# Supplemental Materials

**Table 1:** Calculated imaginary vibration frequencies $(\mathrm{cm}^{-1})$ of the transition states of various ORR elementary steps on pure Pt and modified Pt (Pt/Ni, Pt/Co, and Pt/Fe) (111) surfaces.

|  | Pt | Pt/Ni | Pt/Co | Pt/Fe |
|---|---|---|---|---|
| O–O bond scission |  |  |  |  |
| $O_2^* \longrightarrow O^* + O^*$ | 243 | 368 | 234 | 482 |
| $OOH^* \longrightarrow O^* + OH^*$ | 238 | 341 | 467 | 461 |
| $H_2O_2^* \longrightarrow OH^* + OH^*$ | 327 | 186 | 113 | 134 |
| Protonation |  |  |  |  |
| $O_2^* + H^+ + e^- \longrightarrow OOH^*$ | 627 | 484 | 498 | 482 |
| $OOH^* + H^+ + e^- \longrightarrow H_2O_2^*$ | 386 | 281 | 296 | 335 |
| $O^* + H^+ + e^- \longrightarrow OH^*$ | 679 | 580 | 595 | 403 |
| $OH^* + H^+ + e^- \longrightarrow H_2O^*$ | 143 | 153 | 239 | 321 |

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**Table 2:** Calculated representative bond lengths (in unit of Å) in the initial (I), tranistion (T), and final (F) states of various ORR elementary steps on pure Pt and modified Pt (Pt/Ni, Pt/Co, and Pt/Fe) (111) surfaces. For the three O–O bond scission reactions, the lengths of O–O bond are given. For the four protonation reactions, the lengths of O-H bond are given.

| reactions | Pt | | | Pt/Ni | | | Pt/Co | | | Pt/Fe | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | I | T | F | I | T | F | I | T | F | I | T | F |
| O–O bond scission | | | | | | | | | | | | |
| $O_2^* \longrightarrow O^* + O^*$ | 1.35 | 1.98 | 2.82 | 1.33 | 1.99 | 2.82 | 1.34 | 1.98 | 2.82 | 1.31 | 1.95 | 2.82 |
| $OOH^* \longrightarrow O^* + OH^*$ | 1.47 | 1.81 | 3.43 | 1.46 | 1.88 | 3.41 | 1.46 | 1.85 | 3.43 | 1.46 | 1.88 | 3.41 |
| $H_2O_2^* \longrightarrow OH^* + OH^*$ | 1.47 | 1.66 | 2.82 | 1.47 | 1.47 | 2.82 | 1.47 | 1.47 | 2.83 | 1.47 | 1.47 | 2.81 |
| Protonation | | | | | | | | | | | | |
| $O_2^* + H^+ + e^- \longrightarrow OOH^*$ | 2.74 | 1.53 | 0.99 | 2.87 | 1.88 | 0.99 | 2.92 | 1.88 | 0.98 | 1.31 | 1.85 | 0.98 |
| $OOH^* + H^+ + e^- \longrightarrow H_2O_2^*$ | 2.93 | 1.56 | 0.98 | 3.05 | 1.75 | 0.98 | 3.00 | 1.78 | 0.98 | 2.94 | 1.79 | 0.98 |
| $O^* + H^+ + e^- \longrightarrow OH^*$ | 3.28 | 2.10 | 0.98 | 3.28 | 1.56 | 0.98 | 3.27 | 1.74 | 0.98 | 3.28 | 1.71 | 0.98 |
| $OH^* + H^+ + e^- \longrightarrow H_2O^*$ | 2.69 | 1.81 | 0.98 | 2.68 | 2.00 | 0.98 | 2.69 | 1.93 | 0.98 | 2.70 | 1.93 | 0.98 |