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EUROPIUM HOMODINUCLEAR TRIPLE-STRANDED HELICATES: INSIGHT INTO THE SELF-ASSEMBLY MECHANISMS

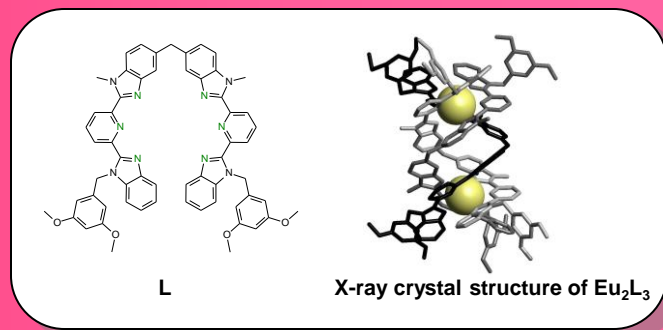
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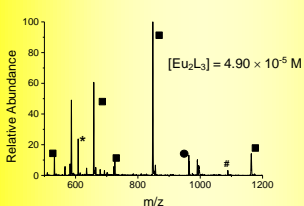
Introduction

The preparation of novel supramolecular structures is a subject of much current interest. The peculiar photophysics of the lanthanides have aroused the development of efficient luminescent sensors with interesting three-dimensional supramolecular topographies. The high potential of applications of the lanthanide helicates in chemical, biochemical and medical domains, increases the interest of thermodynamic and kinetic studies of these compounds. In this work, the speciation studies on the europium(III) complexes formed with the hexadentate receptor L were carried out and the self-assembly mechanism of a triple stranded dimetallic helicate Eu₂L₃ was established.

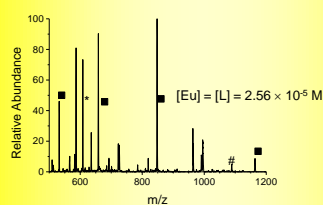


Mass Spectrometry

Electrospray Mass Spectra



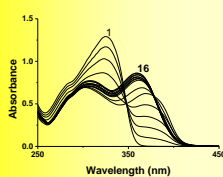
* Eu₂L₂
 # EuL₂
 ■ Eu₂L₃
 ● L



Experimental conditions:
 Solvent: acetonitrile; V_c = 55 V.

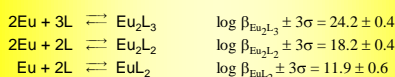
Spectrophotometry

Titration

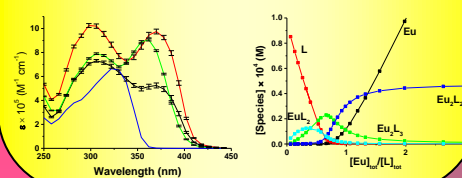


Experimental conditions:
 Solvent: acetonitrile
 T = 25.0 (1)°C; l = 0.2 cm;
 [L]_{tot} = 9.64 × 10⁻⁶ M;
 Spectra 1-16: [Eu]_{tot}/[L]_{tot} = 0 - 6.4

Stability Constants

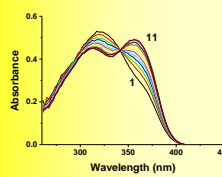


Electronic Spectra and Distribution Curves



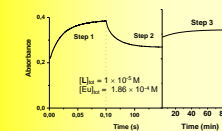
Formation Kinetics

Time-Resolved Absorption Spectra



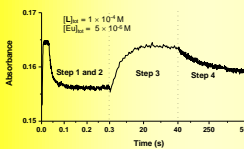
Experimental conditions:
 Solvent: acetonitrile;
 T = 25.0 (1)°C; l = 1 cm;
 [L]_{tot} = 10⁻⁶ M; [Eu]_{tot} = 10⁻⁴ M.
 Spectra: 1.2 ms, 11.280 ms.

Kinetics in Excess of Eu(III)

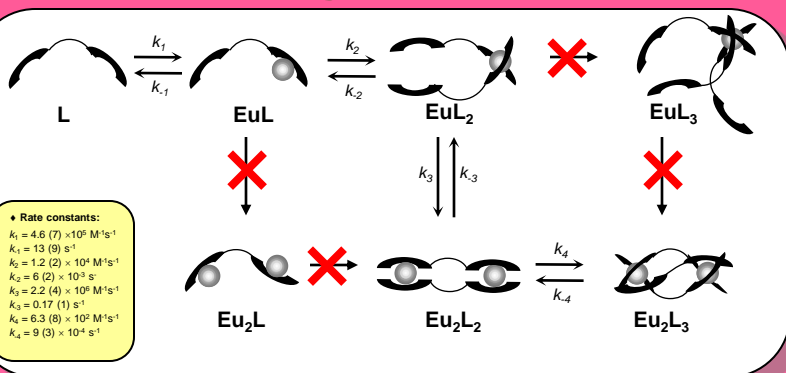


Experimental conditions:
 Solvent: acetonitrile;
 λ = 375 nm; T = 25.0 (1)°C;
 l = 1 cm.

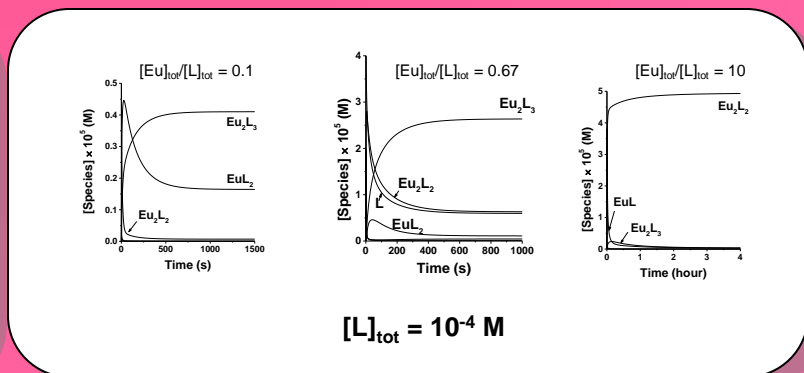
Kinetics in Excess of L



Self-Assembling Mechanism



Kinetic Simulation



Conclusion

The experimental data obtained by a combination of electrospray mass spectrometry (ESMS), absorption spectrophotometry and fast kinetic techniques provided information on the key steps of a self-assembly process of a dinuclear triple-stranded europium helicate Eu₂L₃. It was shown that the supramolecular edifice was totally formed at high concentrations of L ($\cong 10^{-2}$ M) and under strict stoichiometric conditions.

Acknowledgement

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References

(1) Piguet, C.; Bünzli, J.-C. G.; Bernardinelli, G.; Hopfgartner, G.; Williams, A. F., *J. Am. Chem. Soc.*, **1993**, *115*, 8197-8206. (2) Elhabiri, M.; Scopelliti, R.; Bünzli, J.-C. G.; Piguet, C., *J. Am. Chem. Soc.*, **1999**, *121*, 10747-10762. (3) Blanc, S.; Yakir-witch, P.; Leize, E.; Meyer, M.; Libman, J.; Van Dorsselaer, A.; Albrecht-Gary, A.-M.; Shanzer, A., *J. Am. Chem. Soc.*, **1997**, *119*, 4934-4944. (4) Fatim-Rouge, N.; Blanc, S.; Pfeil, A.; Rigault, A.; Albrecht-Gary, A.-M.; Lehn, J.-M., *Helv. Chim. Acta*, **2001**, *84*, 1694-1711.