Particulate Radiations Emitted during Electron Capture and Isomeric Transitions

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Compared with beta emitters, radionuclides that decay by electron capture or solely by isomeric transition are generally considered preferable for nuclear medicine because they do not release particulate radiations from the nucleus during decay, and hence presumably contribute less absorbed dose and biological damage to surrounding tissue. In this regard, the recent article by Rao, Govelitz, and Sastry (*1*) is of considerable interest because it reveals similar radiotoxicities for the electron-capture radionuclide Tl-201 and the beta-emitting radionuclide Tl-204, in spite of the 6:1 ratio of absorbed dose from Tl-204 compared with Tl-201 as computed by conventional dosimetric methods. This observation implies that the radiobiological effectiveness is considerably greater for radiations from Tl-201 than for those from Tl-204.

As the authors point out, the radiations responsible for the local absorbed dose from Tl-201 are principally low-energy electrons resulting from nonradiative Auger and Coster-Kronig transitions. From questions asked by a number of individuals, it appears that these transitions are not familiar to all readers of the *Journal*. Hence, an explanation in relatively nontechnical terms may be useful.

The Tl-201 nucleus decays by electron capture, in which an electron, usually one occupying an inner shell, is captured by the nucleus. This process leaves a vacancy in the electron shell. The capture process reduces the atomic number of the nucleus by one to form Hg-201 in an excited energy state. Decay to the ground state of Hg-201 occurs by isomeric transition, in which energy is released, either as γ (radiative transitions) or through the process of internal conversion (nonradiative transitions). In internal conversion, the released energy is carried from the atom by an electron ejected from an electron shell, usually one near the nucleus. Outside the atom, the kinetic energy E_k of this electron is

$$E_k = E_{IT} - E_B,$$

where E_{IT} is the energy released during the isomeric transition of the nucleus and E_B is the binding energy of the ejected electron.

In both electron capture and internal conversion, a vacancy is created, usually in an inner electron shell of the atom. This vacancy is promptly filled by an electron from an energy level farther from the nucleus, usually (but not always) one level beyond that containing the vacancy. In this process, energy is released either as x rays (radiative transitions) or through emission of one or more Auger electrons (nonradiative transitions). In the latter case, the released energy is carried from the atom by an electron ejected from an energy level beyond that containing the original vacancy. Often the Auger electron and the electron filling the inner vacancy arise from the same energy level.

Outside the atom, the kinetic energy E_k of the Auger electron is

$$E_k = E_{eT} - E_B,$$

where E_{eT} is the energy released during transition of an electron to fill the original vacancy, and E_B is the binding energy of the electron ejected during the Auger process. Auger electrons are one of the types of particulate radiation that contribute to the local absorbed dose from Tl-201.

With the exception of the innermost electron shell (the K shell), all electron energy levels have sublevels with slight differences in electron binding energy. For example, the L shell has three distinct energy levels (subshells) known as L_I, L_{II}, and L_{III}. Theoretically, a vacancy in the K shell could be filled by transition of an L-electron from any of these subshells. Although this statement is true for nonradiative processes (i.e., Auger electron emission), it is not true for radiative transitions (i.e., x-ray emission), at least in the sense that each

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electron has an equal probability of filling the vacancy irrespective of its subshell. Radiative transitions are governed by certain selection rules that make some transitions (termed allowed transitions) more likely than others (called forbidden transitions). In our example, transitions to the K shell are much more likely for electrons in the L_{II} and I_{III} subshells than for electrons in the L_I subshell. Knowledge of the subshells of different energy levels, and the selection rules that govern electron transitions among them, are essential to an understanding of the energies of characteristic x rays and Auger electrons emitted from a specific atomic species.

With the departure of an electron from a subshell of an energy level such as the L shell, M shell, etc., a second vacancy is created that will be filled by a second radiative or nonradiative transition similar to that described above. Hence, electron capture and internal conversion give rise to a series of electron transitions until all internal electron vacancies are filled and the atom regains electronic stability. As part of this cascading process, some interesting rearrangements of electrons occur among sublevels.

In the L shell, electrons in the subshells L_I, L_{II}, and L_{III} differ slightly in binding energy, with electrons in the L_I subshell having the highest binding energy and L_{III} electrons the least. Hence, a vacancy in the L_I shell can be filled by transition of an electron from L_{II} or L_{III}, whereas a vacancy in the L_{II} subshell could be filled by transition of an electron from L_{III}. These transitions are almost always nonradiative; the released energy is quite small and is removed from the atom by release of an electron rather than by emission of electromagnetic radiation.

Because of the small amount of energy released during transition of an electron from one subshell to another within the same energy level, the electron that carries this energy from the atom is required to have a rather small binding energy. Hence, this electron will originate in an energy level rather far from the nucleus and will leave the atom with only a small amount of energy. This energy will be dissipated in the immediate vicinity of the atom and will contribute to the local absorbed dose. Electrons released as a result of transitions among subshells of a given energy level are called Coster-Kronig electrons.

For Tl-201, it is the Auger and Coster-Kronig electrons that create the high local absorbed doses and contribute to the surprisingly high effectiveness of this radionuclide to produce biological damage. The emission of these electrons is entirely predictable from knowledge of the substructure of the electron energy levels, as demonstrated by Rao, Govelitz, and Sastry. We are indebted to these authors for suggesting that decay processes that appear to be rather esoteric sometimes can have practical consequences.

REFERENCES

1. RAO DV, GOVELITZ GF, SASTRY KSR: Radiotoxicity of thallium-201 in mouse testes: Inadequacy of conventional dosimetry. *J Nucl Med* 24:145-153, 1983