Stoichiometry of lanthanide(III) complexes with tripodal aminophosphonic ligands : a new solution to an old problem.

Autorzy

Rafał Janicki

Joanna Gałęzowska

Anna Mondry

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The Eu^{3+} and Gd^{3+} complexes with an *N*-(methylene-2-pyridine)-*N*,*N*-di(methylenephosphonate) ligand (H_4NP_2py), an analogue of nitrilotri(methylphosphonic) acid (H_6NTP), were synthesized and structurally characterized by X-ray single crystal diffraction. The determined crystal structures ($[C(NH_2)_3]_5[Ln(NP_2py)_2]\cdot 12H_2O$) are the first example of a monomeric Ln^{3+} complex encapsulated by two tripodal aminophosphonic ligands. Each of the NP_2py anions coordinates to Ln^{3+} through two oxygen atoms from each monodentate phosphonic group, amine nitrogen and pyridine nitrogen atoms, filling thus 8 coordination sites of Ln^{3+}. The luminescence properties of $[C(NH_2)_3]_5[Eu(NP_2py)_2]\cdot 12H_2O$ crystals were studied and compared with those of Eu–NP_2py complexes in solution. Speciation analysis of Ln–NP_2py complexes (Ln:NP_2py = 1:2), performed by luminescence and potentiometric methods, showed that both $[Ln(NP_2py)]^-$ and $[Ln(NP_2py)_2]^{5-}$species may exist in solution. However, the formation of the latter one occurs in alkaline solutions at pH as high as 8. By implementing the Specific Ion Interaction Theory (SIT) it was possible to calculate the thermodynamic stability constants of the $[Eu(NP_2py)]^-$ and $[Eu(NP_2py)_2]^{5-}$ complexes. The corresponding $\log\beta^0_{Eul}$ and values are 16.3 ± 0.11 and 19.5 ± 0.15, respectively.

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