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Recoil effect on β-decaying *in vivo* generators, interpreted for ^{103}Pd/^{103m}Rh

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ABSTRACT

The use of Auger emitters as potential radiopharmaceuticals is being increasingly investigated. One of the radionuclides of interest is ^{103m}Rh, which can be produced from ^{103}Ru or ^{103}Pd in an *in vivo* generator. A potential problem, however, is the recoil of the ^{103m}Rh out of the carrier molecule and even out of the target cell. In order to determine the likelihood of this happening in the ^{103}Pd/^{103m}Rh, case calculations were made to prove that this does not happen. The equations were generalised for all radionuclides with an atomic mass of 10–240 as a tool for determining the recoil threshold of any β-emitting radionuclide.

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1. Introduction

The well-established use of ^{125}I and ^{111}In Auger-emitting radioisotopes has also stimulated the search for other Auger emitters that may be more practical to use from an availability, physical half-life and cost perspective. Recently the potential of ^{103m}Rh ($t_{1/2} = 56.12$ min) as an Auger emitter in internal radiotherapy was mentioned by Nilsson et al. (2006) and given prominence in the highlights paper of the EANM congress of 2006 (McKillop et al., 2007). Nilsson et al. (2006) suggested that ^{103m}Rh could be derived from ^{103}Ru in a generator and used in the S4 chelator (3,11-dihydroxy-1,5,9,13-tetrathiacyclohexadecane) link to octreotate. ^{103}Ru can be extracted from the fission products of uranium, which implies that, theoretically, large amounts of this nuclide are available.

Another isotope that is well known in radiotherapy is ^{103}Pd ($t_{1/2} = 16.96$ d), although it is normally used in brachytherapy, where it is highly successful in the treatment of prostate cancer, as documented by Sharkey et al. (2002). In this treatment modality the ^{103}Pd is enclosed in a seed and gives a short range of radiation that is effective in treating prostate cancer cells. The decay of ^{103}Pd is via ^{103m}Rh.

$$^{103}_{46}\mathrm{Pd} \xrightarrow[\text{EC branching ratio } 99.974\%]{} {}^{103m}_{45}\mathrm{Rh} \xrightarrow[\gamma\text{-emission}]{} {}^{103}_{45}\mathrm{Rh}$$

As both these nuclides are interesting from the cancer treatment point of view, a combination into one radiopharmaceutical is an option to consider. This would be in the form of an *in vivo* generator system. The concept is not new and is often done with alpha emitters especially. The decay of ^{225}Ac to ^{209}Bi forms five intermediates and produces four alphas and two betas. The release of these decay products from the original carrier molecule and migration out of target cells are well documented (Hassfjell and Brechbiel, 2001). The reason for the release of one. Namely ^{221}Fr (1+ ion and mimicking K^+, an alkali metal ion), is, first, that it requires significantly different chelation chemistry from actinium and, second, that the recoil energy of ^{221}Fr after the alpha particle emission (0.1 MeV) is much greater than the chemical binding energy of a ^{225}Ac conjugate. The change from Pd to Rh is not expected to be so significant, because they both are part of the Pt group of metals. However, the recoil is also a cause of concern for the proposed ^{103}Pd/^{103m}Rh *in vivo* generator. As covalent bonds in the chelate-complex molecule, such as the S4 diol or DOTA, chelates are chemical bonds with a strength estimated to be at least 3 eV (NIST, 2007). The likelihood of bond breaking taking place is investigated in this article. The general calculations presented in this paper can be a useful tool to decide whether there is a recoil effect that is due to the β-decay of the mother radioisotope.

^{103}Pd decays by electron capture (EC), which is a beta transition in which no charged particle is ejected even though the accompanying neutrino of the transition is emitted. However, the same event produces, in addition to the neutrino, a number of Auger electrons and gammas that could contribute to the recoil effect.

Another question that requires attention is what would happen if the chemical bond broke on recoil and how far the product would move in the cell nucleus or mitochondria. A typical animal cell has a diameter of at least 30 μm; a typical mitochondrion or cell nucleus has a length of about 10% of the diameter of the cell, i.e. about 3 μm. Assuming that the ^{103}Pd is uniformly distributed

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in the mitochondrion, one may conclude that the probability of the ^{103}Pd escaping from the mitochondrion upon recoil from a radioactive transition will be small if the range of the recoiling atom in the cell is well below 0.1 μm.

2. Theoretical considerations

From standard expressions of the relativistic relationship between mass, kinetic energy and momentum available in the literature (Beiser, 1969), it follows that the relativistic relationship between momentum p, mass m and kinetic energy T, is

$$p = \frac{\sqrt{2mc^2T + T^2}}{c} \quad (1)$$

A nucleus with mass M_N will recoil with kinetic energy T_N on emitting a particle with mass m_p and kinetic energy T_p. The linear moment of the recoiling nucleus and the emitted particle will be numerically equal, according to the law of the conservation of linear momentum, i.e.

$$\sqrt{2M_Nc^2T_N + T_N^2} = \sqrt{2m_pc^2T_p + T_p^2} \quad (2)$$

and from this it follows that the kinetic energy of the recoiling nucleus (henceforth simply denoted with the symbol E_N) will be

$$E_N = \sqrt{(M_N^2c^4 + E_p^2 + 2m_pc^2E_p} - M_Nc^2 \quad (3)$$

Eq. (3) is a general, fully relativistic expression that holds true for both particles with non-zero mass and for zero-mass energy quanta such as photons. The term $(2m_pE_pc^2)$ will be zero for photon emission, because the mass of a photon is zero; the term $(2m_pE_pc^2)$ will be negligibly small for neutrino emission, because the mass of the neutrino is at present estimated to be of the order of 30 meV, i.e. about 3×10^{-11} a.m.u. For heavy emitted particles, such as alpha and beta particles, the term $(2m_pE_pc^2)$ will dominate and lead to great recoil energies.

When a nucleus de-excites by the emission of a γ-photon or neutrino, the principle of the conservation of linear momentum dictates that this must also be the momentum M_N of the recoil nucleus. The non-relativistic expression for the energy of the recoil nucleus will hence be

$$E_N = \frac{1}{2}M_N\left(\frac{E_\gamma}{M_Nc}\right)^2 \quad (4)$$

The above will also be the recoil energy of an atom that emits an X-ray photon.

For ion range calculations for recoil nuclei that break free from chemical bonds, the initial energy E_0 of the ion will be

$$E_0 = E_{recoil} - E_{bind} \quad (5)$$

where E_{recoil} is the recoil energy and E_{bind} the binding energy of the chemical bond that is broken, i.e. the depth of the potential well from which the recoil process frees the ion to the unbound state.

Table 1
^{103}Pd Auger electron energies, emission abundances and resultant nuclear recoil energies.

Electron energy (keV)	Emission yield	Recoil energy of nucleus (eV)
3.91E+01	1.44E−01	2.17E−01
3.63E+01	7.12E−01	2.02E−01
1.70E+01	1.82E−01	9.44E−02
1.65E+01	9.52E−02	9.18E−02
2.39E+00	1.68E−00	1.32E−02

3. Results and discussion

3.1. Recoil energy of the ^{103}Pd nucleus with Auger electron emission

Auger electrons are emitted at five distinct energies (National Nuclear Data Center, 2007) on the radioactive transition of ^{103}Pd; these electron emissions and consequent recoil energies of the nucleus are summarised in Table 1. The recoil energies of the nucleus were calculated using Eq. (3).

From Table 1 it is clear that all recoil energies are far below 1 eV, so that there is no danger that chemical bonds will be broken as a result of nuclear recoil of ^{103}Pd on Auger electron emission.

3.2. Recoil energy of ^{103}Pd with the emission of photons

Eq. (4) was used to calculate these recoil energies, while the code SRIM-2006 (Ziegler, 2006) was used to obtain a rough estimate of the maximum ranges of such recoil ions, as summarised in Table 2.

In the hypothetical case of a combined emission of all gamma rays in the same direction, $E_{\gamma\_max} = 0.963$ MeV, and the recoil energy of the ^{103}Pd nucleus will be 4.8 eV. It is possible that this recoil energy will cause the chemical bonds to break. However, from Table 2 it is apparent that the emission by ^{103}Pd of an ionising photon with an energy exceeding 0.024 MeV is extremely unlikely—the probability is only ~0.04%. For a yield-weighted average emitted gamma ray energy of less than 0.027 MeV, the recoil energy of the nuclide will be only 0.0038 eV, which is so low that it cannot lead to the chemical bonds breaking. The maximum range of recoil nuclei in water is in the order of 1 nm, which is negligibly low.

Table 2
Recoil energies and approximate maximum recoil ion ranges for ^{103}Pd on emission of gamma photons.

Photon energy (keV)	Emission yield per radioactive transition of parent radionuclide (fraction)	Percentage of total yield (%)	Recoil energy (eV)	Maximum range (μm)
2.007E+01	2.24E−01	29.10	2.1E−03	Negligible
2.022E+01	4.25E−01	55.23	2.1E−03	Negligible
2.270E+01	3.54E−02	4.60	2.7E−03	Negligible
2.272E+01	6.85E−02	8.90	2.7E−03	Negligible
2.317E+01	1.64E−02	2.13	2.8E−03	Negligible
3.574E+02	2.21E−04	0.03	7.0E−01	0.001
4.971E+02	3.98E−05	0.01	***1.3E−00***	0.002
Sum of all photon energies 9.634E+02		Probability of simultaneous emission: 3.31E−12	***4.8E−00***	<0.003

Emissions at which chemical bonds may be severed are printed in bold italics; the probability of these events occurring is very low.

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