Abatement of greenhouse gases using surface-wave microwave discharges sustained at atmospheric pressure

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Outline

- Introduction
- Abatement method
- Working example: abatement of SF_6 in N_2/O_2 gas mixture
- Conclusion

Introduction

- Context of the work
  - ✓ Abatement of greenhouse gases
  - ✓ Destruction of CF_4 and SF_6 (diluted into N_2) from microelectronic fabs
- Conventional methods
  - ✓ Incineration : low conversion rates
  - ✓ Combustion : massive CH_4/H_2 feed flows
- Plasma solution
  - ✓ High destruction rate
  - ✓ Low energy consumption
  - ✓ Selective chemistry
  - ✓ Harmless byproducts

Abatement scheme

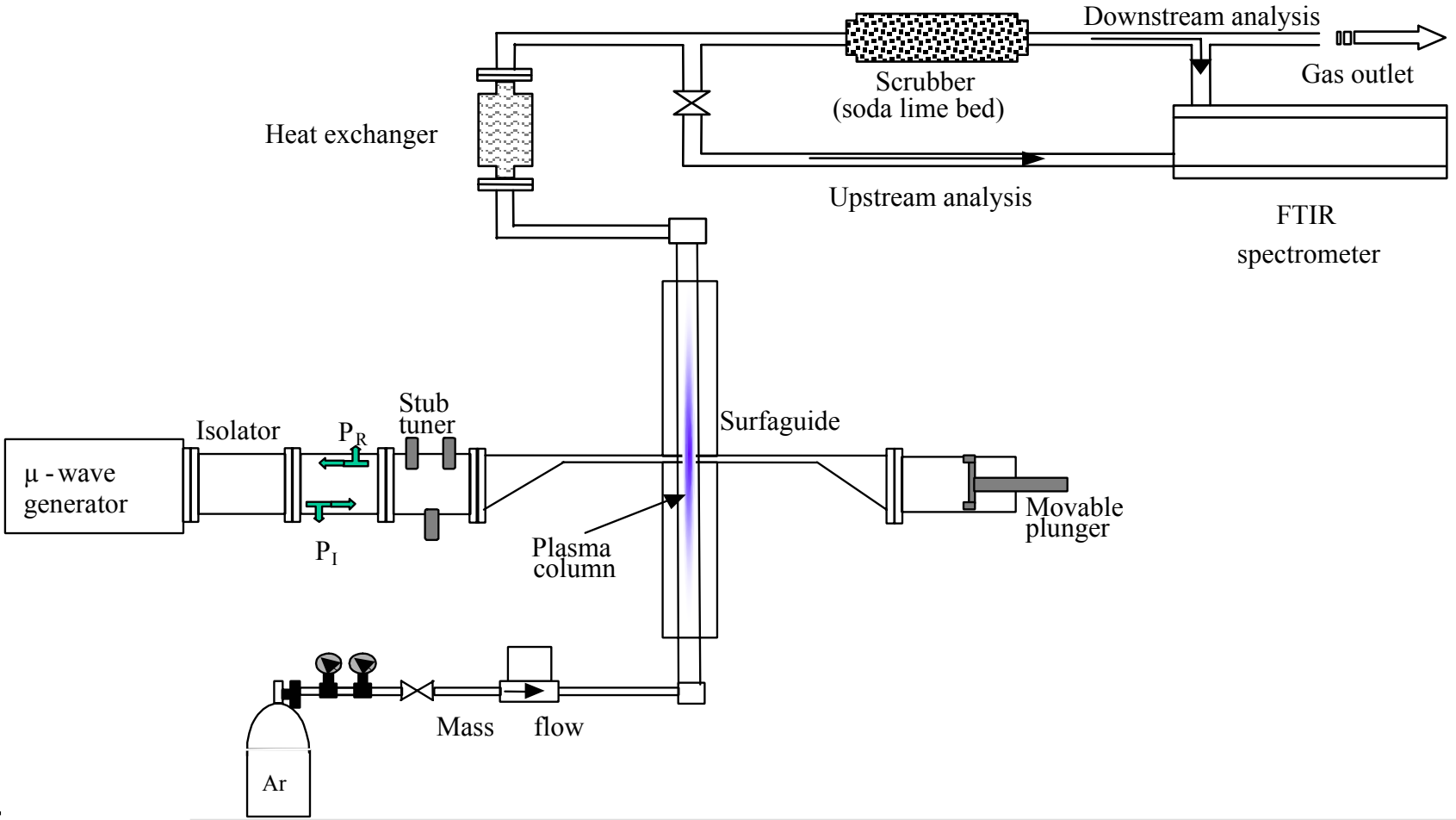
- Non equilibrium plasma :
  - ✓ $T_e >> T_n \cong T_i$
  - ✓ Low enthalpy (T_{gas}~ 1000-5000 K, T_e> 10,000 K)

- A two-step process
  - ✓ electron collisions on PFC molecules and background gas molecules
  - ✓ interaction of molecular fragments and radicals with (added) oxygen atoms: $[O_2] \cong [PFC]$
    - Oxidation: elimination of PFC molecule
    - Non-oxidized by-products recombination: reformation of PFC molecule

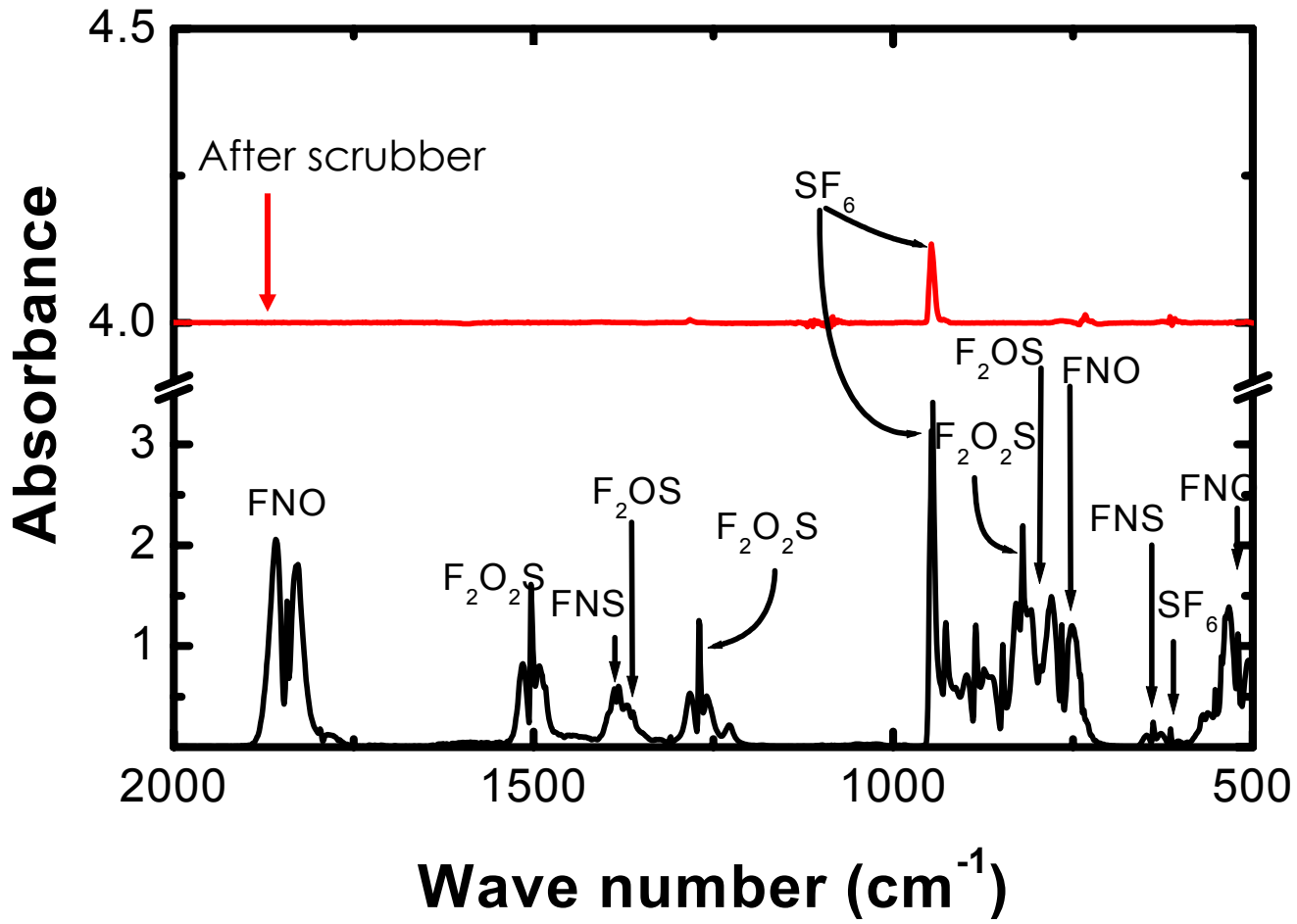
⇨ Trapping of residues on a scrubber
  - ✓ Humidified soda lime (alkaline bed)
  - ✓ No hazardous by-products at exhaust

Experimental arrangement

- 1- Surface-wave discharge (SWD) scheme : *Surfaguide*



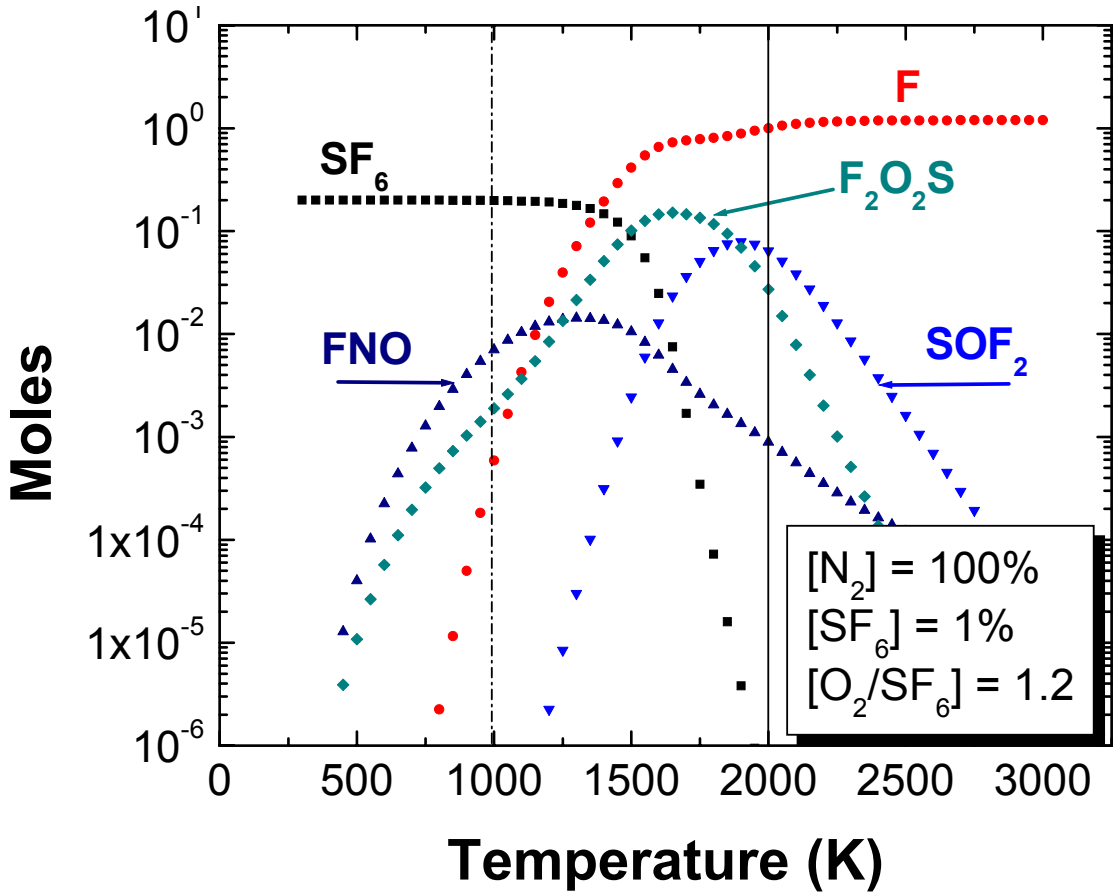
By-products analysis



Main by-product: F_2O_2S

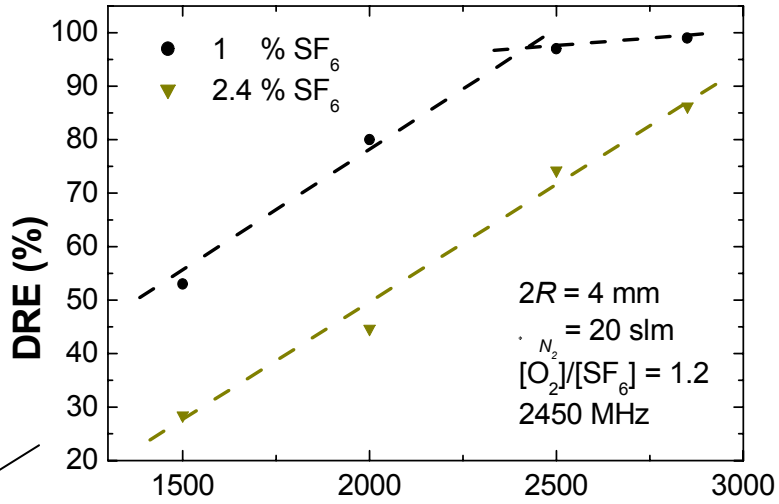
Equilibrium Calculation: SF_6-O_2-N_2

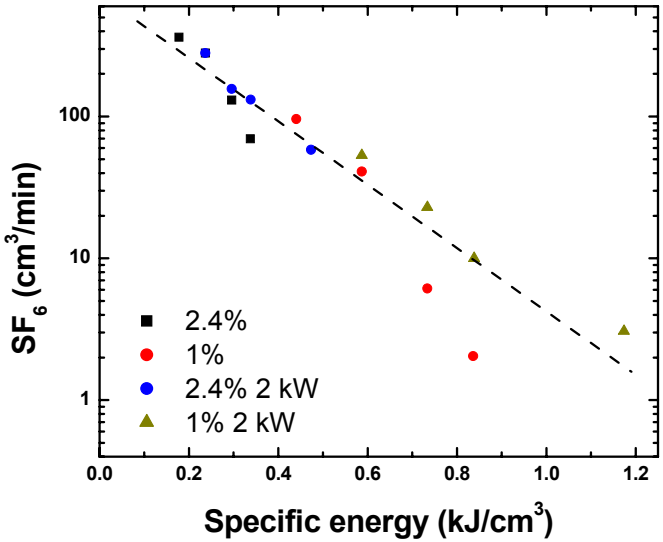
- Molar fraction of SF_6 by-products

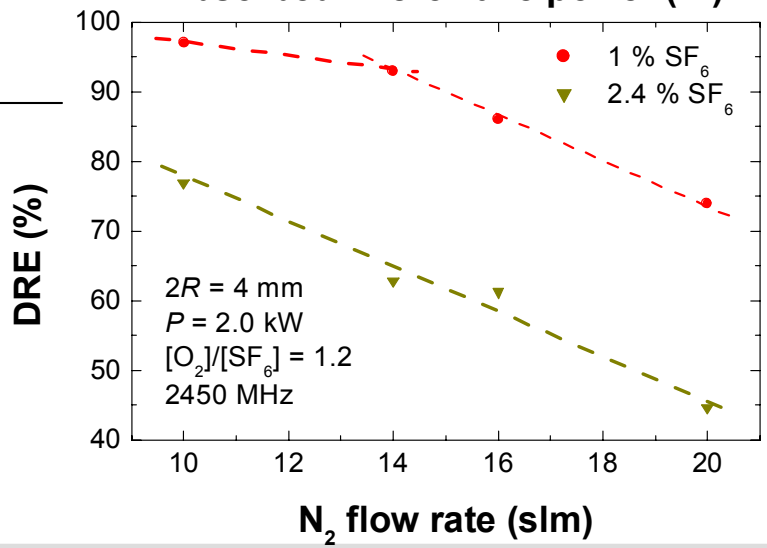


Abatement of SF_6: dissociation

- Influence of microwave power
- Influence of N_2 flow rate
  - ✓ Linear dependence of DRE vs. P
    - SWD property : $P=N\times\theta$
  - ✓ Second slope (1%) : SF_6 reformation

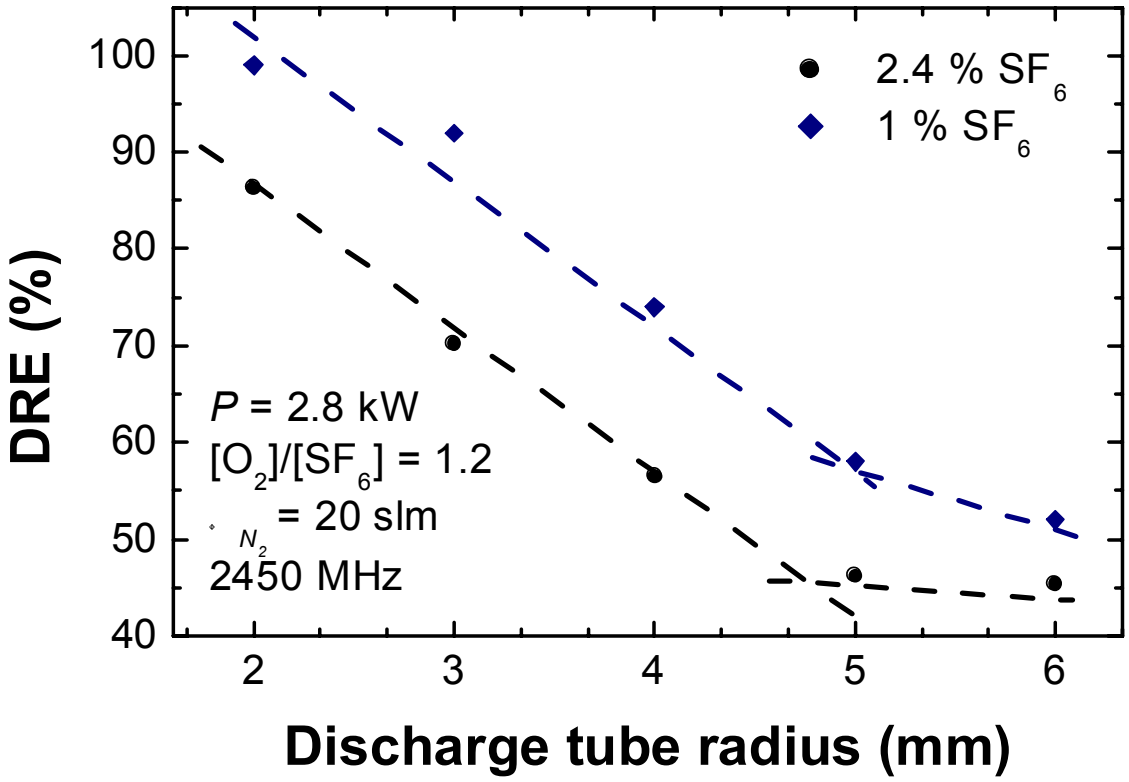


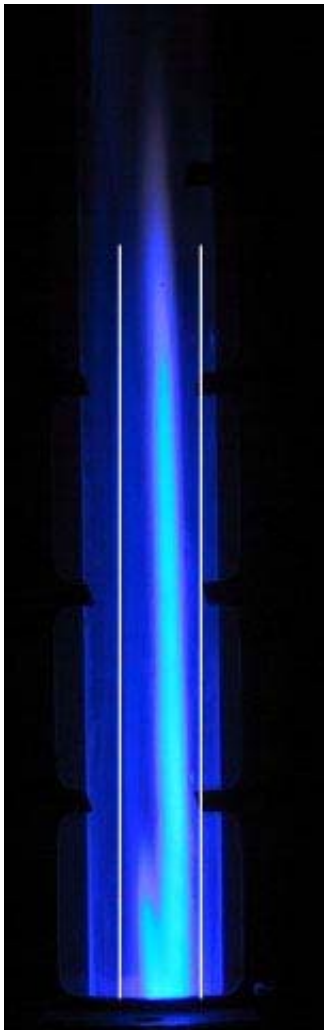




Abatement of SF_6

- Influence of radial contraction

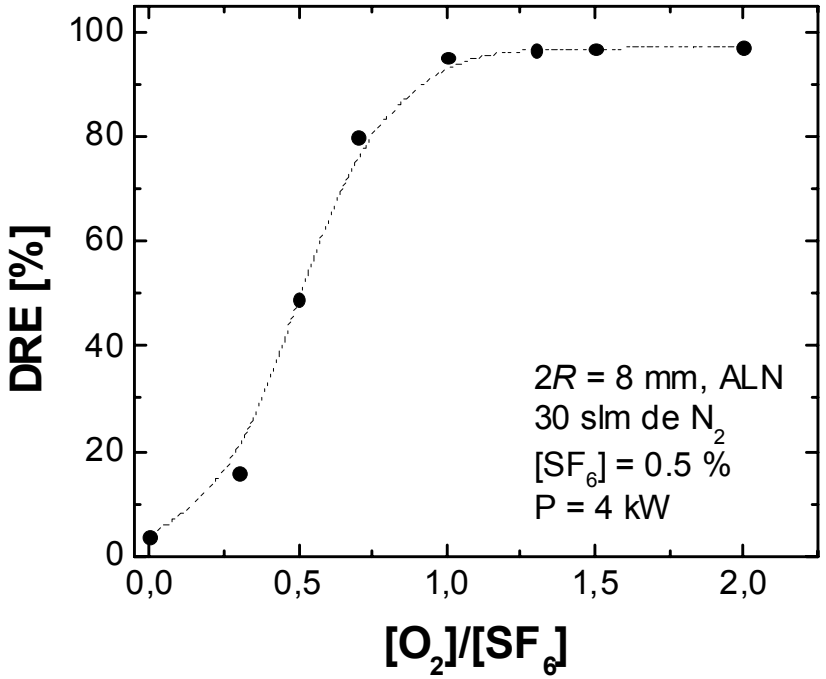




Contraction sets up a limit for tube radius

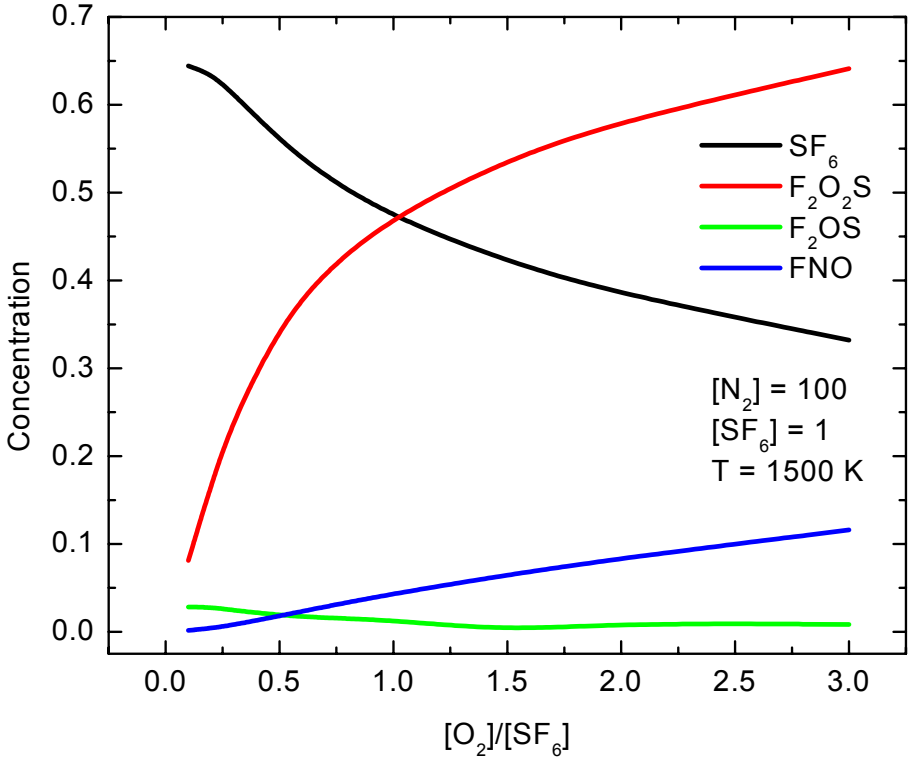
Abatement of SF_6 : oxidation

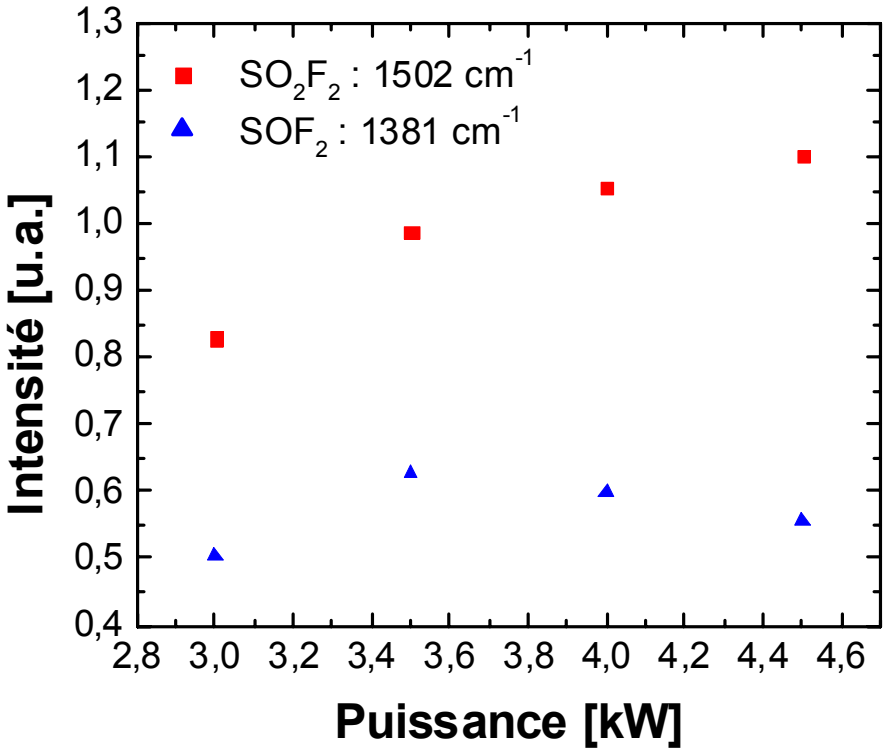
- Influence of the O_2/SF_6 ratio
- Oxidation vs. Reformation
  - ✓ $[O_2]/[SF_6] < 1$: DRE determined by reformation rate
  - ✓ $[O_2]/[SF_6] \geq 1$: full oxidation
- Stoichiometric ratio 2:1 of O to S
  - ✓ Main by-product: F_2O_2S



Thermodynamic calculations: SF_6-N_2-O_2

- Influence of $[O_2]/[SF_6]$ ratio





Gas-phase reactions

- ✓ Electron-impact dissociation
  $e + SF_6 \rightarrow SF_2 + 4F + e$ (high electron density)

- ✓ Gas-phase reactions reaction rates (cm^3/s)

Radical reactions

$SF_2 + O \rightarrow SOF + F$	$\sim 10^{-10}$
$SOF + O \rightarrow SO_2F$	$\sim 10^{-10}$
$SOF + F \rightarrow SOF_2$	$\sim 10^{-12}$
$SF_3 + O \rightarrow SOF_2 + F$	$\sim 10^{-11}$
$SOF_2 + F \rightarrow SOF_3$	$\sim 10^{-10}$

Abatement of SF_6

$SO_2F + F \rightarrow SO_2F_2$	$\sim 10^{-10}$
$SOF_3 + O \rightarrow SO_2F_2 + F$	$\sim 10^{-10}$

Reformation

$SF_5 + F \rightarrow SF_6$	$\sim 10^{-10}$

Not observed with MW plasmas

$SOF_3 + F \rightarrow \boldsymbol{SOF_4}$	$\sim 10^{-10}$
$SF_5 + O \rightarrow \boldsymbol{SOF_4} + F$	$\sim 10^{-10}$

Conclusion

- Abatement of PFCs in SWD at atmospheric pressure
  - ✓ Dissociation and fragmentation ⇒ increase with microwave power (electron density)
  - ✓ Reformation vs. oxidation ⇒ gas temperature

☞ DRE increases with increasing specific energy

- Modeling: by-products composition at plasma exhaust adequately described by equilibrium calculations

- Limiting factors
  - ✓ PFCs reformation
  - ✓ Contraction and filamentation of SWDs (limit for discharge tube radius)

Industrial application: UPAS (Air Liquide)