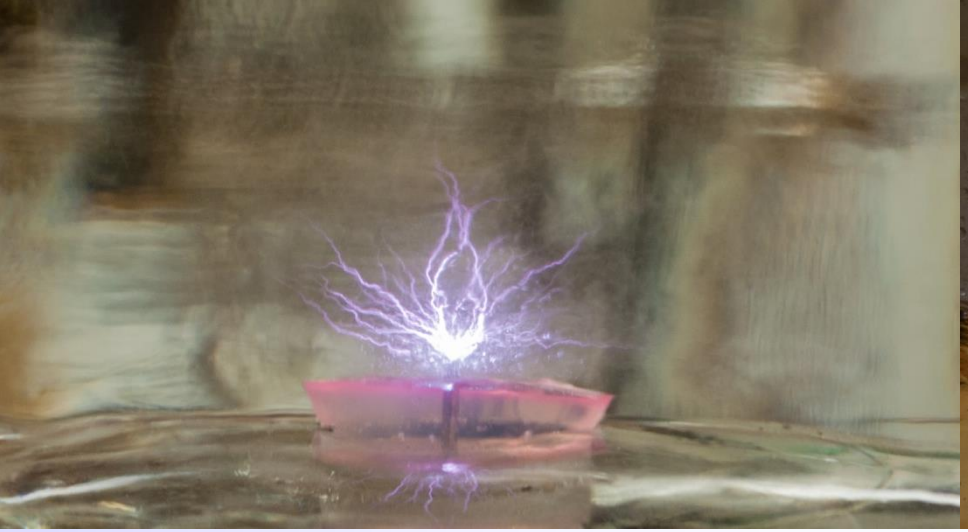

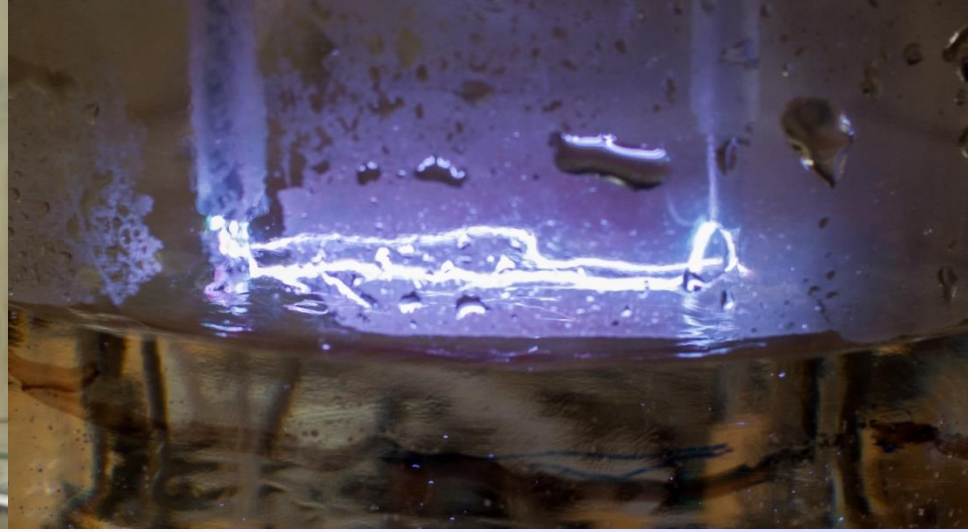

Pursuing a Viable Plasma-Based Water Treatment Process: Identifying Transport Limitations and Investigating the Effect of Reactor Design on the Degradation of Bisphenol A

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Clarkson University, Potsdam, New York

AIChE Annual Meeting, November 2013

Project motivation and objectives

Project motivation:

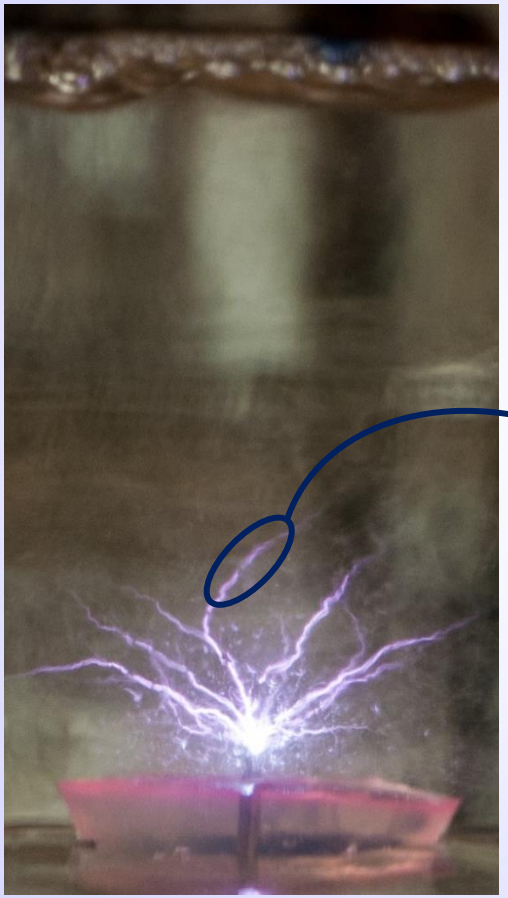
- Current work on plasma-based systems focuses primarily on chemical degradation mechanisms[1,2]
- Very few investigations examine reactor design and process parameter optimization
- EPA grant for developing a pilot-scale plasma-based drinking water treatment system

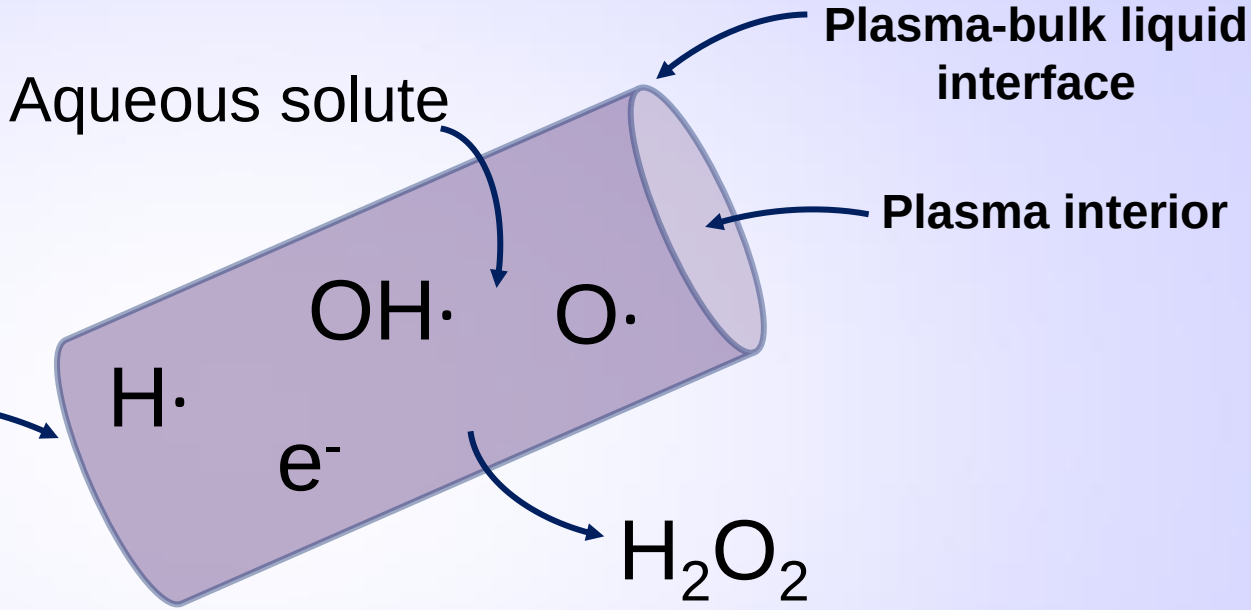
Project objectives:

- Choose appropriate model contaminants
- Optimize the process design
  - Reactor design
  - Electrical parameter optimization

[1]Locke, B.R., Sato, M., Sunka, P., Hoffman, M., Chang, J-S., *Ind. Eng. Chem. Res.*, 2006, 45 (3), pp. 882–905
[2]Joshi, R.P. and Thagard, S.M., *Plasma Chem. Plasma Process*., 2013, 33 (1), pp.17-40

Electrical discharge plasmas in water



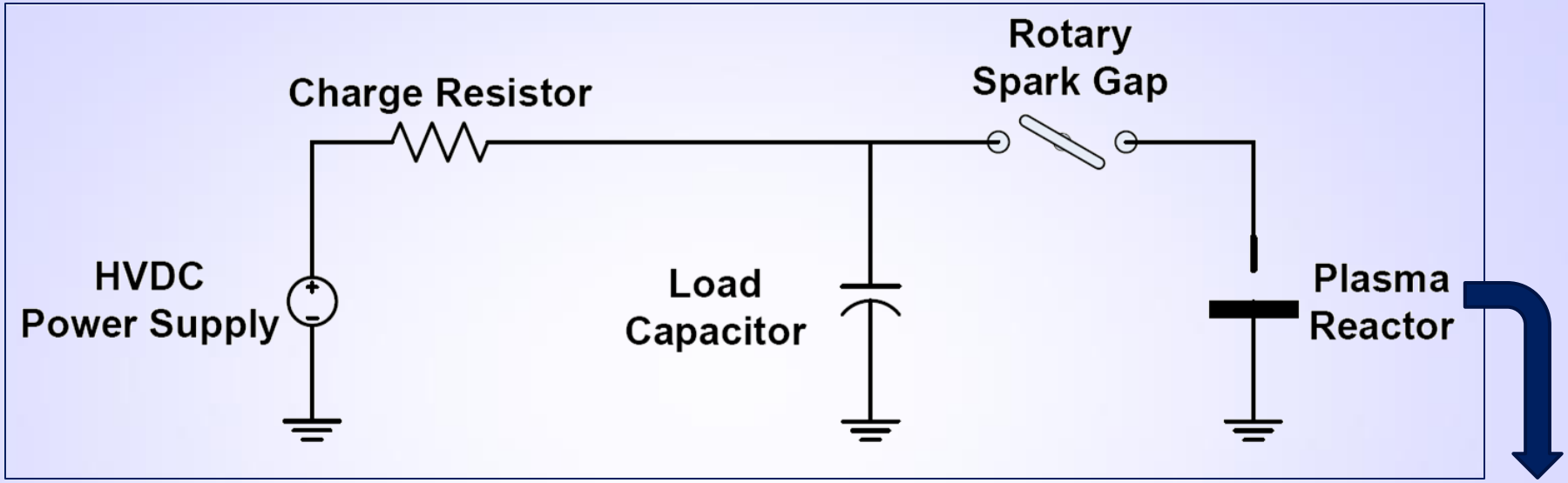


Features of plasma important in treatment process:[3-5]

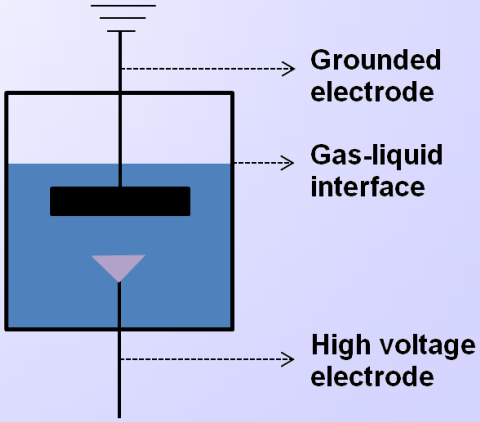
- Oxidative potential of OH: 2.80 V
- Plasma pressure: ~10^9 Pa
- Plasma temperature: >2000 K

[3]Locke, B.R., Sato, M., Sunka, P., Hoffman, M., Chang, J-S., *Ind. Eng. Chem. Res.*, 2006, 45 (3), pp. 882–905
[4]Tarr, M., 2003, CRC Press. ISBN: 0203912551
[5] An, W., Baumung, K., Bluhm, H., *J. Appl. Phys*. ,2007, 101, pp. 053302-053310

Power supply and experimental details



- Source voltage: 16-18 kV
- Discharge frequency: 43 Hz (unless stated otherwise)
- Capacitance: 2 nF (unless stated otherwise)
- Solution volume: 600 mL
- Initial solution conductivity: 300-330 µS/cm
- Initial solution pH: ~5
- Headspace gas: Argon



1. Choose contaminants

Are Some Molecules More Suitable For Plasma Treatment Than Others?

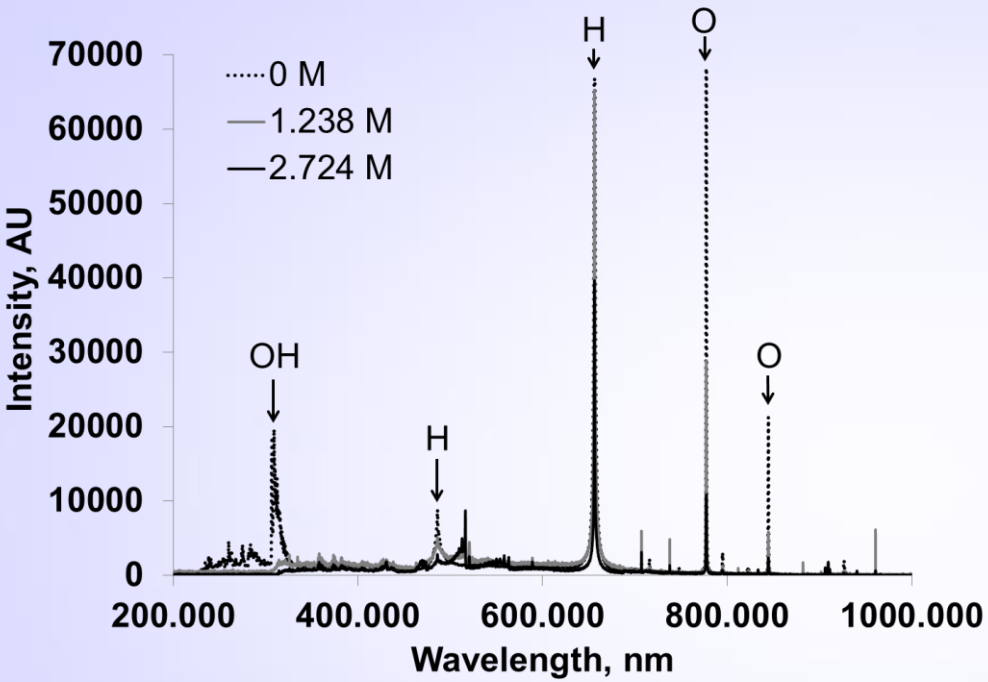
2. Optimize the process design

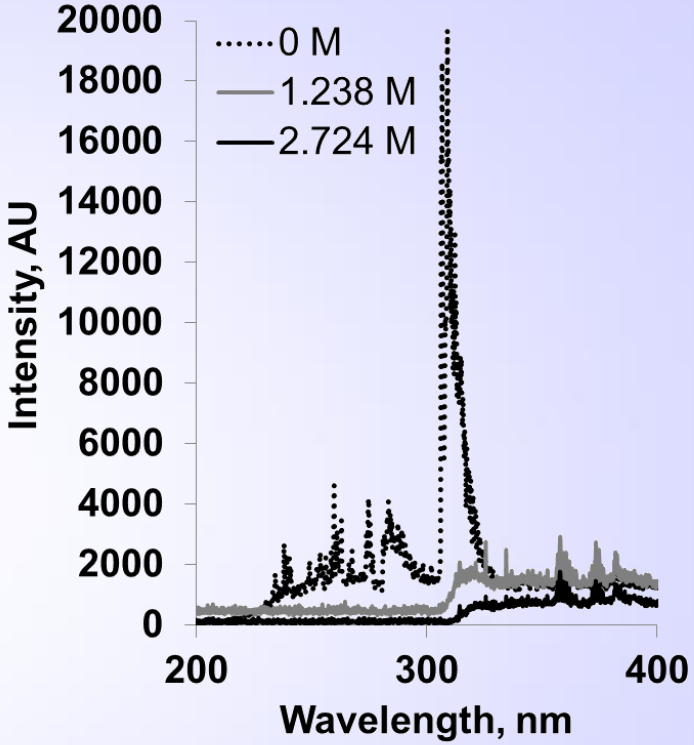
    2.1. Reactor diameter

    2.2. Discharge phase

    2.3. Load capacitance

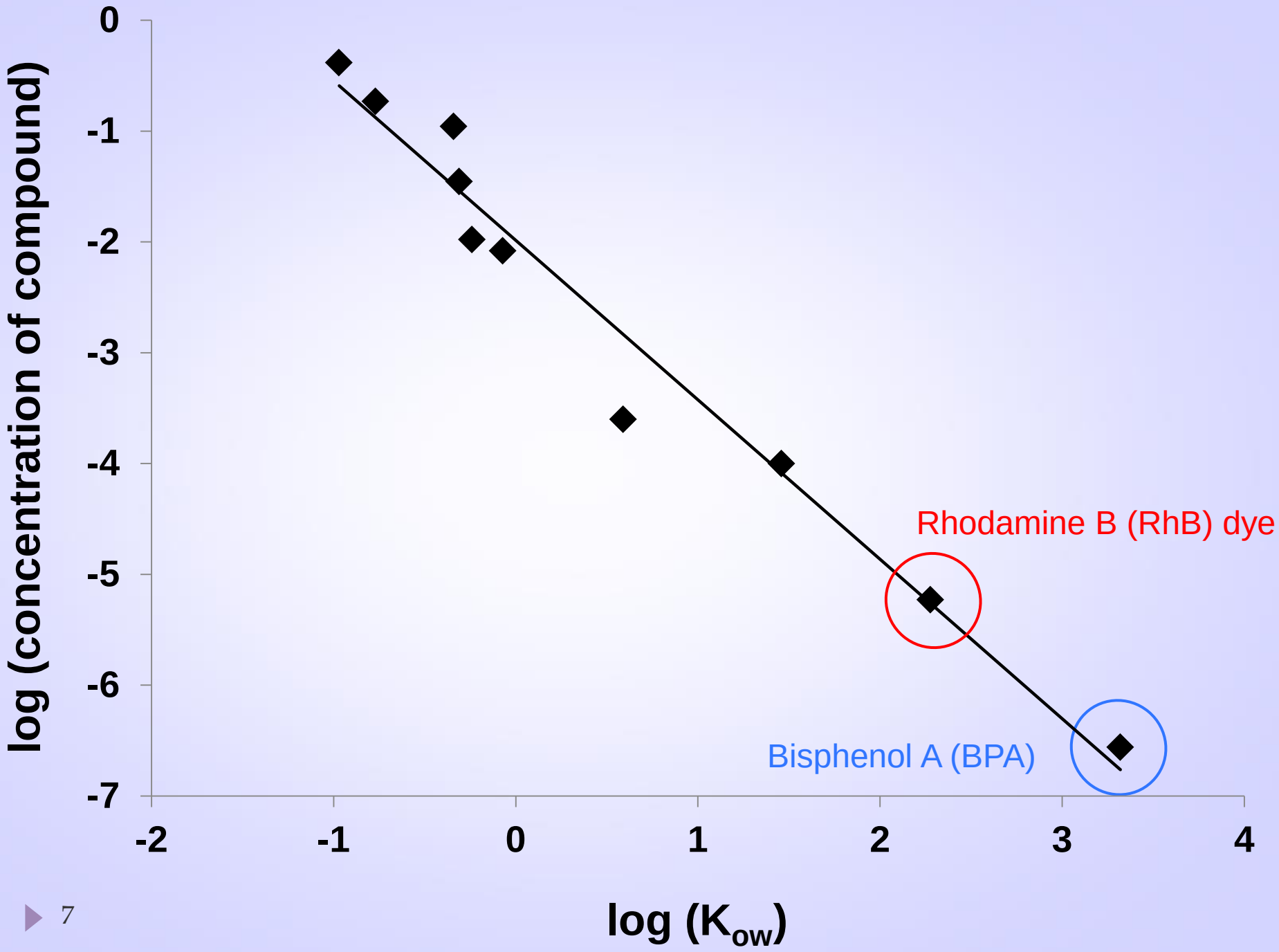
    2.4. Ground plate diameter





Emission spectra of plasma in aqueous solutions containing different concentrations of acetone

OH peak in higher resolution


0
-1
-2
-3
-4
-5
-6
-7
log (concentration of compound)
-2
-1
0
1
2
3
4
log (K_{ow})
Rhodamine B (RhB) dye
Bisphenol A (BPA)

1. Choose contaminants

2. Optimize the process design

    2.1. Reactor diameter

    2.2. Discharge phase

    2.3. Load capacitance

    2.4. Ground plate diameter

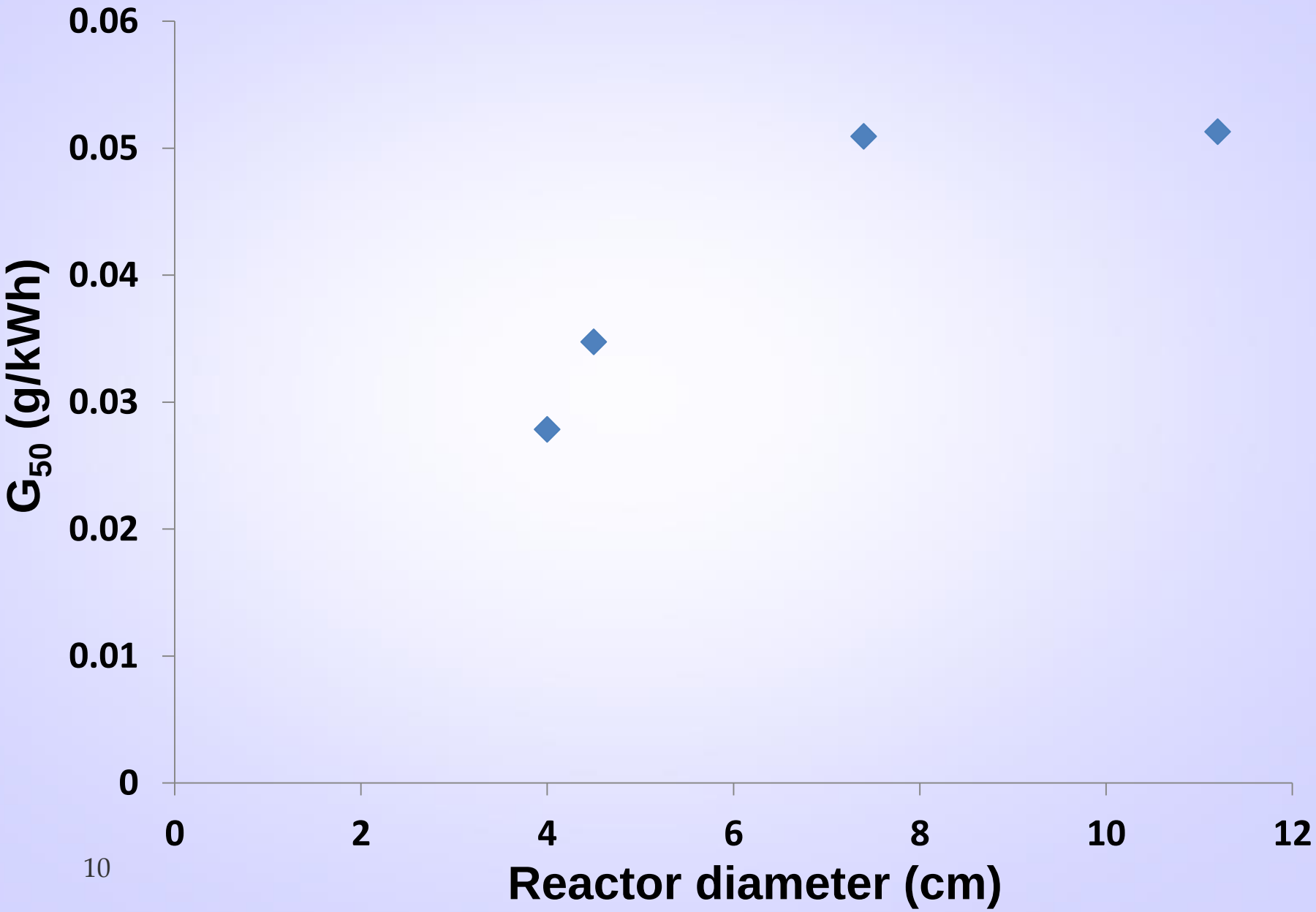
Reactor diameter

The effect of reactor diameter was assessed using the calculated G_{50} value:

$$G_{50} = \frac{0.5 \cdot C_0 \cdot V}{t_{50} \cdot f \cdot E_{pulse}} \left[\frac{g}{kWh} \right]$$

C_0: initial contaminant concentration (g/L)
V: solution volume (L)
t_{50}: time to reduce concentration by 50% (s)
f: discharge frequency (s^{-1})
E_{pulse}: discharge energy per pulse (kWh)

Treatment efficiency vs. reactor diameter



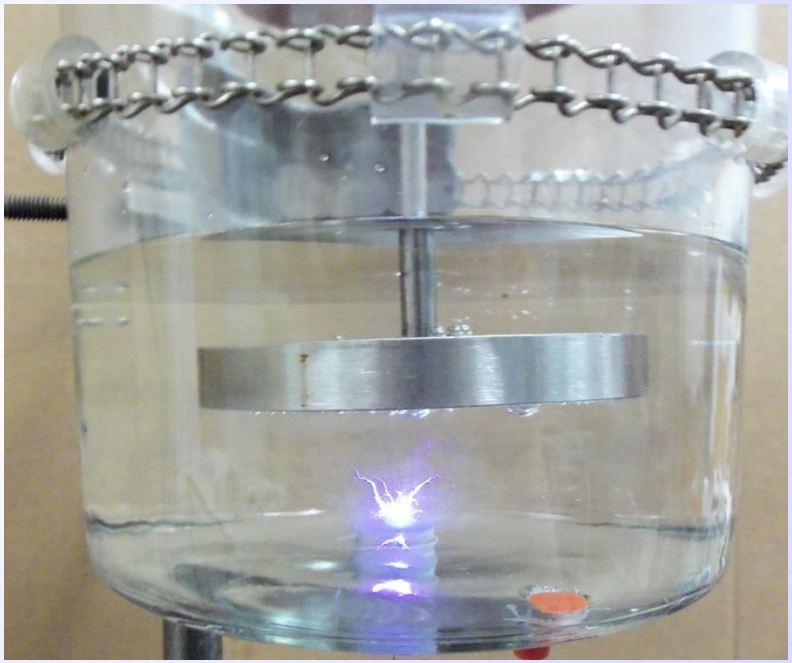
1. Choose contaminants

2. Optimize the process design

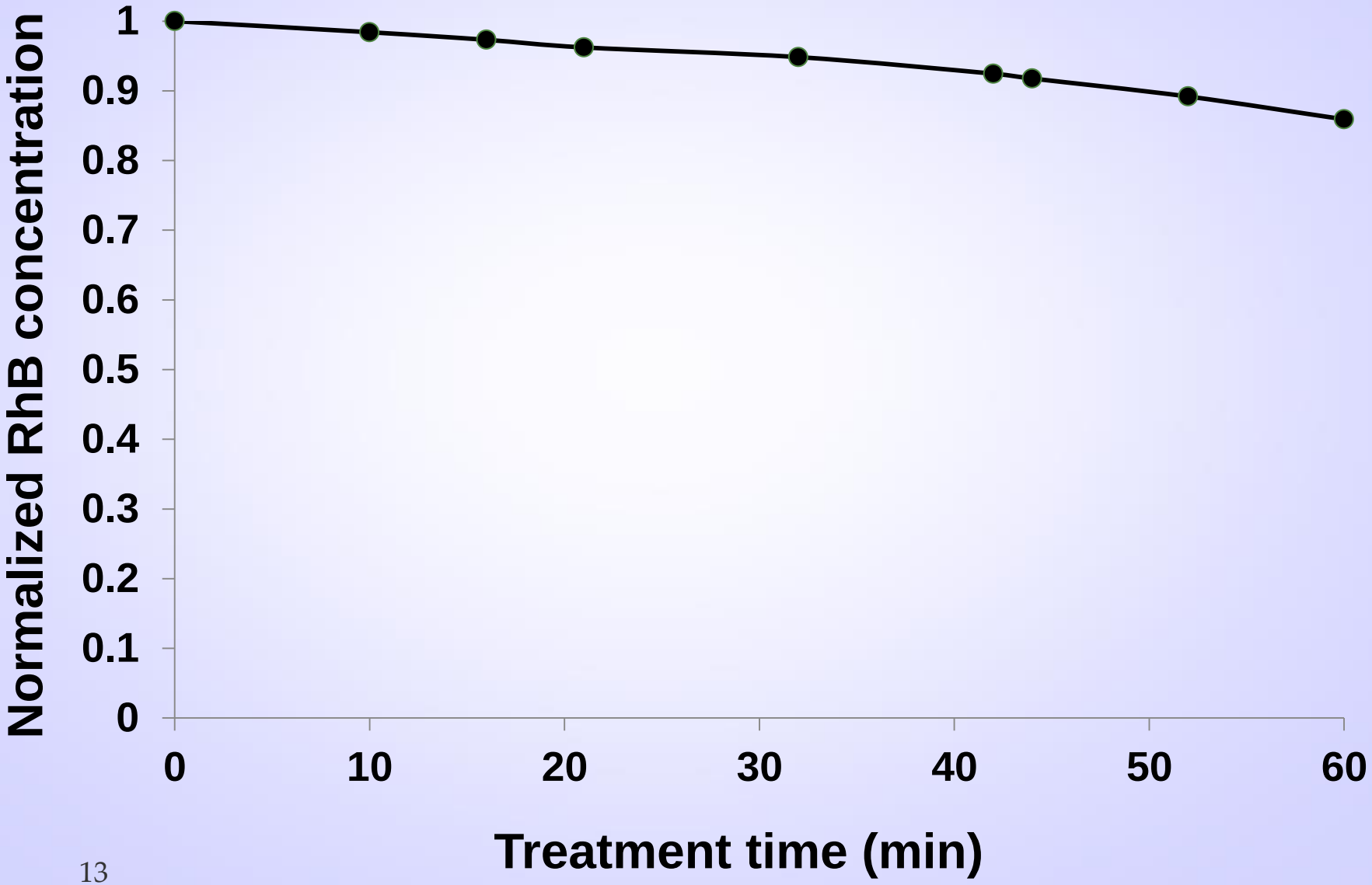
- 2.1. Reactor diameter
- 2.2. Discharge phase
- 2.3. Load capacitance
- 2.4. Ground plate diameter

Point-plane with liquid discharge

- High voltage electrode: nickel chromium (NiCr) (0.8 mm in diameter)
- Ground electrode: stainless steel (SS)

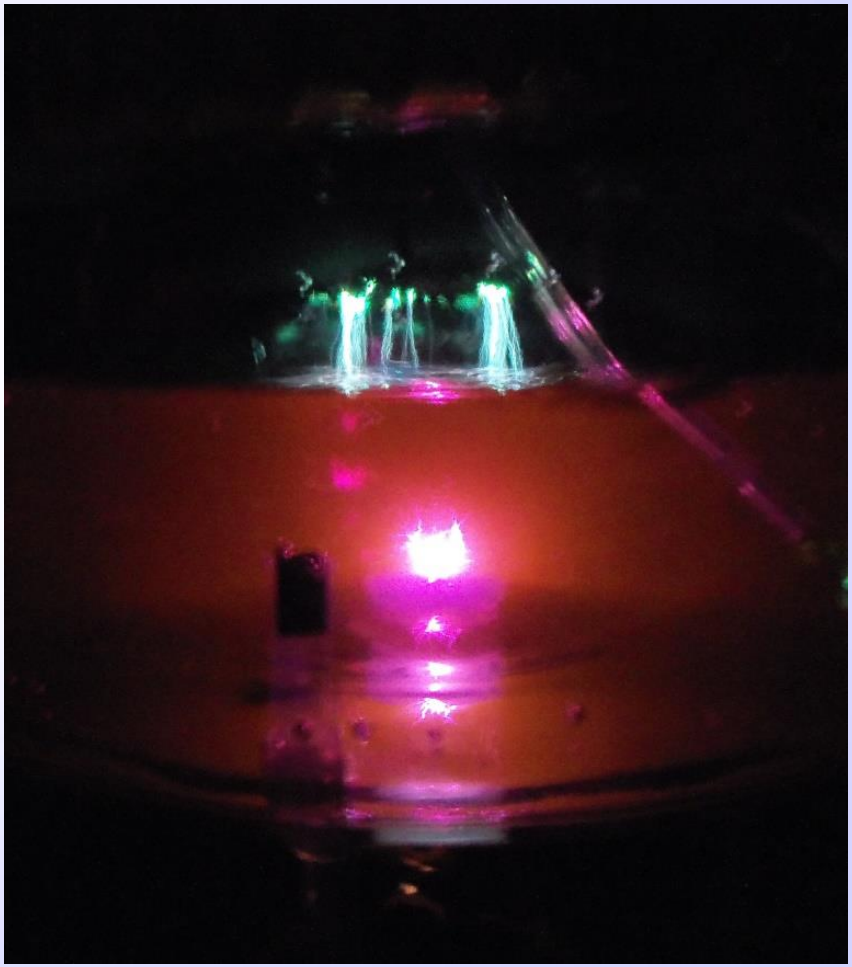


RhB removal for “point-plane with liquid discharge”

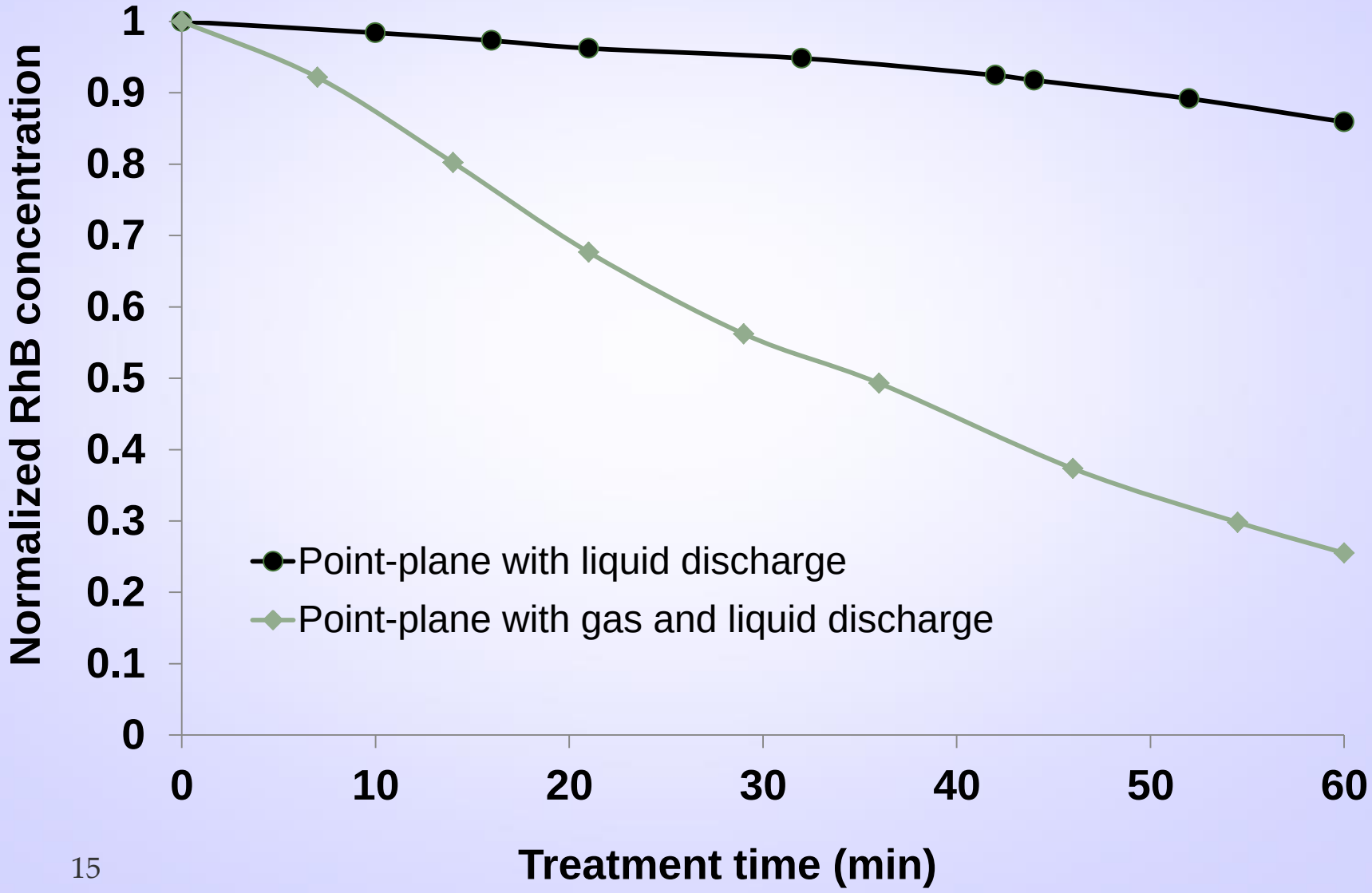


Point-plane with gas and liquid discharge

- High voltage electrode: NiCr
- Ground electrode: reticulated vitreous carbon (RVC)

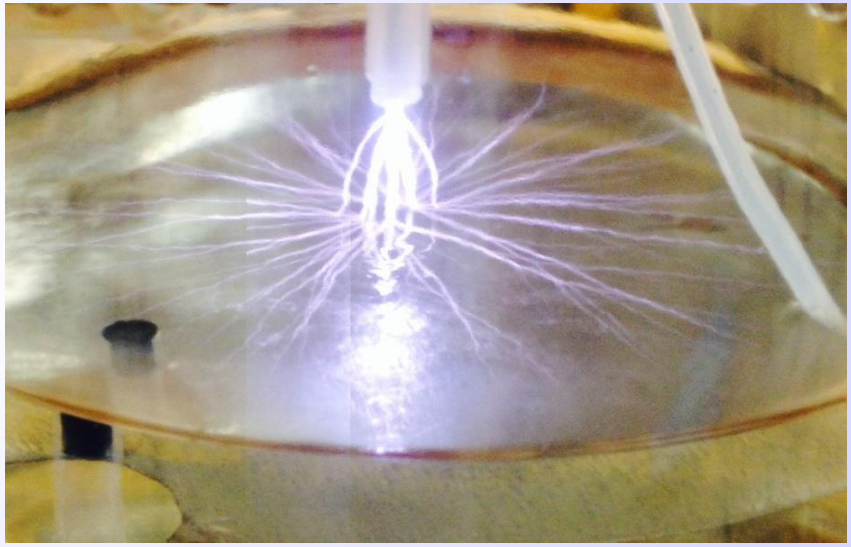


RhB removal for different reactor configurations

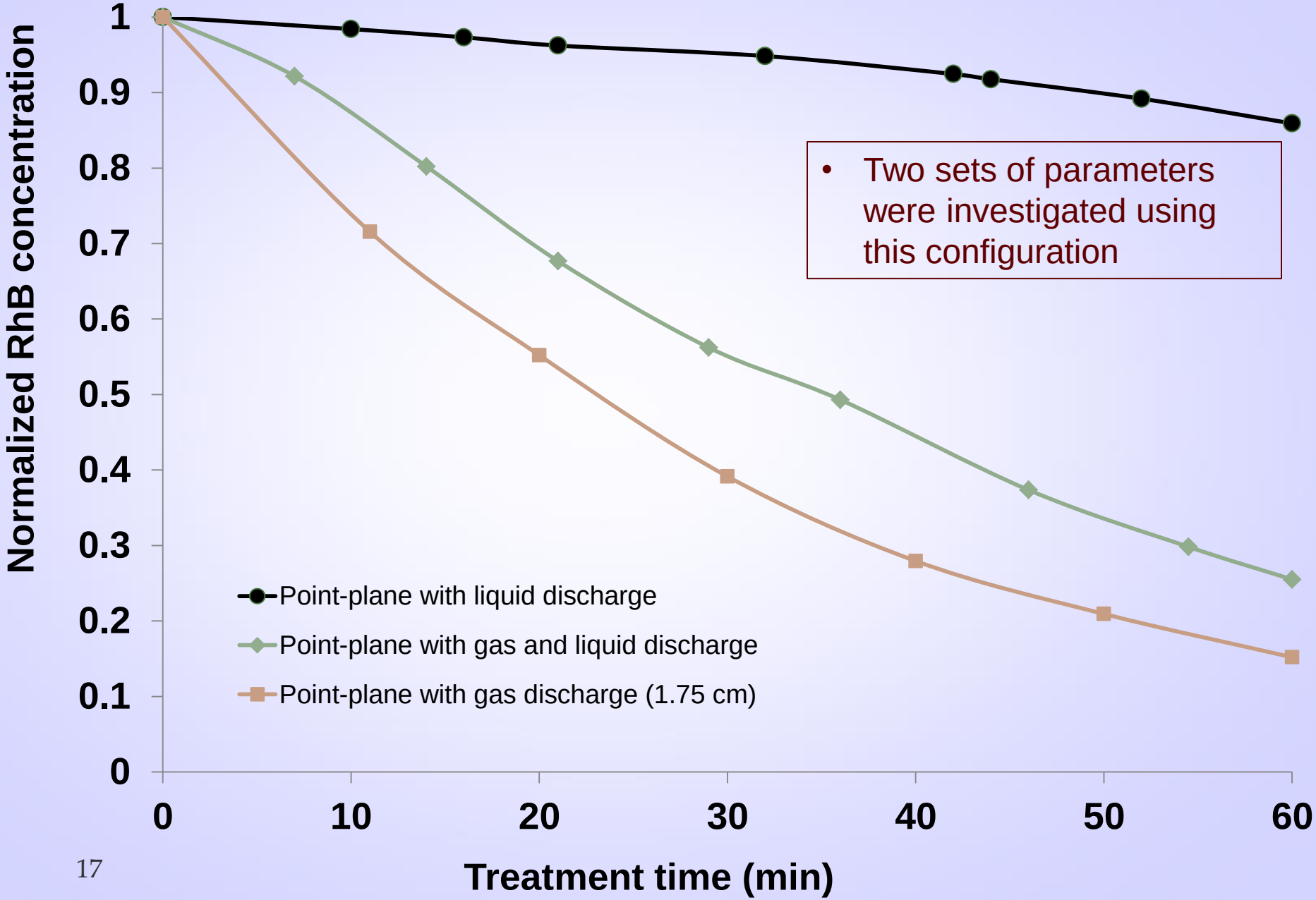


Point-plane with gas discharge

- High voltage electrode: NiCr
- Ground electrode: SS



RhB removal for different reactor configurations



- Two sets of parameters were investigated using this configuration

1. Choose contaminants

2. Optimize the process design

- 2.1. Reactor diameter
- 2.2. Discharge phase
- 2.3. Load capacitance
- 2.4. Ground plate diameter

Load capacitor

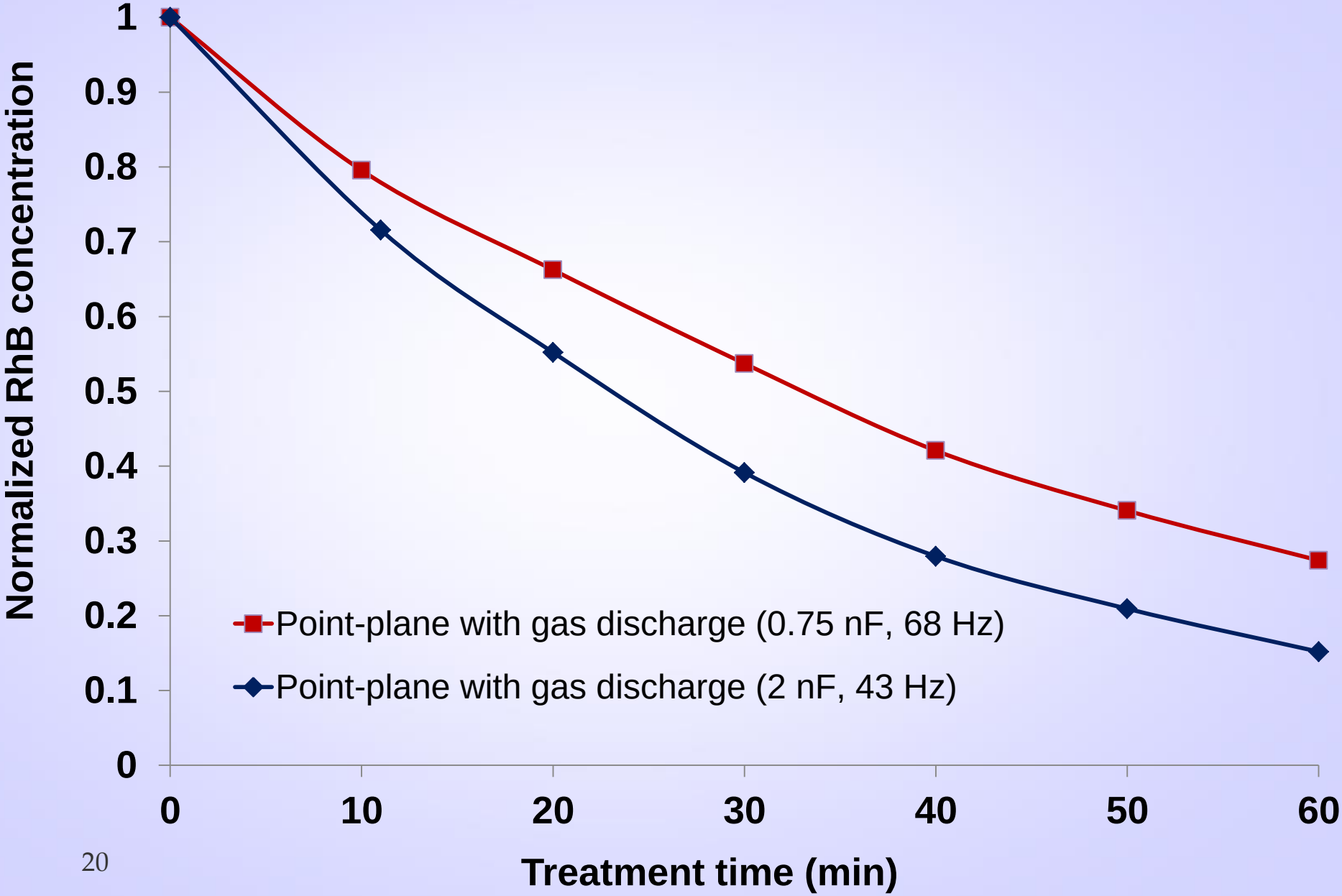
For a limited HVDC power supply output increasing the capacitance will result in a lower achievable discharge frequency for the specified source voltage

$$C = \frac{P_c}{V_c^2 \cdot \ln\left(\frac{V_c}{V_c - V_s}\right)} \frac{1}{f}$$

C: capacitance of load capacitor (F)
f: discharge frequency (Hz)
V_s: voltage at capacitor (source voltage) (V)
P_c: HVDC supply output power (W)
V_c: charge voltage (PS output voltage) (V)

(C and f are inversely related)

RhB removal for different load capacitors



1. Choose contaminants

2. Optimize the process design

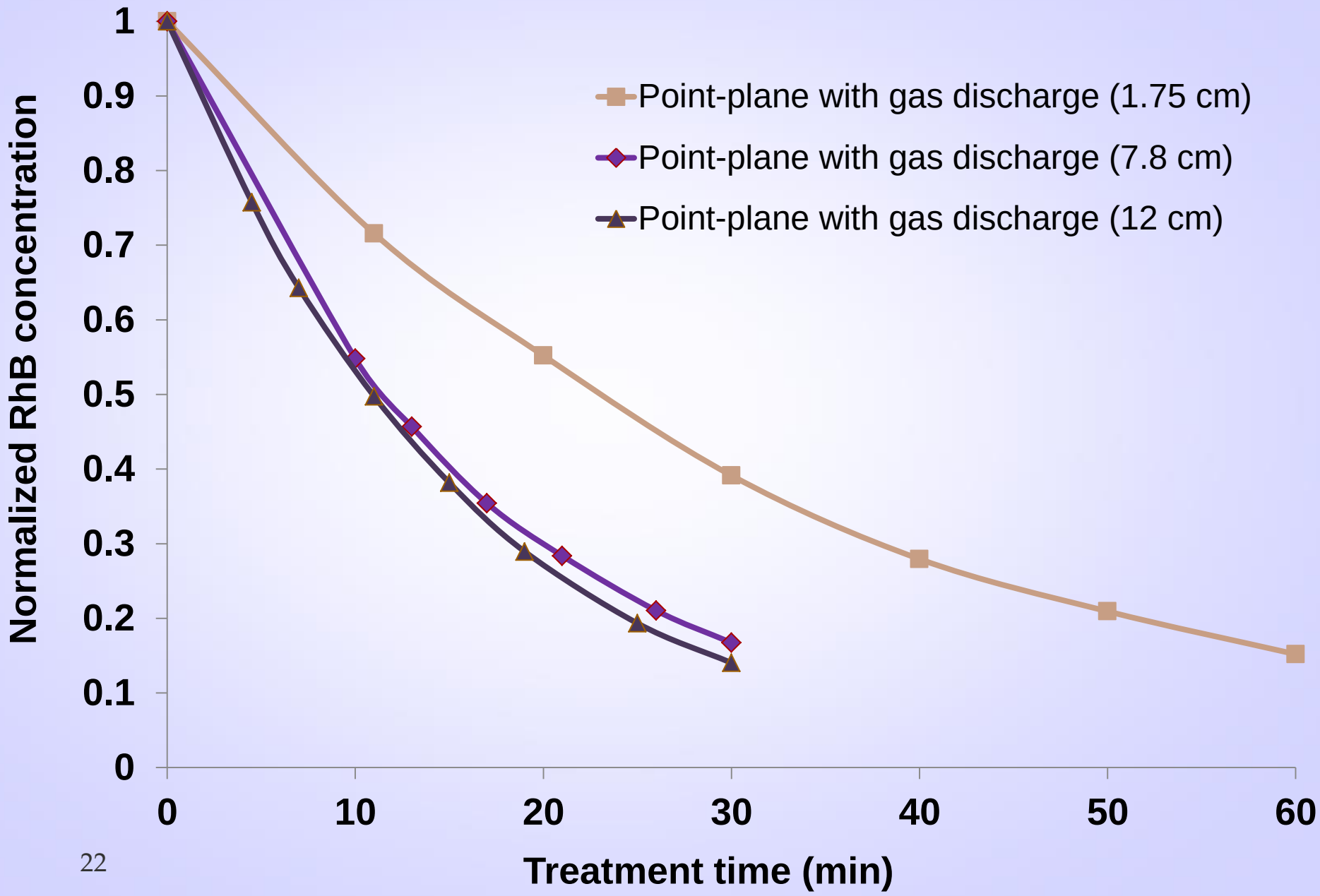
   2.1. Reactor diameter

   2.2. Discharge phase

   2.3. Load capacitance

   2.4. Ground plate diameter

RhB removal for different ground diameters



Ground plate size

Up to 7.8 cm, increasing the ground plate size resulted in better removal

Possible reasons:

- Larger plate enables <u>higher electrical discharge current</u>
- Larger plate causes the plasma to spread wider over the surface, facilitating <u>better contact with the dye</u>

1. Choose contaminants

2. Optimize the process design

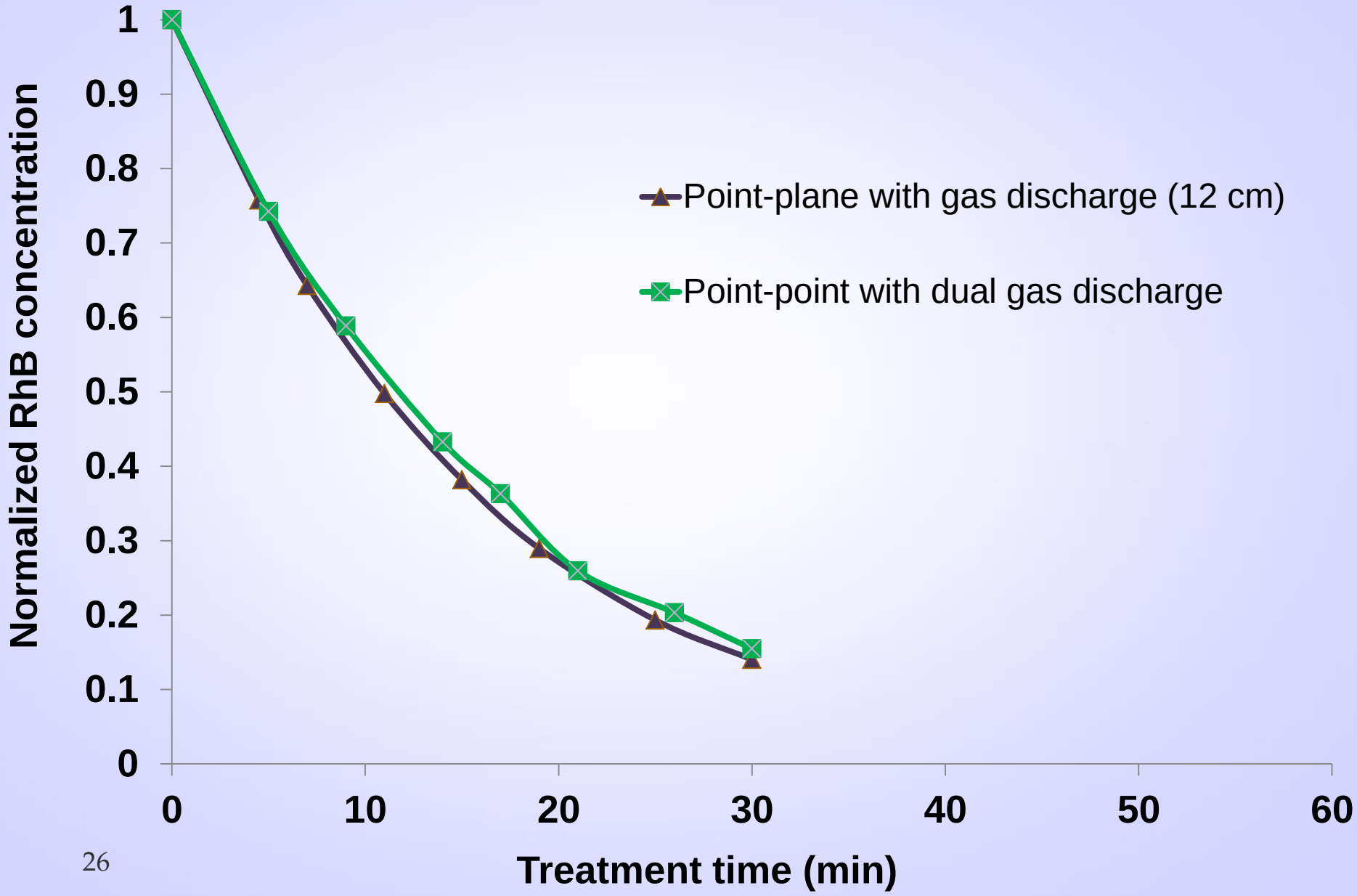
- 2.1. Reactor diameter
- 2.2. Discharge phase
- 2.3. Load capacitance
- 2.4. Ground plate diameter
  - 2.4.1 Electrical current hypothesis
  - 2.4.2 Contact hypothesis

Electrical discharge current hypothesis

Point-point with dual gas discharge

High voltage and ground electrodes: RVC

RhB removal for different reactor configurations



1. Choose contaminants

2. Optimize the process design

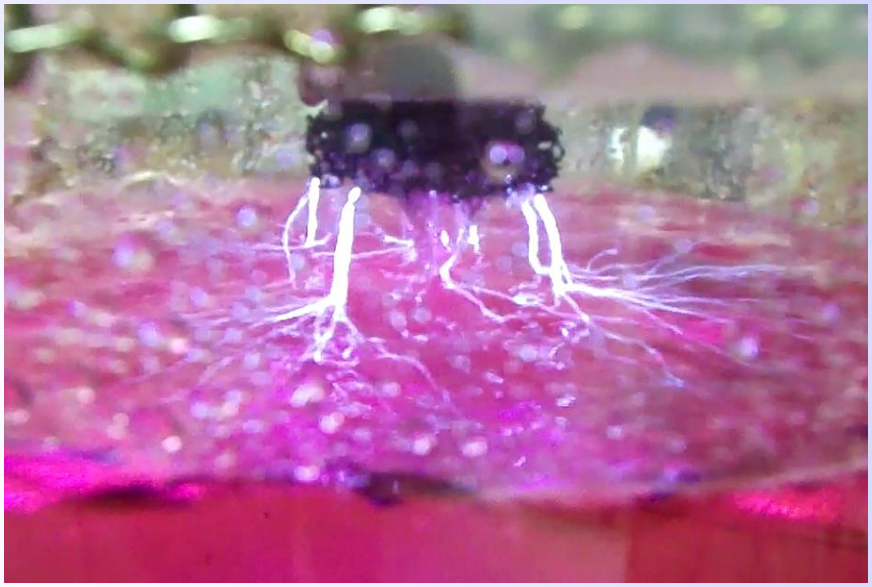
- 2.1. Reactor diameter
- 2.2. Discharge phase
- 2.3. Load capacitance
- 2.4. Ground plate diameter
  - 2.4.1 Electrical current hypothesis
  - 2.4.2 Contact hypothesis

Contact hypothesis

Incorporating liquid feed: 5 configurations in which liquid was pumped through or around the discharge electrodes to improve the contact rate

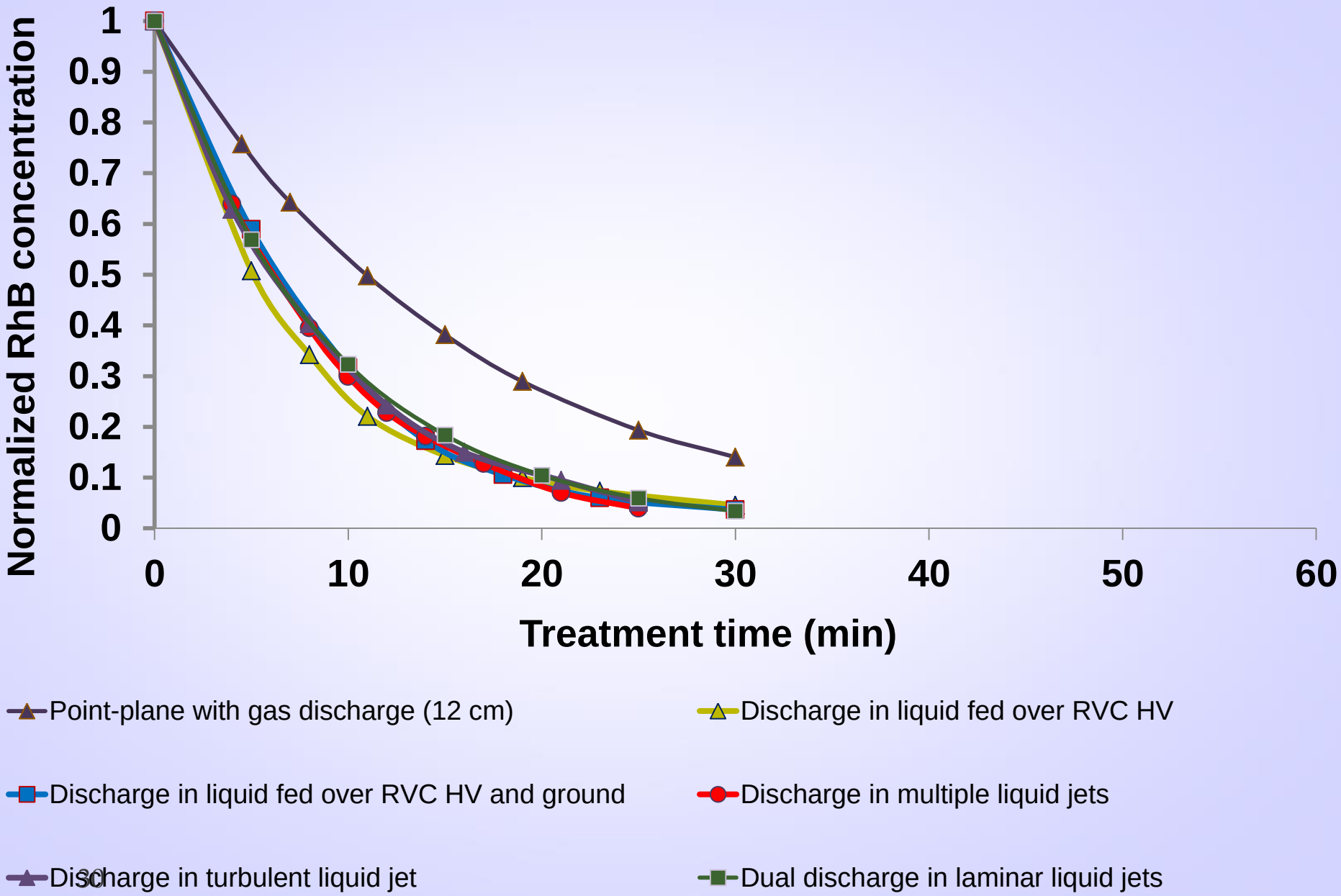
- Discharge in liquid fed through RVC HV
- Dual discharge in liquid fed through RVC HV and ground
- Discharge in turbulent liquid jet
- Dual discharge in laminar liquid jets
- Discharge in multiple liquid jets

Dual discharge in laminar liquid jets

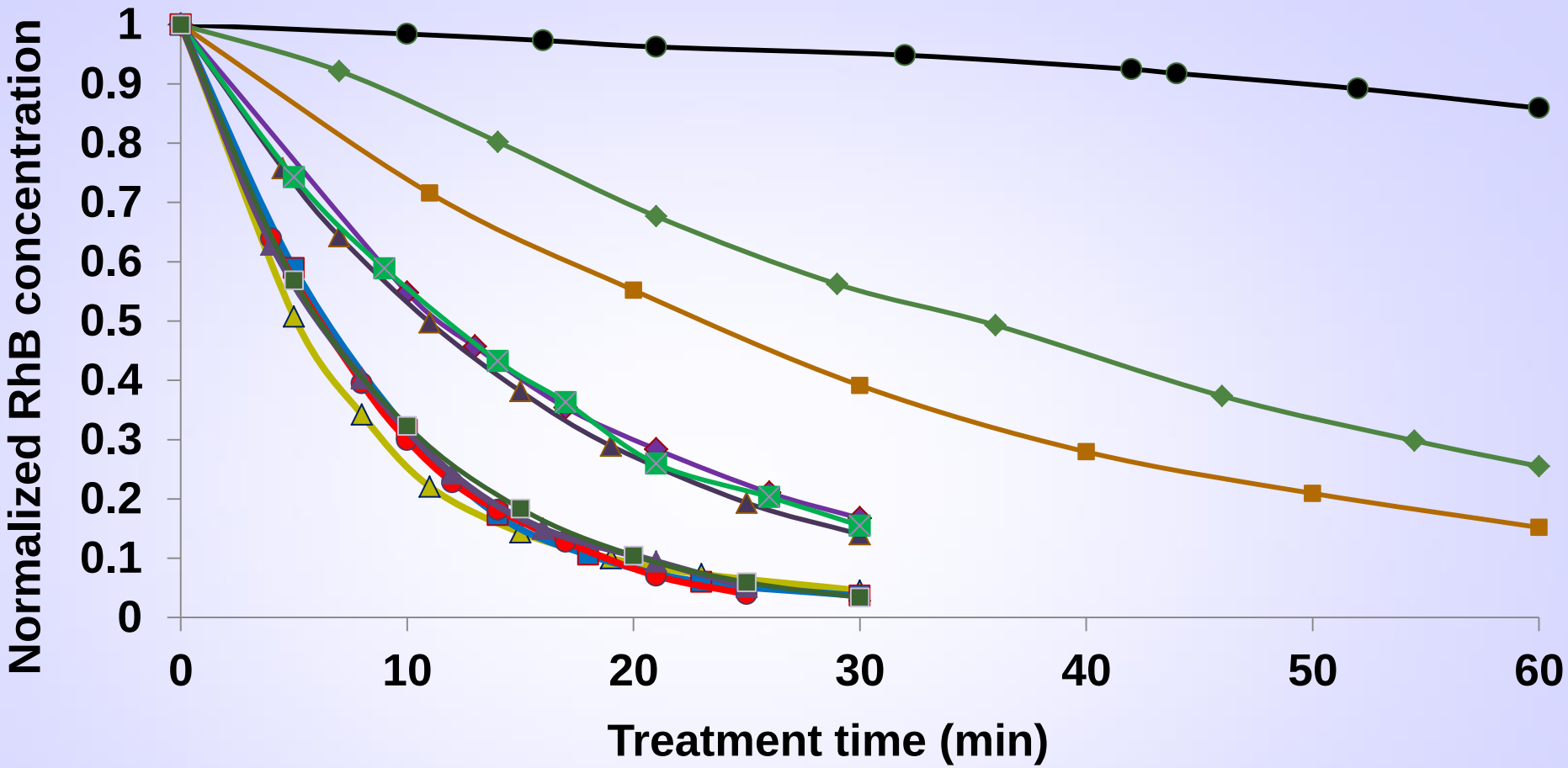


Discharge in liquid fed through RVC HV

RhB removal for different reactor configurations

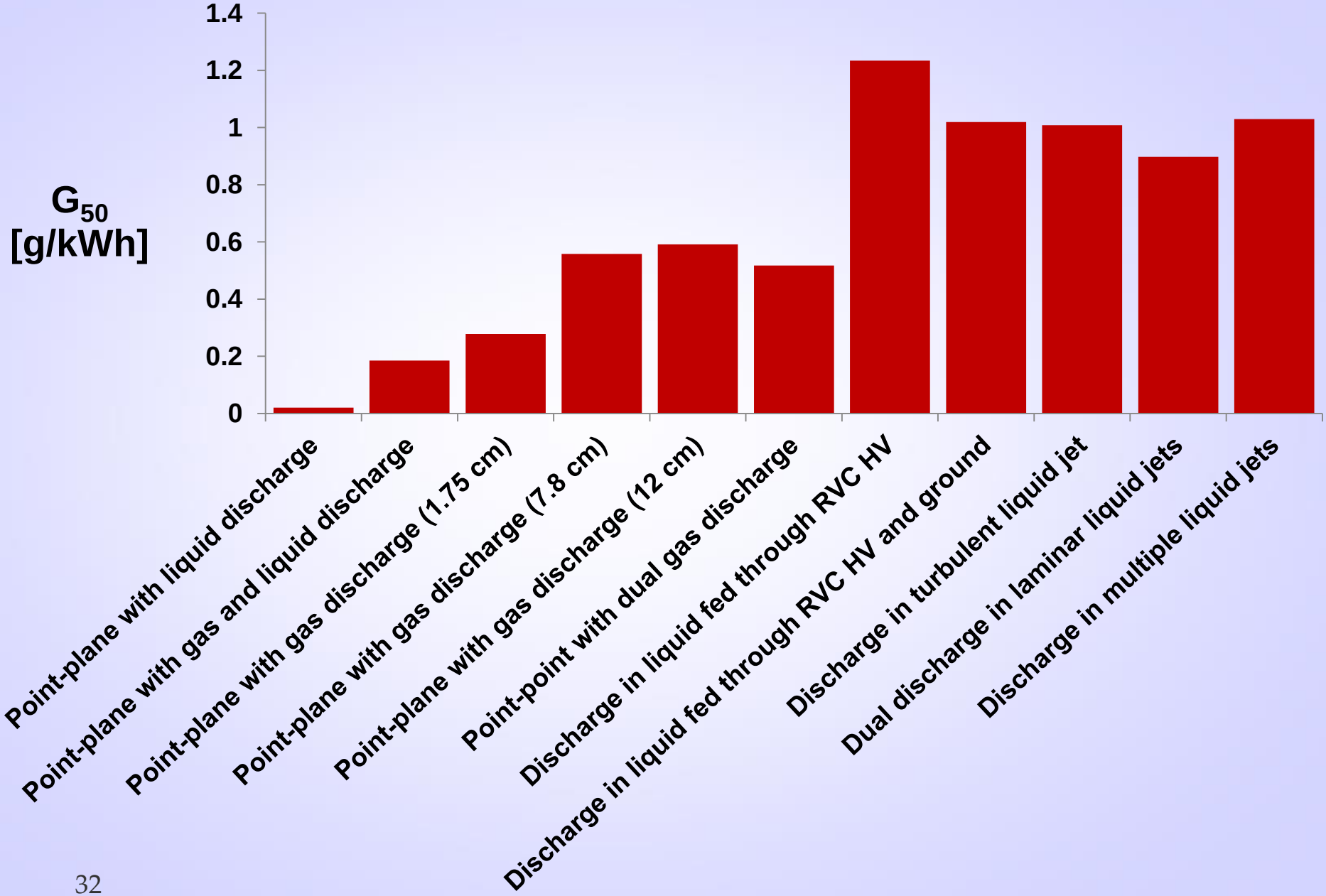


RhB removal for all reactor configurations

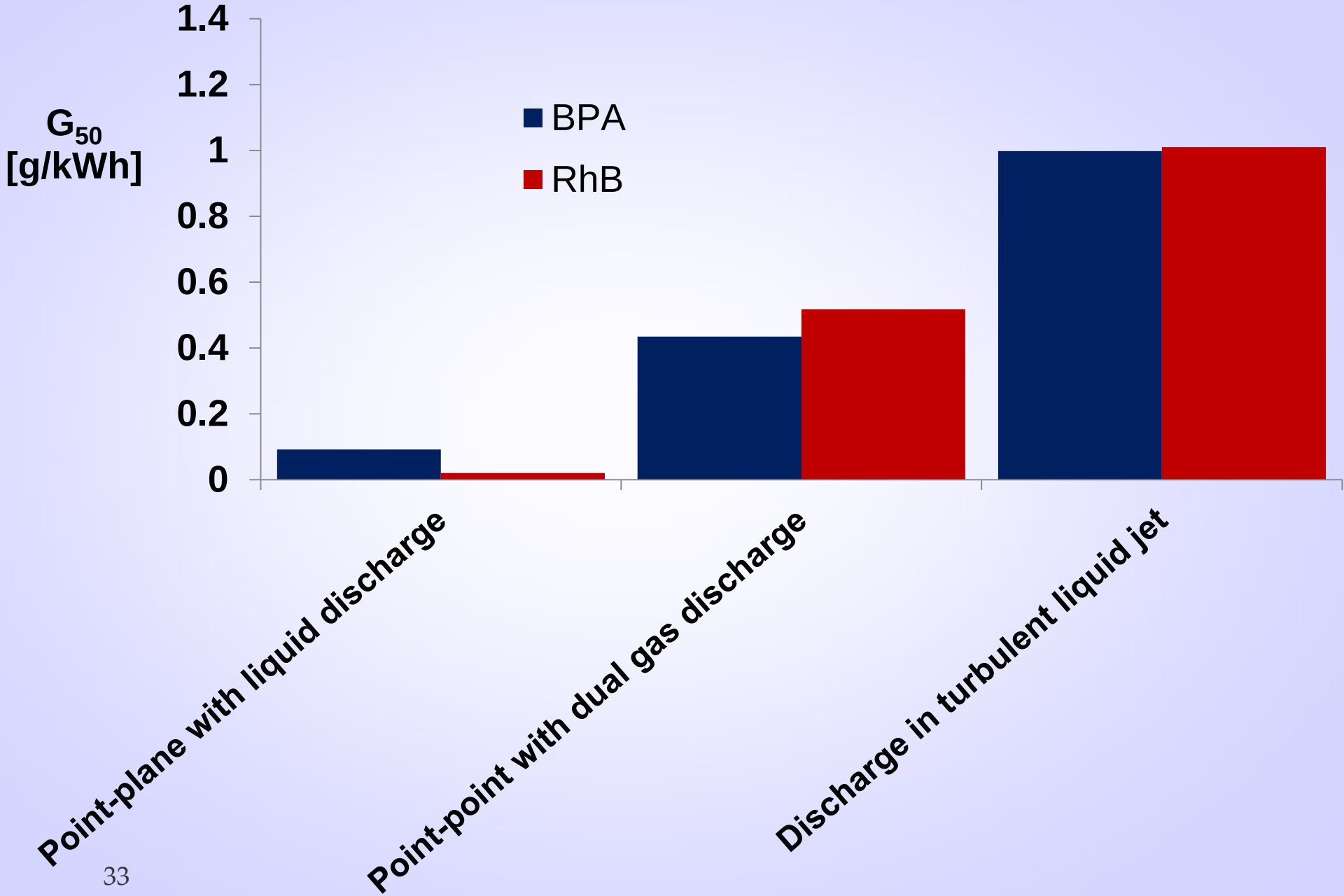


- Point-plane with liquid discharge
- Point-plane with gas and liquid discharge
- Point-plane with gas discharge (1.75 cm)
- Point-plane with gas discharge (7.8 cm)
- Point-plane with gas discharge (12 cm)
- Point-point with dual gas discharge
- Discharge in liquid fed over RVC HV
- Discharge in liquid fed over RVC HV and ground
- Discharge in multiple liquid jets
- Discharge in turbulent liquid jet
- Dual discharge in laminar liquid jets

G_{50} for RhB experiments for all reactors



G_{50} with different reactors for RhB and BPA



Conclusions

Effect of transport properties

- For plasma discharges in liquid, a compound's hydrophobicity determines its ability to diffuse into the plasma channel
- Considering the hydrophobicity of known contaminants may help to determine the applicability of this treatment strategy

Effect of process design

- Increasing the diameter of the reactor and ground plate strongly enhances removal efficiency, until some maximum is reached
- Gas phase discharge is more effective than liquid phase discharge
- Increasing capacitance at expense of frequency yields faster removal
- Discharges directly in feed achieve more rapid removal

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