



Contribution ID: 122

Type: poster

Synthesis and characterization of novel asymmetrical M(III) (M = Re, Tc-99g) complexes as models for the development of new potential tracers for SPECT imaging and radiotherapy

Thursday 22 September 2011 17:30 (1h 30m)

Development of new ^{99m}Tc and $^{186/188}\text{Re}$ radiopharmaceuticals still remain an interesting research topic thanks to their ideal nuclear properties. ^{99m}Tc is the radioisotope of election for SPECT-imaging ($E_\gamma=140$ keV, $t_{1/2}=6.02$ h), and $^{186/188}\text{Re}$ are important radionuclides with therapeutic potential (^{186}Re : $E_\beta=1.07$ MeV, $E_\gamma=137$ keV, $t_{1/2}=90.6$ h; ^{188}Re : $E_\beta=2.12$ MeV, $E_\gamma=155$ keV, $t_{1/2}=17$ h). Despite the rich and diverse coordination chemistry of these two congeners, very few M(III)-complexes are reported in literature and none have clinical applications.

Within we report the synthesis and the characterization of a new series of neutral, asymmetrical, hexacoordinated Re/ ^{99g}Tc -complexes of the type $[\text{MIII}(\text{PS})_2(\text{L})]$ where PS is the strong π -acceptor 2-(diphenylphosphino)ethanethiolate (PS2) or the completely alkylic non- π -acceptor 2-(diisopropyl)ethanethiolate (PSiso) P,S-bidentate ligands, while L is a dithiocarbamate (DTCn) or pyridine-2-thiolate (MPy) ligands.

The $[\text{MIII}(\text{PS})_2(\text{L})]$ complexes were synthesized starting from precursors where the metal is in different oxidation states, involving ligand exchange reactions and/or reduction-ligand exchange reactions. All the complexes were obtained in moderate to good yield and were found to be diamagnetic, neutral and stable and were characterized by elemental analysis, multinuclear NMR, ESI(+)-QTOF-MS, and cyclic voltammetry. X-ray diffraction analysis was performed on crystallized $[\text{ReIII}(\text{PS})_2(\text{DTC1})]$, $[\text{ReIII}(\text{PS})_2(\text{DTC2})]$, $[\text{ReIII}(\text{PS})_2(\text{MPy})]$, $[\text{ReIII}(\text{PS})_2(\text{DTC1})]$ (DTC1 = pyrrolidine-1-carbodithioate; DTC2 = 4-(ethoxycarbonyl)piperidine-1-carbodithioate), and $[\text{ReIII}(\text{PS})_2(\text{MPy})]$. The X-ray structural analysis of $[\text{ReIII}(\text{PS})_2(\text{DTC1})]$, $[\text{ReIII}(\text{PS})_2(\text{DTC2})]$ and $[\text{ReIII}(\text{PS})_2(\text{DTC1})]$ revealed for these complexes a distorted octahedral geometry, where the four Sulfur atoms occupy the equatorial plane and the two Phosphorus atoms span the apical positions. $[\text{ReIII}(\text{PS})_2(\text{DTC1})]$ is isostructural and isomorphous with $[\text{Re}(\text{PS})_2(\text{DTC1})]$. X-rays diffraction data of $[\text{ReIII}(\text{PS})_2(\text{MPy})]$ confirm the presence of the pyridine-2-thiolate moiety instead of the MPyO.

The collected results indicate the possibility to consider this new class of complexes suitable for the development of novel $^{99m}\text{Tc}/^{186/188}\text{Re}$ -agents useful in theragnostic applications. Studies are currently in progress in order to transfer this technology at tracer level.

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Session Classification: Poster Section 2

Track Classification: Radiopharmaceuticals Chemistry