パルス電子ビーム生成大気圧プラズマによるPFC分解処理実験

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■ 電子ビームプラズマによる CF_4 分解処理実験

実験概要、結果、高効率化への課題

■ CF_4 分解反応に関する数値計算（アルゴン希釈）

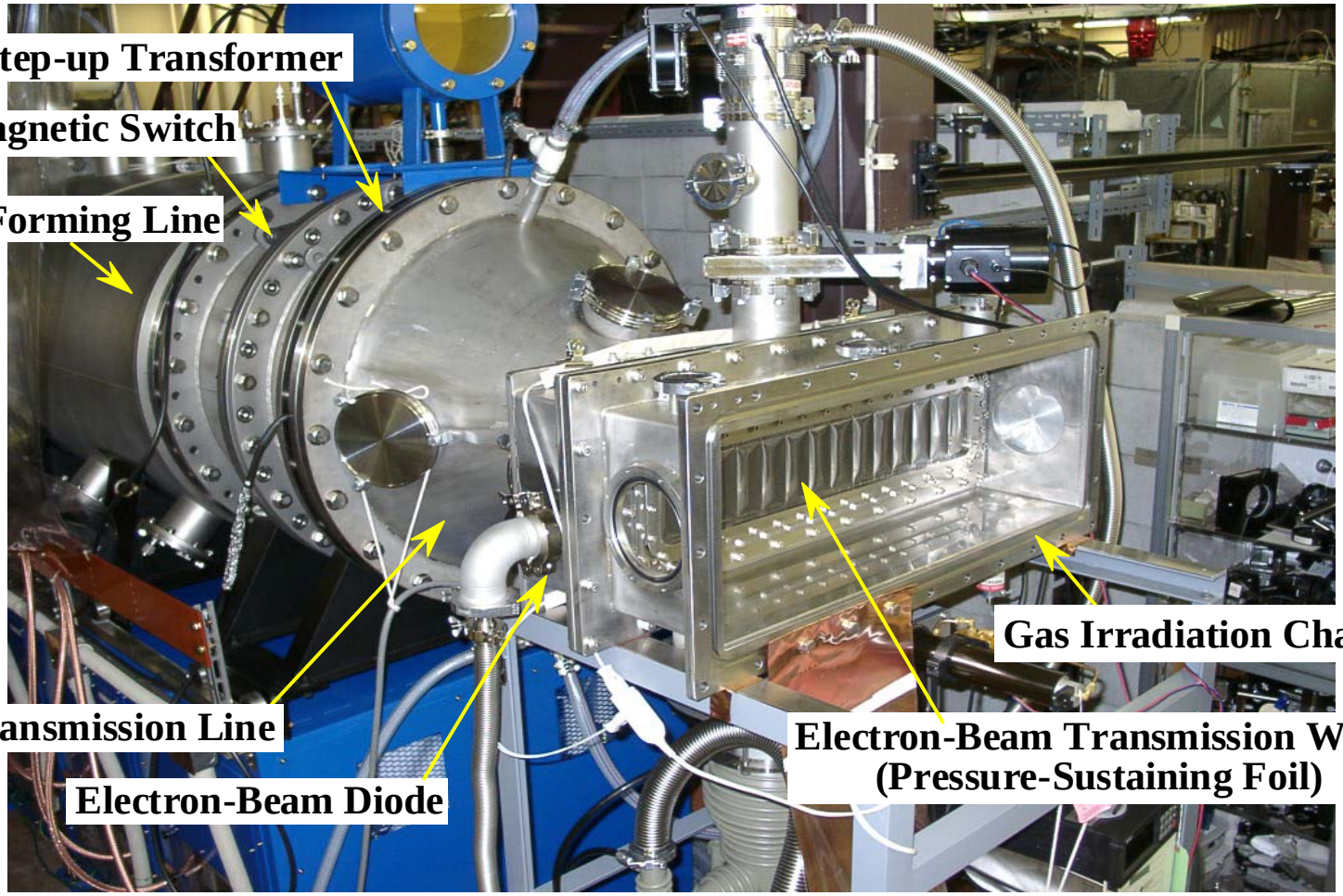
実験データとの比較、処理効率の予測

I. Okuda, et al.: Jpn. J. Appl. Phys. **47**, 5681 (2008), & **47**, 8054 (2008).
S. Kato, et al.: Plasma Fusion Res. **3**, 38 (2008).

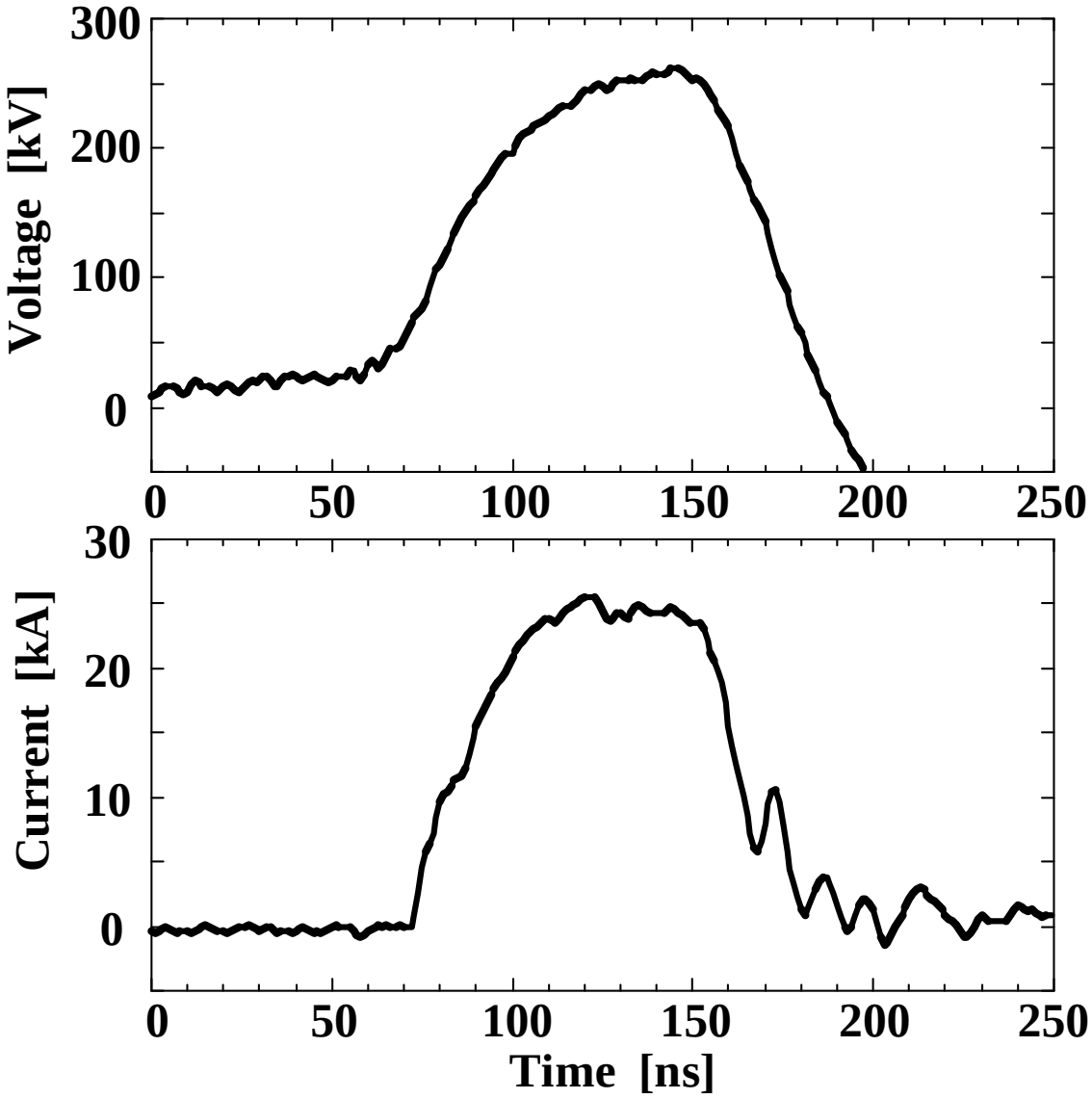
PFC Treatment by Electron-Beam Plasma

- **Atmospheric-pressure operation**
- **High electron energy for efficient PFC decomposition**
  ----- compared to *Discharge Plasma*
- **Fast start, no need for preheat**
  ----- compared to *Combustion*
- **Quick response to change in PFC concentration**
- **Possibility to reduce HF**
  ----- and also reduce NOx ?

High-Repetition-Rate Pulsed-Electron-Beam Generator

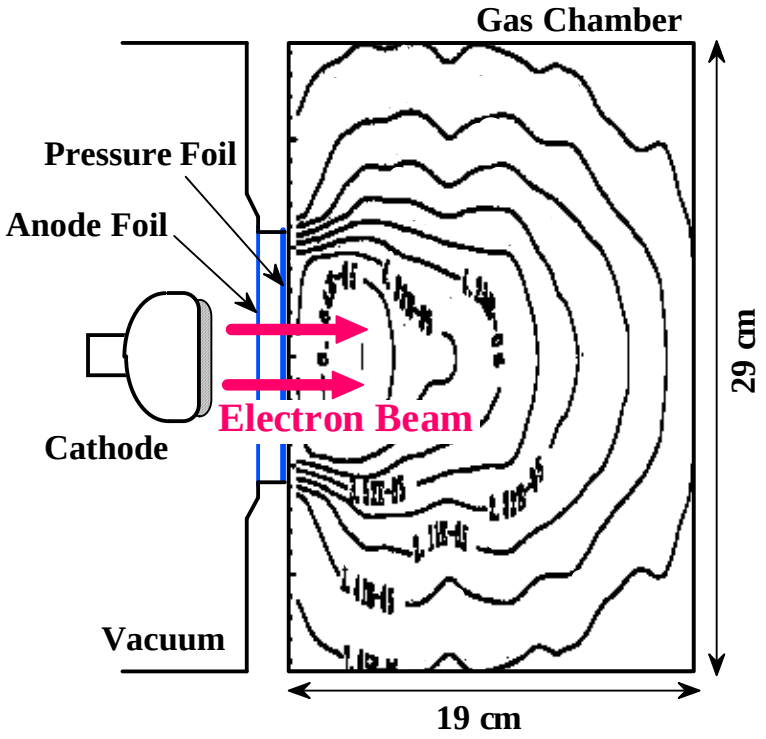


Pulsed-Electron-Beam Voltage and Current Waveforms



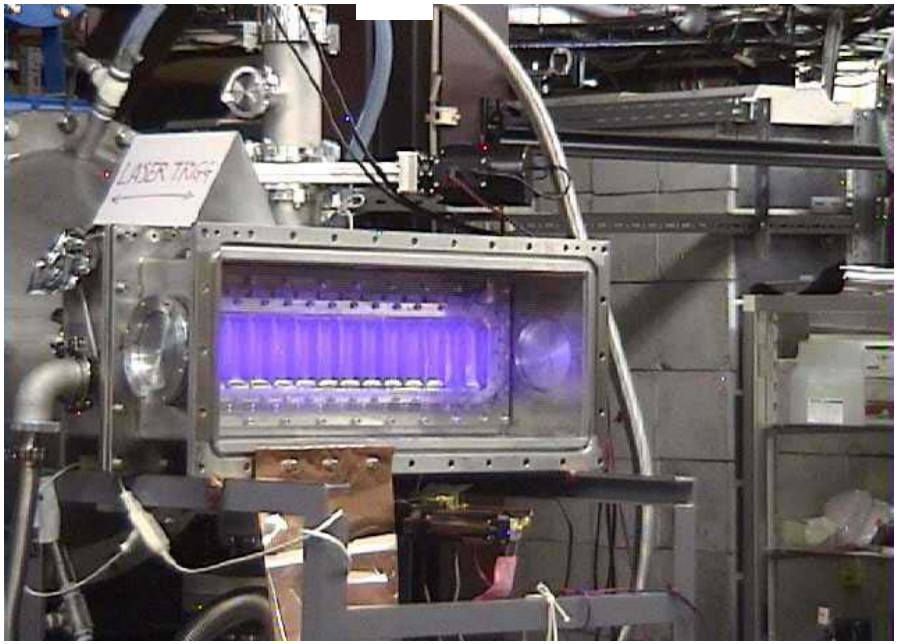
Pulsed-Electron-Beam Emission and Energy Deposition into the Atmospheric-Pressure Gas Mixture

Energy Deposition Distribution (*Simulation*)



800 Torr (Ar: 82%, Kr: 18%)

Pulsed-Electron-Beam Emission to the Air



Voltage: 200 kV
Current: 25 kA
Pulse Width: 80 ns

Repetition Rate: 1 Hz
Exposed to approximately 10 shots

Electron-Beam Energy Deposition in the Gas Mixture

Deposited energy in the gas (ΔE_{dep}) is evaluated on the basis of the pressure jumps (ΔP) due to heating by electron beams and an estimated energy dissipated from the gas in the form of optical radiation (ΔE_{rad}).

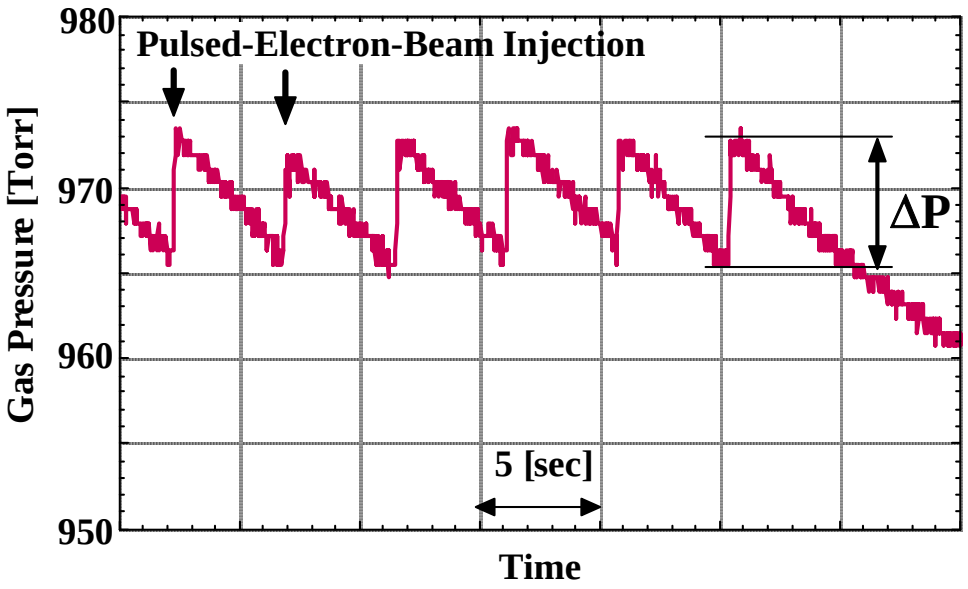
$$\Delta E_{dep} = f_{rad} \frac{1}{\gamma - 1} V_g \Delta P$$

$f_{rad} = \Delta E_{dep} / (\Delta E_{dep} - \Delta E_{rad})$
$= 1.25$ for Ar, 1.11 for N_2 (Assumed)

$\gamma = C_P/C_V = 5/3$ for Ar, $7/5$ for N_2

C_P, C_V : Specific Heat at Constant Pressure, and Constant Volume, respectively

V_g : Gas Volume = 58 L for Ar, 53 L for N_2



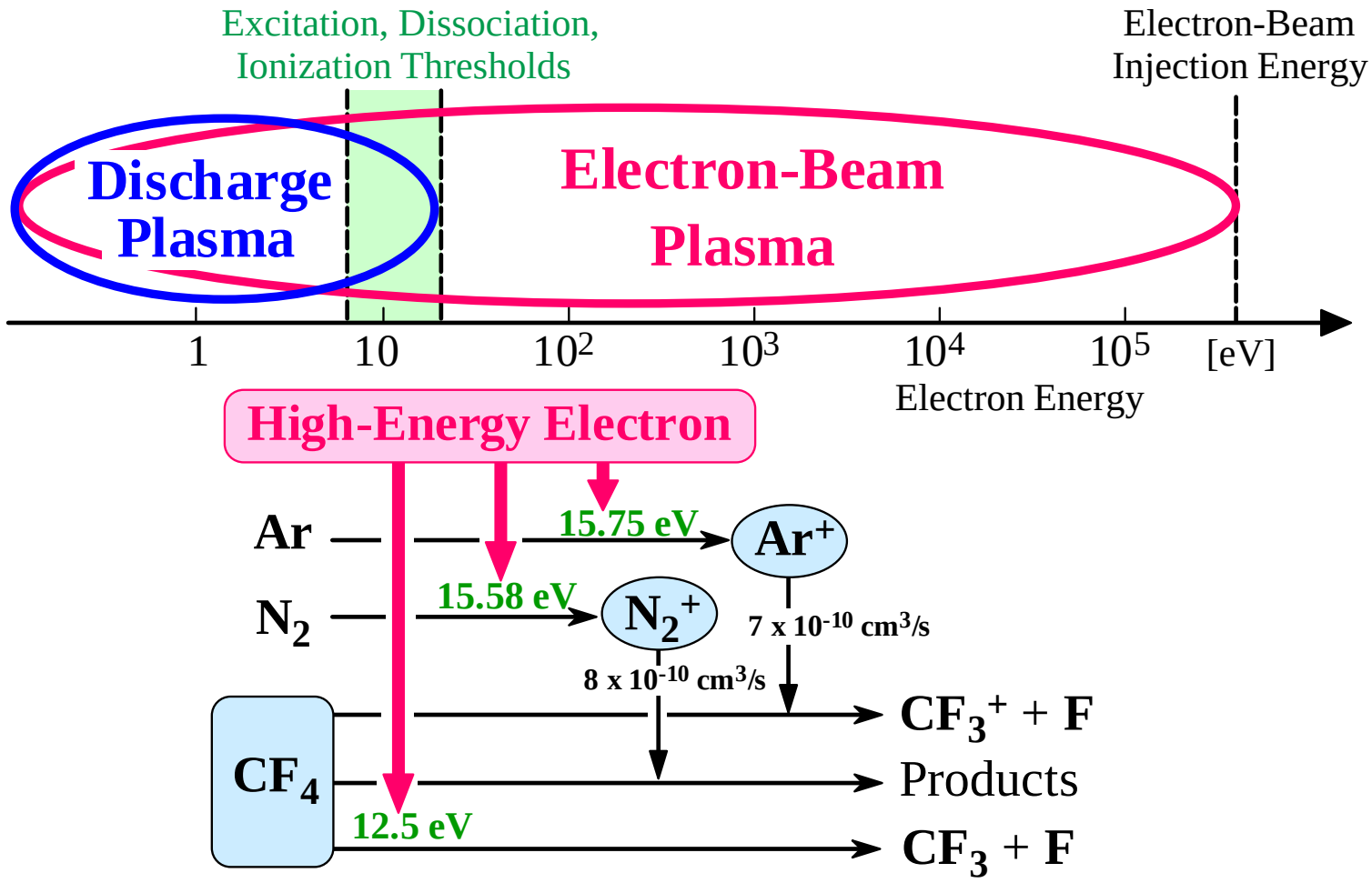
Interval of Pulsed-Electron-Beam Injection : 4.7 sec

Plasma Volume (Irradiated by Electron Beam) : ~ 10 Liter

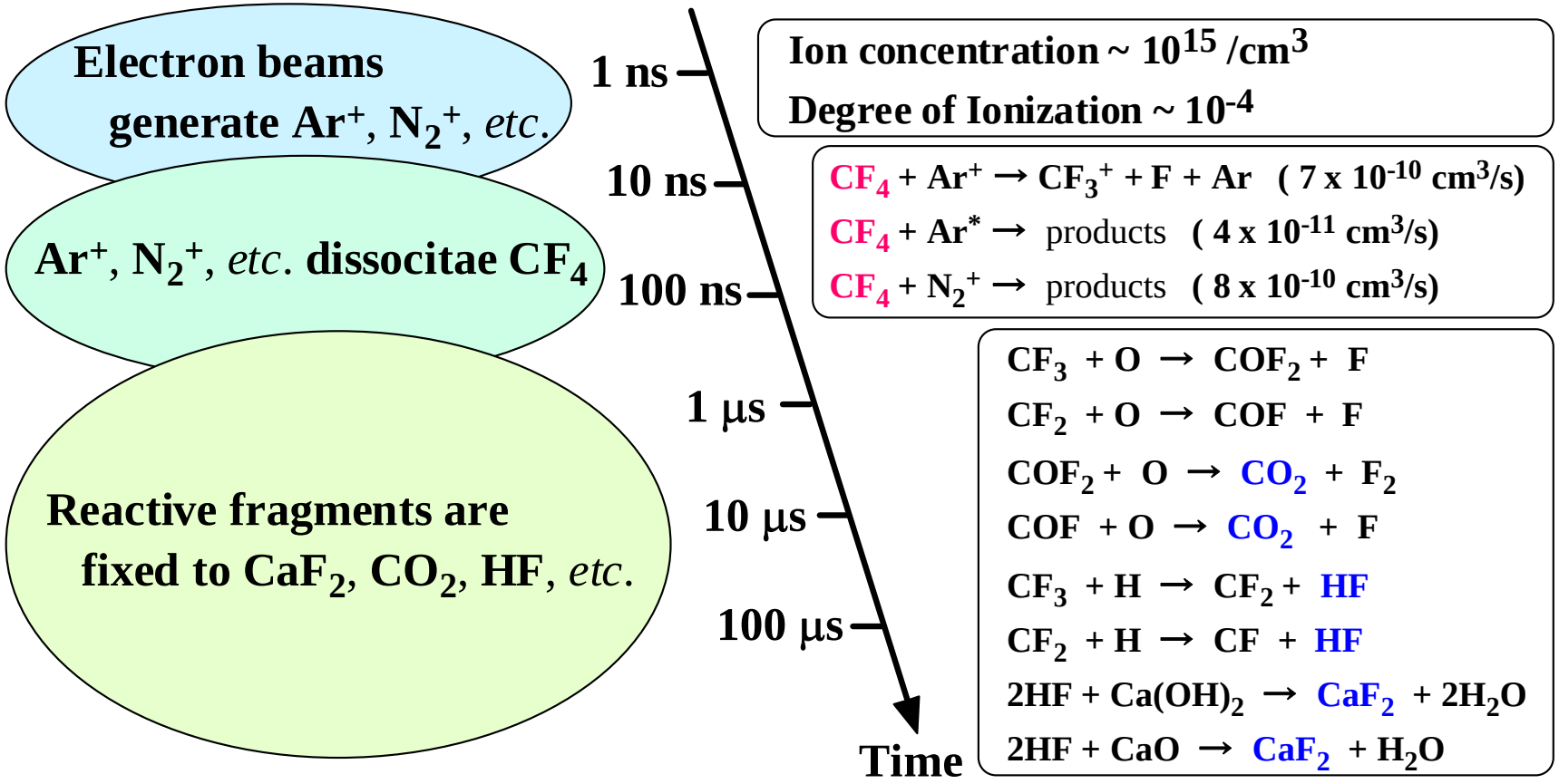
ΔE_{dep} Density/Pulse (Plasma Region) : 10 ~ 30 mJ/cm^3

ΔE_{dep} Density/Pulse (Average over Gas Volume) : 2 ~ 4 mJ/cm^3

High-Energy Electron Beams for Efficient Generation of Active Species to Decompose CF_4

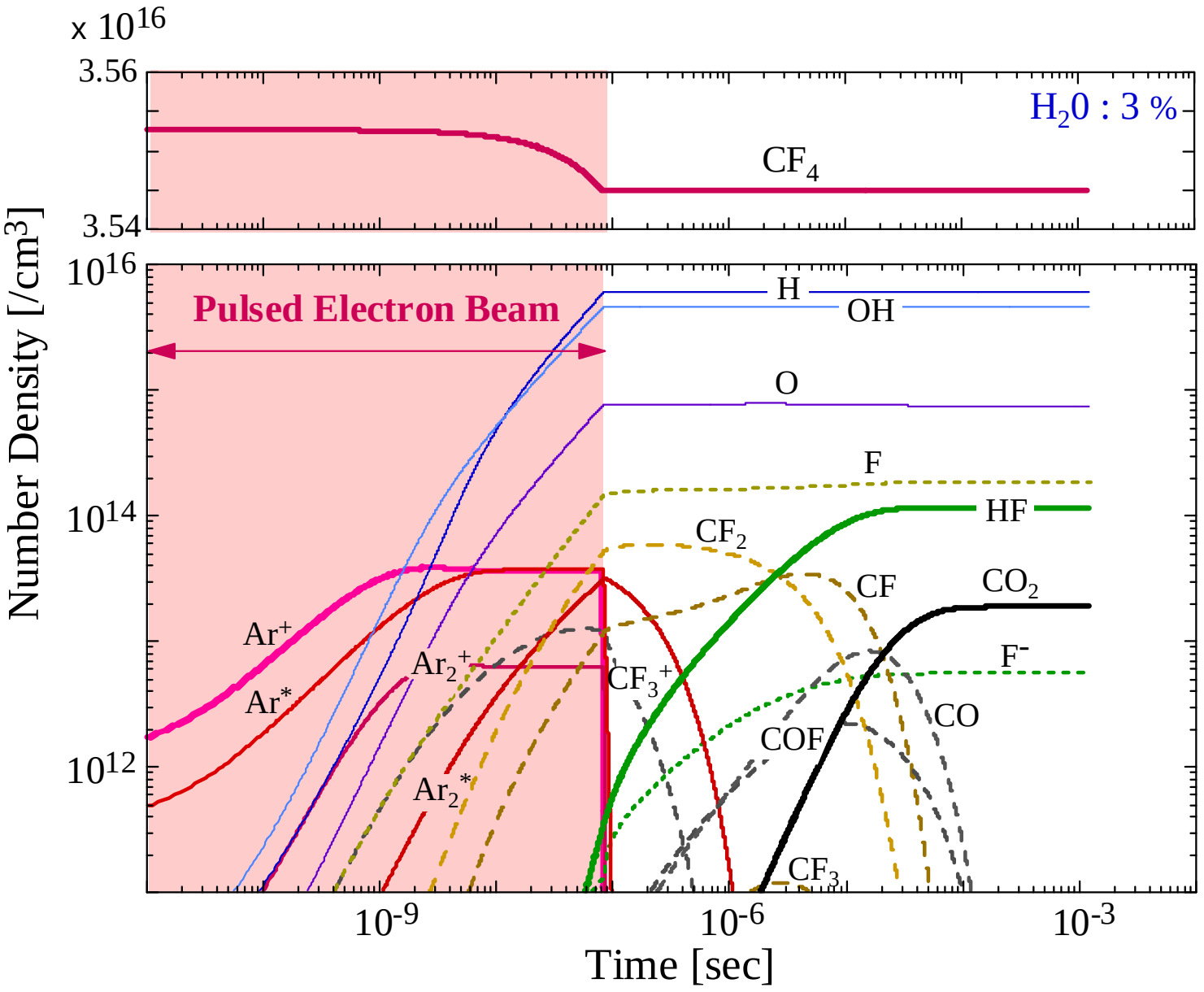


Major Reactions for CF_4 Decomposition and Removal



Reactions proceed sequentially: 1) Generation of active species, 2) Dissociation of CF_4, and 3) Scavenging reactive fragments.

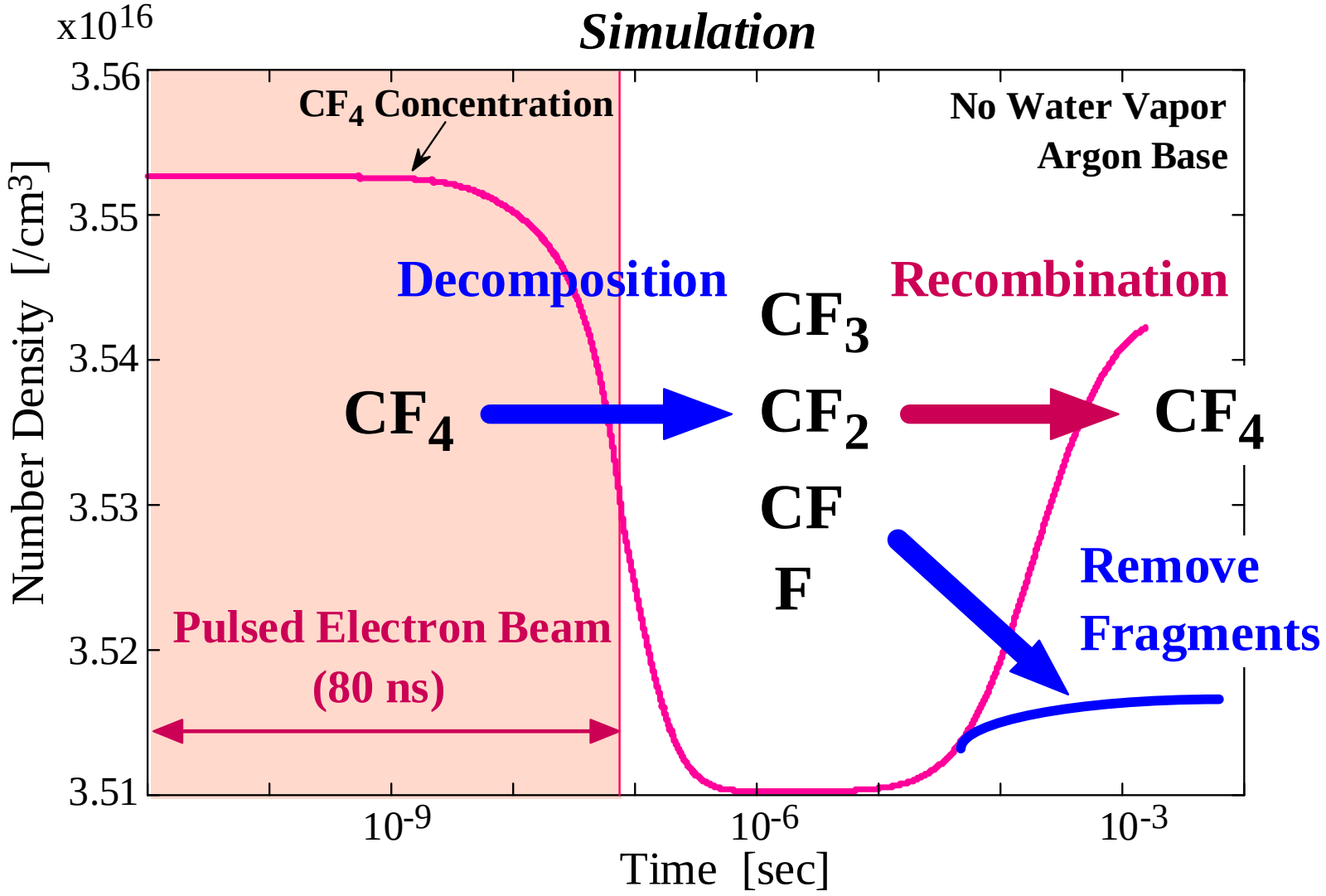
Temporal Evolution of the Reaction Species (Simulation)



CF_4 Decomposition in *Ar-Balanced* Gas Mixtures

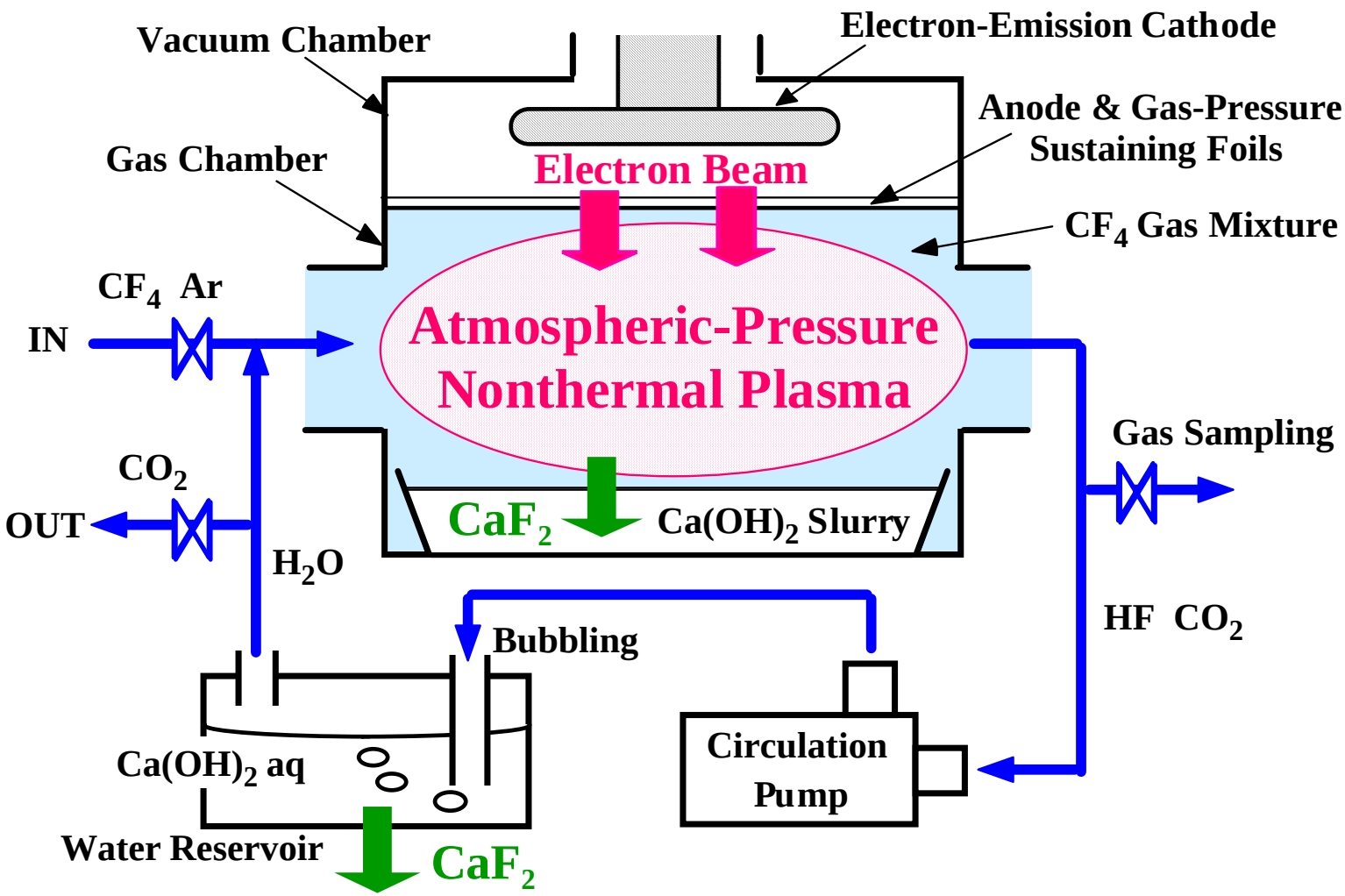
- Argon-balanced gas mixtures at atmospheric pressure
- CF_4 concentration = 1000 ppm
- Scavenge reactive fragments using water vapor and $Ca(OH)_2$
- High energy efficiency obtained

Temporal Evolution of CF_4 Number Density



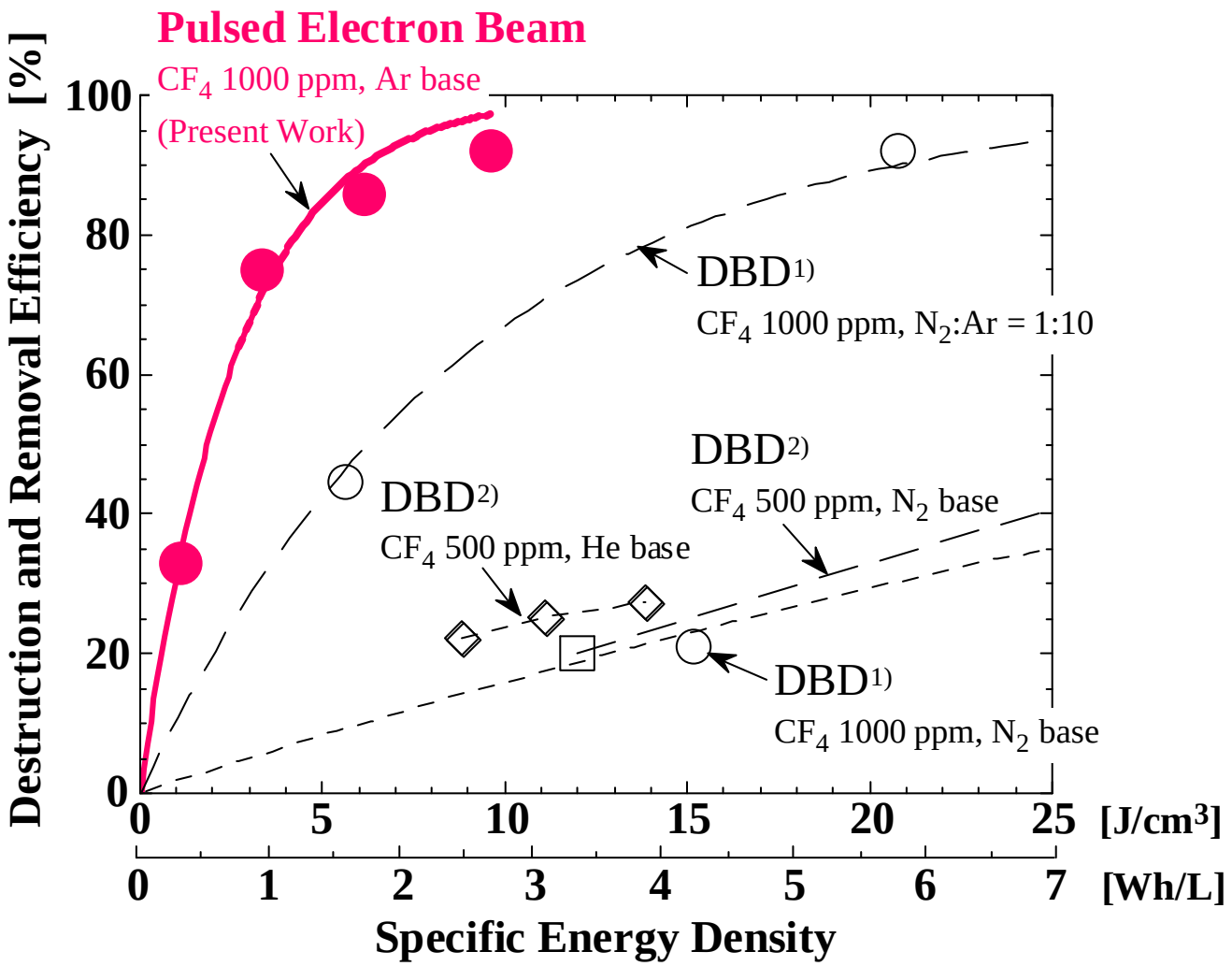
Experimental Setup for CF_4 Decomposition

Ar-Balanced Gas Mixtures with water vapor and $Ca(OH)_2$



Experimental Results of CF_4 Decomposition

Ar-Balanced Atmospheric-Pressure Gas Mixtures

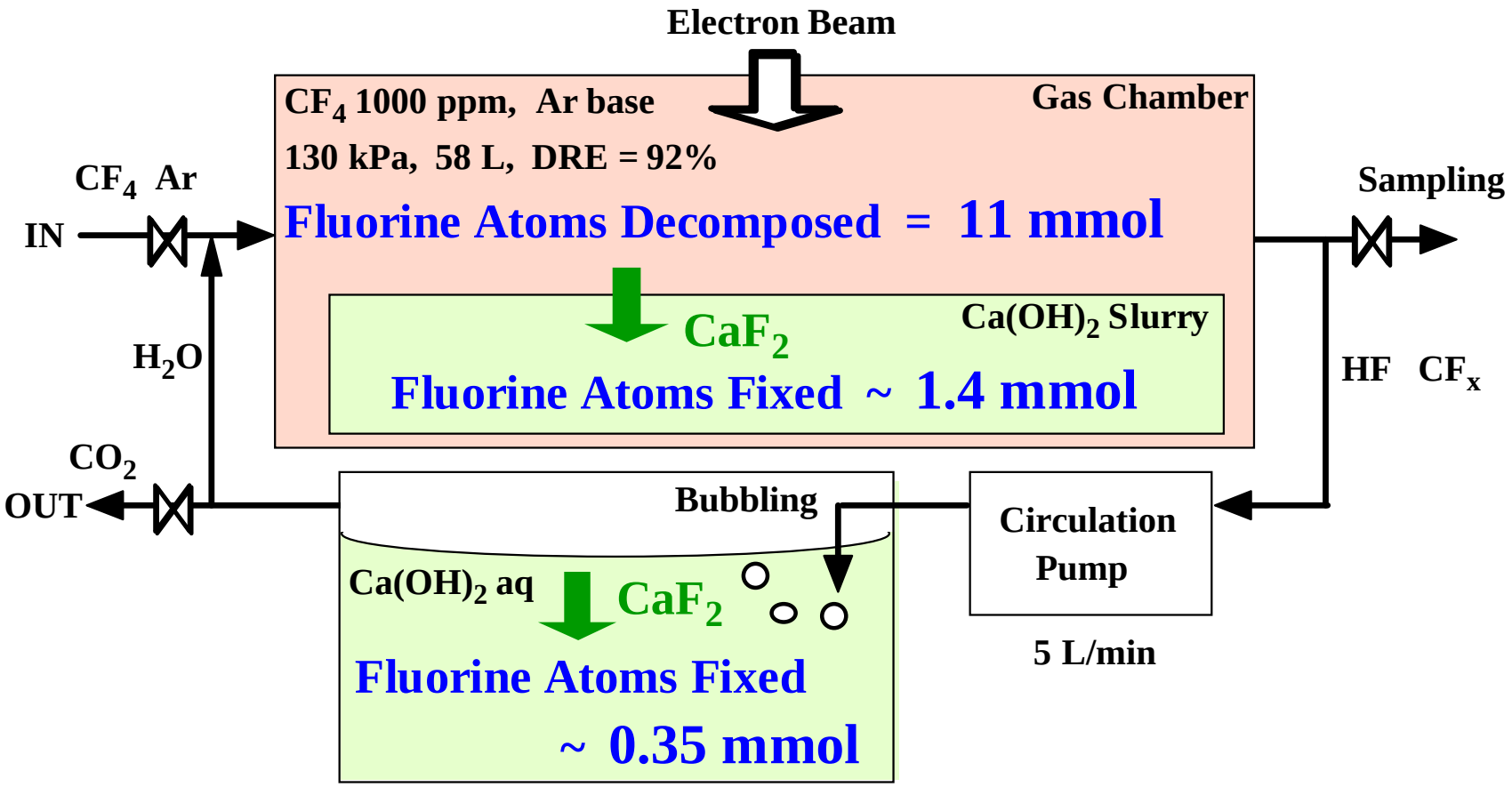


1) G. J. Pietsch, et al.: Proc. 10th Int. Symp. High-Press. Low-Temp. Plasma Chemistry (2006) p.128.

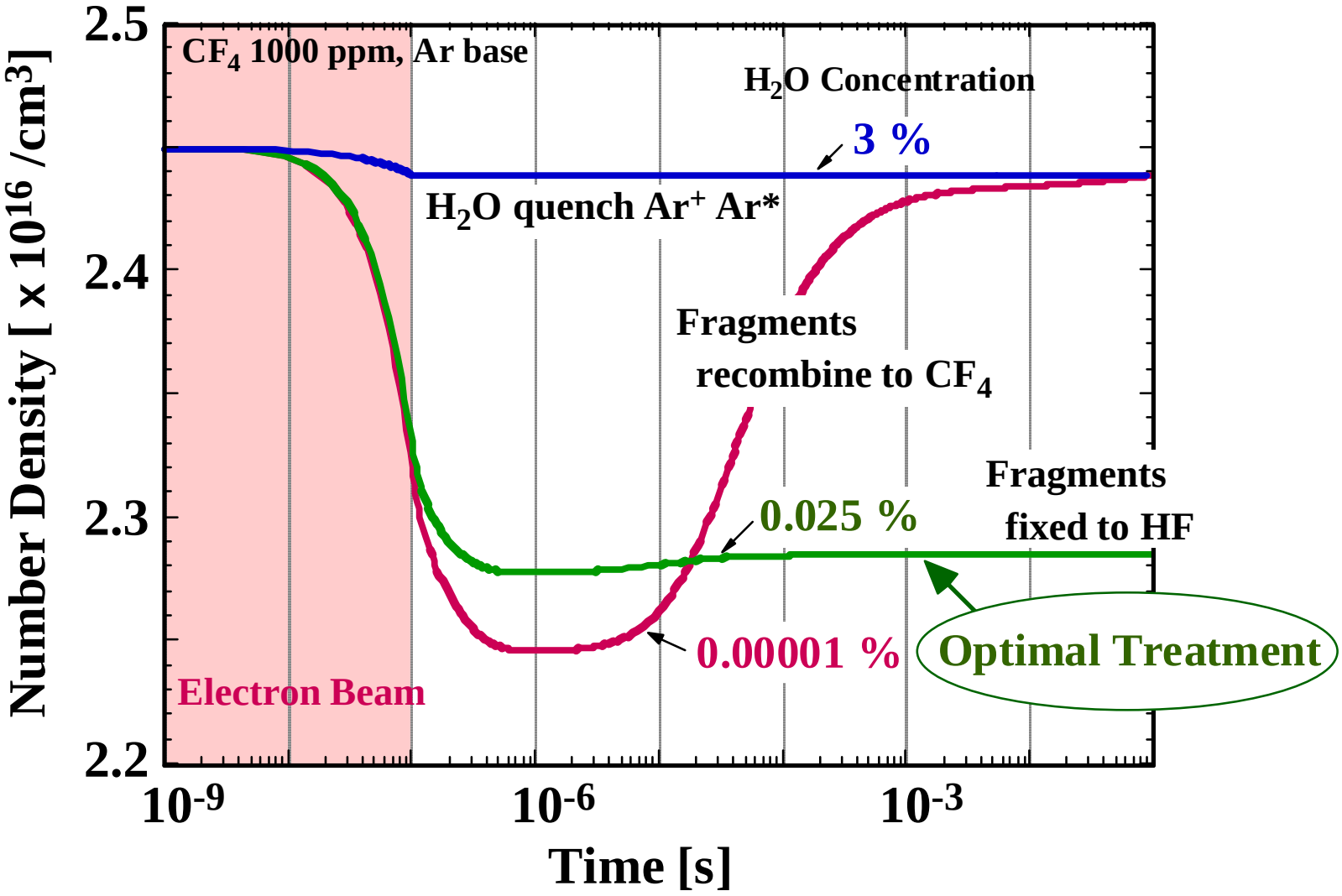
2) Y. Kim, et al.: IEEE Trans. on Plasma Science **33**, 1041 (2005).

Number of Fluorine Atoms Fixed in $Ca(OH)_2$

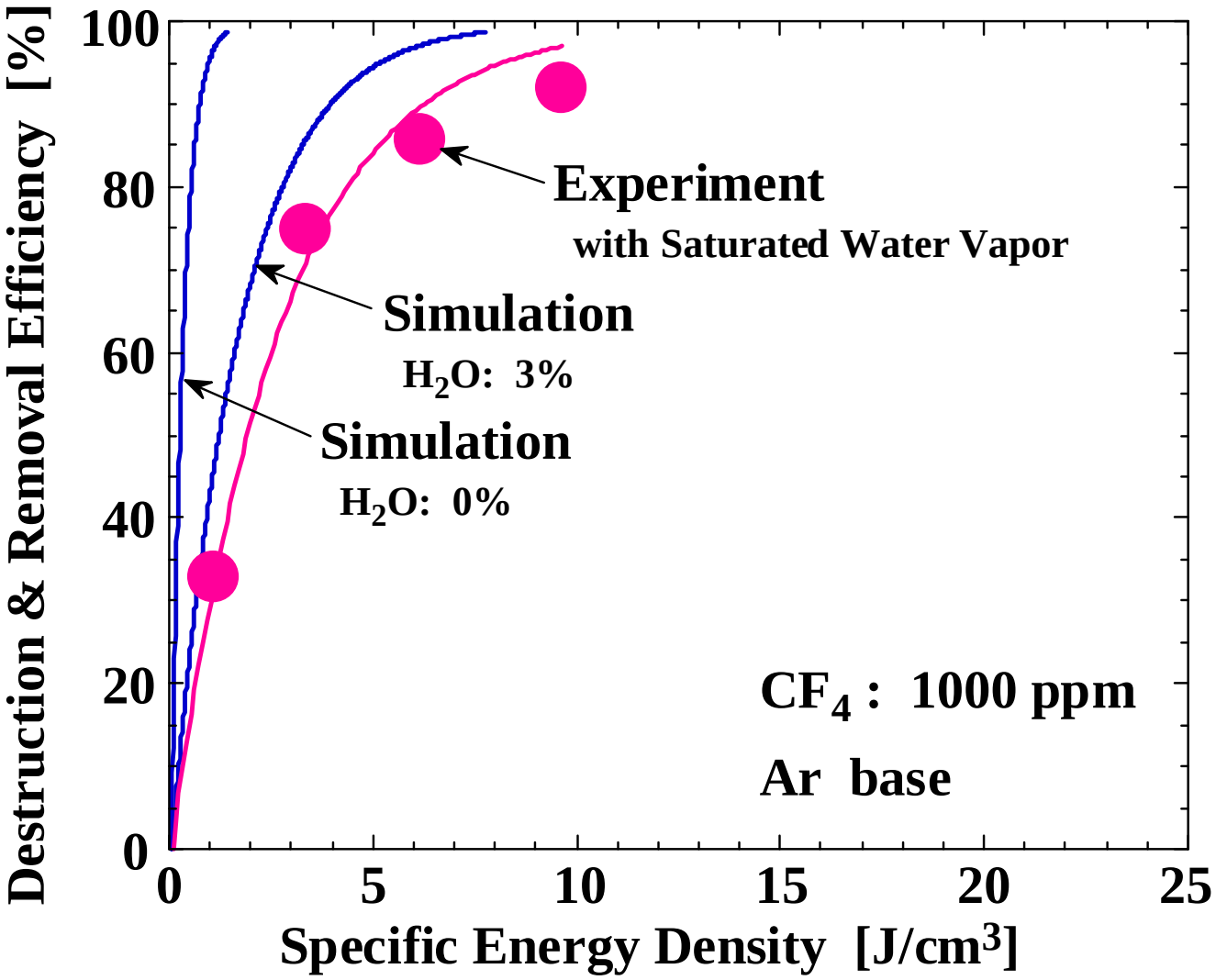
(*Measured in the Ar-Balanced Gas-Mixture Experiments*)



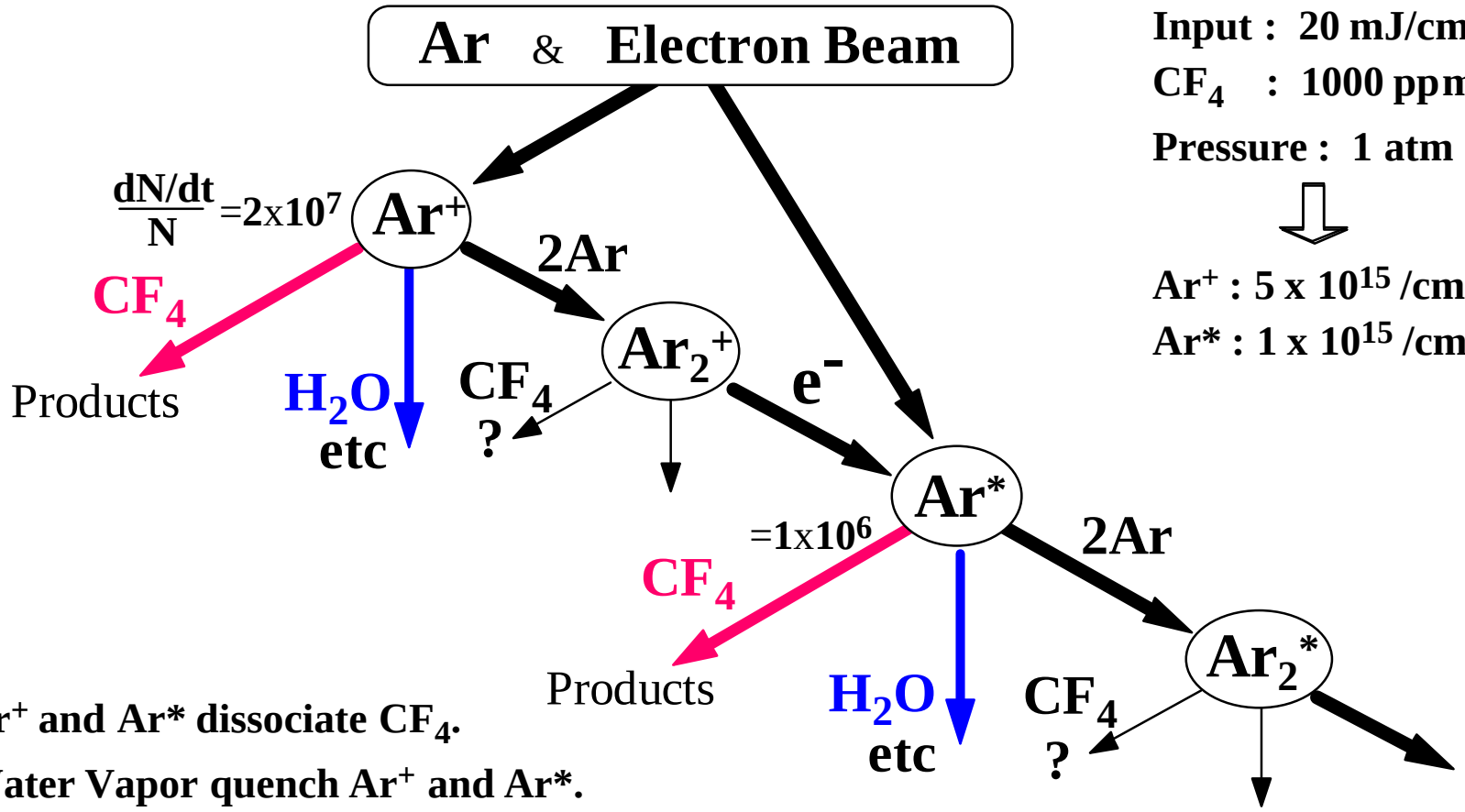
Effect of Water Vapor on CF_4 Number Density



Numerical Simulation and Experimental Results of CF_4 Decomposition and Removal Efficiency

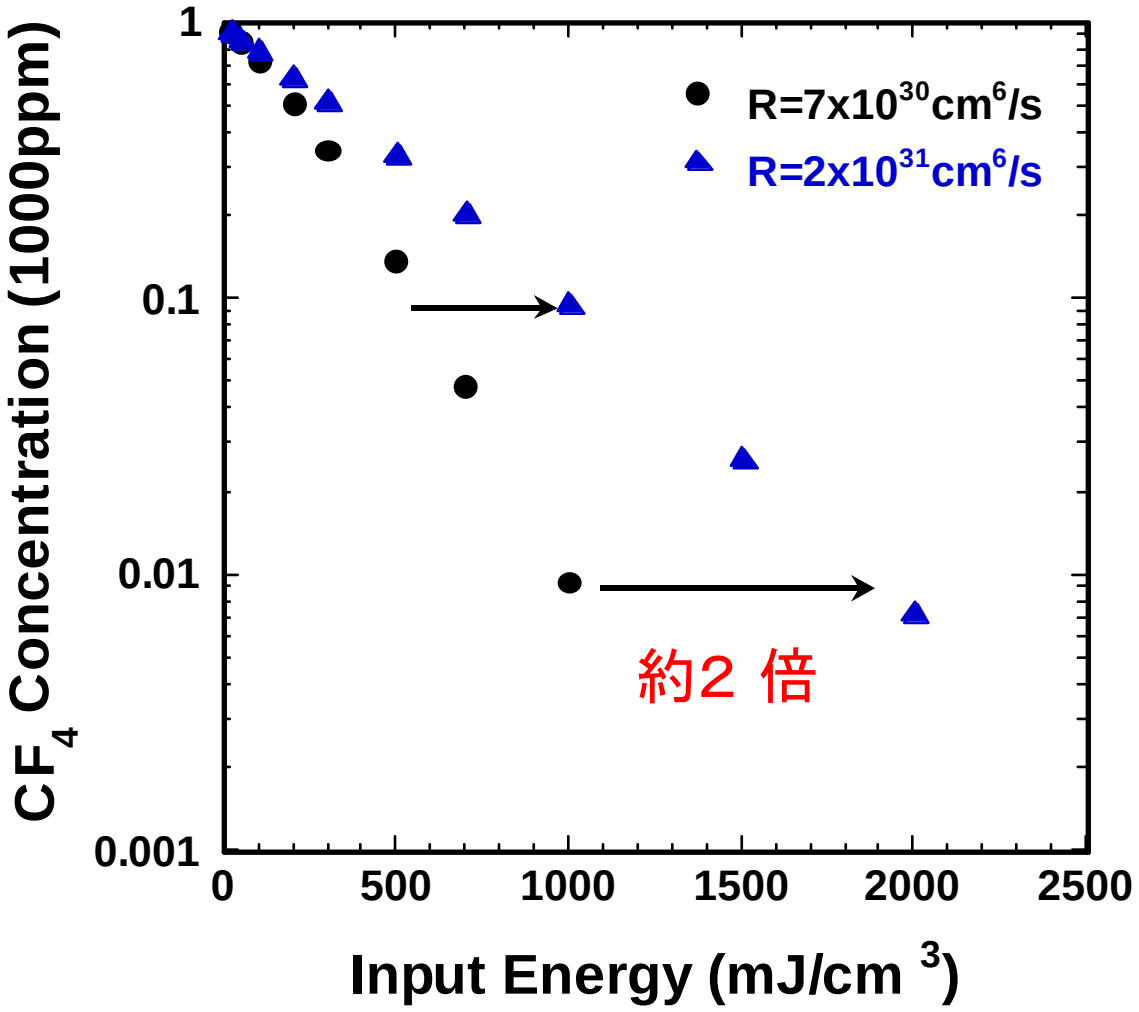


CF_4 Decomposition Paths in Ar-Balanced Gas Mixture



反応レート の違いによる分解効率の違い

（ $Ar^+ + 2Ar \longrightarrow Ar_2^+ + Ar$ ）

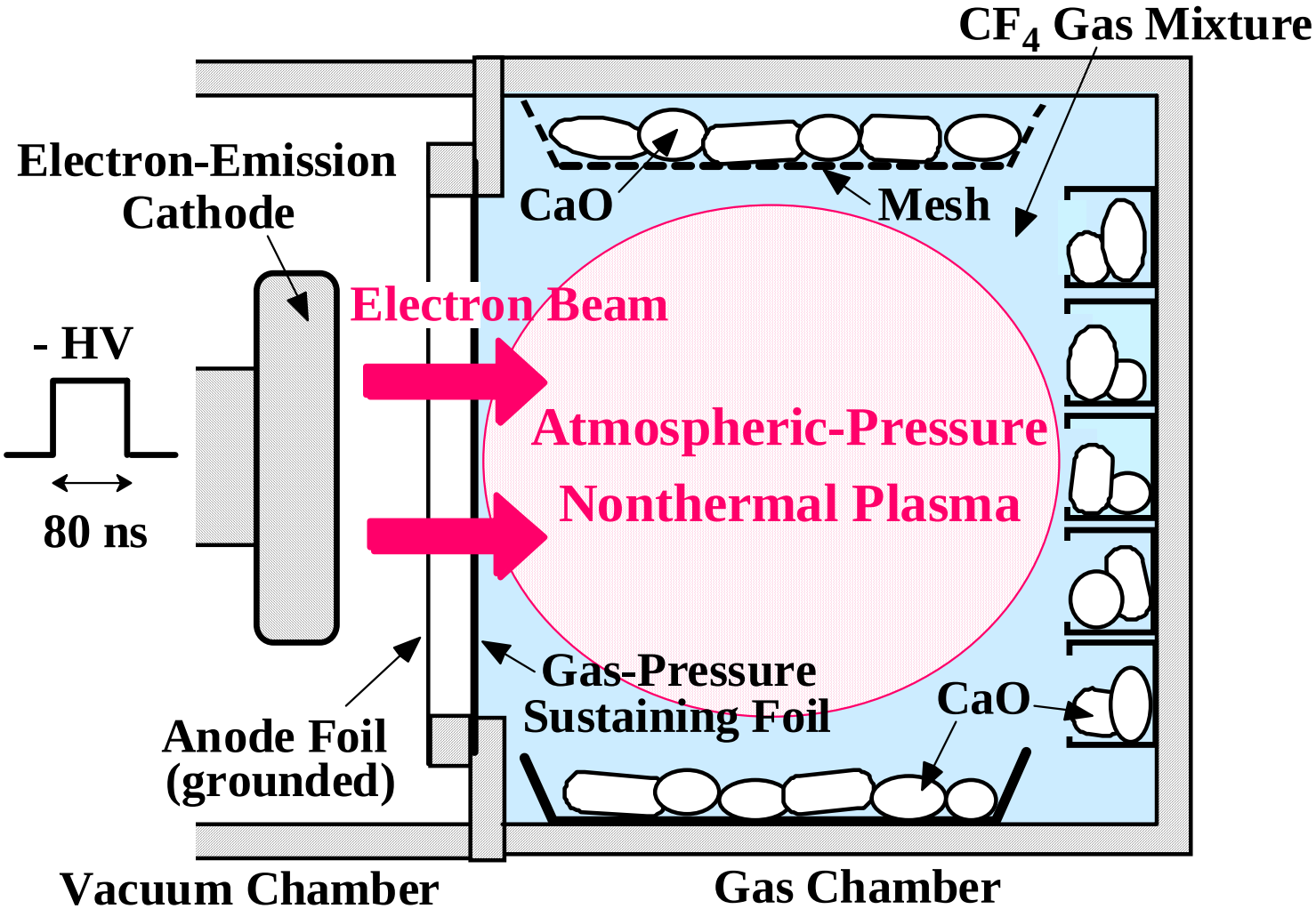


CF_4 Decomposition in *N_2-Balanced* Gas Mixtures

- Nitrogen-balanced gas mixtures at atmospheric pressure
- CF_4 concentration = 900 ppm
- Scavenge reactive intermediates using CaO only
- HF emission reduced using no water vapor as a scavenger

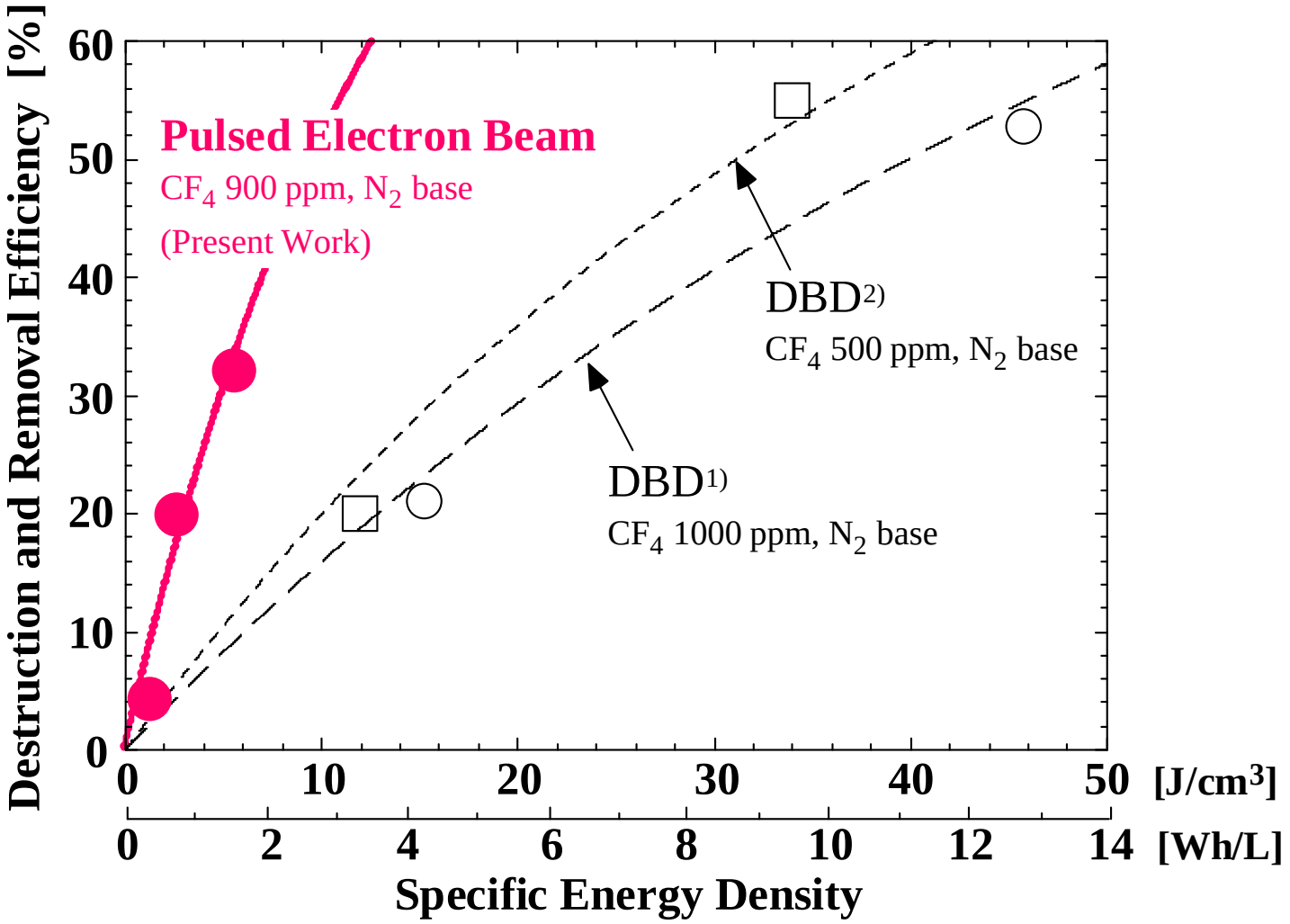
Experimental Setup for CF_4 Decomposition

N_2-Balanced Gas Mixtures with CaO



Experimental Results of CF_4 Decomposition

N_2-Balanced Atmospheric-Pressure Gas Mixtures



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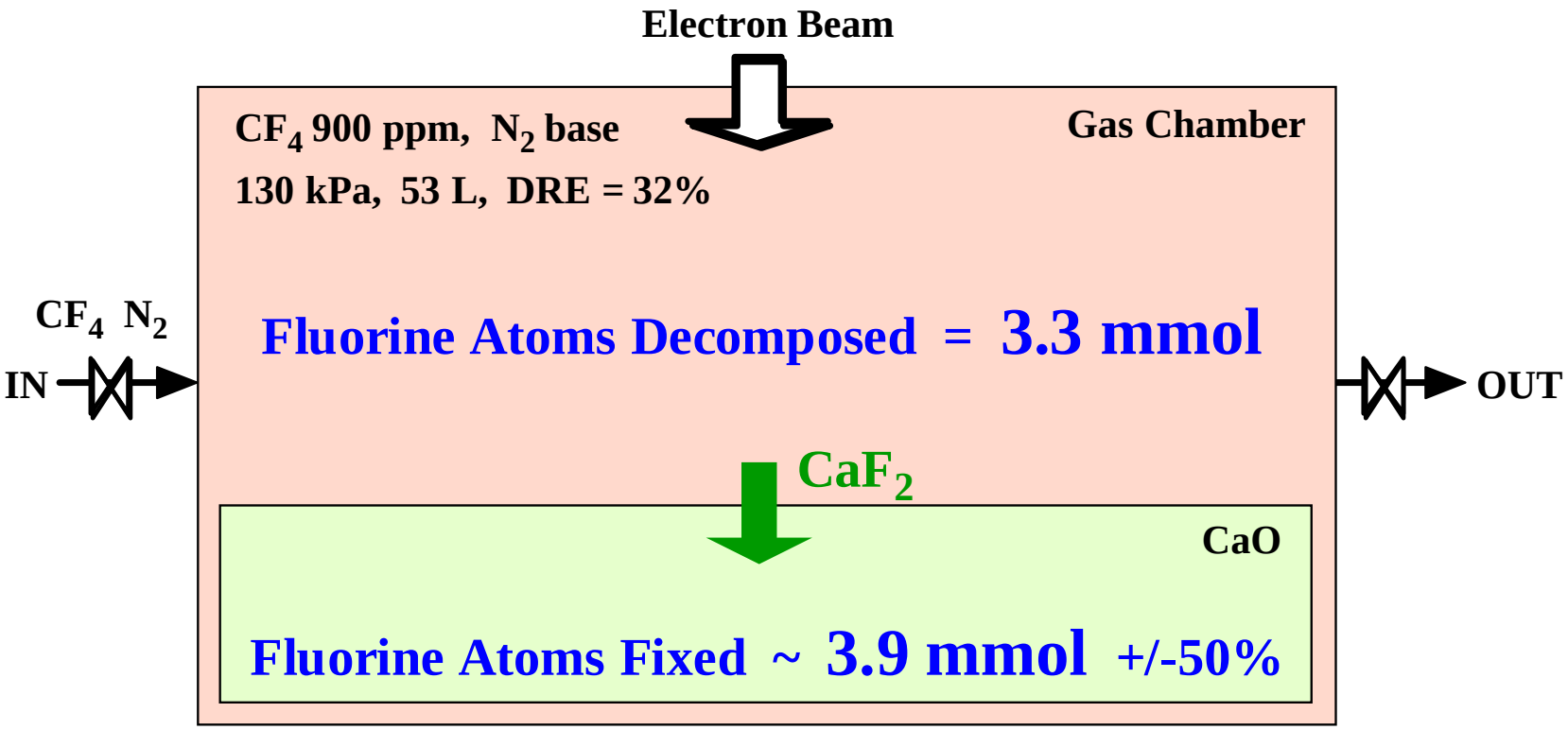
Number of Fluorine Atoms Fixed in CaO

(*Measured in the N_2-Balanced Gas-Mixture Experiments*)



Byproduct gas species with small concentrations

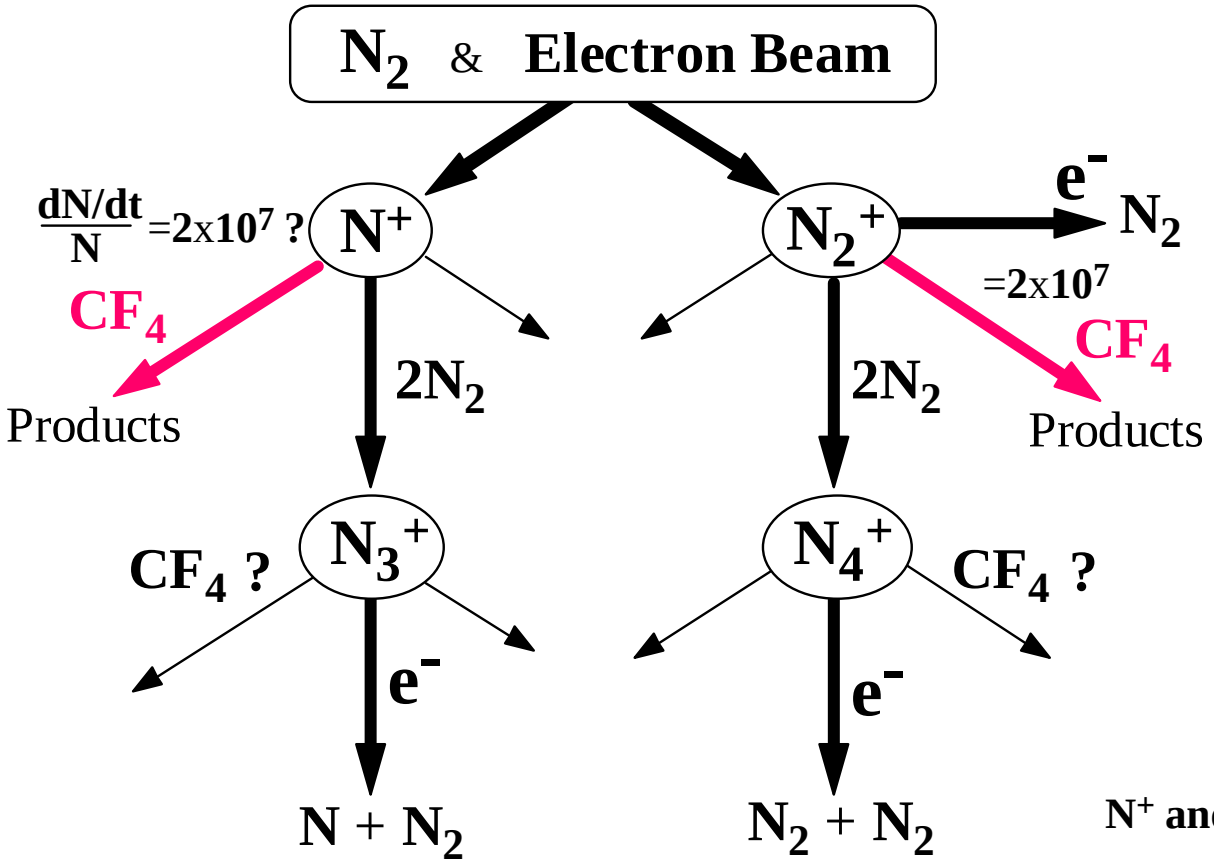
DRE = 26%, SED = 4.1 J/cm^3 (1.1 Wh/L)

* : lower than the detection limits

HF	NF_3	COF_2
< several 100 ppb*	< several ppm*	< several ppm*

CO_2	CO	CN^-	NO_2	NO
< 1 ppm	5 ppm	< 1.5 ppm*	< 1 ppm*	< 1 ppm*

CF_4 Decomposition Paths in N_2-Balanced Gas Mixture



Input : 20 mJ/cm^3

CF_4 : 1000 ppm

Pressure : 1 atm

N^+ : 9 x 10^{14} /cm^3

N_2^+: 3 x 10^{15} /cm^3

N^+ and N_2^+ dissociate CF_4.

Summary

■ 電子ビームプラズマによる CF_4 分解処理実験

・ 放電プラズマに比べて比較的高い分解処理効率を達成
（ CF_4 分解生成物の反応場からの除去がまだ不十分）

■ CF_4 分解反応に関する数値計算（ アルゴン希釈）

・ CF_4 分解生成物の固定・ 除去の最適化により、分解処理の一層の高効率化を予測

・ CF_4 分解反応データの整備の必要性
（ 窒素イオンによる CF_4 分解反応レート の欠如等）