cross-section measurements and evaluations; Auger electrons may become an issue	Half-life is rather long at 11.4 d
⁷⁷ Br	Cross-section evaluations; Auger electrons may become an issue	
⁹⁹ Tc ^m	Auger-electron (E_e < 25 keV) and other low- energy electron (E_e < 1 keV) data for microdosimetry; nuclear decay data evaluated in previous CRP (IAEA-STI/PUB/1287); further needs for cross-section data will arise if produced by charged-particle reactions	Regularly used for diagnosis, but also increased application in therapeutics; Auger- electron studies planned in CRP (2012)
¹⁰³ Pd	Cross sections evaluated in previous CRP (IAEA- TECDOC-1211); decay-data measurements and evaluation	Decay-data evaluation and Auger-electron studies planned in CRP (2012)

Therapeutic Auger-electron emitters (continued)

Radionuclide	Requirements	Comments
123	See IAEA-TECDOC-1211 and IAEA- STI/PUB/1287; Auger electrons	Regularly used for diagnosis, but also increased application in therapeutics; several production reactions and discrepancies to be studied in planned CRP
¹⁴⁰ Nd	Cross-section evaluations of several reactions; Auger electrons may become an issue	Auger and EC decay; <i>in-vivo</i> generator (¹⁴⁰ Pr) – see previous table (β ⁺ emitters: generators)
¹⁷⁸ Ta	¹⁷⁶ Hf(α,2n) ¹⁷⁸ W(EC) ¹⁷⁸ Ta; Auger electrons may become an issue	Auger and EC decay; <i>in-vivo</i> generator (¹⁷⁸ W); Auger-electron studies planned in CRP (2012)
¹⁹³ Pt ^m , ¹⁹⁵ Pt ^m	Cross-section measurements and evaluations; Auger electrons may become an issue	Large number of Auger electrons emitted
¹⁹⁷ Hg	Cross-section and decay-data measurements and evaluations; Auger electrons may become an issue	

Therapeutic α emitters

Radionuclide	Requirements	Comments
¹⁴⁹ Th	Cross-section measurements and	Emission of low-energy alpha
	evaluations of spallation and heavy-	particles (< 4 MeV) – potentially
	ion beam reactions	useful for special applications
²¹¹ At/ ²¹¹ Po	Cross sections and decay data	Well-established therapeutic
	evaluated in previous CRPs	radionuclide
	(IAEA Technical Reports Series No. 473, and "Updated Actinide Decay Data Library (to be published))	
²²⁵ Ac/ ²¹³ Bi	Lack of cross-section data at higher	Potentially important therapeutic
	energies for spallation reaction on	radionuclide
	²³² Th; nuclear decay chain evaluated	
	in previous CRP ("Updated Actinide Decay Data Library" (to be published))	
$^{227}Ac/^{223}Ra$	Inadequate cross-section data for	Impurity in ²²⁵ Ac production
	²³² Th(p,x) production of ²²⁷ Ac –	
	measurements and evaluation; ²²³ Ra	
	nuclear decay data evaluated in	
	previous CRP ("Updated Actinide Decay Data Library" (to be published))	
²³⁰ U/ ²²⁶ Th	Cross-section studies within planned	New decay-data measurements
0 / 111	CRP; decay-data evaluations of	presented at ICRM2011 conference;
	complete α-decay chain	decay-data evaluation planned for
		complete decay chain in CRP (2012)

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Proton beam therapy:

non-elastic cross sections of C, N and O at E_p up to 250 MeV;
activation cross sections of residual nuclei (¹¹C, ¹³N, ¹⁵O, ³⁰P and ³⁸K positron emitters)

Carbon beam therapy – complex fragmentation reactions \rightarrow difficult to prepare data sets

Fragmentation and production of light particles and residues \rightarrow **require more precise models and validated parameter sets Proton beam therapy** \rightarrow **require more precise Monte-Carlo transport calculations** for dose deposition of variations in morphology or in structure arising from bone or implants Alan Nichols

Nuclear Data Needs

Immediate future:

IAEA-NDS CRP launched in late 2012 is dedicated to cross sections and decay data for medical applications based on:

High-Precision Beta-Intensity Measurements and Evaluations for Specific PET Radioisotopes (see IAEA report INDC(NDS)-0535, 2008)

Improvements in Charged-Particle Monitor Reactions and Nuclear Data for Medical Isotope Production (see IAEA report INDC(NDS)-0591, 2011)

Monitor reactions: ^{22,24}Na, ⁴⁶Sc, ^{56,58}Co, ^{62,63,65}Zn, ⁹⁶Tc^{m+g}

Reactions for diagnostic γ emitters: ⁹⁹Tc^m, ¹¹¹In, ¹²³I (¹²³Cs, ¹²³Xe, ¹²¹I production)

Reactions for novel β⁺ **emitters:** ⁵²Fe, ⁵⁵Co, ⁶¹Cu, ^{66,68}Ga, ⁷²As, ⁷³Se, ⁷⁶Br, ⁸⁶Y, ⁸⁹Zr, ⁹⁴Tc^m, ¹¹⁰In^m, ¹²⁰I

Reactions for generators: ⁶²Zn/⁶²Cu, ⁶⁸Ge/⁶⁸Ga, ⁷²Se/⁷²As, ⁸²Sr/⁸²Rb

Reactions for therapeutic isotopes:

- > α emitters ²²⁵Ra and ²²⁵Ac production (\rightarrow ²¹³Bi); ²²⁷Ac impurity
- electron and X-ray emitters ¹³¹Cs (also ¹³¹Ba production)

Decay data evaluations: ⁵²Fe, ^{61,64,67}Cu, ^{62,63}Zn, ⁶⁶Ga, ⁷²As, ⁷³Se, ⁷⁶Br, ⁸⁶Y, ⁸⁹Zr, ⁹⁴Tc^m, ¹⁰³Pd, ¹²⁰I, ²³⁰U decay chain

⁴⁴Ti half-life, Auger electrons: ⁹⁹Tc^m, ¹⁰³Pd, ¹¹¹In, ¹⁷⁸Ta

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Longer term:

Increased dynamic and quantitative positron tomography (PET) coupled with X-ray tomography (CT) and magnetic resonance imaging (MRI) for organ imaging

Assessment of improved internal radiotherapy:

- > PET and therapy involving radioimmuno reactions
- > Auger-electron and α -particle therapy at the cellular level

Positron emitters and therapeutic radionuclides – long-term possibilities:

- ➤ metallic-based positron emitters (e.g., Ti, Ga, Cu radionuclides) → developments in organometallic-complex chemistry
- ➤ improved microdosimetry → requirement to better characterise suitable low-energy Augerelectron emitters

Nuclear Data Needs

Intermediate- and longer-term considerations – 5 to 15/20 years?

Further need for future IAEA-NDS CRP(s) dedicated to cross sections and decay data for medical applications based on recommendations:

Intermediate-term Nuclear Data Needs for Medical Applications: Cross Sections and Decay Data

A.L. Nichols, S.M. Qaim and R. Capote Noy

22-26 August 2011, IAEA Headquarters, Vienna, Austria IAEA Report INDC(NDS)-0596, September 2011

Previous tables refer

Where to get the data



www.nndc.bnl.gov



ie.lbl.gov



www.tunl.duke.edu/nucldata

data for A < 20



www-nds.iaea.org

IAEA-NDS Medical Portal

Repository for and dissemination of nuclear data for medical applications:

IAEA-NDS Medical Portal must be the focal point

http://www-nds.iaea.org/medportal/

α, β⁺, γ and Auger-electron Decay Data in Nuclear Medicine – Experimental Determination, Status and Deficiencies

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> Workshop on Nuclear Data for Science and Technology: Medical Applications ICTP, Trieste, Italy, 30 September – 4 October 2013