# Thermodynamic stability and speciation of Ga(III) and Zr(IV) complexes with high-denticity hydroxamate chelators.

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Streszczenie

Increasing attention has been recently devoted to $^{89}$Zr(IV) and $^{68}$Ga(III) radionuclides, due to their favorable decay characteristics for positron emission tomography (PET). In the present paper, a deep investigation is presented on Ga(III) and Zr(IV) complexes with a series of tri-(**$H_3$L1**, **$H_3$L3**, **$H_3$L4** and desferrioxamine E, **DFOE**) and tetrahydroxamate (**$H_4$L2**) ligands. Herein, we describe the rational design and synthesis of two cyclic complexing agents (**$H_3$L1** and **$H_4$L2**) bearing three and four hydroxamate chelating groups, respectively. The ligand structures allow us to take advantage of the macrocyclic effect; the **$H_4$L2** chelator contains an additional side amino group available for a possible further conjugation with a biomolecule. The thermodynamic stability of Ga(III) and Zr(IV) complexes in solution has been measured using a combination of potentiometric and pH-dependent UV–vis titrations, on the basis of metal–metal competition. The Zr(IV)-**$H_4$L2** complex is characterized by one of the highest formation constants reported to date for a tetrahydroxamate zirconium chelate (log β = 45.9, pZr = 37.0), although the complex-stability increase derived from the introduction of the fourth hydroxamate binding unit is lower than that predicted by theoretical calculations. Solution studies on Ga(III) complexes revealed that **$H_3$L1** and **$H_4$L2** are stronger chelators in comparison to DFOB. The complex stability obtained with the new ligands is also compared with that previously reported for other hydroxamate ligands. In addition to increasing the library of the thermodynamic stability data of Ga(III) and Zr(IV) complexes, the present work allows new insights into Ga(III) and Zr(IV) coordination chemistry and thermodynamics and broadens the selection of available chelators for $^{68}$Ga(III) and $^{89}$Zr(IV).

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