

Supporting Information

Catalyst-Free Ammonia Decomposition in Atmospheric Pressure Nanosecond Pulsed DBD: Energy Deposition Dynamics and Its Role in Hydrogen Production

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This Supporting Information contains additional figure captions, table notes, and associated references

Figure captions

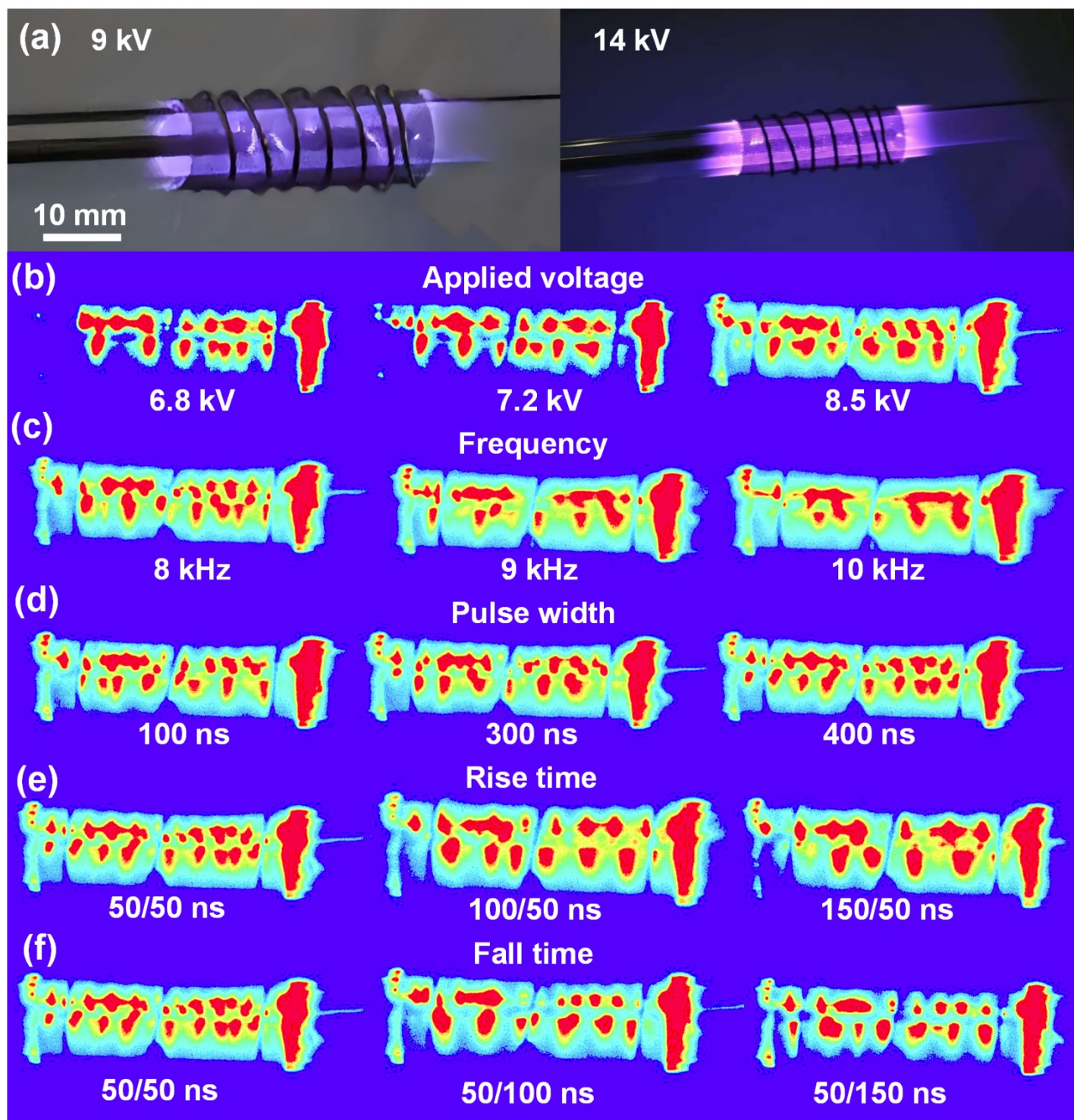


Figure S1. (a) Typical images at different voltages for AC-DBD ($Q = 0.1$ SLM, $f = 9$ kHz). These photographs were recorded using a digital camera with an exposure time of 50 ms, an aperture of $f/3.2$, and an ISO setting of 3200. (b) ICCD images at different voltages ($Q = 0.1$ SLM, $f = 7$ kHz, $T_{pw} = 200$ ns, $T_{ris}/T_{fal} = 50/50$ ns), (c) frequencies ($Q = 0.1$ SLM, $V_{pp} = 11$ kV, $T_{pw} = 200$ ns, $T_{ris}/T_{fal} = 50/50$ ns, $SIE = 9.5$ kJ·L⁻¹), (d) pulse widths ($Q = 0.1$ SLM, $f = 7$ kHz, $V_{pp} = 11$ kV, $T_{ris}/T_{fal} = 50/50$ ns, $SIE = 9.5$ kJ·L⁻¹), (e) rise times ($Q = 0.1$ SLM, $f = 7$ kHz, $V_{pp} = 11$ kV, $T_{pw} = 400$ ns, $SIE = 9.5$ kJ·L⁻¹), (f) fall times ($Q = 0.1$ SLM, $f = 7$ kHz, $V_{pp} = 11$ kV, $T_{pw} = 400$ ns, $SIE = 9.5$ kJ·L⁻¹) for NP-DBD. All ICCD images were acquired with a gate delay of 100 ns and a gate width of 20 ns. The remaining ICCD settings were kept unchanged for comparison.

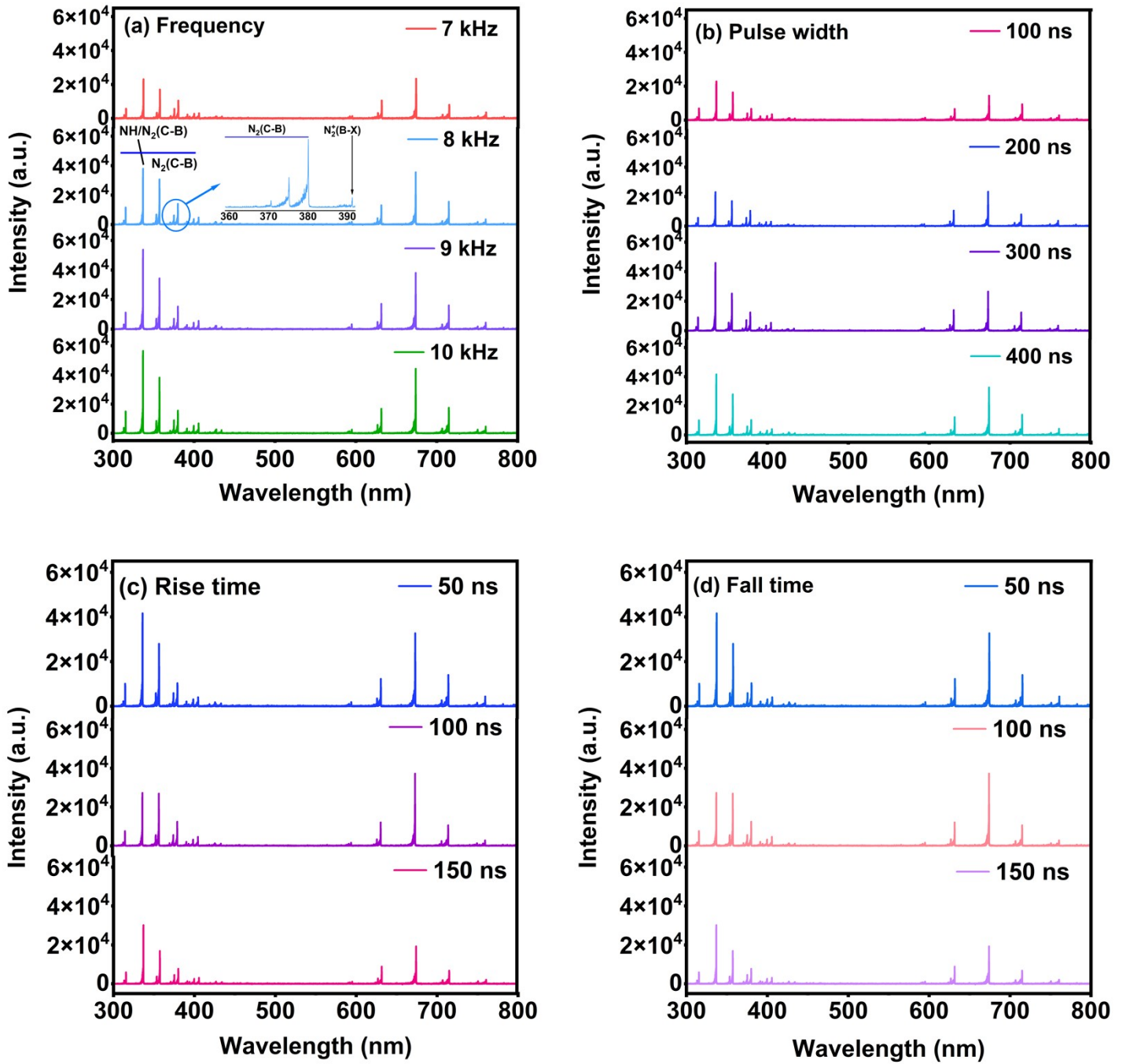


Figure S2. OES of NP-DBD at different (a) frequencies, (b) pulse widths, (c) rise times and (d) fall times ($Q = 0.1$ SLM, $V_{pp} = 8$ kV).

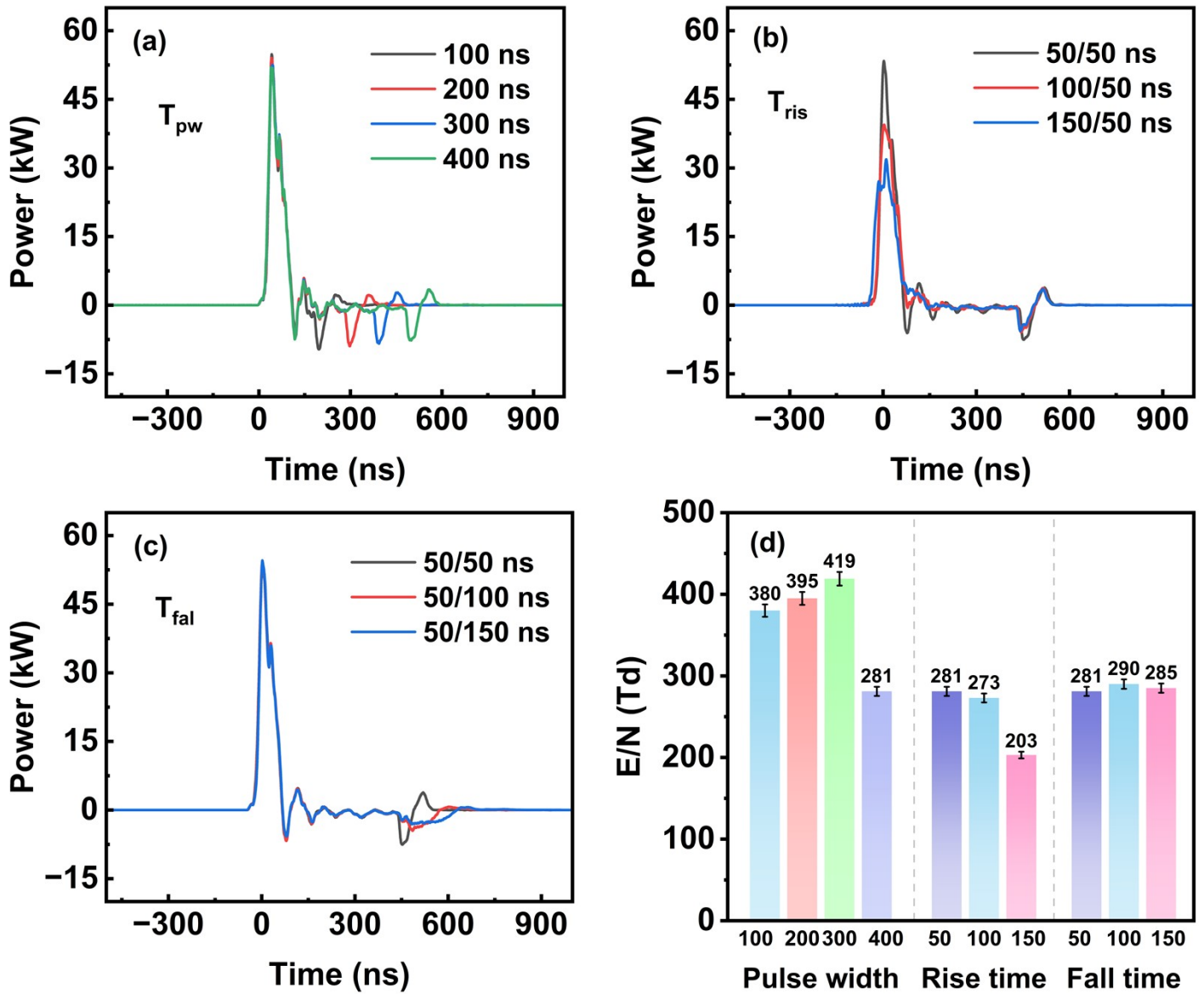


Figure S3. Instantaneous power profiles and corresponding estimated reduced electric fields under representative operating conditions at the same V_{pp} ($f = 7\text{kHz}$, $V_{pp} = 8.5\text{ kV}$). (a) Temporal power profiles $P(t)$ under different T_{pw} . (b) Temporal power profiles under different T_{ris} . (c) Temporal power profiles under different T_{fal} . (d) Estimated E/N under the corresponding conditions.

Table Notes

Table S1 summarizes representative previous studies on non-catalytic NH₃ decomposition in DBD reactors for contextual comparison with the present work. Because substantial differences exist among studies in reactor configuration, feed composition, operating conditions, and performance metric definitions, this comparison is intended mainly as a reference for positioning rather than as a strict direct ranking.

In addition to NH₃ conversion, H₂ production rate (R_{H_2} , g·h⁻¹), energy yield (E_{H_2} , g H₂·kWh⁻¹), and EUE are listed when available. For studies in which H₂ production rate and energy yield were not explicitly reported, they were estimated from the reported feed flow rate, NH₃ concentration, conversion, and input power according to Eqs. (S1) and (S2)¹:

$$R_{H_2} (g \cdot h^{-1}) = Q_{H_2} (L \cdot \min^{-1}) \times \rho_{H_2} (g \cdot L^{-1}) \times 60 (\min \cdot h^{-1}) = \frac{Q_{H_2} (L \cdot \min^{-1}) \times 2 (g \cdot \text{mol}^{-1}) \times 60 (\min \cdot h^{-1})}{22.4 (L \cdot \text{mol}^{-1})} \quad \backslash *$$

MERGEFORMAT (S1)

where Q_{H_2} is the volumetric flow rate of H₂, calculated from Eq. (7).

$$E_{H_2} (gH_2 \cdot kWh^{-1}) = \frac{R_{H_2} (g \cdot h^{-1})}{P_{in} (kW)} \quad \backslash * \text{ MERGEFORMAT (S2)}$$

In this work, the H₂-related energy term used in the EUE calculation is based on the stoichiometric H₂ amount corresponding to converted NH₃ rather than on directly measured hydrogen energy output. Therefore, the EUE reported here is used as a conversion-based comparative metric for energy utilization under different operating conditions. For the present work, the reported single values of H₂ production rate, energy yield, and EUE correspond to the operating condition with the maximum EUE.

Table S1 Comparison of non-catalytic NH₃ decomposition from various studies in DBD.

| Author/Year (Ref.) | Power supply | Rotational temperature (K) | Macroscopic reaction temperature (K) | SIE (kJ·L ⁻¹) | Feed flow rate (L·min ⁻¹) | Feed NH ₃ concentration (%) | NH ₃ conversion (%) | H ₂ production rates (g·h ⁻¹) | Energy yield (g H ₂ ·kWh ⁻¹) | EUE (%) |
|----------------------------|-----------------|----------------------------------|--|------------------------------|---|--|--------------------------------------|--|--|---------|
| Zhao/2013 ² | AC | - | - | 25.3-36.4 | 0.04 | 100 | 32-98 | 0.315 | 12.981 | 31.87 |
| Goto/2017 ³ | AC | - | - | 18 | 1 | 100 | 19 | 1.527 | 5.089 | 14.89 |
| Akiyama/2018 ⁴ | AC | - | 298-523 (Electrode surface temperature) | 0.6-15 | 0.2 | 4.9 | 2-100 | 0.079 | 1.575 | 5.1 |
| Yi/2018 ⁵ | AC | - | 643-773 (Packed bed temperature) | 15.1-24.1 | 0.12 | 100 | 8-23 | 0.222 | 4.601 | 9.1 |
| Bang/2023 ⁶ | AC | - | 300-900 (Reaction gas temperature) | 0.3-1.2 | 1 | 1 | 8-16 | 0.013 | 0.643 | 3.9 |
| Andersen/2023 ⁷ | AC | - | - | 6-18 | 0.05 | 2 | 34-82 | 0.007 | 0.439 | 1.4 |
| Wang/2024 ⁸ | AC | - | 633-743 (Packed bed temperature) | 32.4-42 | 0.04 | 100 | 17-23 | 0.074 | 2.640 | 5.5 |
| Gao/2024 ⁹ | NP | - | 298-898 (Reaction gas temperature) | - | 0.1 | 5 | 26.3-100 | - | - | - |
| This work | AC | 379-633 | 297-479 (Reaction gas temperature) | 3.7- 11.2 | 0.1 | 5 | 13.0-71.1 | 0.029 | 1.530 | 4.41 |
| | NP | 388-597 | 297-395 (Reaction gas temperature) | 5.4-9.5 | 0.1 | 5 | 34.4-99.5 | 0.040 | 2.525 | 7.19 |

Note. For the present work, the reported single values of H₂ production rate, energy yield, and EUE correspond to the operating condition with the maximum EUE.

References

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