Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Thermodynamic Integration Network Study of Electron Transfer: from Proteins to Aggregates

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Supplementary Information

S1. NrfA thermodynamic network analysis output

```
least squares thermodynamic network analysis

5 species

8 equations

extended coefficient matrix

0.00 -1.00 1.00 0.00 0.00 12.75

1.00 0.00 -1.00 0.00 0.00 4.25

0.00 1.00 0.00 0.00 -1.00 -1.10

1.00 -1.00 0.00 0.00 0.00 17.42

0.00 0.00 -1.00 1.00 0.00 -1.39

0.00 0.00 0.00 -1.00 1.00 -13.07

-1.00 0.00 0.00 0.00 1.00 -17.98

0.00 1.00 0.00 0.00 0.00 0.00

solution

i, residue, Delta G (kcal/mol), hole potential (meV)

1 1 17.730 768.8

2 2 0.000 0.0

3 3 13.410 581.5

4 4 12.610 546.8

5 5 0.130 5.6
```

```
eq. no., residual (kcal/mol)
1 0.660
2 0.070
3 0.970
4 0.310
5 0.590
6 0.590
7 0.380
8 0.000

0.96999999999999864 = maximum residual (kcal/mol)
0.87903356022395407 = rms error (kcal/mol)
```

S2. NrfH thermodynamic network analysis output

```
least squares thermodynamic network analysis

4 species

5 equations

extended coefficient matrix

-1.00 1.00 0.00 0.00 -12.35

0.00 -1.00 1.00 0.00 2.57

0.00 0.00 -1.00 1.00 2.37

-1.00 0.00 0.00 1.00 -8.05

0.00 1.00 0.00 0.00 0.00

solution

i, residue, Delta G (kcal/mol), hole potential (meV)

1 1 12.510 542.5

2 2 0.000 0.0

3 3 2.410 104.5

4 4 4.620 200.3
```

```
eq. no., residual (kcal/mol)
1 0.160
2 0.160
3 0.160
4 0.160
5 0.000

0.16000000000000636 = maximum residual (kcal/mol)
0.32000000000000095 = rms error (kcal/mol)
```

S3. NrfAH thermodynamic network analysis output

```
least squares thermodynamic network analysis

9 species

12 equations

extended coefficient matrix

-1.00 0.00 1.00 0.00 0.00 0.00 0.00 0.00 0.00 -6.55

0.00 -1.00 1.00 0.00 0.00 0.00 0.00 0.00 0.00 14.12

0.00 0.00 -1.00 1.00 0.00 0.00 0.00 0.00 0.00 -0.98

0.00 0.00 0.00 -1.00 1.00 0.00 0.00 0.00 0.00 -14.54

-1.00 0.00 0.00 1.00 0.00 0.00 0.00 0.00 0.00 -6.36

0.00 1.00 0.00 0.00 0.00 0.00 0.00 0.00 -1.00 -1.40

0.00 0.00 0.00 0.00 1.00 0.00 0.00 0.00 -1.00 -1.28

0.00 0.00 0.00 0.00 0.00 -1.00 1.00 0.00 0.00 -11.65

0.00 0.00 0.00 0.00 0.00 0.00 -1.00 1.00 0.00 0.85

0.00 0.00 0.00 0.00 0.00 0.00 0.00 -1.00 1.00 0.38

0.00 0.00 0.00 0.00 0.00 -1.00 0.00 0.00 1.00 -9.83

0.00 0.00 0.00 0.00 0.00 0.00 1.00 0.00 0.00 0.00
```

```
solution i, residue, Delta G (kcal/mol), hole potential (meV)
1 1 20.970 909.3
2 2 0.367 15.9
3 3 14.729 638.7
4 4 14.301 620.1
5 5 0.003 0.1
6 6 11.502 498.8
7 7 0.000 0.0
8 8 0.998 43.3
9 9 1.525 66.1

eq. no., residual (kcal/mol)
1 0.309
2 0.242
3 0.551
4 0.242
5 0.309
6 0.242
7 0.242
8 0.148
9 0.148
10 0.147
11 0.147
12 0.000

0.55142857142856938 = maximum residual (kcal/mol)
```

```
0.52183855376014154 = rms error (kcal/mol)
```

Figure 1 Cartoon representation of the NrfHA aggregate and the energetics of the biochemically relevant electron transfer steps between the NrfHA dimer hemes studied here. Data are in kcal/mol, they originate from a least squares thermodynamic integration network analysis.