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镧系水合离子的密度泛函理论研究

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摘要 用密度泛函理论(DFT)方法研究了镧系水合离子[Ln(H2O9)]^3+(Ln=Ce,Pr,Nd,Pm,Ho,Er,Tm,Yb)的几何构型、电荷分布和Ln^3+与水的结合能，计算结果与实验基本符合，表明DFT方法也适用于计算镧系离子与中性配体形成的化合物，对计算结果的分析表明，Ln^3+与H2O之间主要通过Ln5d轨道与氧孤对电子相互作用成键而结合，其余轨道起的作用比较小，用镧系化合物成键模型解释了镧系离子与水的结合能从La到Lu逐渐增加的事实。

关键词 镧系元素化合物 水合离子 构型 电荷分布 结合能 密度函数

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Study on hydrated lanthanide ions by means of density functional theory

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Abstract The lanthanide hydrated ions [Ln(H2O9)]^3+(Ln=Ce, Pr, Nd, Pm, Ho, Er, Tm, Yb) have been studied by means of density functional theory (DFT). The fully optimized geometrical structure and the binding energy between lanthanide ion and H2O of these complexes have been obtained. The calculated results are in fair good agreement with experiments, showing that DFT method can also give reliable results for the calculations of the complex systems forming from lanthanide ions and neutral molecules. Based on analysis of the calculated results it is shown that the binding between Ln^3+ and H2O is mainly due to the interaction of Ln 5 d orbitals and the lone pairs in oxygen atoms. The contribution to the binding energy by other lanthanide orbitals is small. This conclusion is in accordance with the bonding model of lanthanide compounds proposed by one of the authors before. This bonding model can also be used to explain the fact that the binding energy between Ln^3+ and H2O increases from La to Lu, while the bonding energy of the neutral lanthanide compounds decreases along the lanthanide series.

Key words LANTHANIDE SERIES COMPOUNDS HYDRATED ION CONFIGURATION CHARGE DISTRIBUTION BINDING ENERGY

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