Supporting Information

A Model for Radiolysis in a Flowing-Water Target during High-Intensity Proton Irradiation

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Figure S1. Schematic diagram of the isotope harvesting system. Reprinted with permission from ref 2. Copyright 2020 Elsevier.
Figure S2. The passage of the proton beam through the layers of the flowing-water target. After passing through the Ti64 front window, the proton beam penetrates 1.2 mm deep into the first water layer and deposits the majority of its power within the Bragg peak at the end of its path. The Stopping and Range of Ions in Matter compilation (SRIM 2013) was used to estimate the energy of the protons as they travel through the layers of Ti64 and water.¹

Estimation of the H₂, H₂O₂, and O₂ levels

The concentration levels of H₂, H₂O₂, and O₂ were computed by numerical integration of the following differential equations using time steps of 100 seconds.

\[ \frac{d[H₂]_{water}}{dt} = \frac{P(t) \times G \times 10^4}{N_A} - \frac{[H₂]_{water} \times \beta_1}{V_{water}} + \frac{[H₂]_{headspace} \times \beta_1 \times K_H \times R \times T \times 10^{-6}}{V_{headspace}} \]

(S1)

where \( P(t) \) represents the power of the \(^1\)H⁺ beam at time \( t \) (eV/s) and \( G \) is the escape \( H_2 \) yield (molecules/100 eV). The kinetics of the concurrent exchange of \( H_2 \) between water and headspace are described by the factor \( \beta_1 \) (≈ 0.05 L/s) while the amount of \( H_2 \) in the headspace is represented by \([H_2]_{headspace}\). \( R \) is the ideal gas constant (8.21 x 10⁻² L*atm/K*mol), \( K_H \) is Henry’s law constant for \( H_2 \) (780 μM/atm) and \( T \) is the bulk water temperature (≈298 K). The volumes of the water \((V_{water})\) and headspace \((V_{headspace})\) amount to 36 L and 10 L, respectively. To determine the \( H_2 \) levels at the end of the gas loop, the \( H_2 \) produced in the water first passes into the headspace, then to the traps, before it is analyzed by the sensor (% \( H_2 \) at sensor). The equations for these three steps are outlined in the following three equations.
**H₂ levels in the headspace**

\[
\frac{[\text{H}_2]_{\text{headspace}}}{dt} = \frac{[\text{H}_2]_{\text{water}} \times \beta_1}{V_{\text{water}}} - \frac{[\text{H}_2]_{\text{headspace}} \times \beta_1 \times K_H \times R \times \tau \times 10^{-6}}{V_{\text{headspace}}} - \frac{[\text{H}_2]_{\text{headspace}} \times f}{V_{\text{headspace}}} \quad (S2)
\]

To approximate the amount of H₂ in the headspace, \([\text{H}_2]_{\text{headspace}}\), a gas exchange between the water and the headspace and a removal by the He-based gas flow (\(f\), in L/s) are considered. The volumes of the water (\(V_{\text{water}}\)) and headspace (\(V_{\text{headspace}}\)) amount to 36 L and 10 L, respectively.

**H₂ levels in the traps**

\[
\frac{[\text{H}_2]_{\text{traps}}}{dt} = \frac{[\text{H}_2]_{\text{headspace}} \times f}{V_{\text{headspace}}} - \frac{[\text{H}_2]_{\text{traps}} \times f}{V_{\text{traps}}} \quad (S3)
\]

The H₂ amount in the traps, \([\text{H}_2]_{\text{traps}}\), is approximated by considering a transport from the headspace and a concurrent removal with the He gas flow. The volume of the traps (\(V_{\text{traps}}\)) is estimated to be 1.4 L.

**H₂ levels at the sensor**

\[
[\text{H}_2]_{\text{sensor}} \% = \frac{[\text{H}_2]_{\text{traps}} \times V_m \times 10^{-4}}{V_{\text{traps}}} \quad (S4)
\]

The H₂ levels at the sensor, \([\text{H}_2]_{\text{sensor}}\), are given in % H₂ and consider the H₂ transport from the traps. \(V_m\) is the molar gas volume of 22.4 L.

**H₂O₂ levels in the water**

The net balance of H₂O₂ is governed by a beam-induced formation (\(P(t) \times G\)) and a continuous peroxide decomposition by the catalytic converter after the third irradiation period. These approximations lead to equation S5, which is used to predict the hydrogen peroxide concentration \([\text{H}_2\text{O}_2]\) (\(\mu\text{M}\)) in the water:

\[
\frac{d[\text{H}_2\text{O}_2]}{dt} = \frac{P(t) \times G \times 10^4}{N_A \times V_{\text{water}}} - k_{\text{KLC}}[\text{H}_2\text{O}_2] \quad (S5)
\]

where \(N_A\) is the Avogadro constant (6.022 x 10²³ particles/mol), the total water volume (\(V_{\text{water}}\)) is 36 L and \(k_{\text{KLC}}\) is the hydrogen peroxide decomposition rate of the catalytic converter (3.34 x 10⁻⁵ s⁻¹).²

**Dissolved O₂ levels in the water**

\[
\frac{d[\text{O}_2]}{dt} = \frac{P(t) \times G \times 10^4}{N_A \times V_{\text{water}}} - a[\text{O}_2] + \frac{k_{\text{KLC}}[\text{H}_2\text{O}_2]}{2} \quad (S6)
\]

The amount of dissolved oxygen in the water \([\text{O}_2]\) (\(\mu\text{M}\)) was estimated by using the beam power-dependent production (\(P(t) \times G\)) with a constant deaeration of O₂ from the water. Though the deaeration rate, \(a\), was previously measured offline, it was allowed to vary in our model as fitting parameter (2.50 x 10⁻⁴ s⁻¹). This rate is specific to the operation conditions of our system and can be slightly influenced by the experimental conditions. The input of additional oxygen from the catalytic H₂O₂ decomposition is described by the decomposition rate of the catalytic converter, \(k_{\text{KLC}}\).
Table S1. The determined power functions describing the steady-state concentrations of selected radical, ionic, and molecular radiolytic species (in μM) in dependency of the dose rate (D_R, in Gy/s), delivered by 9.7 MeV protons.

| Species          | Power function                                      |
|------------------|-----------------------------------------------------|
| \([H_2]_{SS}\)   | \(= 2.917 \times 10^{-5} \times D_R^{0.372}\)       |
| \([H_2O_2]_{SS}\)| \(= 2.348 \times 10^{-5} \times D_R^{0.372}\)       |
| \([O_2]_{SS}\)   | \(= 2.736 \times 10^{-6} \times D_R^{0.371}\)       |
| \([OH]_{SS}\)    | \(= 6.008 \times 10^{-11} \times D_R^{0.626}\)      |
| \([e_{aq}]_{SS}\)| \(= 3.699 \times 10^{-13} \times D_R^{0.569}\)      |
| \([O_2^-]_{SS}\) | \(= 1.811 \times 10^{-7} \times D_R^{0.332}\)       |
| \([HO_2]_{SS}\)  | \(= 2.211 \times 10^{-9} \times D_R^{0.660}\)       |
| \([H]_{SS}\)     | \(= 3.493 \times 10^{-12} \times D_R^{0.624}\)      |

Table S2. The steady-state concentrations of \(H_2\) and \(H_2O_2\) were computed for each irradiation segment (\([H_2]_{SS, segment}\) and \([H_2O_2]_{SS, segment}\) in μM) by applying the determined regression equations (Supporting Information, Table S1) together with the calculated average dose rates. The average dose rates were computed by considering the average beam power deposited into the beam strike volume. A complete dissolution of molecular hydrogen at steady-state concentrations exceeding its solubility at standard conditions was possible because of the positive pressure within the harvesting system.

| Irradiation segment # | av. Beam current (μA) | av. Dose rate (kGy/s) | \([H_2]_{SS, segment}\) (μM) | \([H_2O_2]_{SS, segment}\) (μM) |
|-----------------------|------------------------|-----------------------|-----------------------------|-----------------------------|
| 1                     | 5.5                    | 1.29                  | 418                         | 337                         |
| 2                     | 18.2                   | 4.23                  | 645                         | 524                         |
| 3                     | 27.1                   | 6.30                  | 753                         | 607                         |
| 4                     | 32.6                   | 7.60                  | 808                         | 651                         |
Table S3 a–d. The effective G values (G_{eff}) of H₂, H₂O₂, and O₂ for four different beam current ranges, i.e. (a) 0.5–7.5 μA, (b) 7.5–15.0 μA, (c) 15.0–24.5 μA and (d) 24.5–34.0 μA. The computation was performed with the simulation model for the flowing-water target for 9.7 MeV protons. The [H₂], [H₂O₂], and [O₂] represent the baseline and in-flowing solution concentrations used to compute the respective effective yields of H₂, H₂O₂, and O₂. The following data sets were used to determine the functional dependencies of G_{eff} on the beam current (I) and the concentrations of H₂, H₂O₂, and O₂.

Table S3 a. Beam current range: 0.5–7.5 μA

| # | I (μA) | [H₂] (μM) | [H₂O₂] (μM) | [O₂] (μM) | G_{eff}(H₂) | G_{eff}(H₂O₂) | G_{eff}(O₂) |
|---|-------|-----------|-------------|-----------|-------------|--------------|-------------|
| 1 | 0.50  | 45.00     | 107.50      | 47.50     | 0.516       | 0.119        | 0.092       |
| 2 | 1.00  | 10.00     | 30.00       | 30.00     | 0.591       | 0.464        | 0.009       |
| 3 | 1.50  | 22.50     | 33.75       | 20.00     | 0.550       | 0.405        | 0.035       |
| 4 | 1.50  | 20.00     | 22.50       | 30.00     | 0.569       | 0.496        | -0.008      |
| 5 | 2.00  | 50.00     | 100.00      | 75.00     | 0.546       | 0.329        | 0.056       |
| 6 | 2.25  | 22.50     | 53.75       | 33.75     | 0.558       | 0.371        | 0.058       |
| 7 | 2.25  | 67.50     | 53.75       | 33.75     | 0.497       | 0.362        | 0.030       |
| 8 | 2.25  | 67.50     | 161.25      | 33.75     | 0.558       | 0.078        | 0.204       |
| 9 | 2.25  | 67.50     | 161.25      | 33.75     | 0.496       | 0.077        | 0.171       |
| 10| 2.25  | 22.50     | 53.75       | 61.25     | 0.575       | 0.436        | 0.025       |
| 11| 2.25  | 67.50     | 53.75       | 61.25     | 0.527       | 0.434        | -0.002      |
| 12| 2.25  | 22.50     | 161.25      | 61.25     | 0.570       | 0.156        | 0.164       |
| 13| 2.25  | 67.50     | 161.25      | 61.25     | 0.517       | 0.160        | 0.133       |
| 14| 2.50  | 35.00     | 40.00       | 25.00     | 0.531       | 0.391        | 0.040       |
| 15| 3.00  | 30.00     | 45.00       | 20.00     | 0.531       | 0.361        | 0.059       |
| 16| 3.00  | 70.00     | 120.00      | 50.00     | 0.508       | 0.240        | 0.098       |
| 17| 4.00  | 0.00      | 107.50      | 47.50     | 0.578       | 0.282        | 0.121       |
| 18| 4.00  | 90.00     | 107.50      | 47.50     | 0.489       | 0.272        | 0.078       |
| 19| 4.00  | 45.00     | 0.00        | 47.50     | 0.543       | 0.542        | -0.031      |
| 20| 4.00  | 45.00     | 215.00      | 47.50     | 0.534       | 0.090        | 0.192       |
| 21| 4.00  | 45.00     | 107.50      | 20.00     | 0.511       | 0.211        | 0.127       |
| 22| 4.00  | 45.00     | 107.50      | 75.00     | 0.546       | 0.329        | 0.075       |
| 23| 4.00  | 45.00     | 107.50      | 47.50     | 0.531       | 0.276        | 0.099       |
| 24| 4.00  | 80.00     | 161.25      | 47.50     | 0.499       | 0.175        | 0.132       |
| 25| 4.00  | 22.50     | 100.00      | 47.50     | 0.554       | 0.294        | 0.102       |
| 26| 4.00  | 61.25     | 90.00       | 20.00     | 0.491       | 0.244        | 0.100       |
| 27| 4.00  | 30.00     | 210.00      | 20.00     | 0.536       | 0.031        | 0.227       |
| 28| 5.00  | 60.00     | 160.00      | 45.00     | 0.514       | 0.190        | 0.136       |
| 29| 5.00  | 40.00     | 130.00      | 40.00     | 0.529       | 0.230        | 0.125       |
| 30| 5.75  | 22.50     | 53.75       | 33.75     | 0.537       | 0.363        | 0.067       |
| 31| 5.75  | 67.50     | 53.75       | 33.75     | 0.497       | 0.356        | 0.049       |
| 32| 5.75  | 22.50     | 161.25      | 33.75     | 0.540       | 0.180        | 0.158       |
| 33| 5.75  | 67.50     | 161.25      | 33.75     | 0.500       | 0.176        | 0.140       |
| #  | \( I \) (\( \mu A \)) | \([\text{H}_2]\) (\( \mu M \)) | \([\text{H}_2\text{O}_2]\) (\( \mu M \)) | \([\text{O}_2]\) (\( \mu M \)) | \( G_{\text{eff}}(\text{H}_2) \) (molecules/100 eV) | \( G_{\text{eff}}(\text{H}_2\text{O}_2) \) (molecules/100 eV) | \( G_{\text{eff}}(\text{O}_2) \) (molecules/100 eV) |
|----|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 34 | 5.75            | 22.50           | 53.75           | 61.25           | 0.553           | 0.408           | 0.047           |
| 35 | 5.75            | 67.50           | 53.75           | 61.25           | 0.519           | 0.403           | 0.032           |
| 36 | 5.75            | 22.50           | 161.25          | 61.25           | 0.551           | 0.230           | 0.136           |
| 37 | 5.75            | 67.50           | 161.25          | 61.25           | 0.516           | 0.227           | 0.118           |
| 38 | 6.00            | 90.00           | 40.00           | 20.00           | 0.460           | 0.350           | 0.037           |
| 39 | 6.25            | 40.00           | 107.50          | 33.75           | 0.521           | 0.265           | 0.107           |
| 40 | 7.00            | 67.50           | 200.00          | 53.75           | 0.511           | 0.177           | 0.144           |
| 41 | 7.00            | 80.00           | 200.00          | 65.00           | 0.507           | 0.194           | 0.132           |
| 42 | 7.00            | 70.00           | 180.00          | 50.00           | 0.507           | 0.195           | 0.133           |
| 43 | 7.50            | 45.00           | 107.50          | 47.50           | 0.521           | 0.292           | 0.094           |

**Table S3 b.** Beam current range: 7.5–15.0 \( \mu A \)
| #  | I   | [H₂] | [H₂O₂] | [O₂]  | G_{eff}(H₂) | G_{eff}(H₂O₂) | G_{eff}(O₂) |
|----|-----|------|--------|-------|--------------|---------------|-------------|
|    | (μA)| (μM)| (μM)   | (μM)  | (molecules/100 eV) | (molecules/100 eV) | (molecules/100 eV) |
| 27 | 11.25 | 100.00 | 187.50 | 75.00 | 0.494 | 0.243 | 0.106 |
| 28 | 11.25 | 25.00 | 37.50 | 37.50 | 0.515 | 0.364 | 0.061 |
| 29 | 12.000 | 50.00 | 150.00 | 62.50 | 0.513 | 0.268 | 0.105 |
| 30 | 13.125 | 37.50 | 62.50 | 25.00 | 0.496 | 0.311 | 0.079 |
| 31 | 13.125 | 112.50 | 62.50 | 25.00 | 0.452 | 0.302 | 0.061 |
| 32 | 13.125 | 37.50 | 187.50 | 25.00 | 0.503 | 0.193 | 0.140 |
| 33 | 13.125 | 112.50 | 187.50 | 25.00 | 0.462 | 0.186 | 0.123 |
| 34 | 13.125 | 37.50 | 62.50 | 75.00 | 0.520 | 0.366 | 0.060 |
| 35 | 13.125 | 112.50 | 62.50 | 75.00 | 0.485 | 0.361 | 0.044 |
| 36 | 13.125 | 37.50 | 187.50 | 75.00 | 0.521 | 0.250 | 0.118 |
| 37 | 13.125 | 112.50 | 187.50 | 75.00 | 0.486 | 0.246 | 0.102 |
| 38 | 14.000 | 85.00 | 190.00 | 70.00 | 0.495 | 0.242 | 0.110 |
| 39 | 14.500 | 77.50 | 160.00 | 80.00 | 0.501 | 0.277 | 0.095 |
| 40 | 15.000 | 75.00 | 125.00 | 50.00 | 0.488 | 0.275 | 0.091 |

**Table S3 c.** Beam current range: 15.0–24.5 μA
| #  | I (μA) | [H₂] (μM) | [H₂O₂] (μM) | [O₂] (μM) | G_{eff}(H₂) | G_{eff}(H₂O₂) | G_{eff}(O₂) |
|----|--------|----------|-------------|----------|-------------|--------------|-------------|
| 23 | 20.00  | 70.00    | 200.00      | 70.00    | 0.492       | 0.242        | 0.110       |
| 24 | 22.00  | 180.00   | 355.00      | 130.00   | 0.474       | 0.203        | 0.118       |
| 25 | 22.125 | 75.00    | 277.50      | 70.00    | 0.489       | 0.201        | 0.130       |
| 26 | 22.125 | 165.00   | 277.50      | 70.00    | 0.459       | 0.197        | 0.116       |
| 27 | 22.125 | 75.00    | 432.50      | 70.00    | 0.494       | 0.123        | 0.170       |
| 28 | 22.125 | 165.00   | 432.50      | 70.00    | 0.464       | 0.121        | 0.156       |
| 29 | 22.125 | 75.00    | 277.50      | 110.00   | 0.499       | 0.230        | 0.119       |
| 30 | 22.125 | 165.00   | 277.50      | 110.00   | 0.471       | 0.228        | 0.106       |
| 31 | 22.125 | 75.00    | 432.50      | 110.00   | 0.502       | 0.153        | 0.159       |
| 32 | 22.125 | 165.00   | 432.50      | 110.00   | 0.475       | 0.152        | 0.145       |
| 33 | 23.00  | 165.00   | 320.00      | 120.00   | 0.474       | 0.215        | 0.114       |
| 34 | 23.50  | 180.00   | 450.00      | 90.00    | 0.465       | 0.134        | 0.150       |
| 35 | 23.50  | 120.00   | 355.00      | 120.00   | 0.487       | 0.200        | 0.128       |
| 36 | 24.00  | 200.00   | 355.00      | 120.00   | 0.464       | 0.199        | 0.117       |
| 37 | 24.50  | 120.00   | 355.00      | 90.00    | 0.479       | 0.181        | 0.135       |

**Table S3 d.** Beam current range: 24.5–34.0 μA
Net water decomposition

With the deposition of ionizing radiation, bond breakages of water molecules are induced, resulting in the formation of water decomposition products. As a consequence, the material balance of H and O atoms for the escape yields of all primary radiolytic products equals the amount of decomposed water. For example, when multiplying the escape yield of every primary species created with 10 MeV protons (see Table 1 in publication) by the number of H and O atoms present in each compound, the net amount of decomposed water molecules equals 2.92. Analogously, the material balance between the number of H and O atoms in all radiolytic species, generated throughout the homogeneous regime, is equivalent to the net water decomposition. When a solution, consisting of 45 μM H₂, 107.5 μM H₂O₂, and 75 μM O₂, is irradiated with a 4 μA proton beam (scenario presented in Table S3 a, #22), the sum of H and O atoms calculated from the effective yields of all formed species, yields a net decomposition of 0.902 H₂O molecules. For a better understanding, an example calculation is given in Table S4. With this simple test, the validity of all computed effective G values (Supporting Information, Table S3 a-d) could be corroborated.
**Table S4.** Calculation of the net water decomposition of an irradiated solution consisting of 45 μM H₂, 107.5 μM H₂O₂ and 75 μM O₂ (proton beam intensity of 4 μA). For H₂, H₂O₂, and O₂ first the respective solution concentrations were subtracted from the apparent steady-state concentrations, whereas for all other species the calculation was performed directly with the apparent steady-state concentrations.

|                | OH    | H₂    | H     | HO₂⁻ | HO₂  | OH⁻   | H₂O₂  | O₂⁻   |
|----------------|-------|-------|-------|-------|-------|--------|-------|-------|
| Conc. (M)      | 7.97x10⁻⁷ | 5.26x10⁻⁵ | 1.78x10⁻⁸ | 9.81x10⁻⁸ | 7.31x10⁻⁷ | 7.61x10⁻⁸ | 3.18x10⁻⁵ | 3.26x10⁻⁶ |
| No. of molecules | 2.00x10¹⁶ | 1.32x10¹⁸ | 4.47x10¹⁴ | 2.46x10¹⁵ | 1.84x10¹⁶ | 1.91x10¹⁵ | 7.97x10¹⁷ | 8.19x10¹⁶ |
| G-eff (molec./ 100 eV) | 8.26x10⁻³ | 0.546 | 1.85x10⁻⁴ | 1.02x10⁻³ | 7.58x10⁻³ | 7.89x10⁻⁴ | 0.329 | 3.38x10⁻² |
| No. of H atoms  | 8.26x10⁻³ | 1.091 | 1.85x10⁻⁴ | 1.02x10⁻³ | 7.58x10⁻³ | 7.89x10⁻⁴ | 0.659 | -   |
| No. of O atoms  | 8.26x10⁻³ | -     | -     | 2.03x10⁻³ | 1.52x10⁻² | 7.89x10⁻⁴ | 0.659 | 6.77x10⁻² |

|                | O₂    | H⁺    | O⁻    | O₃⁻    | O₃    | HO₃   | Σ (No. of H/O atoms) | No. of H₂O molecules |
|----------------|-------|-------|-------|--------|-------|-------|----------------------|----------------------|
| Conc. (M)      | 7.19x10⁻⁶ | 3.45x10⁻⁶ | 4.35x10⁻¹⁰ | 4.16x10⁻¹⁰ | 1.31x10⁻¹⁰ | 1.24x10⁻¹² | - | - |
| No. of molecules | 1.81x10¹² | 8.65x10¹⁶ | 1.09x10¹³ | 1.04x10¹³ | 3.29x10¹² | 3.12x10¹⁰ | - | - |
| G-eff (molec./ 100 eV) | 7.46x10⁻² | 3.57x10⁻² | 4.51x10⁻⁶ | 4.31x10⁻⁶ | 1.36x10⁻⁶ | 1.29x10⁻⁸ | - | - |
| No. of H atoms  | -     | 3.57x10⁻² | -     | -     | -     | 1.29x10⁻⁸ | 1.803 | 0.902 |
| No. of O atoms  | 0.149 | -     | 4.51x10⁻⁶ | 1.29x10⁻⁵ | 4.08x10⁻⁶ | 3.86 x10⁻⁸ | 0.902 | - |

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Table S5 a–d. The functional dependencies of the effective G values (molecules/100 eV) are described by polynomial equations, and in the following tables the relevant terms and coefficients are represented for four different beam current ranges, i.e. (a) 0.5–7.5 μA, (b) 7.5–15.0 μA, (c) 15.0–24.5 μA and (d) 24.5–34.0 μA. In the example below the polynomial equation is illustrated for $G_{\text{eff}}(H_2)$ in the beam current range of 0.5–7.5 μA. To display the computed $G_{\text{eff}}(H_2)$ in molecules/100 eV the beam current (I) is inserted in μA and the concentrations of $H_2$, $H_2O_2$, and $O_2$ in μM.

$$G_{\text{eff}}(H_2) = 5.77 \times 10^{-1} - 2.87 \times 10^{-3} \times (I) - 1.94 \times 10^{-3} \times [H_2] - 9.58 \times 10^{-5} \times [H_2O_2] + 1.09 \times 10^{-3} \times [O_2] - 8.98 \times 10^{-4} \times (I)^2 + 5.00 \times 10^{-7} \times [H_2O_2]^2 - 5.65 \times 10^{-6} \times [O_2]^2 + 1.38 \times 10^{-4} \times I \times [H_2] + 1.35 \times 10^{-5} \times I \times [H_2O_2] + 7.27 \times 10^{-6} \times [H_2] \times [O_2] - 2.21 \times 10^{-6} \times [H_2O_2] \times [O_2]$$

Table S5 a. Beam current range: 0.5–7.5 μA

| Term         | Coefficient | Term         | Coefficient | Term         | Coefficient |
|--------------|-------------|--------------|-------------|--------------|-------------|
| $G_{\text{eff}}(H_2)$ |             | $G_{\text{eff}}(H_2O_2)$ |             | $G_{\text{eff}}(O_2)$ |             |
| Constant     | 5.77 x 10^{-1} | Constant     | 4.77 x 10^{-1} | Constant     | 9.13 x 10^{-3} |
| $I$          | -2.87 x 10^{-3} | $I$          | 2.58 x 10^{-2} | $I$          | 8.21 x 10^{-3} |
| $H_2$        | -1.94 x 10^{-3} | $H_2$        | -1.38 x 10^{-3} | $H_2$        | -6.98 x 10^{-4} |
| $H_2O_2$     | -9.58 x 10^{-5} | $H_2O_2$     | -4.28 x 10^{-3} | $H_2O_2$     | 1.96 x 10^{-3} |
| $O_2$        | 1.09 x 10^{-3}  | $O_2$        | 2.44 x 10^{-3} | $O_2$        | -1.80 x 10^{-3} |
| $I \times I$| -8.98 x 10^{-4} | $I \times I$| -4.55 x 10^{-3} | $I \times I$| -3.68 x 10^{-4} |
| $H_2O_2 \times H_2O_2$ | 5.00 x 10^{-7} | $H_2 \times H_2$ | 6.98 x 10^{-6} | $H_2O_2 \times H_2O_2$ | -1.56 x 10^{-6} |
| $O_2 \times O_2$ | -5.65 x 10^{-6} | $H_2O_2 \times H_2O_2$ | 3.97 x 10^{-6} | $O_2 \times O_2$ | 3.52 x 10^{-6} |
| $I \times H_2$ | 1.38 x 10^{-4} | $I \times H_2O_2$ | 3.02 x 10^{-4} | $I \times H_2$ | 5.79 x 10^{-5} |
| $I \times H_2O_2$ | 1.35 x 10^{-6} | $I \times O_2$ | -2.14 x 10^{-4} | $I \times H_2O_2$ | -1.34 x 10^{-4} |
| $H_2 \times O_2$ | 7.27 x 10^{-6} | $H_2 \times O_2$ | 1.12 x 10^{-5} | $I \times O_2$ | 1.42 x 10^{-4} |
| $H_2O_2 \times O_2$ | -2.21 x 10^{-6} |             |             |             |             |
**Table S5 b. Beam current range: 7.5–15.0 μA**

| Term     | Coefficient | Term     | Coefficient | Term     | Coefficient |
|----------|-------------|----------|-------------|----------|-------------|
| Constant | 5.41 x 10^{-1} | Constant | 3.92 x 10^{-1} | Constant | 5.46 x 10^{-2} |
| I        | -3.16 x 10^{-3} | I        | -5.12 x 10^{-4} | I        | 6.16 x 10^{-5} |
| H₂       | -1.08 x 10^{-3} | H₂       | -1.72 x 10^{-4} | H₂       | -4.55 x 10^{-4} |
| H₂O₂     | 8.79 x 10^{-5} | H₂O₂     | -2.05 x 10^{-3} | H₂O₂     | 1.03 x 10^{-3} |
| O₂       | 8.94 x 10^{-4} | O₂       | 2.16 x 10^{-3} | O₂       | -8.04 x 10^{-4} |
| H₂ x H₂  | 2.18 x 10^{-7} | I x I    | -1.66 x 10^{-4} | I x I    | 3.68 x 10^{-5} |
| O₂ x O₂  | -2.14 x 10^{-6} | H₂O₂ x H₂O₂ | 9.63 x 10^{-7} | H₂ x H₂  | 1.08 x 10^{-7} |
| I x H₂   | 2.92 x 10^{-5} | O₂ x O₂  | -2.86 x 10^{-6} | H₂O₂ x H₂O₂ | -4.53 x 10^{-7} |
| I x O₂   | -1.57 x 10^{-5} | I x H₂O₂ | 6.41 x 10^{-5} | O₂ x O₂  | 5.49 x 10^{-7} |
| H₂ x H₂O₂ | 1.79 x 10^{-7} | I x O₂   | -6.49 x 10^{-5} | I x H₂   | 1.44 x 10^{-7} |
| H₂ x O₂  | 2.67 x 10^{-6} | H₂ x H₂O₂ | 2.36 x 10^{-7} | I x H₂O₂ | -3.06 x 10^{-5} |
| H₂O₂ x O₂ | -1.40 x 10^{-6} | H₂ x O₂  | 1.20 x 10^{-6} | I x O₂   | 3.02 x 10^{-5} |
|          |              |          |              | H₂O₂ x O₂ | 3.48 x 10^{-7} |
|          |              |          |              |           | -7.00 x 10^{-7} |

**Table S5 c. Beam current range: 15.0–24.5 μA**

| Term     | Coefficient | Term     | Coefficient | Term     | Coefficient |
|----------|-------------|----------|-------------|----------|-------------|
| Constant | 5.34 x 10^{-1} | Constant | 2.89 x 10^{-1} | Constant | 1.00 x 10^{-1} |
| I        | -2.07 x 10^{-3} | I        | 4.62 x 10^{-3} | I        | -2.41 x 10^{-3} |
| H₂       | -6.40 x 10^{-4} | H₂       | -1.05 x 10^{-4} | H₂       | -2.94 x 10^{-4} |
| H₂O₂     | 3.16 x 10^{-5} | H₂O₂     | -1.09 x 10^{-3} | H₂O₂     | 5.57 x 10^{-4} |
| O₂       | 4.28 x 10^{-4} | O₂       | 1.34 x 10^{-3} | O₂       | -5.01 x 10^{-4} |
| H₂ x H₂  | 1.02 x 10^{-7} | I x I    | -1.40 x 10^{-4} | I x I    | 4.79 x 10^{-5} |
| H₂O₂ x H₂O₂ | 1.85 x 10^{-8} | H₂O₂ x H₂O₂ | 2.74 x 10^{-7} | H₂O₂ x H₂O₂ | -1.36 x 10^{-7} |
| O₂ x O₂  | -4.57 x 10^{-7} | O₂ x O₂  | -6.32 x 10^{-7} | I x H₂   | 6.03 x 10^{-6} |
| I x H₂   | 9.71 x 10^{-6} | I x H₂O₂ | 1.76 x 10^{-5} | I x H₂O₂ | -8.61 x 10^{-6} |
| I x O₂   | -2.99 x 10^{-6} | I x O₂   | -2.32 x 10^{-5} | I x O₂   | 1.16 x 10^{-5} |
| H₂ x H₂O₂ | 5.95 x 10^{-8} | H₂ x H₂O₂ | 1.26 x 10^{-7} | H₂ x O₂  | 1.05 x 10^{-7} |
| H₂ x O₂  | 6.77 x 10^{-7} | H₂ x O₂  | 4.33 x 10^{-7} | H₂O₂ x O₂ | -1.50 x 10^{-7} |
| H₂O₂ x O₂ | -2.97 x 10^{-7} |          |              |           |              |
Table S5 d. Beam current range: 24.5–34.0 μA

| Term       | Coefficient | Term       | Coefficient | Term       | Coefficient |
|------------|-------------|------------|-------------|------------|-------------|
| $G_{\text{eff}}(\text{H}_2)$ |             | $G_{\text{eff}}(\text{H}_2\text{O}_2)$ |             | $G_{\text{eff}}(\text{O}_2)$ |             |
| Constant   | $5.16 \times 10^{-1}$ | Constant   | $2.56 \times 10^{-1}$ | Constant   | $1.13 \times 10^{-1}$ |
| I          | $-1.41 \times 10^{-3}$ | I          | $3.03 \times 10^{-3}$ | I          | $-1.73 \times 10^{-3}$ |
| $\text{H}_2$ | $-4.53 \times 10^{-4}$ | $\text{H}_2$ | $-3.37 \times 10^{-5}$ | $\text{H}_2\text{O}_2$ | $3.84 \times 10^{-4}$ |
| $\text{H}_2\text{O}_2$ | $3.61 \times 10^{-5}$ | $\text{H}_2\text{O}_2$ | $-7.51 \times 10^{-4}$ | $\text{H}_2\text{O}_2$ | $3.84 \times 10^{-4}$ |
| $\text{O}_2$ | $3.48 \times 10^{-4}$ | $\text{O}_2$ | $1.06 \times 10^{-3}$ | $\text{O}_2$ | $-4.02 \times 10^{-4}$ |
| $\text{H}_2 \times \text{H}_2$ | $4.32 \times 10^{-6}$ | $\text{I} \times \text{I}$ | $-5.69 \times 10^{-5}$ | $\text{I} \times \text{I}$ | $2.21 \times 10^{-5}$ |
| $\text{O}_2 \times \text{O}_2$ | $-2.89 \times 10^{-7}$ | $\text{H}_2\text{O}_2 \times \text{H}_2\text{O}_2$ | $1.22 \times 10^{-7}$ | $\text{H}_2 \times \text{H}_2$ | $2.45 \times 10^{-8}$ |
| $\text{I} \times \text{H}_2$ | $4.69 \times 10^{-6}$ | $\text{O}_2 \times \text{O}_2$ | $-5.28 \times 10^{-7}$ | $\text{H}_2\text{O}_2 \times \text{H}_2\text{O}_2$ | $-5.79 \times 10^{-8}$ |
| $\text{I} \times \text{O}_2$ | $-1.80 \times 10^{-6}$ | $\text{I} \times \text{H}_2$ | $-8.82 \times 10^{-7}$ | $\text{O}_2 \times \text{O}_2$ | $1.33 \times 10^{-7}$ |
| $\text{H}_2 \times \text{H}_2\text{O}_2$ | $2.37 \times 10^{-8}$ | $\text{I} \times \text{H}_2\text{O}_2$ | $8.51 \times 10^{-6}$ | $\text{I} \times \text{H}_2$ | $2.90 \times 10^{-6}$ |
| $\text{H}_2 \times \text{O}_2$ | $3.36 \times 10^{-7}$ | $\text{I} \times \text{O}_2$ | $-1.21 \times 10^{-5}$ | $\text{I} \times \text{H}_2\text{O}_2$ | $-4.18 \times 10^{-6}$ |
| $\text{H}_2\text{O}_2 \times \text{O}_2$ | $-1.61 \times 10^{-7}$ | $\text{H}_2 \times \text{H}_2\text{O}_2$ | $4.93 \times 10^{-8}$ | $\text{I} \times \text{O}_2$ | $5.68 \times 10^{-6}$ |
|                                     | $\text{H}_2 \times \text{O}_2$ |                                     | $2.01 \times 10^{-7}$ | $\text{H}_2 \times \text{H}_2\text{O}_2$ | $-1.57 \times 10^{-8}$ |
|                                     | $\text{H}_2 \times \text{O}_2$ |                                     |                                     | $\text{H}_2 \times \text{O}_2$ | $5.07 \times 10^{-8}$ |
|                                     | $\text{H}_2\text{O}_2 \times \text{O}_2$ |                                     |                                     |                                     | $-6.09 \times 10^{-8}$ |

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