Computational studies of modified [Fe3S4] clusters: Why iron is optimal - DTU Orbit (14/12/2018)

**Computational studies of modified [Fe3S4] clusters: Why iron is optimal**

This work reports density functional computations of metal-substituted models of biological [Fe3S4] clusters in oxidation states [MFe2S4]+/0/−1 (M = Mn, Fe, Co, Ni, Cu, Zn, and Mo). Geometry optimization with a dielectric screening model is shown to provide a substantial improvement in structure, compared to earlier used standard procedures. The error for average Fe–S bonds decreased from 0.038 Å to 0.016 Å with this procedure. Four density functionals were compared, B3LYP, BP86, TPSS, and TPSSh, and to a lesser extent TPSSh energies were inconsistent with experiment for the oxidized [Fe3S4]+ cluster. BP86 (and to a slightly lesser extent TPSS) was within expected theoretical and experimental uncertainties for all oxidation states, the only qualitative error being 5 kJ/mol in favor of the MS = 3/2 configuration for the [Fe3S4]+ cluster, so BP86 was used for quantitative results. Computed reorganization energies and reduction potentials point directly towards the [Fe3S4] cluster as the superior choice of electron carrier, with the [ZnFe2S4] cluster a close second. In addition, partially and fully Mo-substituted clusters were investigated and found to have very low reorganization energies but too negative reduction potentials. The results provide a direct rationale why any substitution weakens the cluster as an electron carrier, and thus why the [Fe3S4] composition is optimal in the biological clusters.

**General information**

State: Published
Organisations: Physical Chemistry, Department of Chemistry
Contributors: Kepp, K. P.
Pages: 87-100
Publication date: 2008
Peer-reviewed: Yes

**Publication information**

Journal: Journal of Inorganic Biochemistry
Volume: 102
Issue number: 1
ISSN (Print): 0162-0134
Ratings:
BFI (2018): BFI-level 1
Web of Science (2018): Indexed yes
BFI (2017): BFI-level 1
Scopus rating (2017): CiteScore 3.18 SJR 0.743 SNIP 0.916
Web of Science (2017): Impact factor 3.063
Web of Science (2017): Indexed yes
BFI (2016): BFI-level 1
Scopus rating (2016): CiteScore 3.12 SJR 0.742 SNIP 0.936
Web of Science (2016): Impact factor 3.348
BFI (2015): BFI-level 1
Scopus rating (2015): CiteScore 3.39 SJR 0.952 SNIP 1.086
BFI (2014): BFI-level 1
Scopus rating (2014): CiteScore 3.42 SJR 1.026 SNIP 1.111
Web of Science (2014): Impact factor 3.444
BFI (2013): BFI-level 1
Scopus rating (2013): CiteScore 3.32 SJR 0.924 SNIP 1.112
Web of Science (2013): Impact factor 3.274
ISI indexed (2013): ISI indexed yes
BFI (2012): BFI-level 1
Scopus rating (2012): CiteScore 3.39 SJR 0.99 SNIP 1.243
Web of Science (2012): Impact factor 3.197
ISI indexed (2012): ISI indexed yes
Web of Science (2012): Indexed yes
BFI (2011): BFI-level 1
Scopus rating (2011): CiteScore 3.44 SJR 1.021 SNIP 1.308
Web of Science (2011): Impact factor 3.354
ISI indexed (2011): ISI indexed yes
Web of Science (2011): Indexed yes
BFI (2010): BFI-level 1
Scopus rating (2010): SJR 0.955 SNIP 1.216
Web of Science (2010): Impact factor 3.317
Web of Science (2010): Indexed yes
BFI (2009): BFI-level 1
Web of Science (2009): Indexed yes
BFI (2008): BFI-level 1
Scopus rating (2008): SJR 1.445 SNIP 1.358
Web of Science (2008): Indexed yes
Scopus rating (2007): SJR 1.543 SNIP 1.435
Web of Science (2007): Indexed yes
Scopus rating (2006): SJR 0.891 SNIP 1.194
Web of Science (2006): Indexed yes
Scopus rating (2005): SJR 0.992 SNIP 1.235
Web of Science (2005): Indexed yes
Scopus rating (2004): SJR 0.837 SNIP 1.113
Scopus rating (2003): SJR 0.879 SNIP 1.118
Scopus rating (2002): SJR 0.731 SNIP 1.041
Web of Science (2002): Indexed yes
Scopus rating (2001): SJR 0.617 SNIP 0.801
Web of Science (2001): Indexed yes
Scopus rating (2000): SJR 0.492 SNIP 0.843
Scopus rating (1999): SJR 0.55 SNIP 0.912
Original language: English
Keywords: Density functional theory, Reorganization energy, Electron transfer, Iron–sulfur proteins, Reduction potential
DOIs:
10.1016/j.jinorgbio.2007.07.025
URLs:
http://www.sciencedirect.com/science?_ob=ArticleURL&_udi=B6TGG-4P961WP-1&_user=10&_rdoc=1&_fmt=&_orig=search&_sort=d&view=c&acct=C000050221&_version=1&_urlVersion=0&_userid=10&md5=42034d473dcd78eb147d6705a9783ce7
Source: orbit
Source-ID: 222378
Research output: Research - peer-review ; Journal article – Annual report year: 2008