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^9^9^mTcO^3^+和^9^9^mTcN^2^+的二胺基二硫醇配合 物在脑中滞留机制的量子 化学研究

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摘要 N,N'-二(2-巯乙基)-乙二胺(DTEN)作为二胺基二硫醇类配体的模型化合物，CH~3SH作为还原型谷胱甘肽(GSH)的模拟物，用从头算分子轨道理论研究了这两种配合物与CH~3S^-的反应产物。结果表明，CH~3S^-配位到TcO-DTEN的Tc原子上，而将其转化为不能穿越血脑屏障的[TcO-DTEN-SCH~3]^-离子。与此相反，CH~3S^-不能与TcN-DTEN中的Tc配位，后者仍保持可以穿越血脑屏障的中性分子状态。以此解释了^9^9^mTcO^3^+的二胺基二硫醇配合物比相应的^9^9^mTcN^2^+配合物在脑中有较高的滞留。还用从头算分子轨道理论研究了胺基配体与TcO^3^+及TcN^2^+配位时从胺基N上脱去质子的规律。

关键词 硫醇 从头计算法 分子轨道理论 锝 放射性核素滞留

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A quantum chemical study on the cerebral retention of ^9^9^mTcO^3^+ and ^9^9^mTcN^2^+ complexes with diaminodithiol ligands

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Abstract In order to examine the effectiveness of reduced glutathione (GSH) attack mechanism for cerebral retention of ^9^9^mTcO^3^+- or ^9^9^mTcN^2^+- labeled diaminodithiols (N~2S~2), an ab initio MO study was performed on a model system in which N,N'-di (mercaptoethyl) ethylendiamine (DTEN) was taken as a model compound for diaminodithiols, and CH~3S^- anion as the mimic of GSH. The calculation results show that CH~3S^- anion can coordinate to the ^9^9^mTc of ^9^9^mTcO-DTEN, converting the latter to an anionic species [^9^9^mTcO-DTEN-SCH~3]^- that can not penetrate the intact brain blood barrier (BBB). Similar reaction does not occur in the case of ^9^9^mTcN-DTEN. Thus the retention of ^9^9^mTcN-N~2S~2 complex by GSH attack can not be expected. Th effectiveness of GSH attack mechanism for the cerebral retention of ^9^9^mTcO-N~2S~2 is sensitive to the substitutions for protons on the N~2S~2 backbone. The rule governing the deprotonation from the amino groups in a N~2S~2 ligand when that ligand coordinates to TcO^2^+ core is also discussed based on ab initio MO calculations.

Key words MERCAPTAN AB INITIO CALCULATION MOLECULAR ORBITAL THEORY TECHNETIUM RADIONUCLIDE RETENTION

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