Contribution ID: **216** Type: **Verbal**

In vivo generators –recoil and conversion electrons considerations

Thursday, 22 April 2010 10:00 (30 minutes)

The use of radionuclides as potential therapeutic radiopharmaceuticals is increasingly investigated. An important aspect is the delivery of the radionuclide to the target, i.e. the radionuclide is not lost from the chelating agent. For in vivo generators it is not only the log K of complexation between the metal ion and the chelator that is important but whether the daughter radionuclide stays inside the chelator after decay of the parent radionuclide. In our previous work [1] we showed that the classical recoil effect only is applicable for decays with a Q value higher than 0.6 MeV (in the atomic mass range around 100). However Rosch et al [2] published result for Nd-140/Pr-140 (Q = 0.222 MeV) which indicated that >95% of the daughter (Pr-140) was lost by a DOTA chelator upon decay of Nd-140. This was explained to be due to a "post-effect"by the authors.
Their experiment was repeated with the Dy-166/Ho-166 potential in vivo generator to confirm that our calculations were correct. It was found that indeed some of the daughter (Ho-166) was liberated from the DOTA chelator, therefore indicating that the "post effect"does exist in contrast to our recoil calculations. Upon further investigation we determined that one should not only consider recoil energy levels but also the mode of decay (decay of the parent nuclide and the height of excitation states of the daughter nucleus). If decay takes place via electron capture (EC), Auger electrons are emitted resulting in a daughter radionuclide with a very high oxidation state due to the loss of these electrons from the electron shells. From the well known described biological effects of I-125 [3] which decays to (Te-125)21+ one knows that chemical bonds in the vicinity of the decay are broken. It is therefore postulated that the EC decay of Nd-140/Pr-140 results in a Nd-140 daughter that destroys the DOTA chelate and is therefore released from this chelator. In the case of Dy-166/Ho-166 (Q = 0.486) the decay mode is via β- decay (which according to our recoil calculations is not strong enough to break the bond with the chelator) followed by dissipation of excitation energy of the excited state of the formed daughter nuclide via a branched decay - γ radiation and conversion electron process. The percentage of decay via the conversion electron process was found to match the percentage of the amount of Ho-166 that was measured to be released from the DOTA chelator in our experiment.
It is therefore concluded that the recoil effect below 0.6 MeV is not sufficient to rule out release of the daughter nuclide from chelators but one needs to consider the mode of decay as well. This knowledge has important implications for the design of future radiopharmaceuticals.
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Session Classification: Nuclear Methods in Medicine, Radiopharmaceuticals and Diagnostics, Labelled Compounds 2

Track Classification: Nuclear Methods in Medicine, Radiopharmaceuticals and Radiodiagnostics, Labelled Compounds