# Supporting Information for "Quantum Mechanics / Molecular Mechanics Method combined with Hybrid All-Atom and Coarse-Grained Model: Theory and Application on Redox Potential Calculations"

Lin Shen and Weitao Yang*

Department of Chemistry, Duke University, Durham, North Carolina 27708, USA

Table 1: Average restraint energies per TIP3P water molecule of the aqueous ruthenium and iron in the reduced and oxidized states calculated with QM/TIP3P/PW and QM/TIP3P/BMW methods by varying $\varepsilon_{\mathrm{rmix}}$(AA-CG). Energy units are in kcal/mol.

| | TIP3P/PW | | | TIP3P/BMW | | |
|---|---|---|---|---|---|---|
| $\varepsilon_{\mathrm{rmix}}$ | 1.45 | 1.75 | 2.50 | 1.00 | 1.10 | 1.30 |
| $\mathrm{Ru}\,(\mathrm{H_2O})_6^{2+}$ | 0.06 | 0.02 | 0.01 | 0.12 | 0.11 | 0.06 |
| $\mathrm{Ru}\,(\mathrm{H_2O})_6^{3+}$ | 0.06 | 0.03 | 0.01 | 0.13 | 0.11 | 0.05 |
| $\mathrm{Fe}\,(\mathrm{H_2O})_6^{2+}$ | 0.08 | 0.02 | 0.01 | 0.14 | 0.11 | 0.05 |
| $\mathrm{Fe}\,(\mathrm{H_2O})_6^{3+}$ | 0.09 | 0.03 | 0.01 | 0.13 | 0.11 | 0.05 |

Table 2: External electrostatic potentials generated by solvent molecules at the center of the QM subsystems of the aqueous ruthenium and iron in the reduced and oxidized states calculated with QM/TIP3P/PW and QM/TIP3P/BMW methods by varying $\varepsilon_{\mathrm{rmix}}$(AA-CG). The results obtained with QM/(AA+CG) method are also listed as a comparison. Energy units are in hartree.

| | AA + CG | TIP3P/PW | | | TIP3P/BMW | | |
|---|---|---|---|---|---|---|---|
| $\varepsilon_{\mathrm{rmix}}$ | | 1.45 | 1.75 | 2.50 | 1.00 | 1.10 | 1.30 |
| $\mathrm{Ru}\,(\mathrm{H_2O})_6^{2+}$ | -0.3067 | -0.2960 | -0.2803 | -0.2544 | -0.3032 | -0.2855 | -0.2722 |
| $\mathrm{Ru}\,(\mathrm{H_2O})_6^{3+}$ | -0.4571 | -0.4447 | -0.4212 | -0.3937 | -0.4561 | -0.4386 | -0.4215 |
| $\mathrm{Fe}\,(\mathrm{H_2O})_6^{2+}$ | -0.3158 | -0.3146 | -0.2927 | -0.2663 | -0.3174 | -0.2973 | -0.2842 |
| $\mathrm{Fe}\,(\mathrm{H_2O})_6^{3+}$ | -0.4711 | -0.4624 | -0.4390 | -0.4067 | -0.4723 | -0.4527 | -0.4364 |

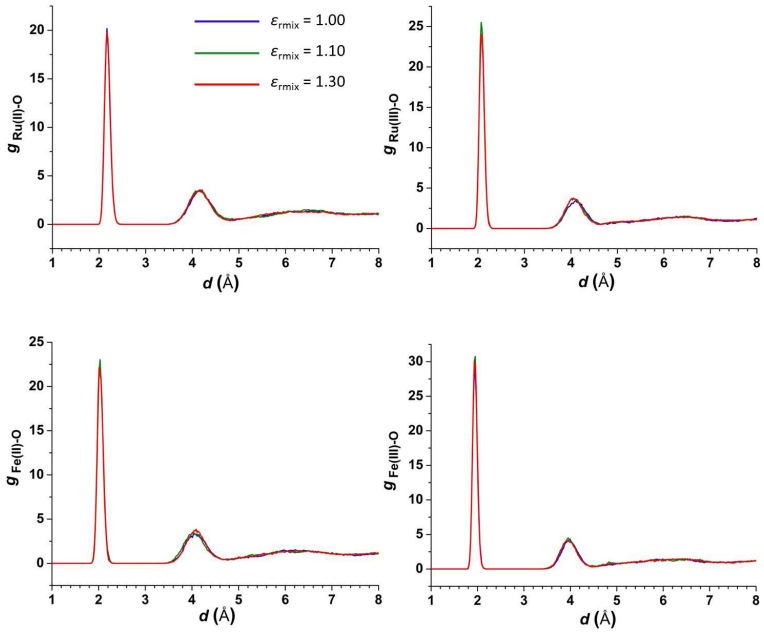


Figure 1: Metal–oxygen radial distribution functions of aqueous ruthenium and iron in the reduced and oxidized states with QM/TIP3P/BMW model with different relative dielectric permittivities between AA and CG.

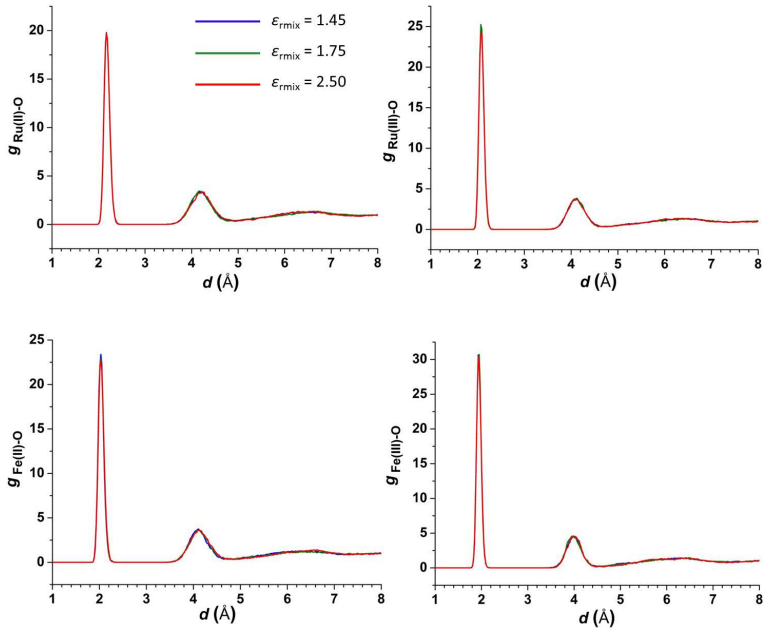


Figure 2: Metal–oxygen radial distribution functions of aqueous ruthenium and iron in the reduced and oxidized states with QM/TIP3P/PW model with different relative dielectric permittivities between AA and CG.

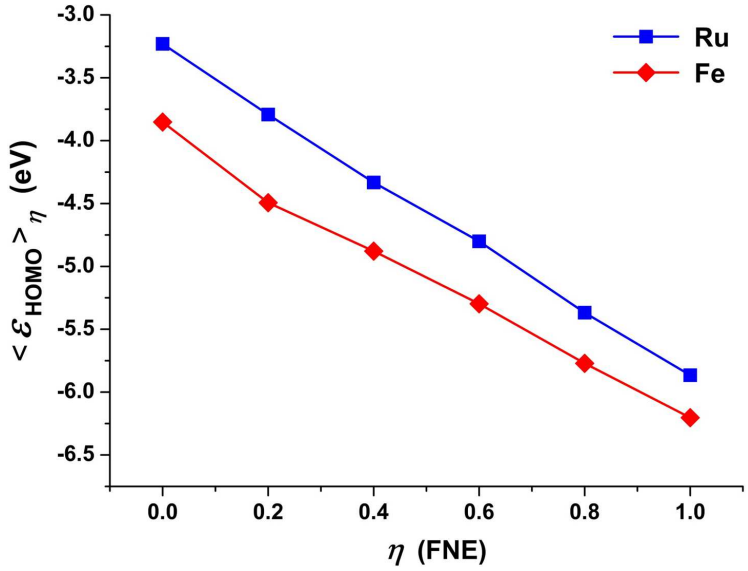


Figure 3: Ensemble average of $\varepsilon_{\mathrm{HOMO}}$ at different fractional numbers of electrons with QM/(AA+CG) model.

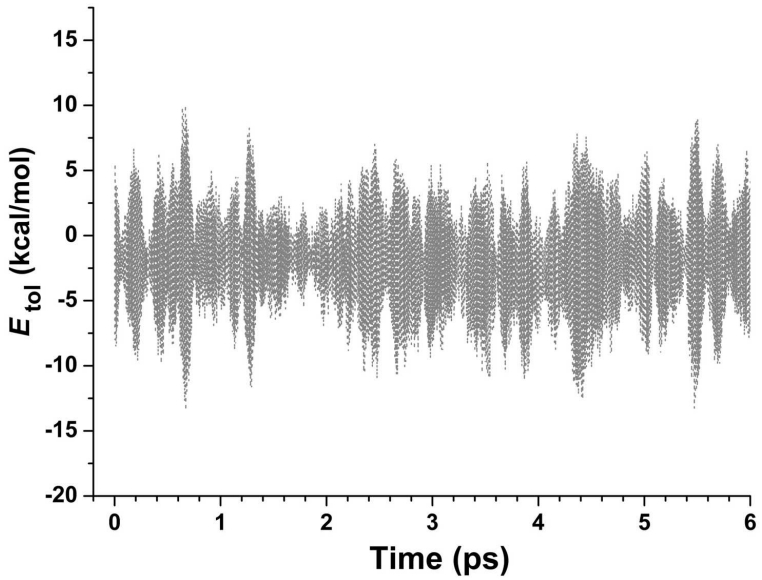


Figure 4: Total energy as a function of time for one representative trajectory in aqueous ruthenium(II) by using QM/(AA+CG) model for NVE simulation with integration time step as 1 fs.