Simulation of Auger Electron Emission Spectra for ^{99m}Tc and ^{123}I by Validated Monte Carlo Codes

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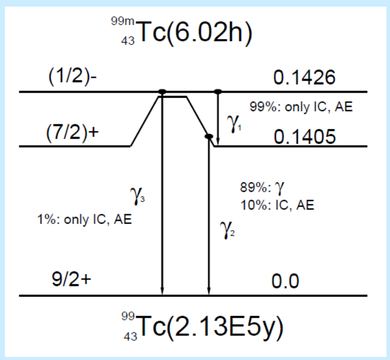
Introduction

Radionuclides decaying by electron capture (EC) and/or by internal conversion (IC) are of special interest in radiobiology (^{111}In, ^{123}I, ^{125}I) and nuclear medicine (^{99m}Tc). Incorporated into the DNA structure, they can cause severe molecular and cellular damage. These findings open a unique possibility for an application of these nuclides in tumour therapy. Therefore, to utilise them effectively, the understanding of their radiation action mechanism is essential, which requires first of all a precise knowledge of the nuclides' electron emission spectra.

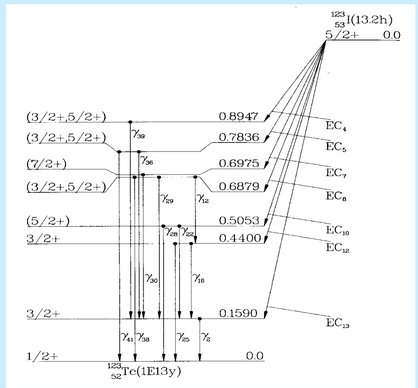
Due to a lack of experimental data for the particular nuclides of interest, computer simulations have become necessary. A decay by EC and/or IC induces an inner electron shell vacancy, the starting point of a complex cascade of photon (radiative) and Auger (non-radiative) transitions within the atomic energy levels. Because of the stochastic nature of these transitions, the Monte Carlo technique is an appropriate tool for the study and simulation of these processes and for providing emission spectra. Those spectra are needed, e.g., as input for track structure calculations.

Decay Schemes



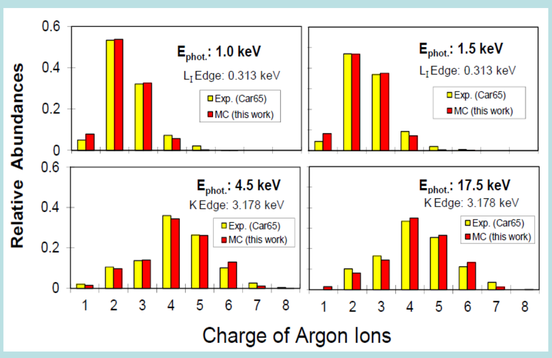
^{99m}Tc: Internal Conversion (IC) induces a chain of Auger and X-ray transitions within the electronic shells of the decaying atom (Auger cascade).

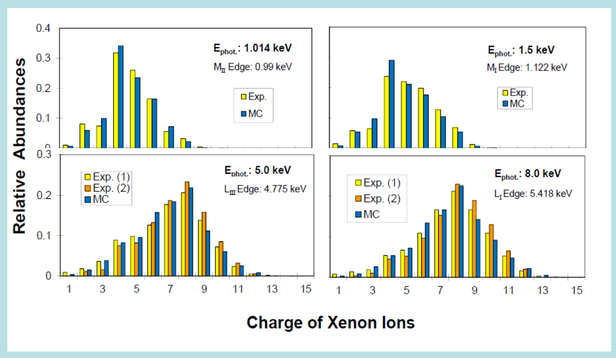
^{123}I: The decay is triggered by electron capture (EC) which initiates an Auger cascade followed by γ transition (84%) or by IC (16%) which gives rise for a second cascade.

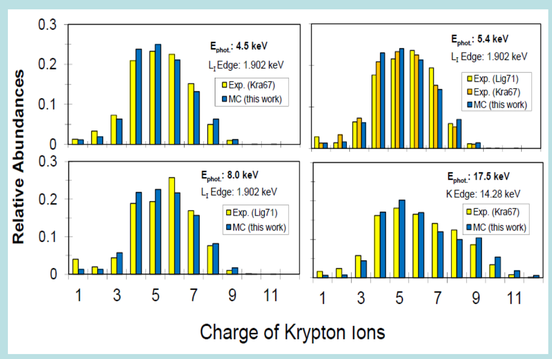


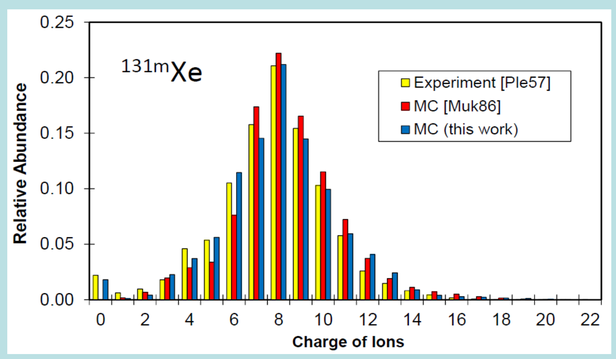
Methods and Validation

Auger cascades induced by photoionisation in noble gas atoms and by IC in ^{131m}Xe were simulated by a Monte Carlo computer code. For these cases many available experimental data allow a comparison of the results and a validation of the simulation code. In view of quite rough assumptions used here, a remarkable good agreement was obtained, e.g., with the experimentally found distributions of charges left on the atoms after the Auger cascades have finished. On this reasonable basis, the computer code was used to calculate electron energy spectra for ^{99m}Tc and ^{123}I.









Results: Mean Numbers of Emitted Particles per Decay

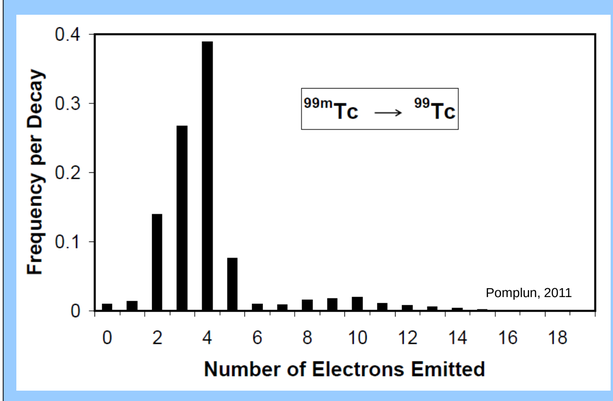
Emitted Particles	^{99m}Tc			^{123}I			
	ICRP (1983)[1]	Howell (1992)[2+3]	Pomplun (2011)[2]	ICRP (1983)[1]	Humm (1984)[2+3]	Howell (1992)[2+3]	Pomplun (2011)[2]
Auger and Coster-Kronig Electrons	1.2	4.0	2.5	2.9	12.3[a]	14.9	6.4
Internal Conversion Electrons	1.1	1.1	1.1	0.15		0.15	0.15
Shake-off Electrons	-	-	0.5	-	-	-	0.9
γ-Rays	0.89	0.89	0.91	0.86	1.86[a]	0.87	0.87
X-Rays	0.07	0.08	0.07	0.86		0.93	0.92

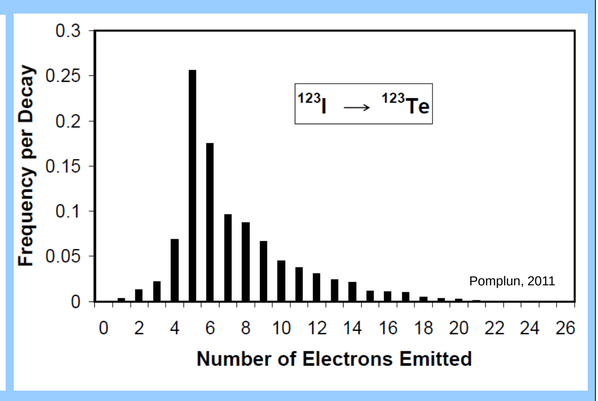
1: no transition within N or higher shells; **2**: neutralization of all vacancies during metastable states; **3**: instantaneous neutralization of valence shell vacancies during cascade; **a**: total values for both Auger and IC electrons as well as for γ and X-rays

Results: Energies Released per Decay

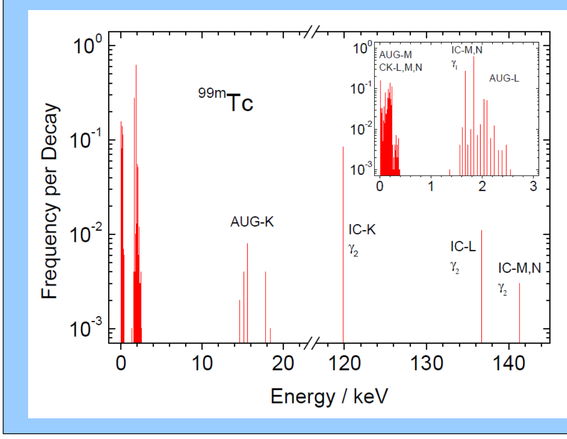
Energy Released (keV)	^{99m}Tc			^{123}I		
	ICRP (1983)	Howell (1992)	This work	ICRP (1983)	Howell (1992)	This work
Total Energy	142	142.65	142.61	199	200.4	197.0
Auger and Coster-Kronig Electrons	1.0	0.9	0.76	7.48	7.42	6.48
Internal Conversion Electrons	15.24	15.38	15.85	20.62	20.25	19.49
γ-Rays	125	125.0	126.6	147.25	148.6	146.64
X-Rays	1.27	1.37	1.23	24.21	24.13	23.86
Ionization Potential	-	-	0.12			0.56

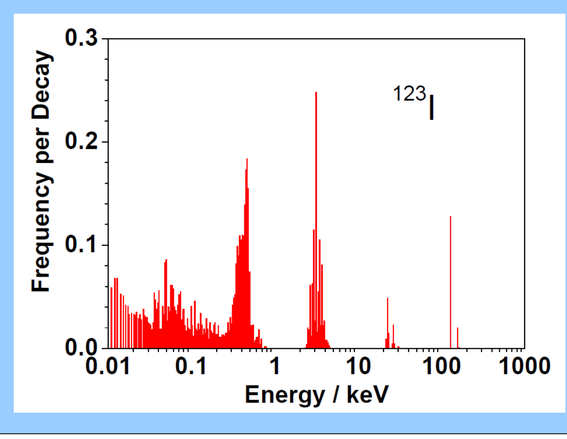
Results: Electron Distribution Spectra





Results: Electron Energy Spectra





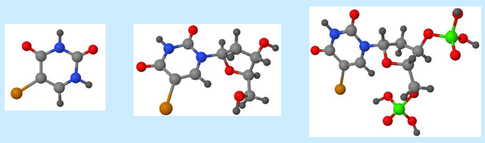
Does Charge Build-Up by DNA Incorporated ^{123}I Induce Strand Breaks?

Methods

- **Gaussian software:** Geometry optimization of various cations at the B3LYP level of density functional theory
- Basis sets: **Lanl2DZ** for Te, **6-31G(d,p)** for the other atoms, Lanl2DZ also for an effective core potential of Te
- Calculation of vibrational frequencies and eigenvectors to discriminate between stable and transitional states
- Molecular charge was changed incrementally by one, starting with the neutral molecule. Optimized structure for one charge level was used as the initial structure for the geometry optimization of the next charge level.

Results

Molecular dissociation at charges greater than:



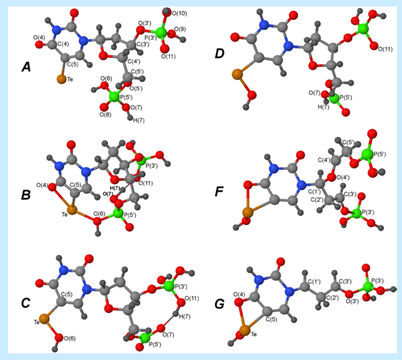
Base	Nucleoside	Nucleotide
5+	2+	7+

Fraction of all ^{123}I decays leading to corresponding charges after the first Auger cascade:

~ 0.60	~ 0.98	~ 0.25

Nucleoside: The early break occurs within the deoxyribose part.

5-telluro-2'-deoxyuridine-3',5'-biphosphate with increasing charges



More stable than 5-telluro-2'-deoxyuridine, the two phosphate groups have a strongly stabilizing effect.

Structure G is the result of a DNA single strand break.

Conclusions

MC-simulated Auger Cascades:

- Multistep stochastic processes, great variety of possible pathways
- Great differences between individual cascades, e.g. number of e^-, e^- energy emitted, etc.
- Comparison with experimental data: reasonable good agreement, differences are due to
  - Experimental X-ray irradiation conditions, which could not be simulated in the required detail.
  - Only transition rates for singly ionized atoms are available.

Consequences of Charge Build-up:

Not every decay of DNA-incorporated ^{123}I necessarily leads to a Coulomb explosion.

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