Advanced Redox Technology (ART) Lab
고도산화환원 환경공학 연구실

http://artlab.re.kr

AOPs Based on Electrochemical Reactions and Electrical Discharge

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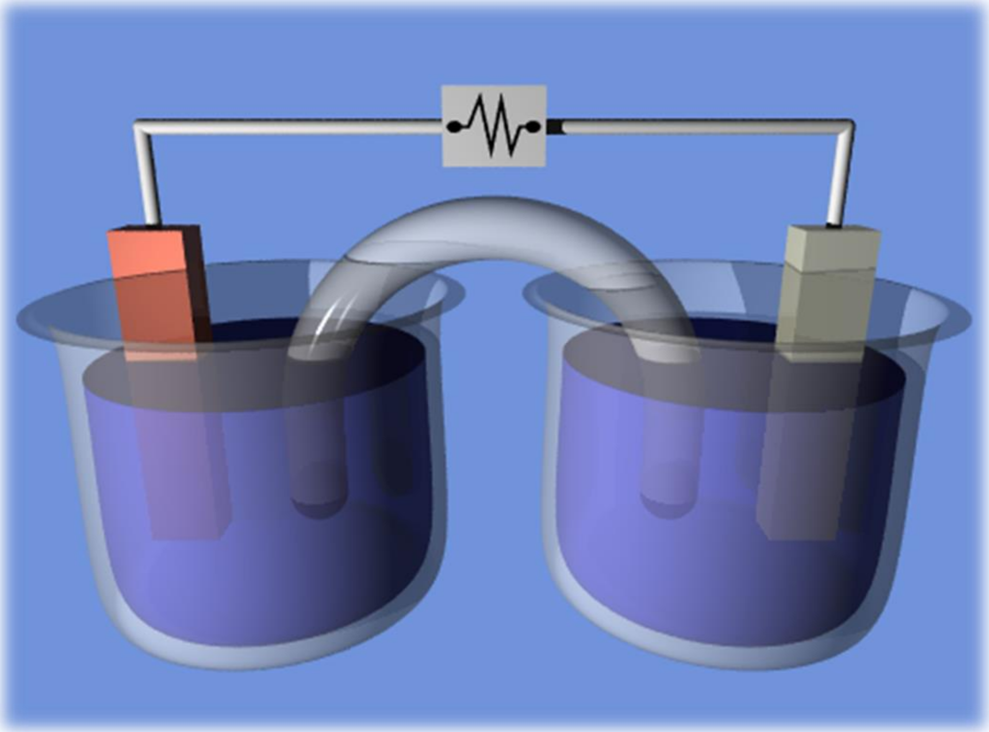
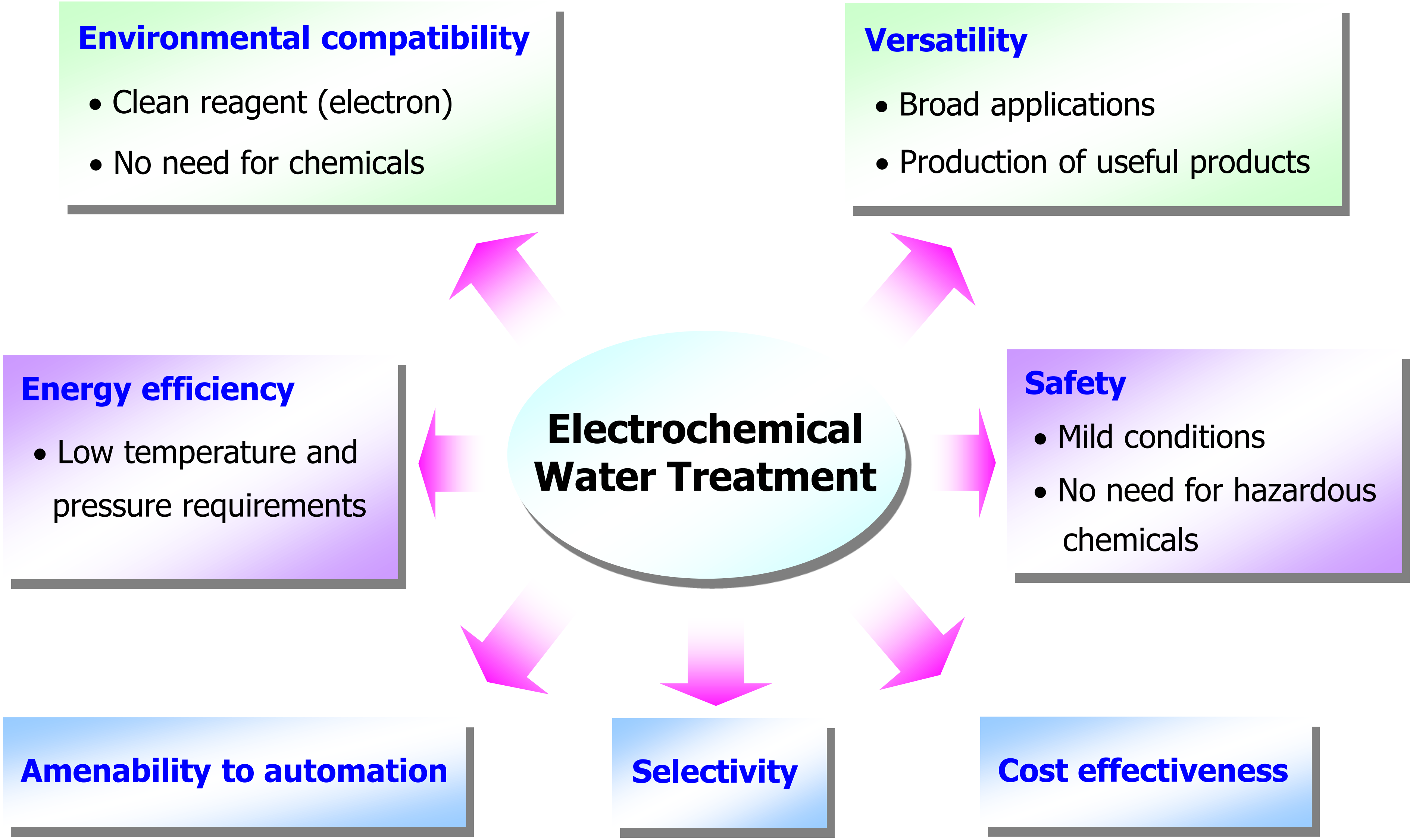
Electrochemical Processes



Characteristics of Electrochemical Water Treatment



Electrochemical Water Treatment

Environmental compatibility

- Clean reagent (electron)
- No need for chemicals

Versatility

- Broad applications
- Production of useful products

Energy efficiency

- Low temperature and pressure requirements

Safety

- Mild conditions
- No need for hazardous chemicals

Amenability to automation

Selectivity

Cost effectiveness

Electrolysis

- **Bulk electrolysis**

Electrochemical alteration of the composition of bulk solution, an analyte is quantitatively converted from its original oxidation state to a new oxidation state (i.e., either reduced or oxidized)

- **Modes of bulk electrolysis**

Controlled-potential

- The most desirable for bulk electrolysis

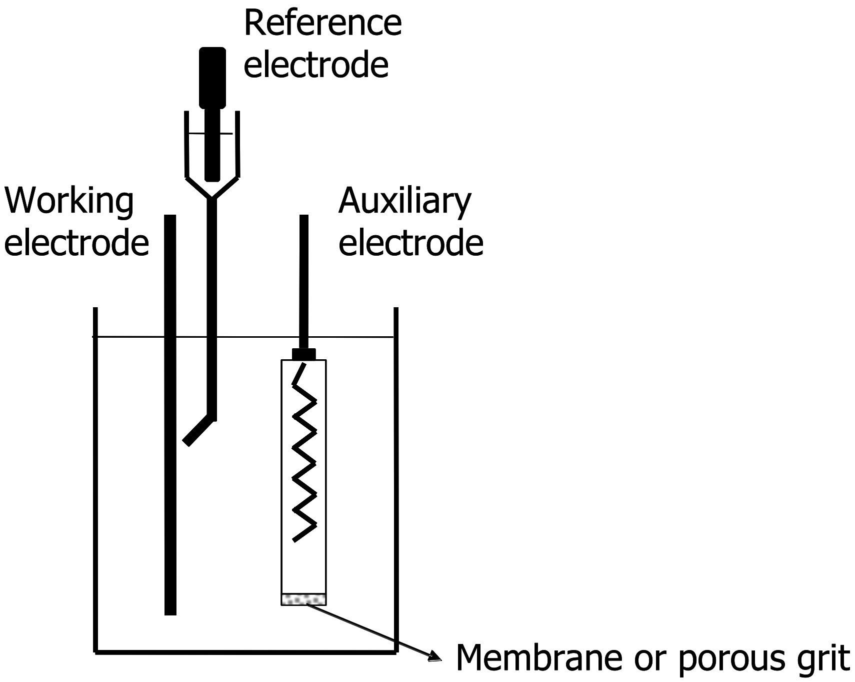
- Requires a **potentiostat** with large output current and voltage capabilities and **reference electrode**

- **The auxiliary electrode** is placed in a separate compartment isolated from the working electrode compartment.

Controlled-current

- Applicable to large-scale and flow electrosynthesis, to galvanic cells.

Three Electrode System



General scheme of
the three-electrode cell configuration

Electrodes

1) Working electrode

The electrode at which the reaction of interest occurs.

2) Counter electrode (auxiliary electrode)

The electrode which facilitates to pass the current of working electrode.

3) Reference electrode

The electrode which standardizes the potential of working electrode.

Separator

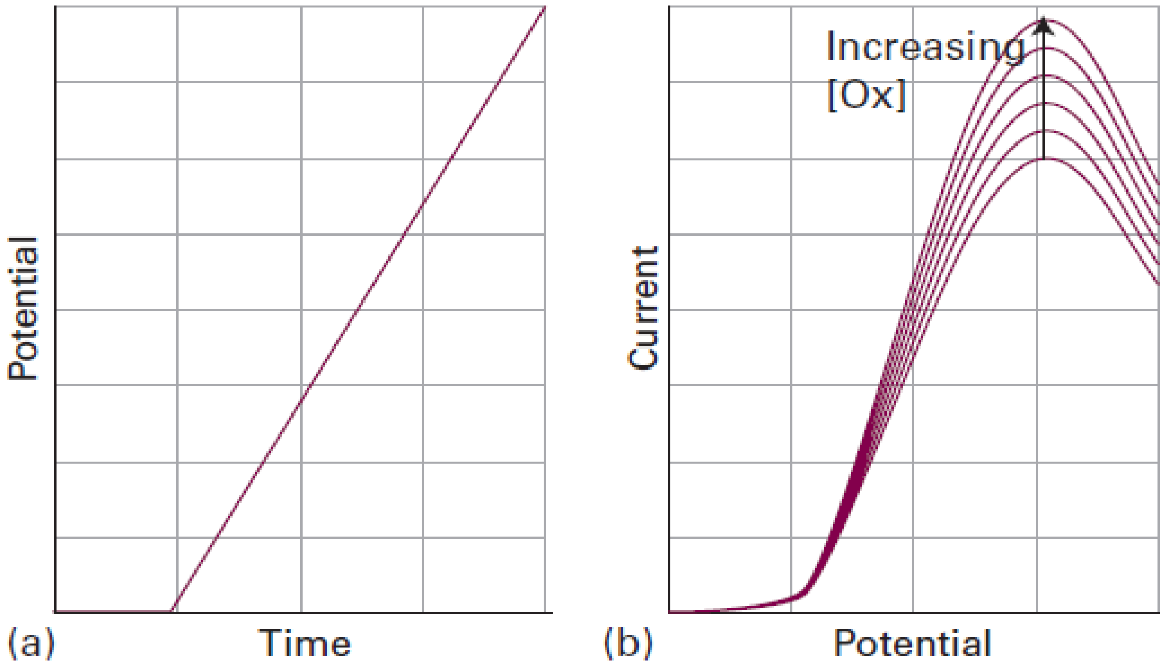
The unit which separates the compartment with a working electrode to the other with a counter electrode.

Voltammetry

- **Voltammetry**

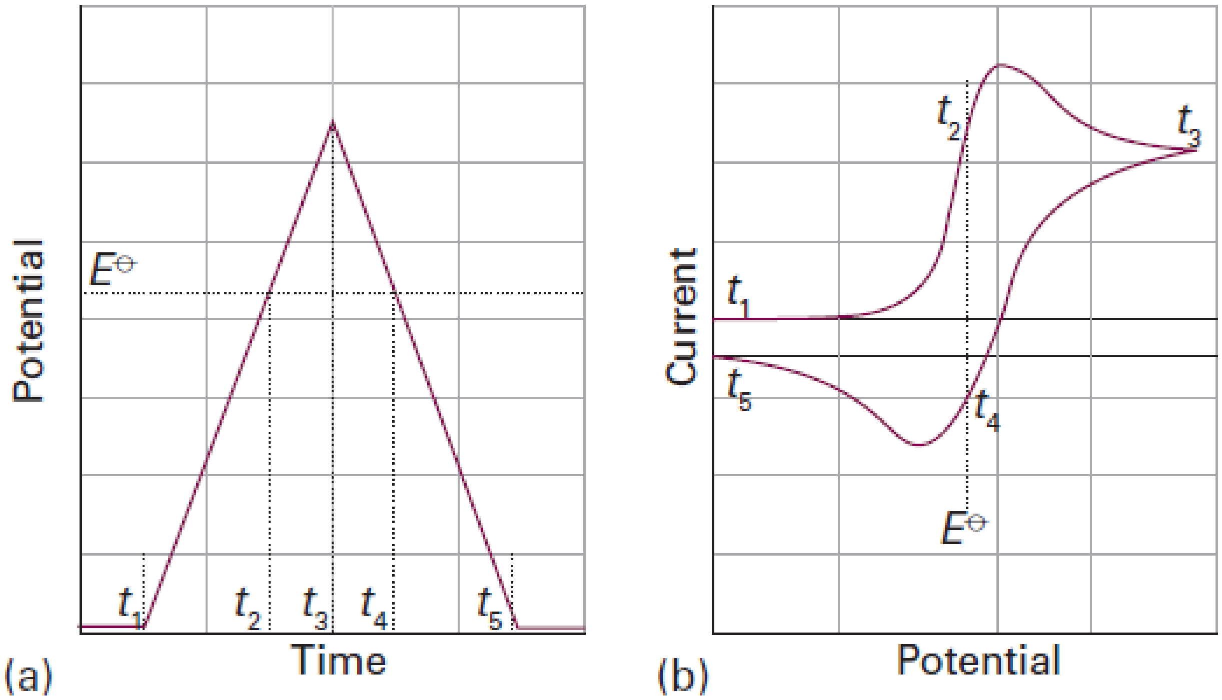
  The study of the current through an electrode as a function of the applied potential difference

Linear-sweep voltammetry

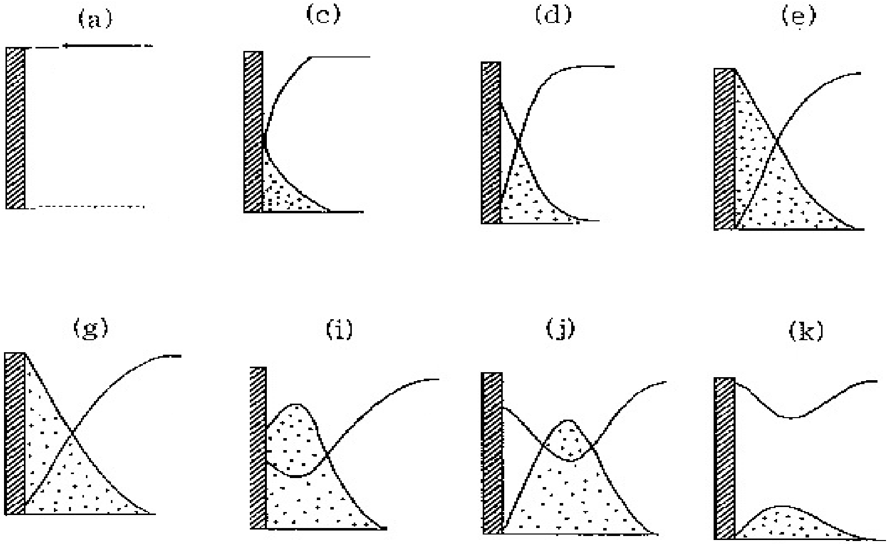


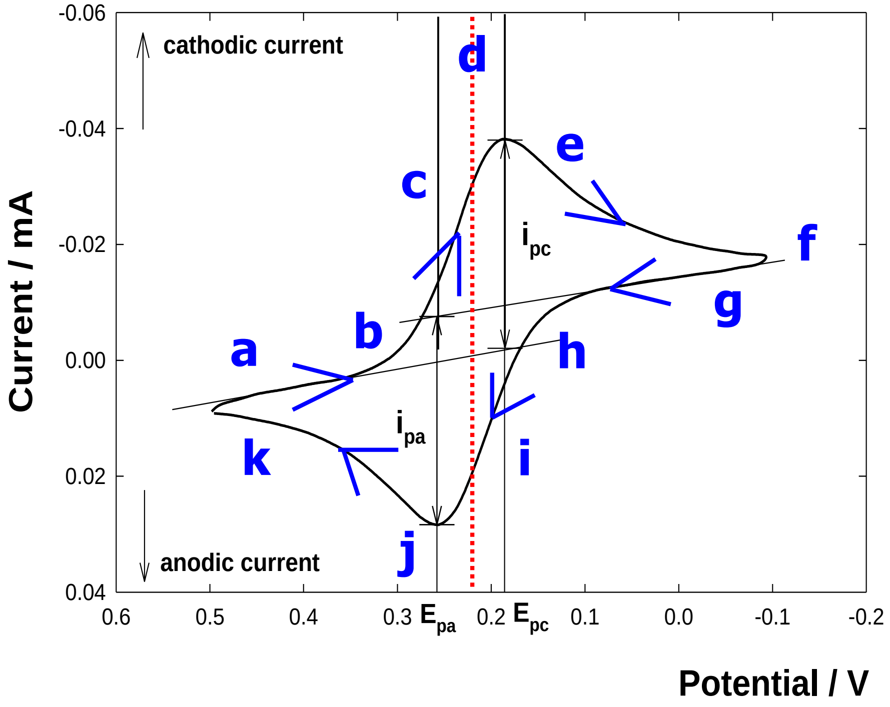
Voltammetry

Cyclic voltammetry



Voltammetry





Concentration profiles

Resulting cyclic voltammogram

$(Fe(CN)_6^{3-} + e^- \rightarrow Fe(CN)_6^{4-})$

Typical Methods of Electrochemical Processes

- **Direct electrolysis**

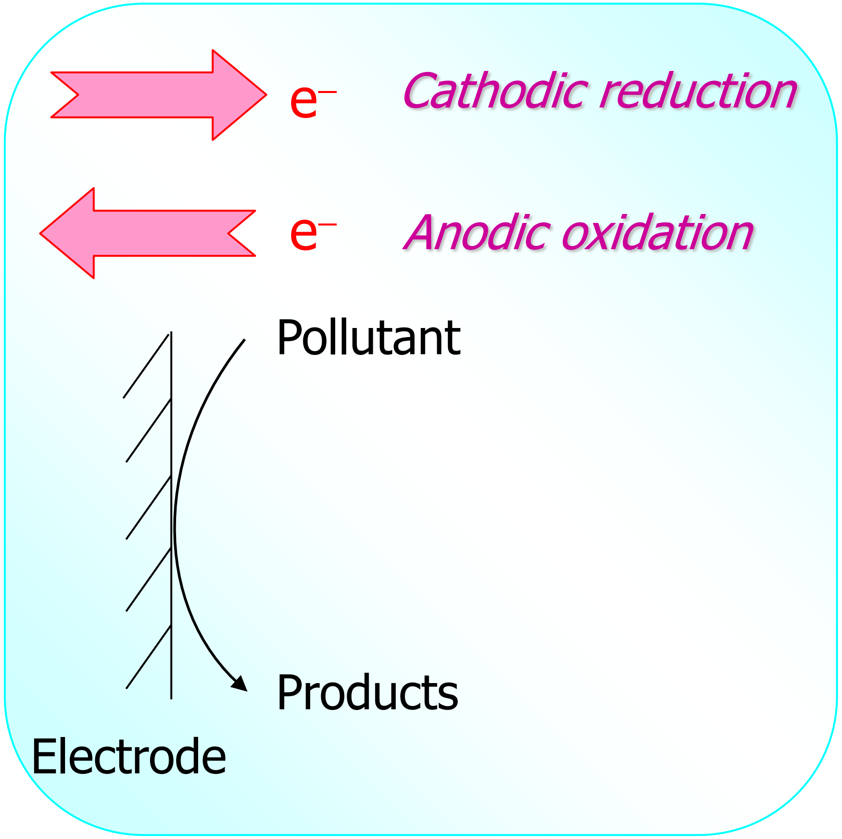
⇨ Oxidation or reduction occur by direct electron transfer reactions between target contaminants and the electrode surface without involving other mediators.

- **Indirect electrolysis**

⇨ Oxidation or reduction of target contaminants occur by reactions with electrochemically generated redox species (serving as chemical reagents or catalysts).

Direct Electrolysis

Direct electron transfer reaction



e⁻ *Cathodic reduction*

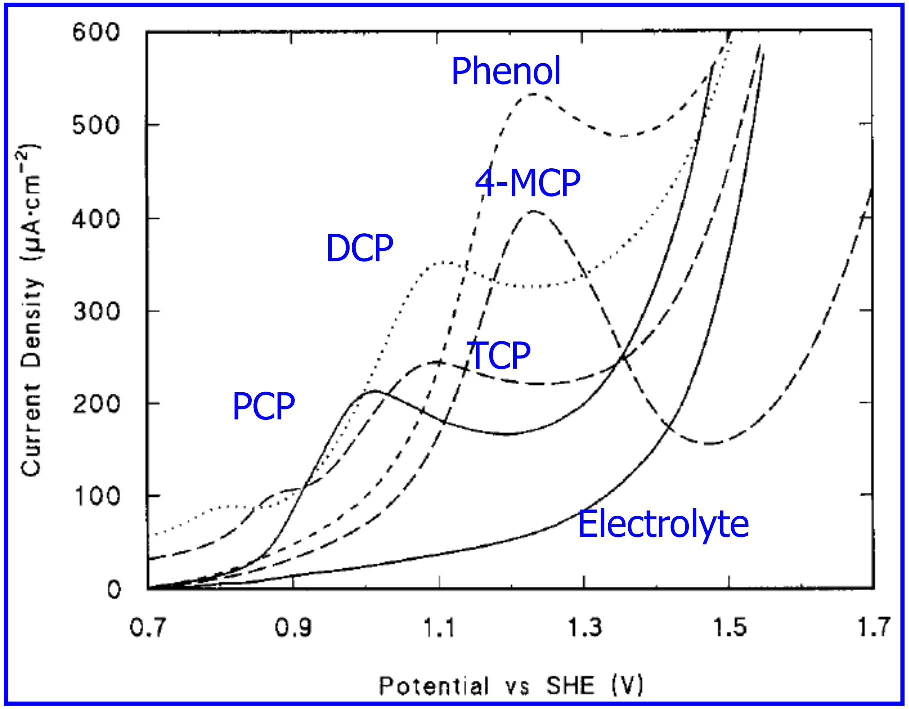
e⁻ *Anodic oxidation*

Pollutant

Products

Electrode

Anodic oxidation of phenol species



Linear sweep voltammograms for the oxidation of several chlorophenols (C_0 = 1 mM, pH 6)

Direct Electrolysis

Li and Hoffman, 1999 (*J. Phys. Chem. B*)

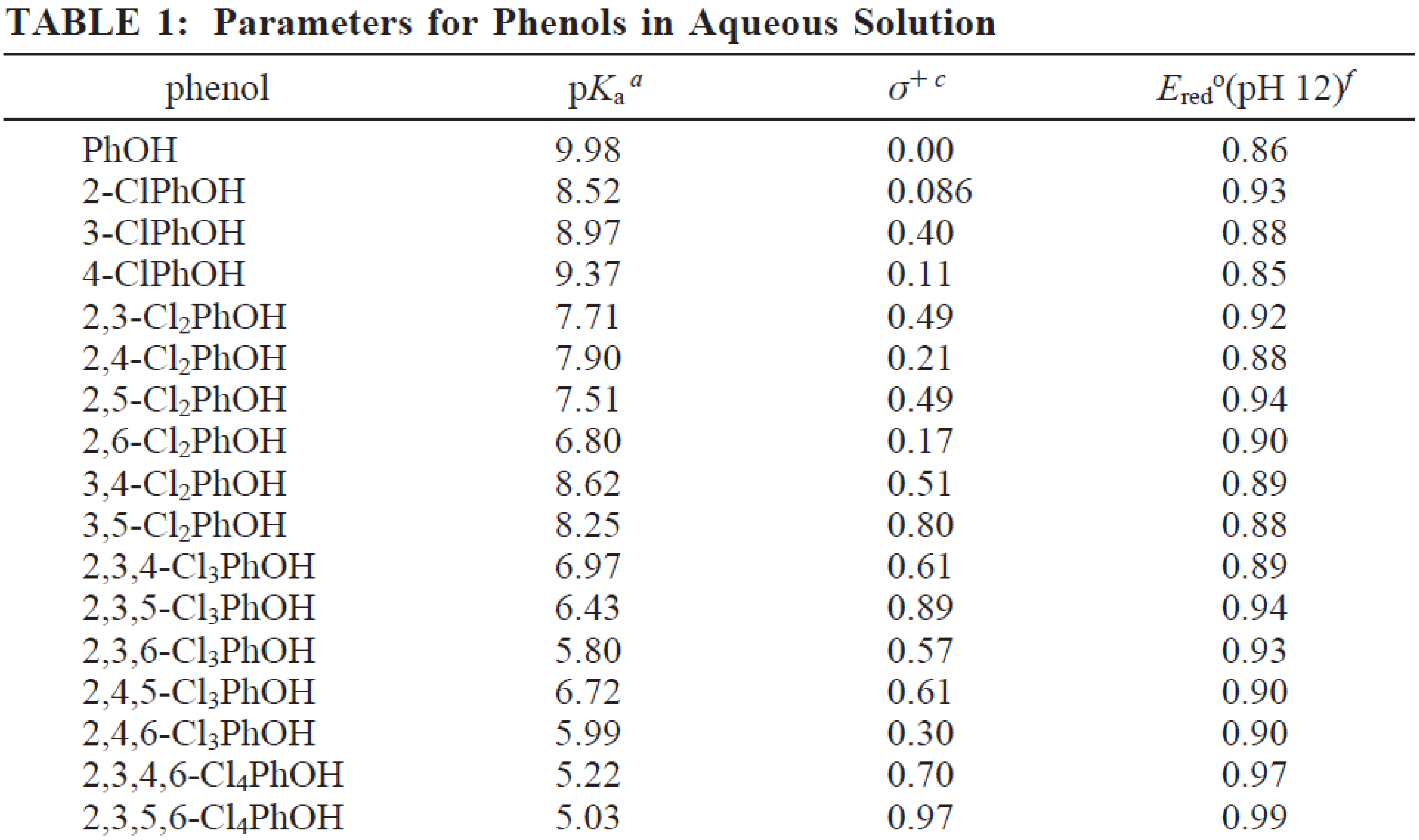


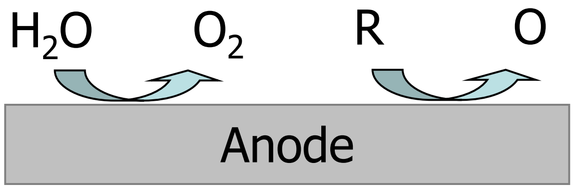
TABLE 1: Parameters for Phenols in Aqueous Solution

phenol	pK_a [a]	σ^+ [c]	E_{red}^o(pH 12) [f]
PhOH	9.98	0.00	0.86
2-ClPhOH	8.52	0.086	0.93
3-ClPhOH	8.97	0.40	0.88
4-ClPhOH	9.37	0.11	0.85
2,3-Cl_2PhOH	7.71	0.49	0.92
2,4-Cl_2PhOH	7.90	0.21	0.88
2,5-Cl_2PhOH	7.51	0.49	0.94
2,6-Cl_2PhOH	6.80	0.17	0.90
3,4-Cl_2PhOH	8.62	0.51	0.89
3,5-Cl_2PhOH	8.25	0.80	0.88
2,3,4-Cl_3PhOH	6.97	0.61	0.89
2,3,5-Cl_3PhOH	6.43	0.89	0.94
2,3,6-Cl_3PhOH	5.80	0.57	0.93
2,4,5-Cl_3PhOH	6.72	0.61	0.90
2,4,6-Cl_3PhOH	5.99	0.30	0.90
2,3,4,6-Cl_4PhOH	5.22	0.70	0.97
2,3,5,6-Cl_4PhOH	5.03	0.97	0.99

Direct Electrolysis

- **Side reactions**

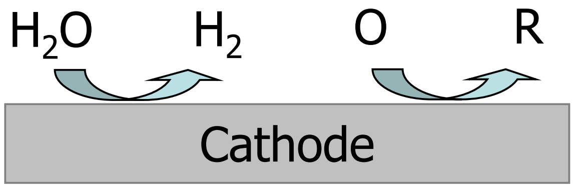
Anodic oxidation



R → O + ne^-

$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$

Cathodic reduction



O + ne^- → R

$2H_2O + 2e^- \rightarrow H_2 + 2OH^-$

Instantaneous Current Efficiency: ICE

In anodic oxidation, a major side reaction is the oxygen evolution.

By measuring the O_2 flow rate during the electrolysis in the presence ($V_{O2,\ org}$) and absence (V_{O2}) of the organic contaminant, the instantaneous current efficiency (ICE) at time t can be obtained as,

$$ICE = \frac{V_{O_2} - V_{O_2,org}}{V_{O_2}} = \frac{[(COD)_t - (COD)_{t+\Delta t}]}{32000 \times I\Delta t} 4FV_s$$

$V_{O2,org}$: Oxygen flow rate during electrolysis in the presence of organic species

V_{O2}: Oxygen flow rate during electrolysis in the absence of organic species

$(COD)_t$: Chemical oxygen demand at time t (mg/L)

V_s: Volume of electrolyte (L)

I: Current (A)

F: Faraday constant

Electrochemical Oxidability Index: EOI

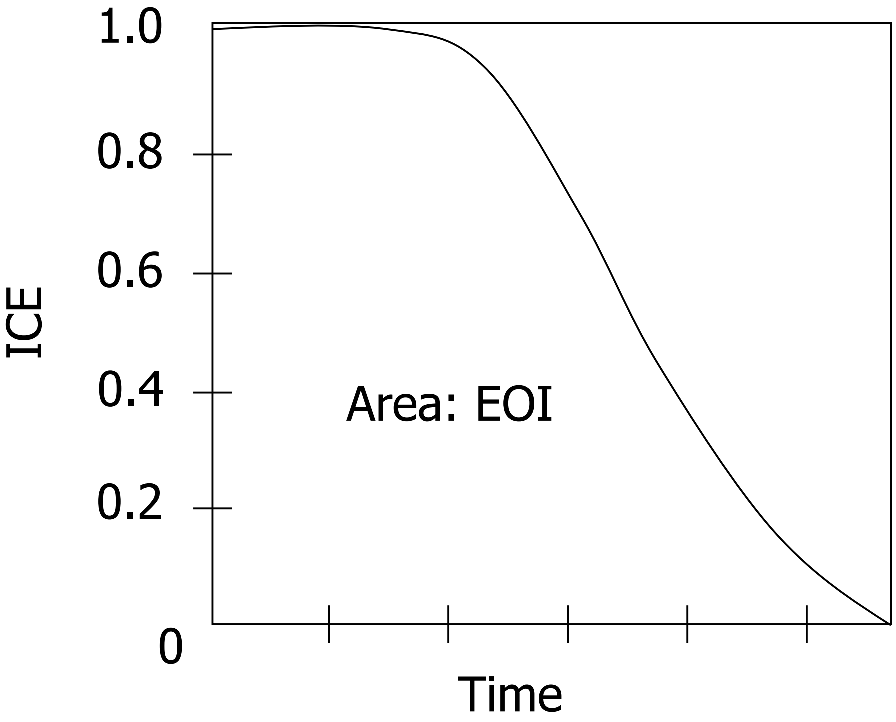
$$EOI = \frac{\int_0^{\tau} ICE\,dt}{\tau}$$

Average current efficiency

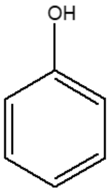
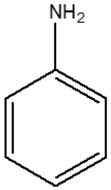
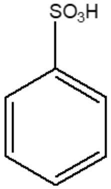
EOI ↑

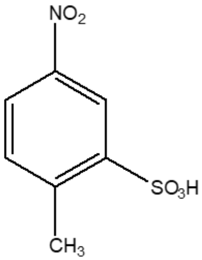
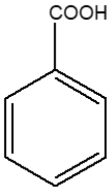
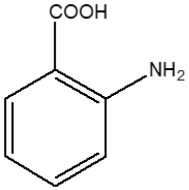
⇓

Oxidation affinity ↑



Electrochemical Oxidability Index: EOI

Substrate	EOI
	0.2
	0.56
	<0.05

Substrate	EOI
	0.1
	<0.05
	0.55

Electron-withdrawing groups (e.g., -SO$_3$H, -COOH) → **EOI** ⬇

Electron-donating groups (e.g., -NH$_2$) → **EOI** ⬆

Electrochemical Oxygen Demand: EOD

- **Electrochemical Oxygen Demand (EOD)**

The fraction of current used in oxidation of organic compounds can be expressed as the mount of evolved O_2.

In the absence of an organic contaminant

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$$

$$\text{mol } O_{2(\text{theor})} = \left(\frac{1 \text{ mol } O_2}{4 \text{ mol } e^-}\right)\left(\frac{1 \text{ mol } e^-}{F}\right) It = \frac{It}{4F}$$

In the presence of an organic contaminant

$$\text{Org} \rightarrow \text{Oxidation products} + ne^-$$

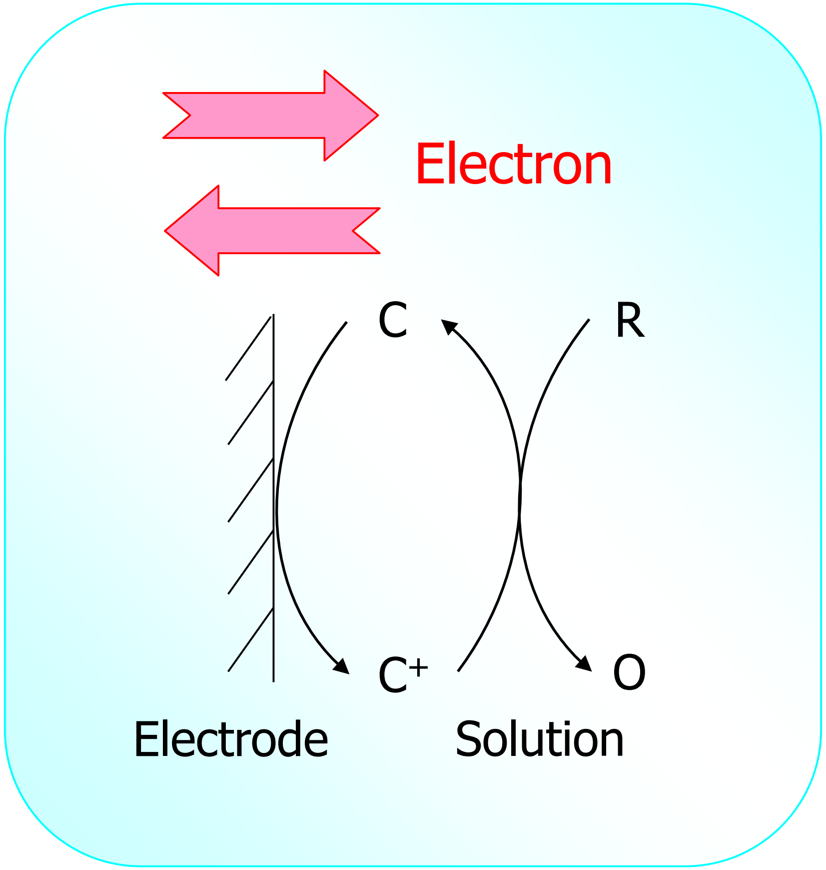
$$\text{EOD} = \text{mol } O_2 \text{ consumed for the contaminant oxidation}$$

$$= \text{mol } O_{2(\text{theor})} \bullet \text{EOI} = \frac{It}{4F} \bullet \text{EOI}$$

Indirect Electrolysis

Indirect electron transfer reactions

Reversible process



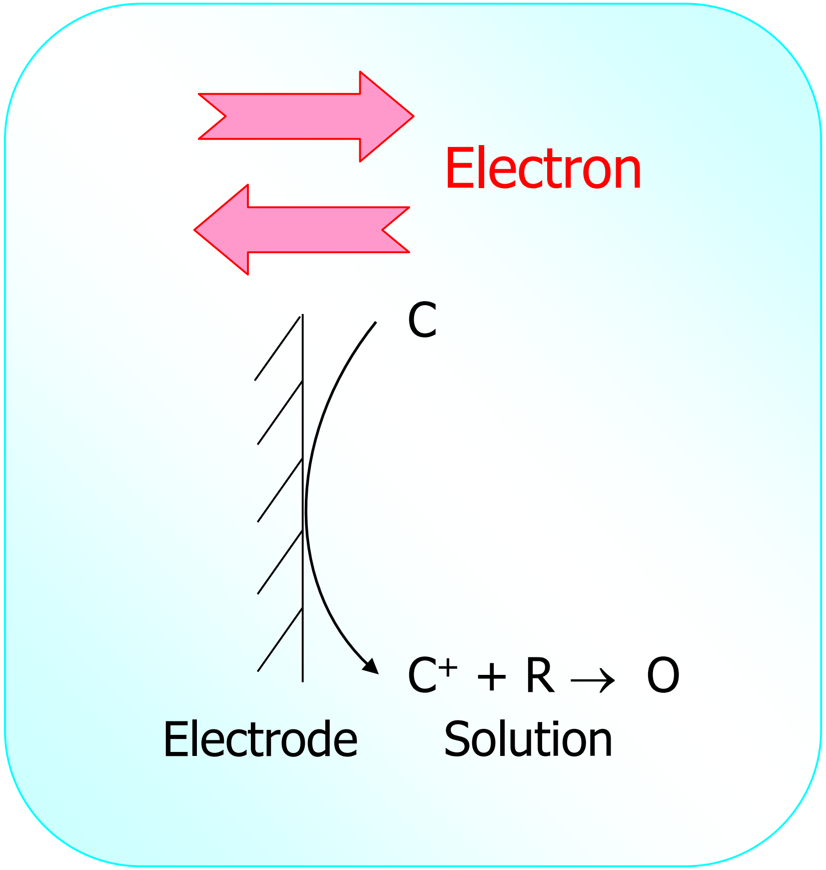
Electron

C R

C^+ O

Electrode Solution

Irreversible process



Electron

C

$C^+ + R \rightarrow O$

Electrode Solution

Indirect Electrolysis

- Electron mediator for reversible process

Mediator Couple	Standard Reduction Potential V versus NHE
Ag(I/II)	1.98
Co(II/III)	1.82
Ce(IV/III)	1.44
Fe(II/III)	0.77

Reversible Process

Ag(I)/Ag(II) couple

$Ag^+ \rightarrow \mathbf{Ag^{2+}} + e^-$

$\mathbf{Ag^{2+}} + R \rightarrow O + Ag^+$

(Example)

$Ag^+ \rightarrow Ag^{2+} + e^-$

$Ag^{2+} + NO_3^- \rightarrow AgNO_3^+$

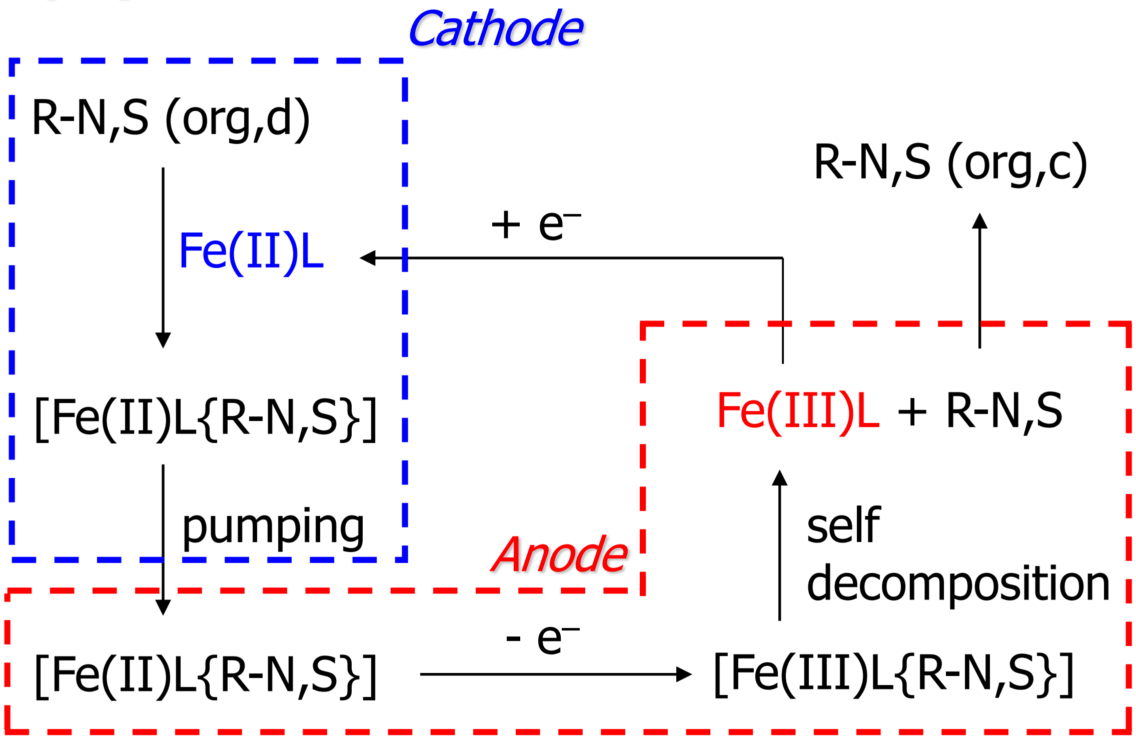
$a\ AgNO_3^+ + \text{Organics} + b\ H_2O \rightarrow a\ Ag^+ + n\ CO_2 + a\ HNO_3$

Reversible Process

Fe(II)/Fe(III) couple

$Fe^{3+} + e^- \rightarrow Fe^{2+}$ $E^0 = 0.77$ V vs NHE

(Example)



Cathode

R-N,S (org,d)

Fe(II)L

[Fe(II)L{R-N,S}]

pumping

$+ e^-$

R-N,S (org,c)

Fe(III)L + R-N,S

self decomposition

Anode

[Fe(II)L{R-N,S}]

$- e^-$

[Fe(III)L{R-N,S}]

Irreversible Process

- **Various oxidants generated by water electrolysis**

Reagents		**Standard Redox Potential (V vs NHE)**
$\bullet OH$	**/ H_2O**	2.80
O	**/ H_2O**	2.42
O_3	**/ O_2**	2.08
H_2O_2	**/ H_2O**	1.78
$HO_2\bullet$	**/ H_2O_2**	1.70
Cl_2	**/ Cl^-**	1.39

Irreversible Process

•OH production

$H_2O \rightarrow \mathbf{\bullet OH_{(ads)}} + H^+ + e^-$

$\mathbf{\bullet OH_{(ads)}} \rightarrow 1/2O_2 + H^+ + e^-$

(Overall reaction)

$H_2O \rightarrow 1/2O_2 + 2H^+ + 2e^-$

Chlorine evolution

$2Cl^- \rightarrow \mathbf{Cl_2} + 2e^-$

$\mathbf{Cl_2} + H_2O \rightarrow HCl + HOCl$

($K = 4\times10^{-4}$ at 25 °C)

$HOCl \Leftrightarrow H^+ + OCl^-$

($pK_a = 7.54$ at 25 °C)

Irreversible Process

Ozone evolution

$H_2O \rightarrow \bullet OH_{(ads)} + H^+ + e^-$

$\bullet OH_{(ads)} \rightarrow \bullet O_{(ads)} + H^+ + e^-$

$2\bullet O_{(ads)} \rightarrow (O_2)_{(ads)} \rightarrow O_2$

$\bullet O_{(ads)} + (O_2)_{(ads)} \rightarrow O_3$

(Overall reaction)

$3H_2O \Leftrightarrow \mathbf{O_3} + 6H^+ + 6e^-$

(Requirement for O_3 generation)

1) Anodes with high O_2 overpotential (poor oxygen evolution kinetics)

2) No significant competitive redox reactions of electrolytes (anions and cations)

3) Anodes present in their highest oxidation state or kinetically resistant to further oxidation.

Selection of Electrode Material

- **Consideration points for choosing the electrode material**

  - Composition and nature of the electrolytes in solution
  - Stability of the electrode material
  - Effects of cathodic reactions on the overall efficiency
  - Available range of potential and pH
  - Cost
  - Selectivity
  - Environmental compatibility

Selection of Electrode Material

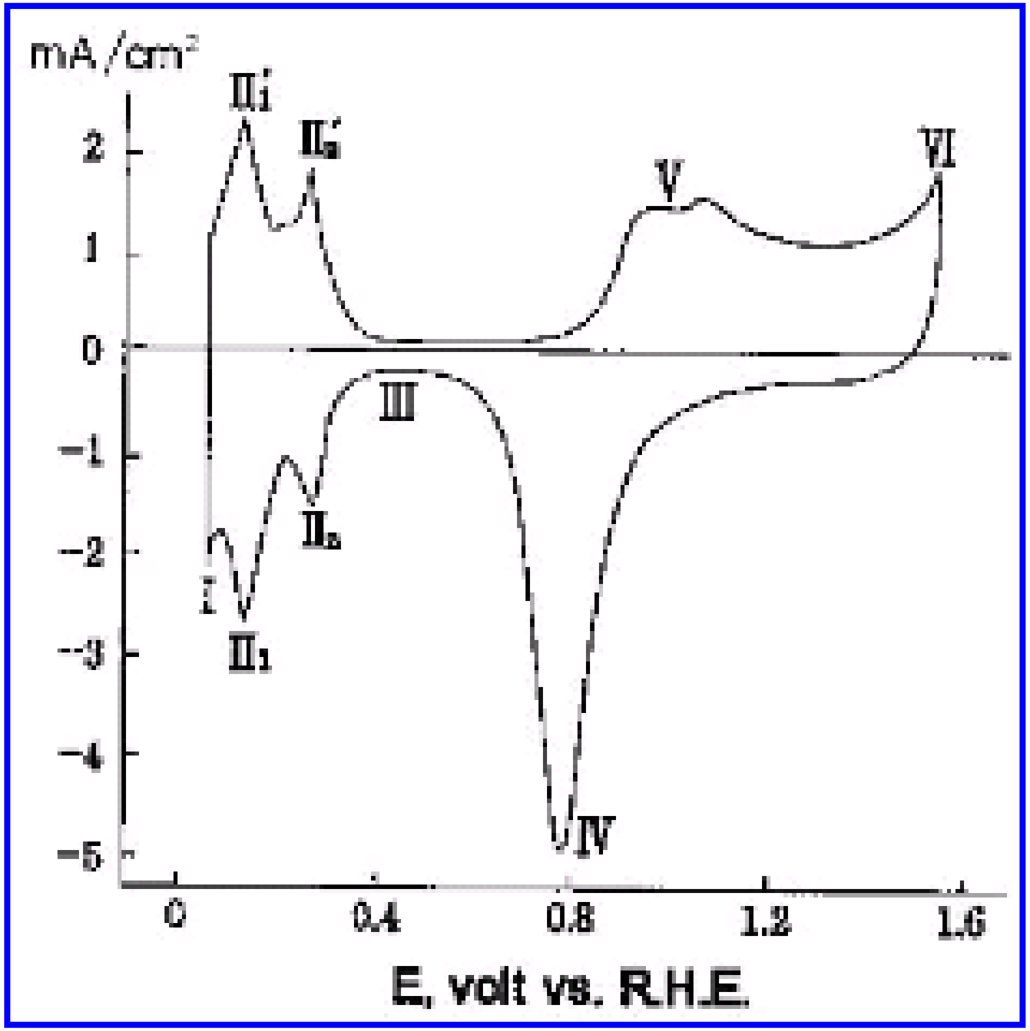
Noble metal

- Platinum (Pt), Gold (Au)
- Fast electron transfer kinetics
- Wide positive potential window
- High background current
- Very expensive

Metal

- Stainless steel, nickel
- Available in only a limited range of potential and pH

Selection of Electrode Material



Cyclic voltammogram of Pt in 1N H_2SO_4

I: H_2 gas evolution

II_1, II_2: $H^+ + e^- \rightarrow H_{ad}$

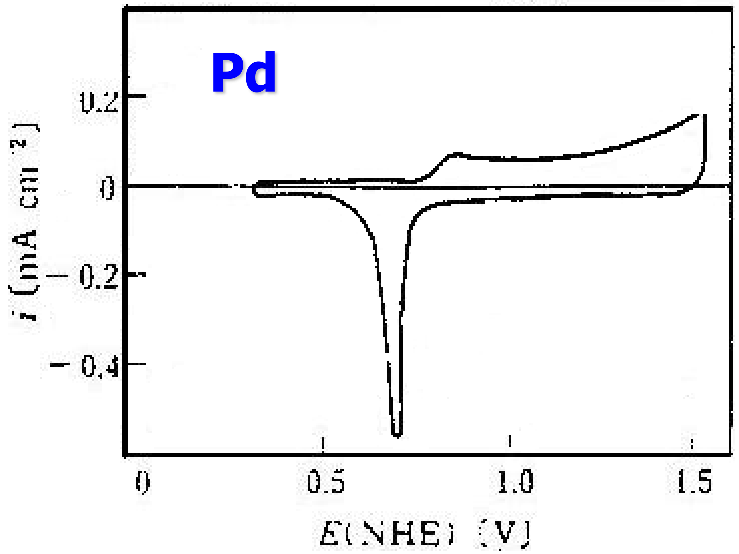
III: Charged current in double layer

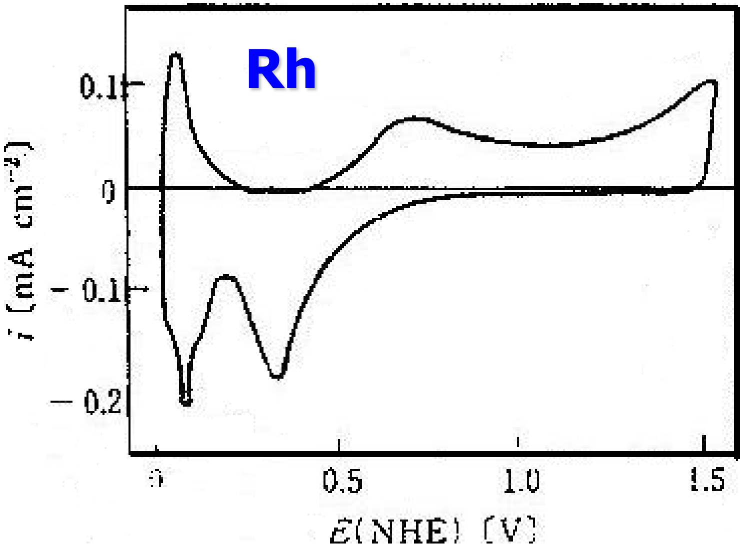
IV: Oxide reduction

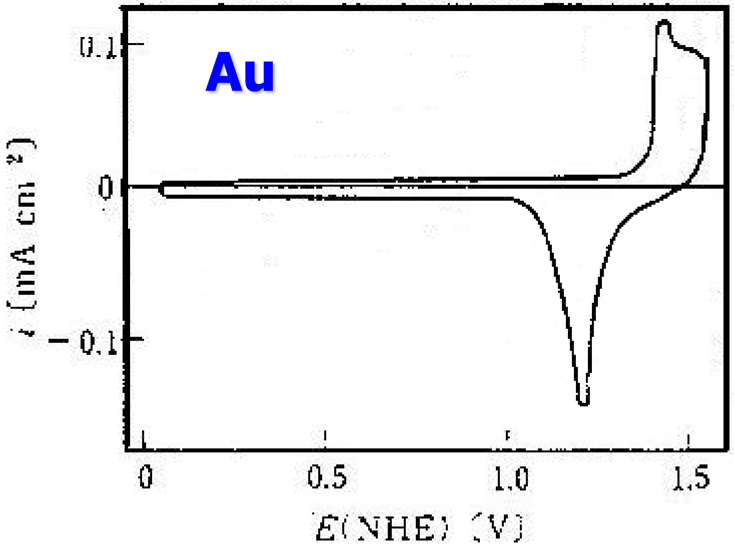
V: Oxide formation

II_1', II_2' : $H_{ad} \rightarrow H^+ + e^-$

Selection of Electrode Material







Selection of Electrode Material

Metal oxide

- Pt, Ir, Ru, Pb oxide
- High resistance to severe conditions
- High O_2 evolution overpotential
- Very suitable for degrading organic contaminants by anodic oxidation

Carbon based electrode

- Graphite, Carbon felt, Glassy carbon, Reticulated vitreous carbon (RVC)
- High surface area
- High chemical inertness
- Commercial availability at a reasonable cost

Reticulated Vitreous Carbon (RVC)

Selection of Electrode Material

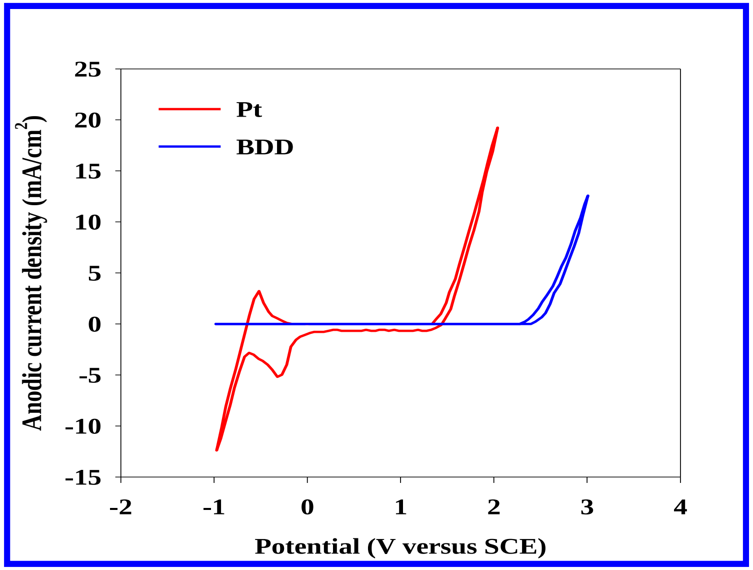
Boron-doped diamond electrode

- High overpotential for both H_2 and O_2 evolution

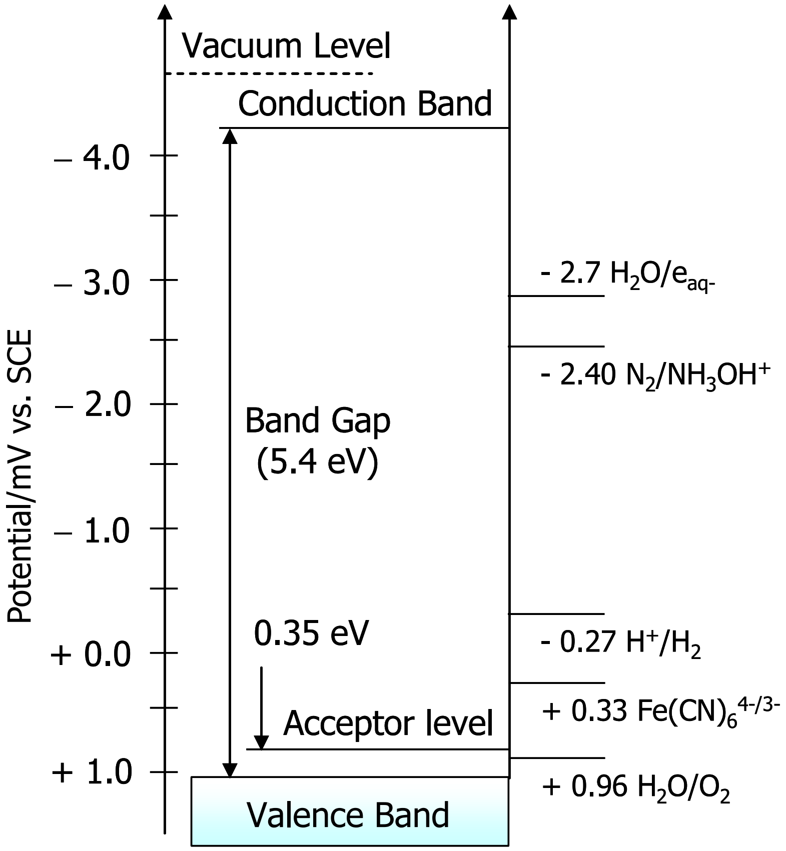
  (a wide potential window)

- Low background current

- High stability under strong oxidizing conditions



Cyclic voltagram for BDD and Pt
(0.2 M KH_2PO_4, ν = 50 mV/sec)



Band gap of boron-doped diamond electrode

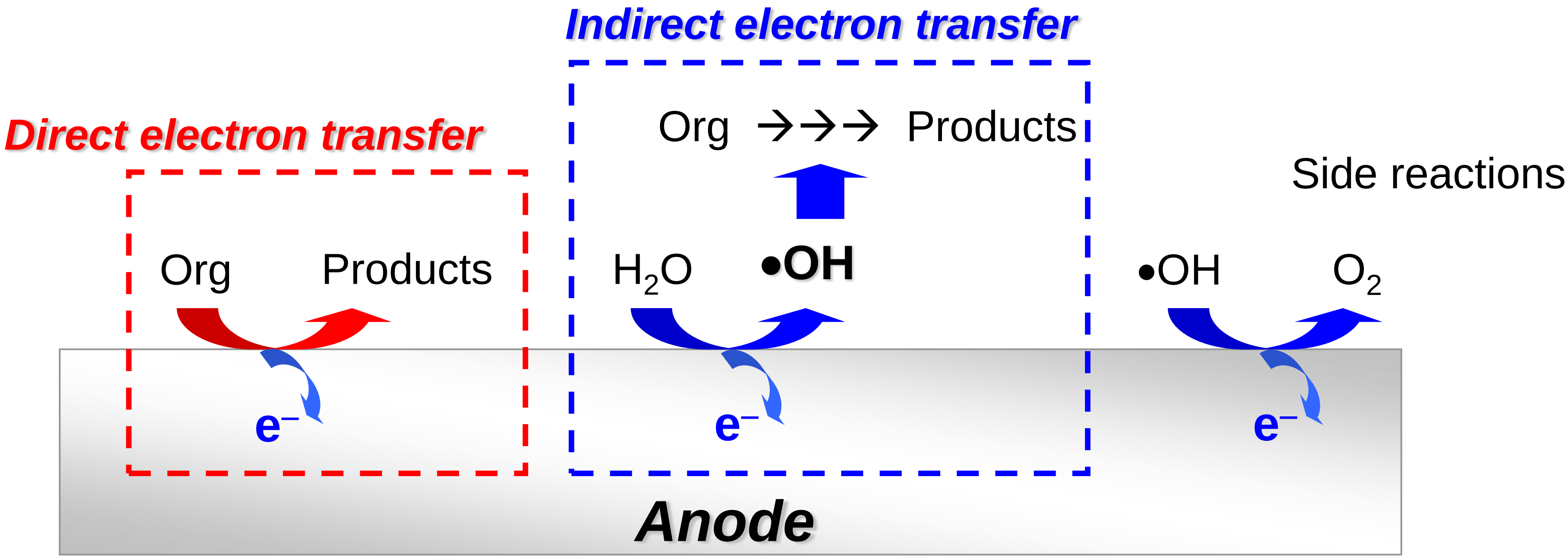
Electrochemical Water Treatment Processes

- Anodic oxidation process
- Cathodic reduction process
- Electrocoagulation process
- Electrochemical generation of Fenton’s reagents

Anodic Oxidation Process

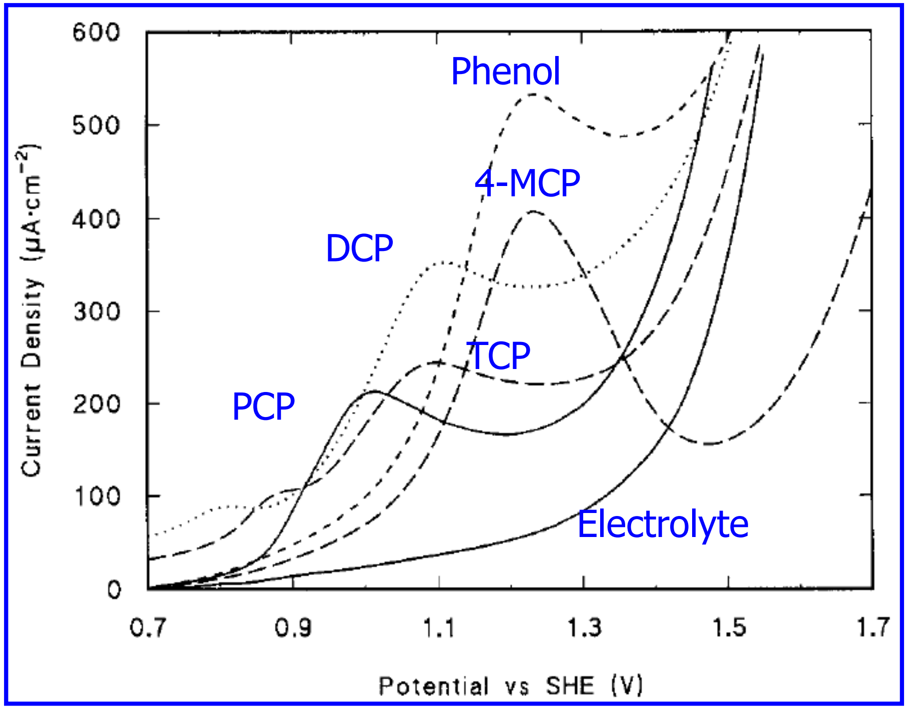
- Electrochemical oxidation of contaminants by direct and indirect reactions occurring at the anodic part

Dual reaction pathways in anodic oxidation process

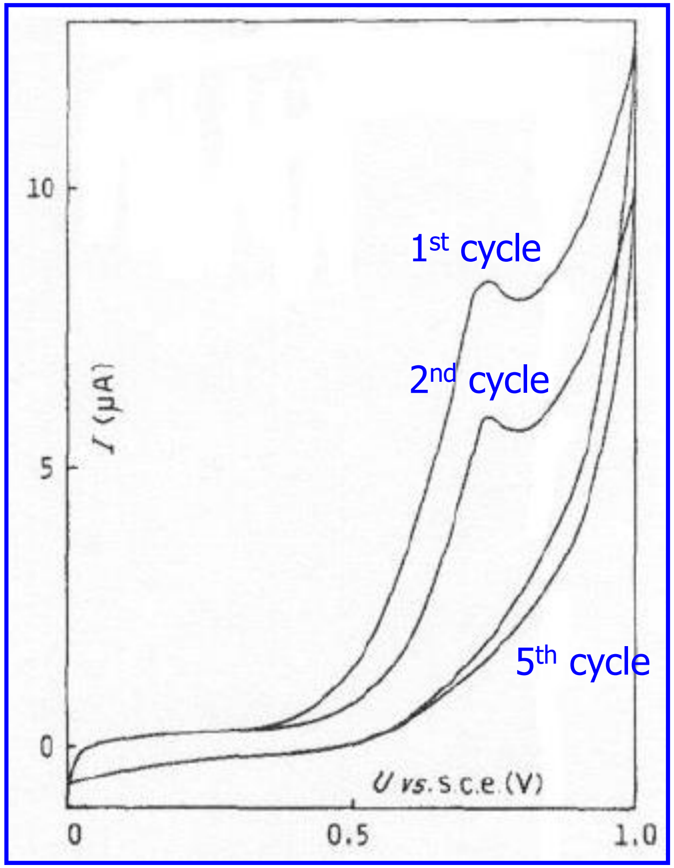


Anodic Oxidation Process

Examples of direct anodic oxidation of water contaminants



Linear sweep voltammograms for the oxidation of several chlorophenols (C_0 = 1 mM, pH 6)

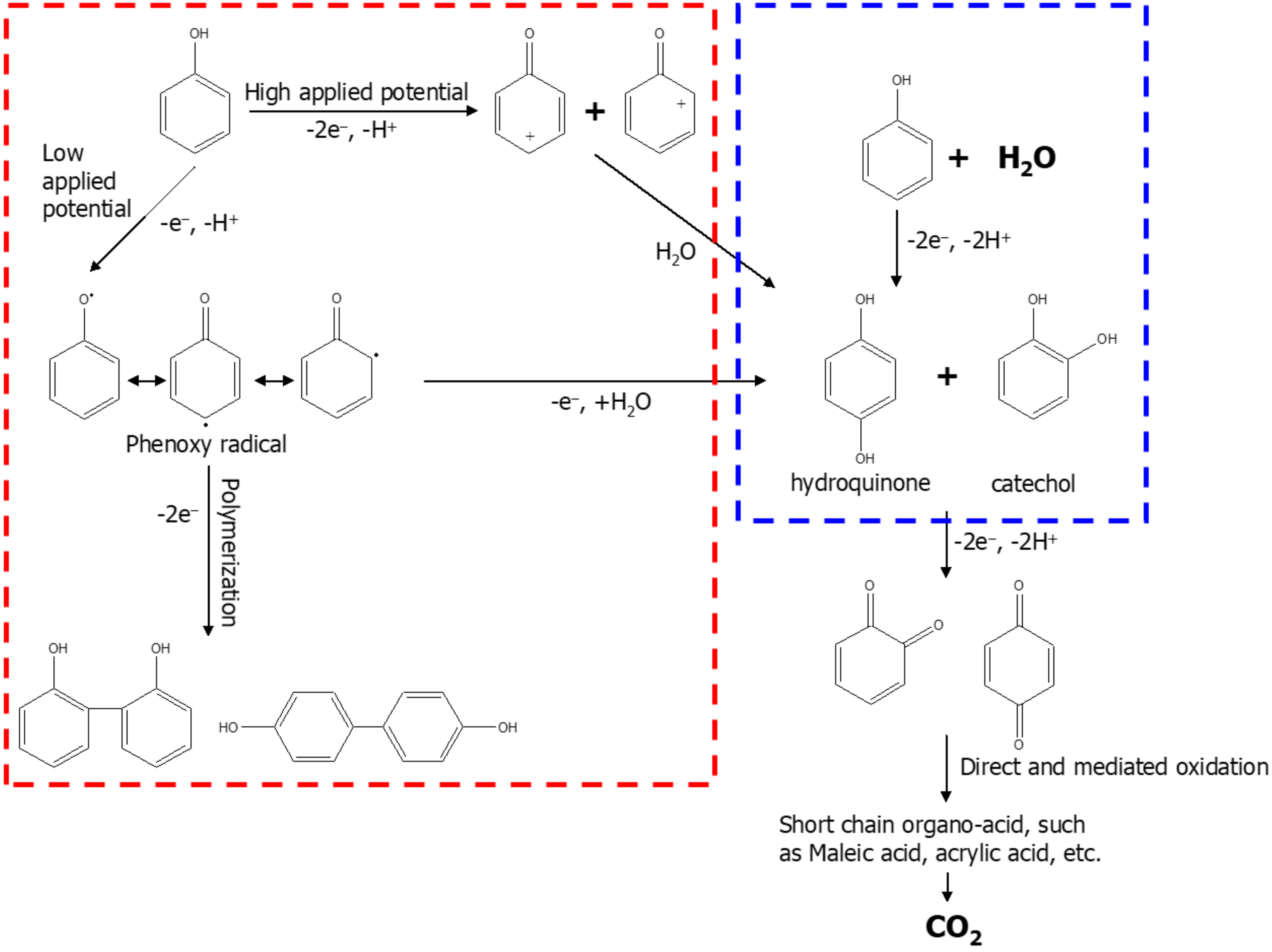


Cyclic voltammograms for whole cells of *S. cerevisiae* (C_0 = 1.28×10^8 CFU/ml, 0.1 M phosphate buffer (pH 7)

Dual Pathways for Anodic Oxidation of Phenol

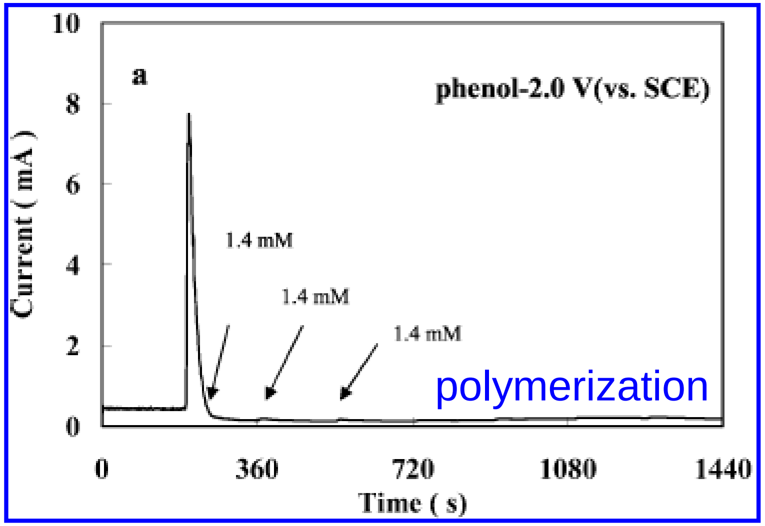
Direct oxidation pathway

•OH mediated oxidation pathway



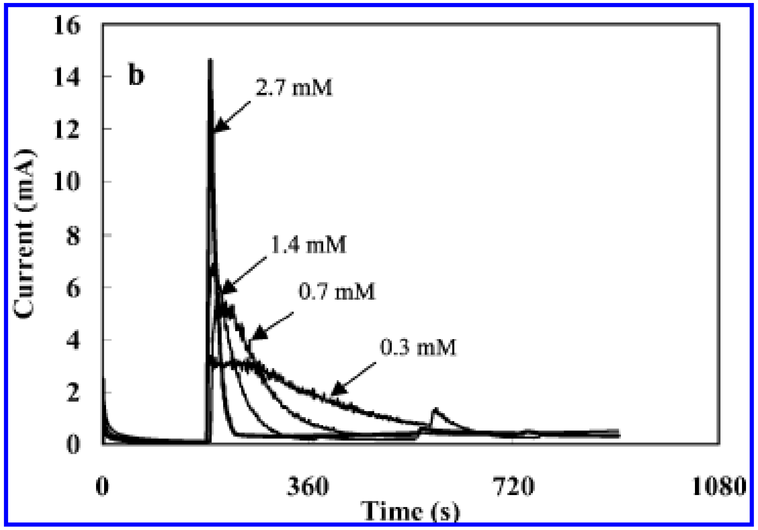
Dual Pathways for Anodic Oxidation of Phenol

Direct oxidation pathway



polymerization

Chronoamperometric response to step-by-step injections of phenol



Current decay curves at various injection concentrations of phenol

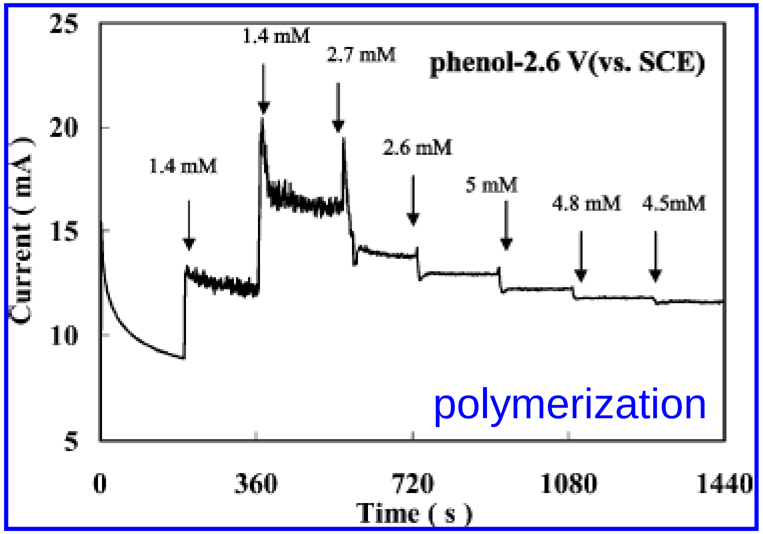
At 2.0 V vs. SCE

Phenol → phenoxy radical + e^- →→ Polymerization **(O)** → **Current decay**

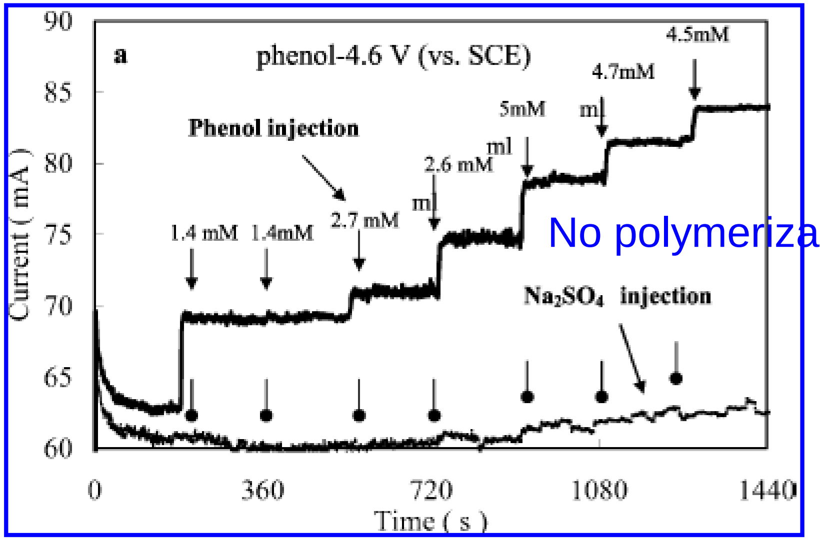
H_2O → •OH + H^+ + e^- **(X)**

Dual Pathways for Anodic Oxidation of Phenol

Direct and indirect oxidation pathways



polymerization



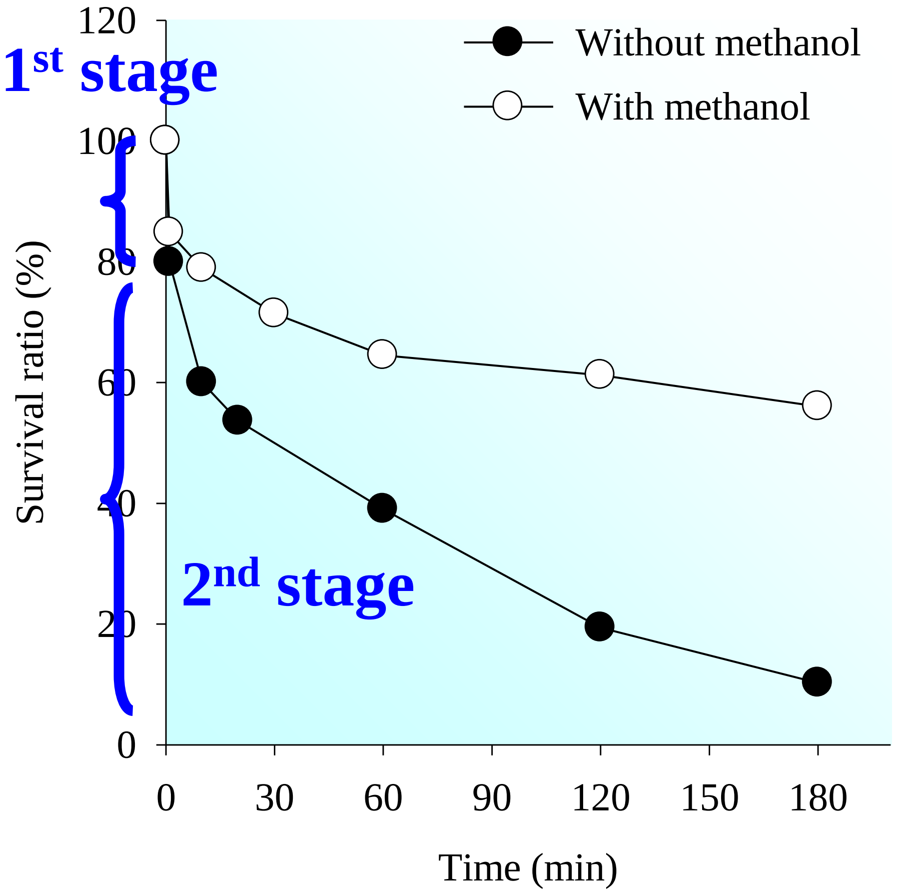
No polymerization

At above 2.6 V vs. SCE

Phenol → hydroquinone / catechol $+ 2e^-$ **(O)** ➡ **Steady state current**

$H_2O \rightarrow \bullet OH + H^+ + e^-$ **(O)** ➡ **Removal of electrode fouling**

Anodic Oxidation for Disinfection



[Methanol]$_0$ = 0 or 30 mM, [*E. coli*]$_0$ = 1×10^3 CFU/ml
j = 100 mA/cm^2

1st stage

E. coli →→→ inactivation

CoA (coenzyme A) →
dimeric CoA + e^-

2nd stage

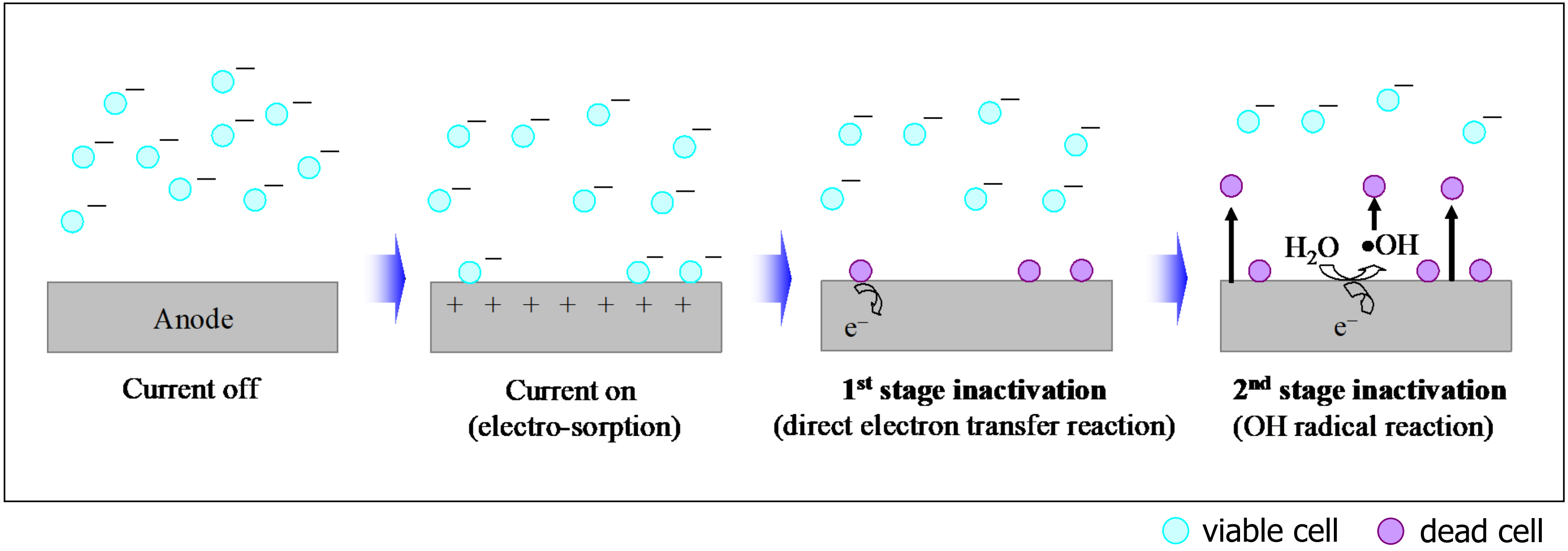
H_2O → •OH + H^+ + e^-

E. coli + •OH →→→ inactivation

MeOH + •OH → scavenging

Anodic Oxidation for Disinfection

Two stages inactivation of *E. coli* in anodic oxidation process



Cathodic Reduction Process

Chlorinated organics

Effective dechlorination of compounds that have a low reactivity to •OH

$R\text{-}Cl + 2H^+ + 2e^- \rightarrow R\text{-}H + HCl$ (E = -1 ~ -3 V vs. SCE)

$C_{12}H_{10\text{-}n}Cl_n$ (PCBs) $+ 2ne^- + nH^+ \rightarrow C_{12}H_{10} + nCl^-$

Oxynitrogen ions

$2NO_3^- + 2H_2O + 4e^- \rightarrow 2NO_2^- + 4OH^-$

$2NO_2^- + 4H_2O + 6e^- \rightarrow N_2 + 8OH^-$

Cathodic Reduction Process

Metal removal

Conventional process for metal removal

- Precipitation as hydroxides, sulfides, or oxalates
- Filtration
- Chemical or electrochemical ion exchange
- Reverse osmosis
- Chemical or physical adsorption
- Stabilization or solidification
- Chemical reduction
- Biological remediation
- **Electrochemical deposition**

Cathodic Reduction Process

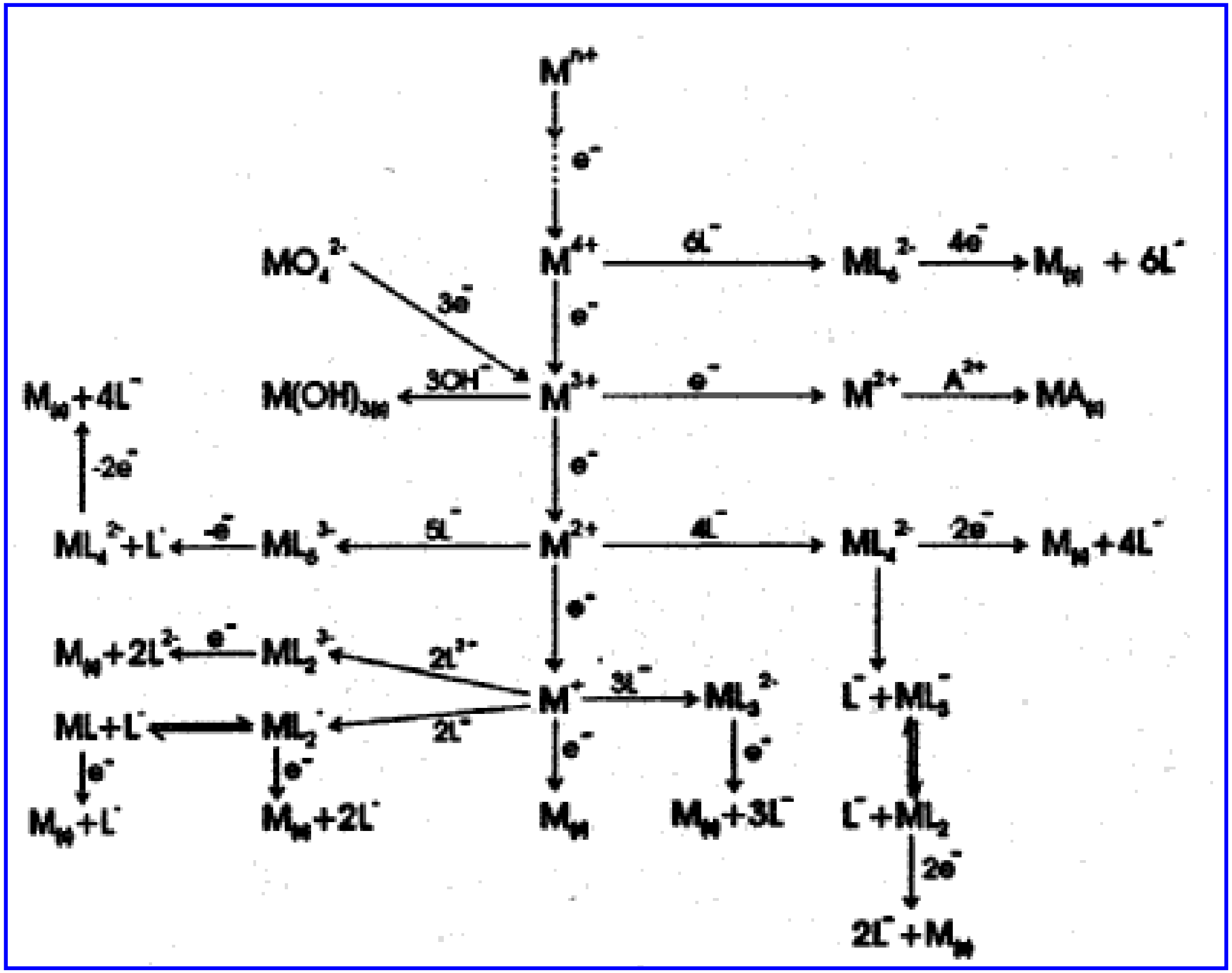
Advantages of the electrochemical method for metal removal

- The metal is produced in its most valuable form
- No extra reagents are added. (treated water can be recycled)
- Control of pH can be electrochemically achieved.
- Sludge production is minimized
- Selective deposition of one metal in mixture may be possible.
- The operating cost is low
- The corresponding anodic reaction can be additionally used.

  (e.g. other organics degradation (ligands), high-purity O_2 production)
- Simple and compact

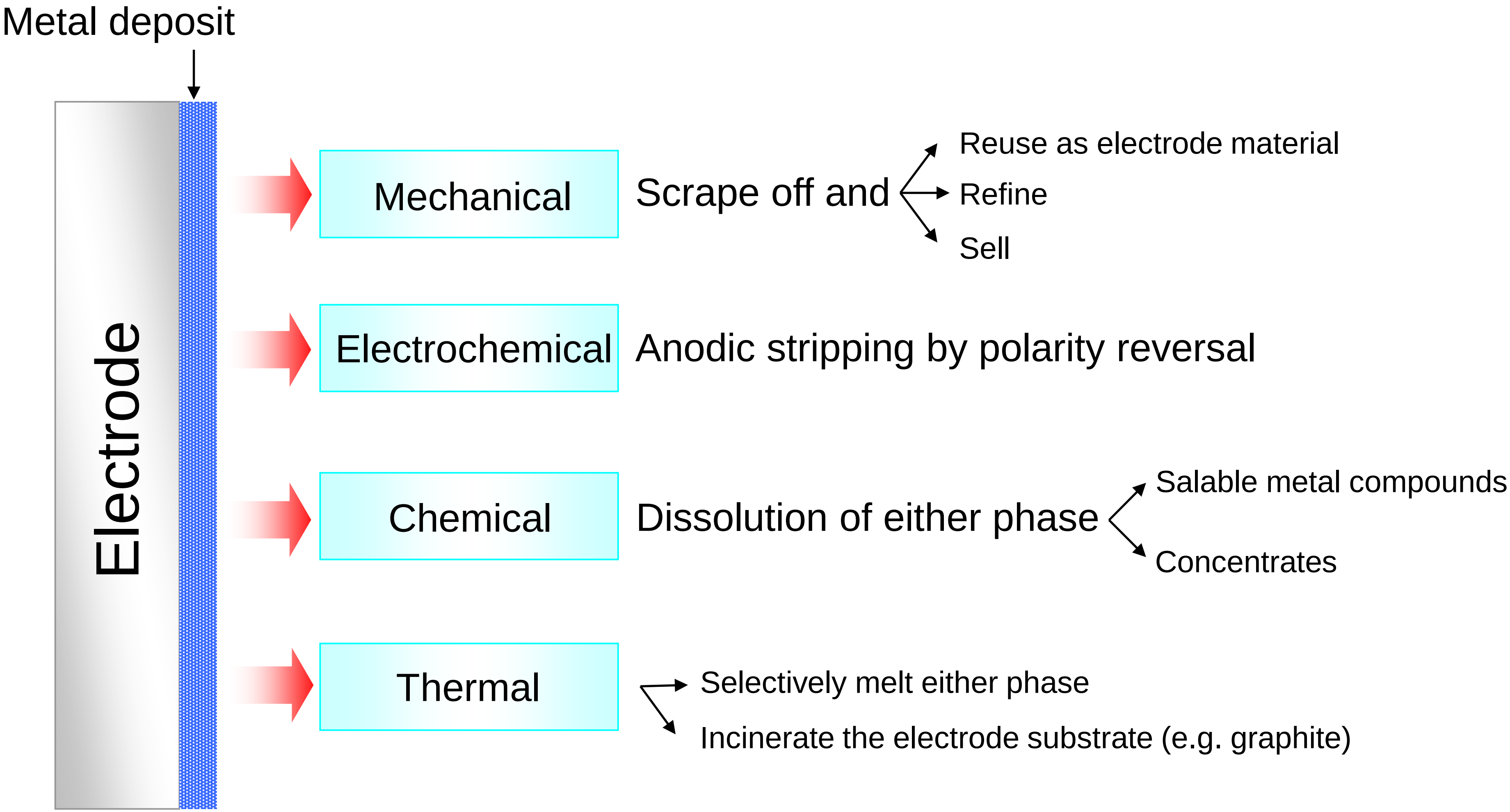
Cathodic Reduction Process

Pathways for the electrochemical metal reduction



Cathodic Reduction Process

Recovery methods of metal deposit



Metal deposit

Electrode

Mechanical: Scrape off and
- Reuse as electrode material
- Refine
- Sell

Electrochemical: Anodic stripping by polarity reversal

Chemical: Dissolution of either phase
- Salable metal compounds
- Concentrates

Thermal
- Selectively melt either phase
- Incinerate the electrode substrate (e.g. graphite)

Electrocoagulation Process

Mechanisms

- Case **I**

Anode: $4\,Fe(s) \rightarrow 4\,Fe^{2+}(aq) + 8\,e^{-}$

Chemical: $4\,Fe^{2+}(aq) + 10\,H_2O(l) + O_2(g) \rightarrow 4\,Fe(OH)_3(s) + 8\,H^{+}(aq)$

Cathode: $8\,H^{+}(aq) + 8\,e^{-} \rightarrow 4\,H_2(g)$

Overall: $4\,Fe(s) + 10\,H_2O(l) + O_2(g) \rightarrow 4\,Fe(OH)_3(s) + 4\,H_2(g)$

- Case **II**

Anode: $Fe(s) \rightarrow Fe^{2+}(aq) + 2\,e^{-}$

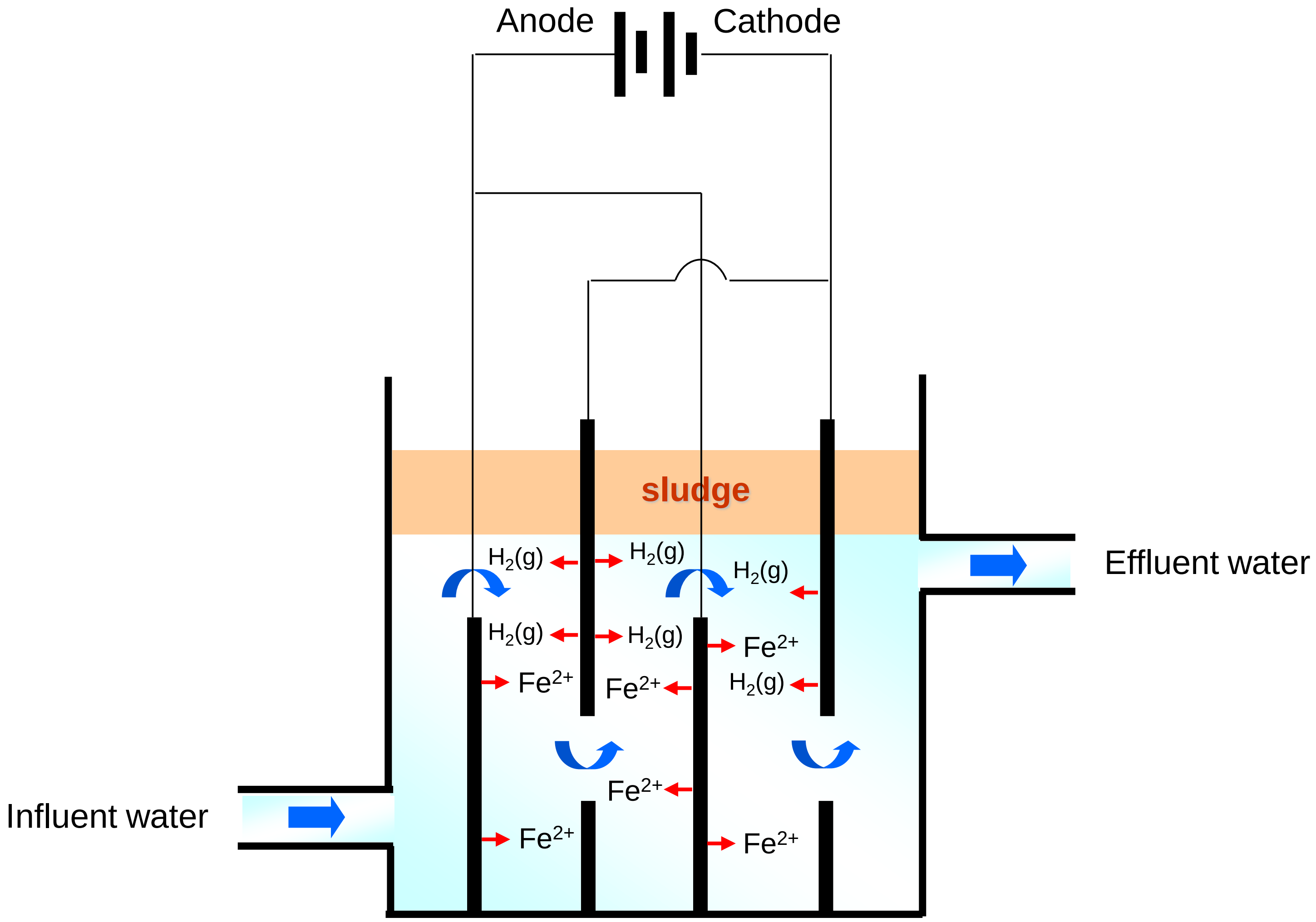
Chemical: $Fe^{2+}(aq) + 2\,OH^{-}(aq) \rightarrow Fe(OH)_2(s)$

Cathode: $2\,H_2O(l) + 2\,e^{-} \rightarrow H_2(g) + 2\,OH^{-}(aq)$

Overall: $Fe(s) + 2\,H_2O(l) \rightarrow Fe(OH)_2(s) + H_2(g)$

Electrocoagulation Process

Schematic diagram of the electrocoagulation process



Electrochemical Generation of Fenton Reagents

Electro-Fenton®

- In-situ electrogeneration of Fenton's reagent (Fe^{2+}, H_2O_2)

- Cathodic part

$O_2 + 2H^+ + 2e^- \rightarrow H_2O_2$ ($E^0 = 0.68$ V)

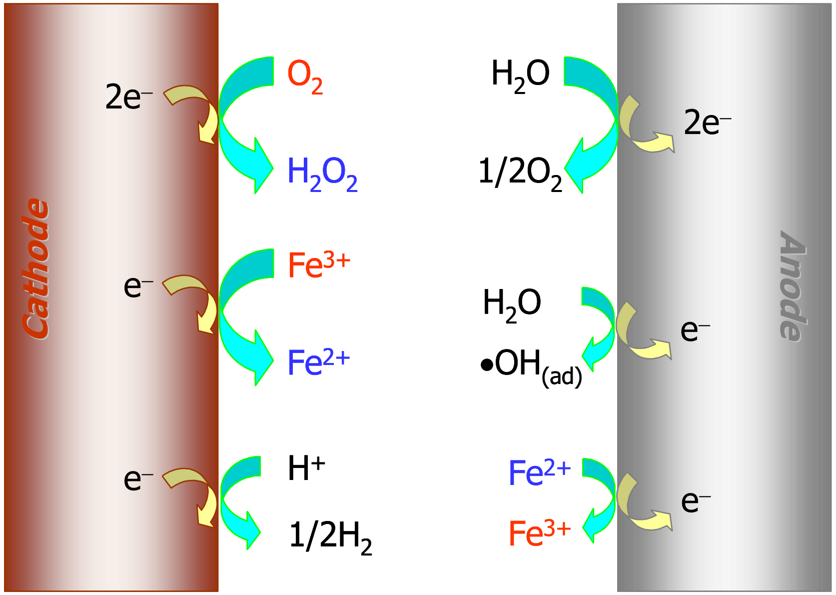
$Fe^{3+} + e^- \rightarrow Fe^{2+}$ ($E^0 = 0.77$ V)

- Anodic part

$2H_2O \Leftrightarrow O_2 + 4H^+ + 4e^-$ ($E^0 = 1.23$ V)

- Overall

$\frac{1}{2}O_2 + H_2O + e^- \rightarrow 2OH\bullet$



Schematic diagram for electro-fenton

Electrochemical Generation of Fenton Reagents

Anodic Fenton

- Cathodic part

$2H_2O + 2e^- \Leftrightarrow H_2 + 2OH^-$ ($E^0 = -0.8277$ V)

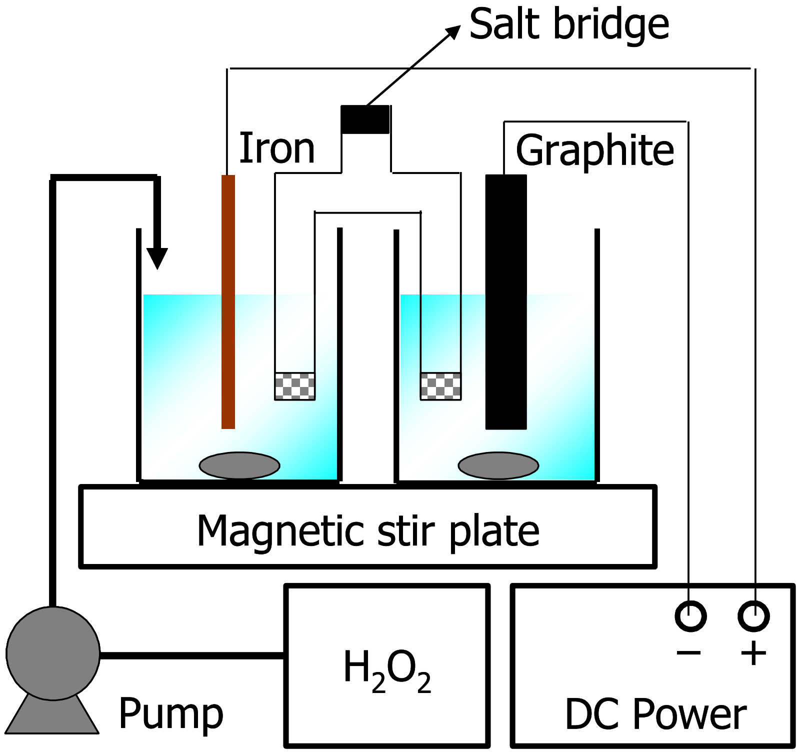
- Anodic part

Fe (sacrificial anode) $\rightarrow Fe^{2+} + 2e^-$

($E^0 = 0.447$ V)

- Overall

$Fe + 2H_2O \rightarrow Fe(OH)_2 + H_2$



Electrochemical cell for the anodic Fenton system

Electrochemical Generation of Fenton Reagents

Full-scale Electro-Fenton process in Taiwan

Outer view of Electro-Fenton process

An electrolytic tank with 100 anodes and cathodes

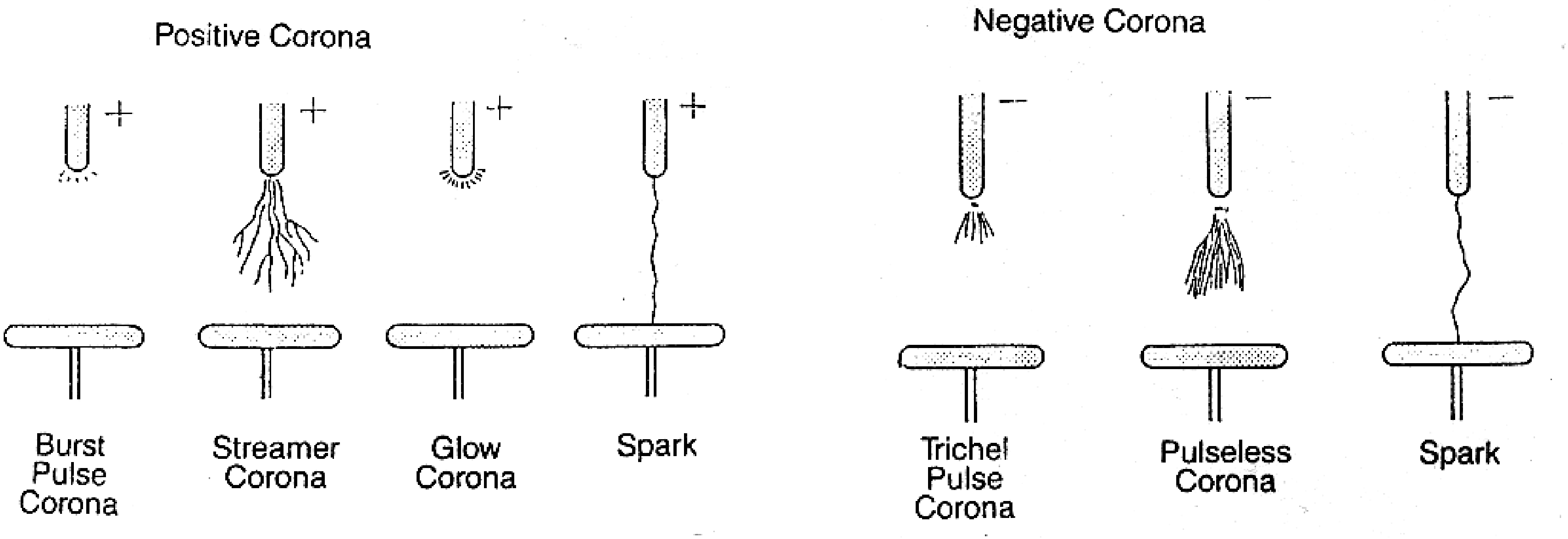
Electrochemical Generation of Fenton Reagents

Full-scale Electro-Fenton process in Taiwan

Type of wastewater	Influent COD, mg/l	Effluent COD, mg/l	COD removal efficiency (%)
Oil/ink waste liquor in a chemical plant	74,600	2,390	97
Hexamine wastewater in a chemical plant	29,600	40	>99
Electroless nickel wastewater in a electro-plating plant	27,900	1,940	93
Black liquor in a pulp/paper plant	30,900	350	99
Acrylonitrile wastewater in a latex plant	5,800	560	90
Resin processing wastewater in a chemical plant	2,500	350	86
Catalyst regenerate wastewater in a manmade fiber plant	24,900	620	97
Waste liquor of laboratory in a college	23,900	4,780	80

High-Voltage
Electric Discharge Processes

Types of Electrical (Corona) Discharge

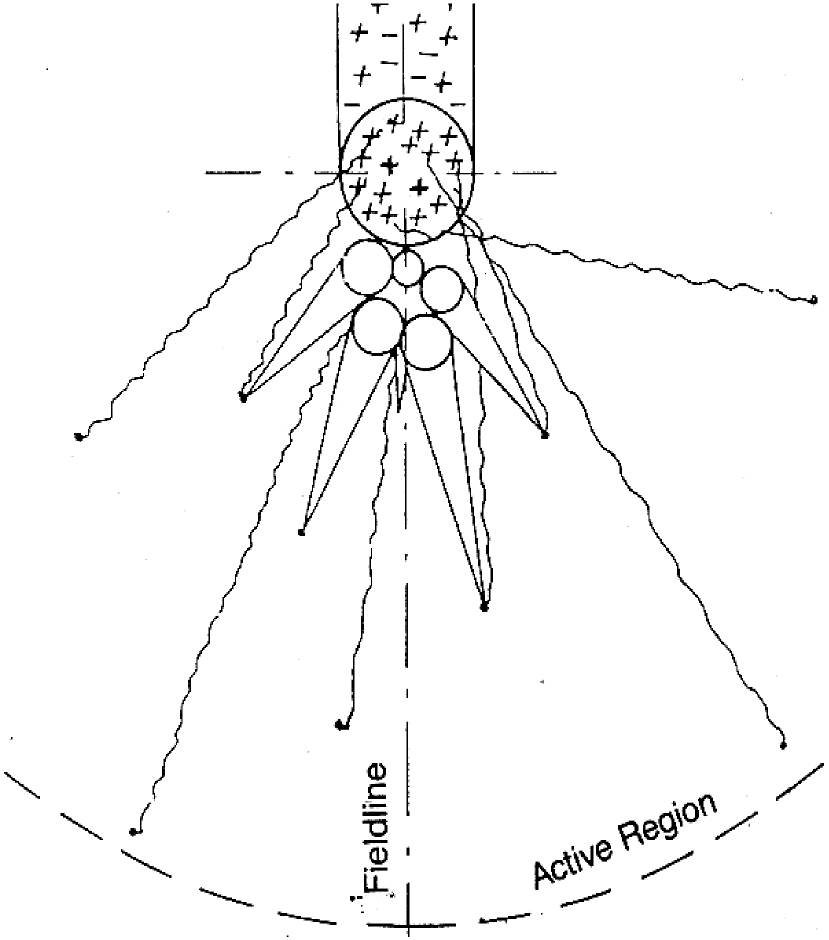


from Chang et al., 1991

Type of corona discharge depend on

- **Polarity of the field**
- **Electrode configuration**
- **Applied voltage**

Streamer Corona Discharge



Streamer corona ⇒

Large active volume

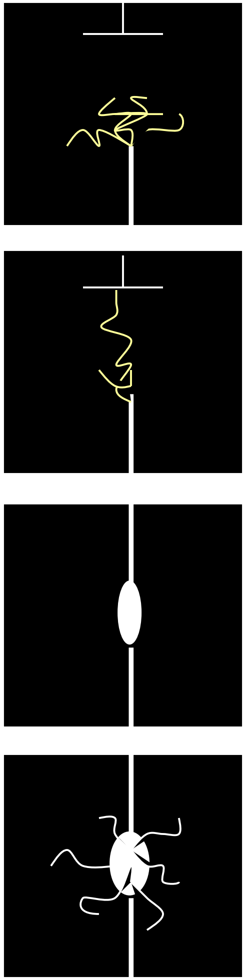
Low temperature (100K) plasma

⇑ Schematic of the streamer corona charge distributions

from Chang et al., 1991

Electrical Discharge Modes in Water

- **Corona discharge**
- **Spark discharge**
- **Arc discharge**
- **Electrohydraulic discharge**



Higher pulse energy

Higher peak current

Larger conductance

Narrow sparkgap

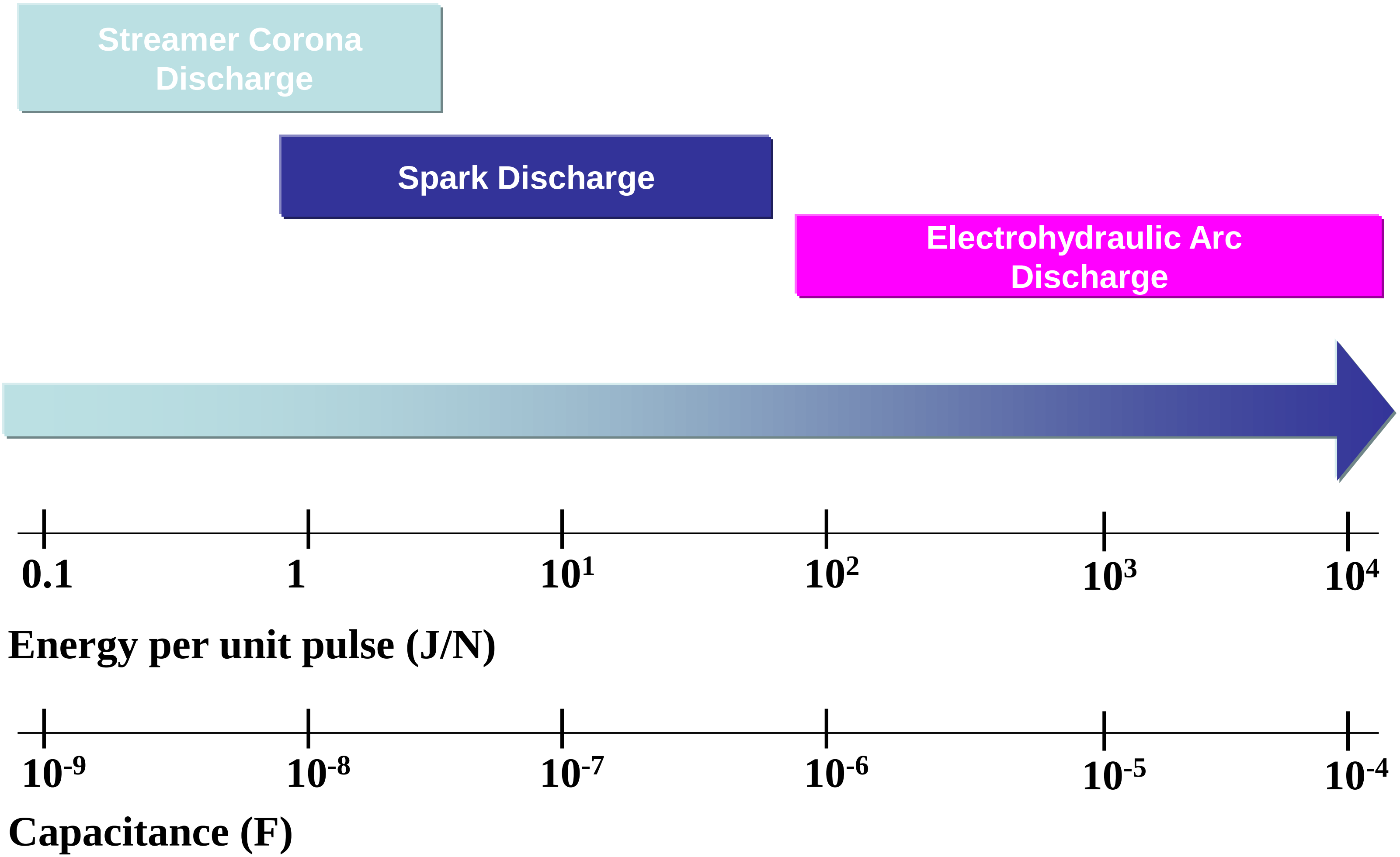
Higher coductivity in sol’n

…

Electrical Discharge Modes in Water

Pulse energy	
~ 1 J	➢ **Corona discharge** (peak current : < 10 A)
10 J	➢ **Spark discharge** (peak current : 10 ~ 100 A)
100 J	➢ **Arc discharge** (peak current : 100 A ~ 1 kA)
~ 10 kJ	➢ **Electrohydraulic(EHD) discharge** (peak current : 1 ~ 100 kA) (intensive UV, shockwave, temperature increase, local supercritical state…)

Electrical Discharge Modes in Water



Streamer Corona Discharge

Spark Discharge

Electrohydraulic Arc Discharge

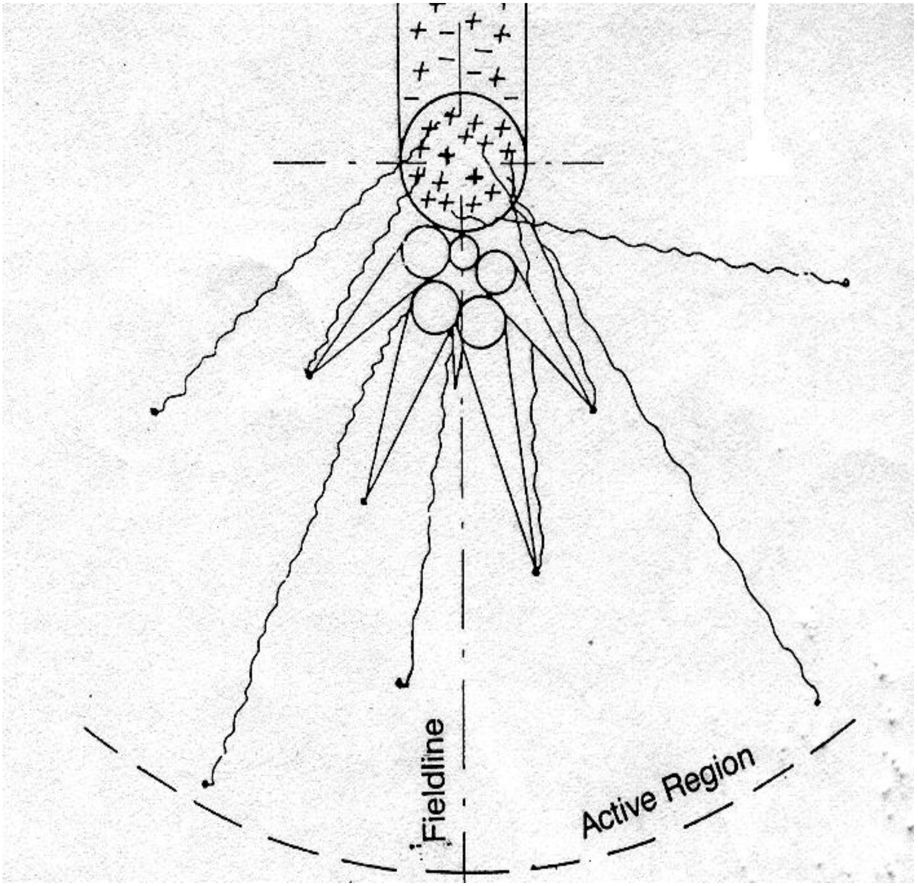
0.1 1 10^1 10^2 10^3 10^4

Energy per unit pulse (J/N)

10^{-9} 10^{-8} 10^{-7} 10^{-6} 10^{-5} 10^{-4}

Capacitance (F)

Formation of Plasma Channel



Chang et al., 1991

- **Plasma :**

High energy state in which molecules are ionized, and exist as radicals, electrons, and cations

- **Plasma channel temperature :**

~ 10000 K

- **Plasma channel volume :**

1 nl ~ 5 ml
vary with specifications of the system

What Happens in the Plasma Channel?

- **Water dissociation :**

$H_2O \rightarrow H\cdot + \cdot OH$

$H_2O \rightarrow 1/2H_2O_2 + 1/2H_2$

$2H_2O \rightarrow H_3O^+ + e^-_{aq} + \cdot OH$

- **Intensive UV/visible light emission** :

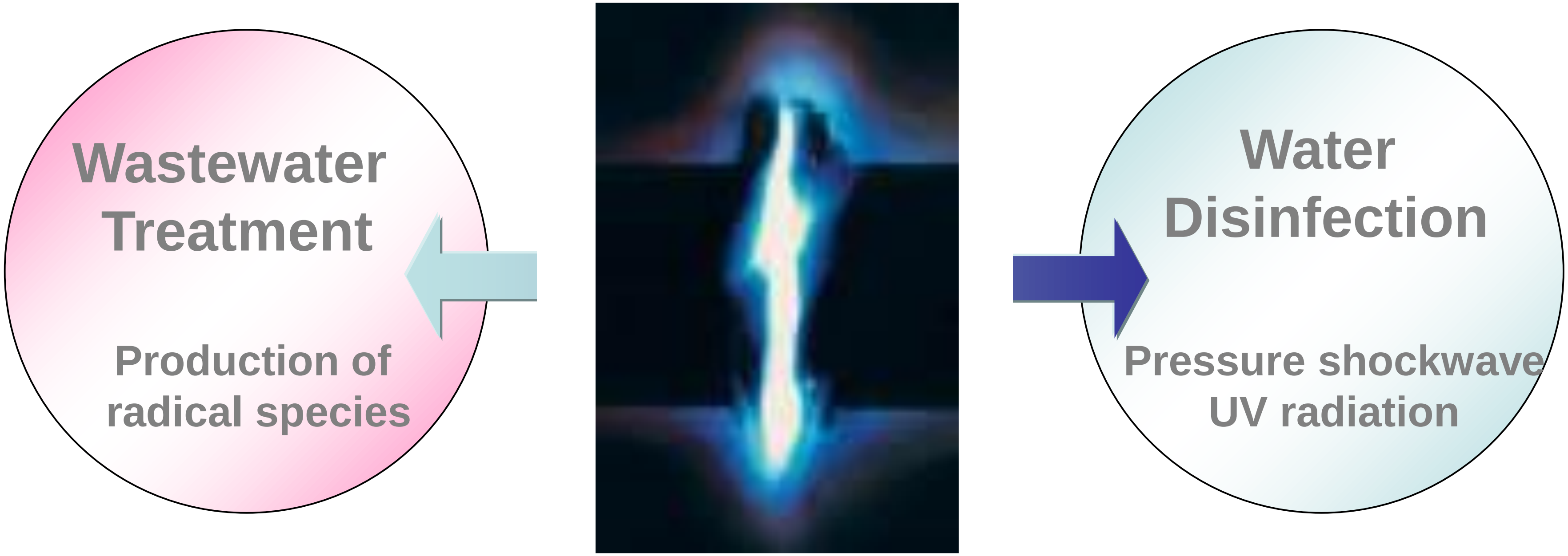
(including VUV emissions)

$h\nu + H_2O \rightarrow H\cdot + \cdot OH$

- **Pyrolysis in the plasma channel**
- **Intensive shockwave : 5 ~ 20 kbar**
- **Electrohydraulic cavitation**
- **Supercritical water oxidation**

Application to Water Treatment

Electrical Discharge Process



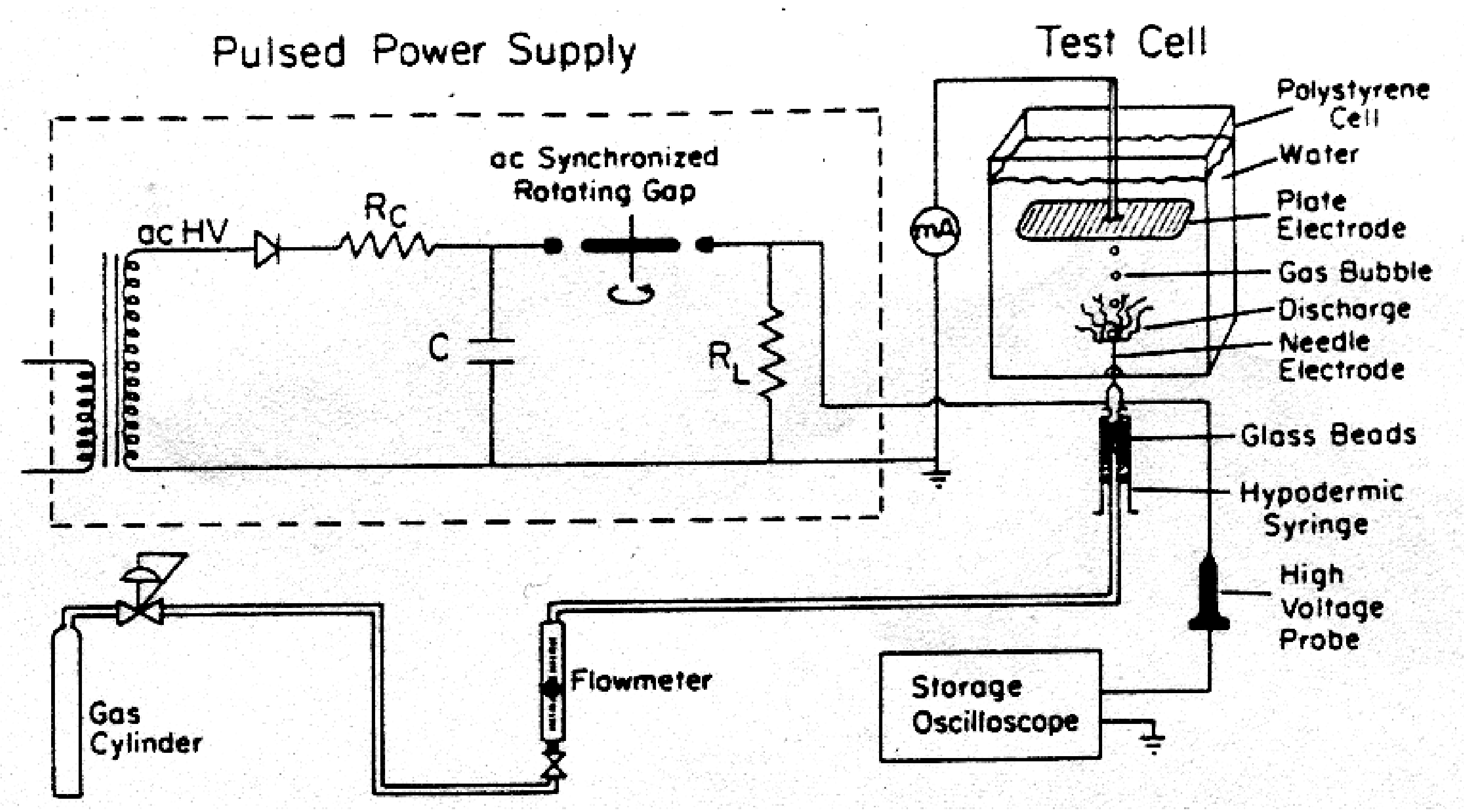
Wastewater Treatment

Production of radical species

Water Disinfection

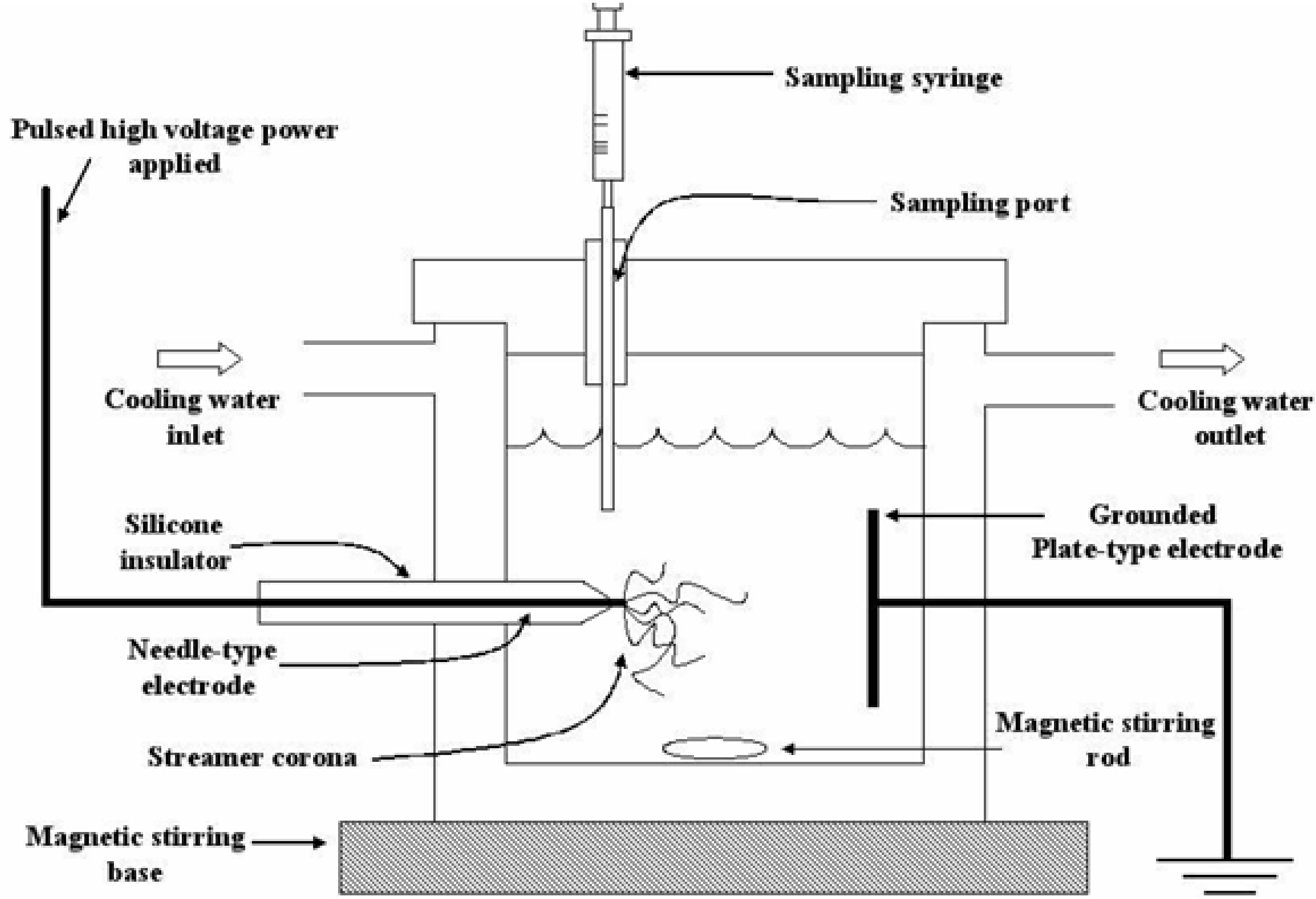
Pressure shockwave
UV radiation

Experimental Apparatus



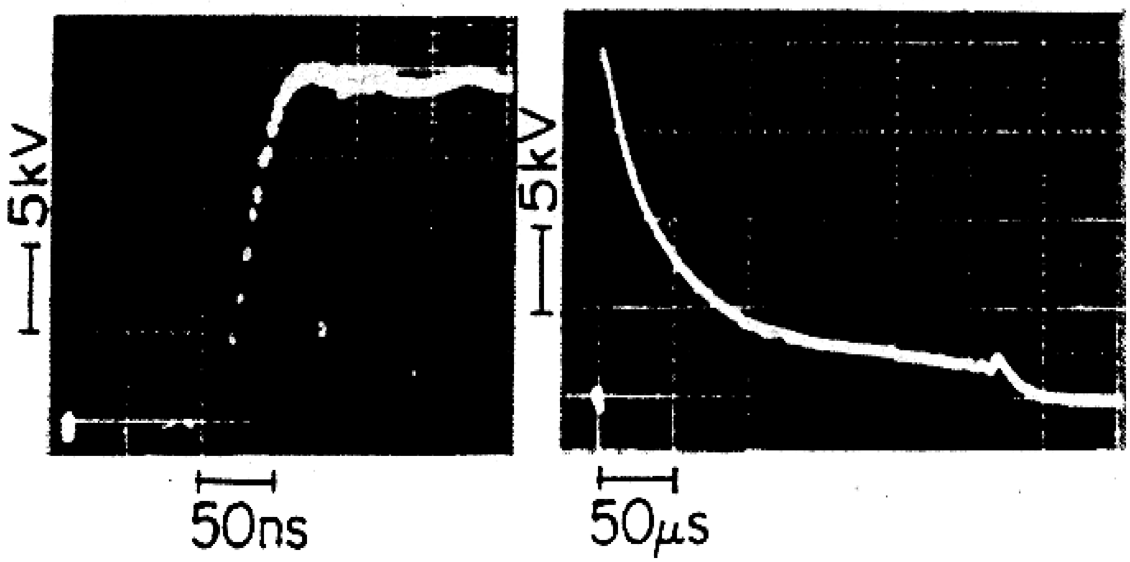
⇑ **Schematic diagram of pulsed discharge reactor**

Experimental Apparatus



⇑ **Schematic diagram of streamer corona reactor.**

Voltage Waveform of Pulsed Discharge



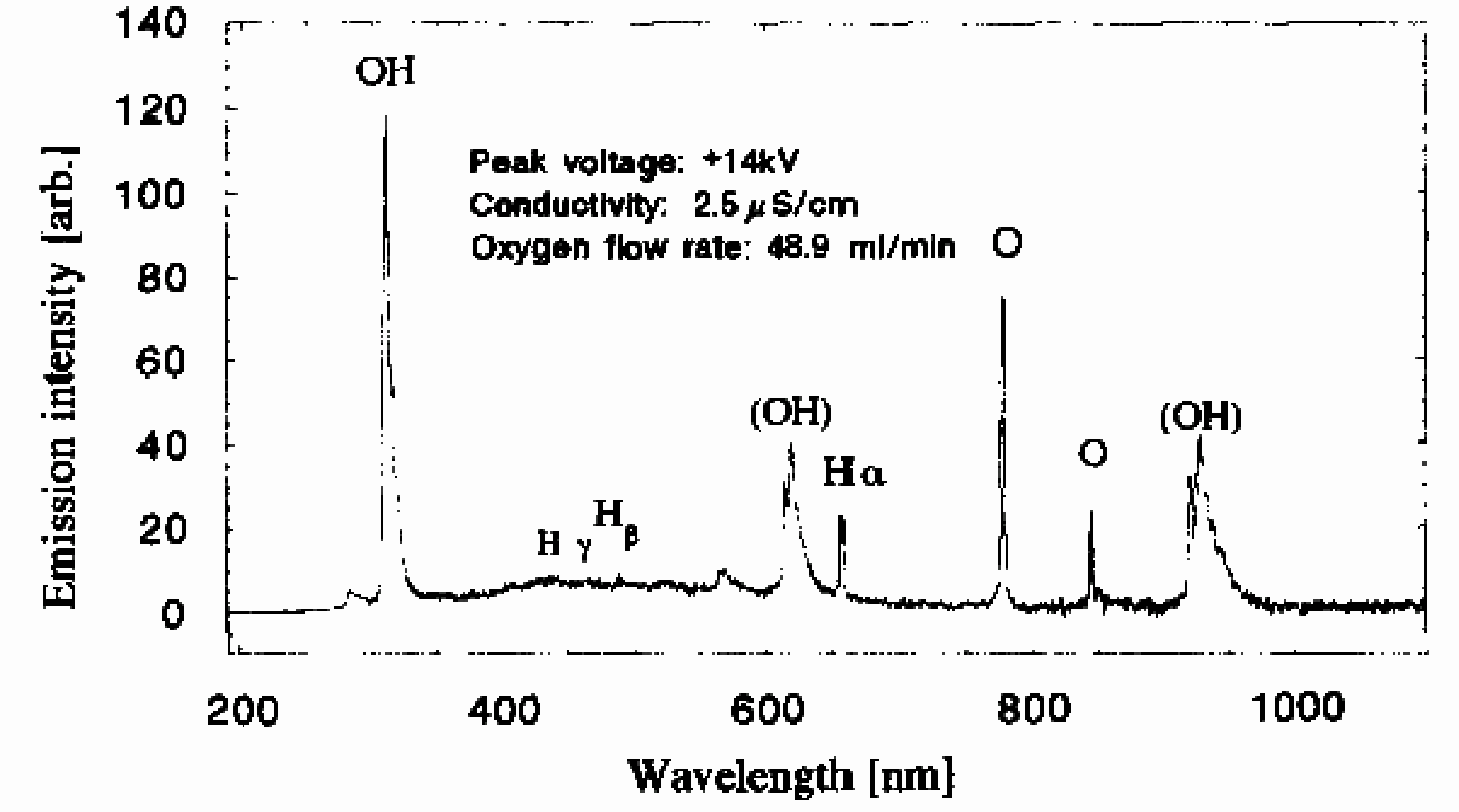
(a) (b)

Applied voltage (6~30 kV)
Frequency (60 discharges/s)
Time constant (1~1000 µs)

⇑ (a) Rise time of applied voltage pulse
(b) Exponential decay of voltage pulse

$$e^{-t/\tau}\quad , \tau : \text{time constant}$$

Light Emission Spectrum

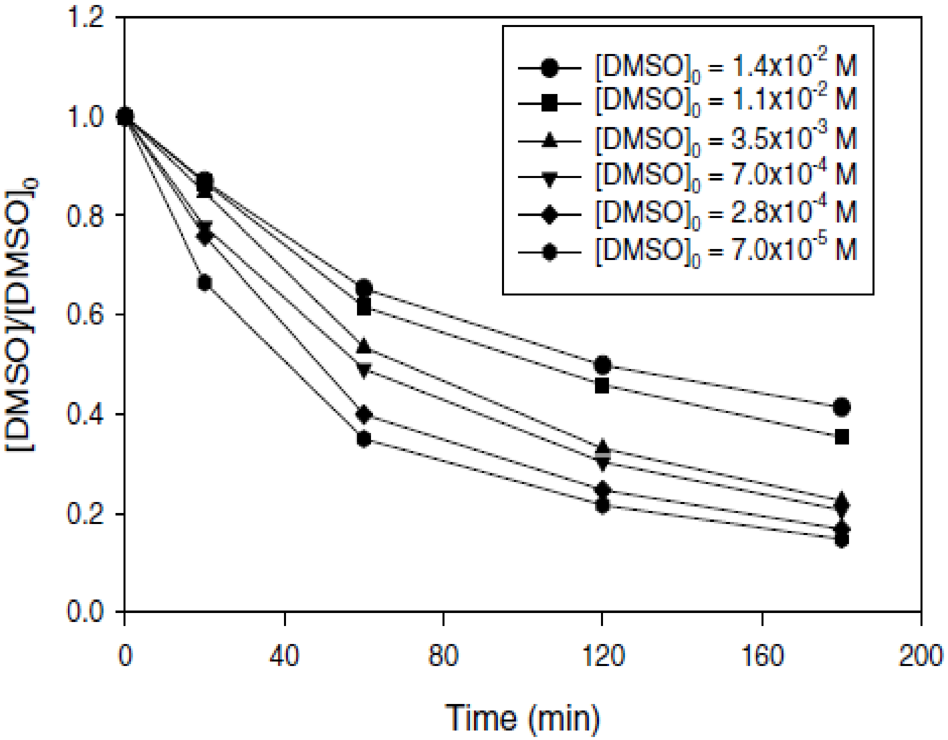


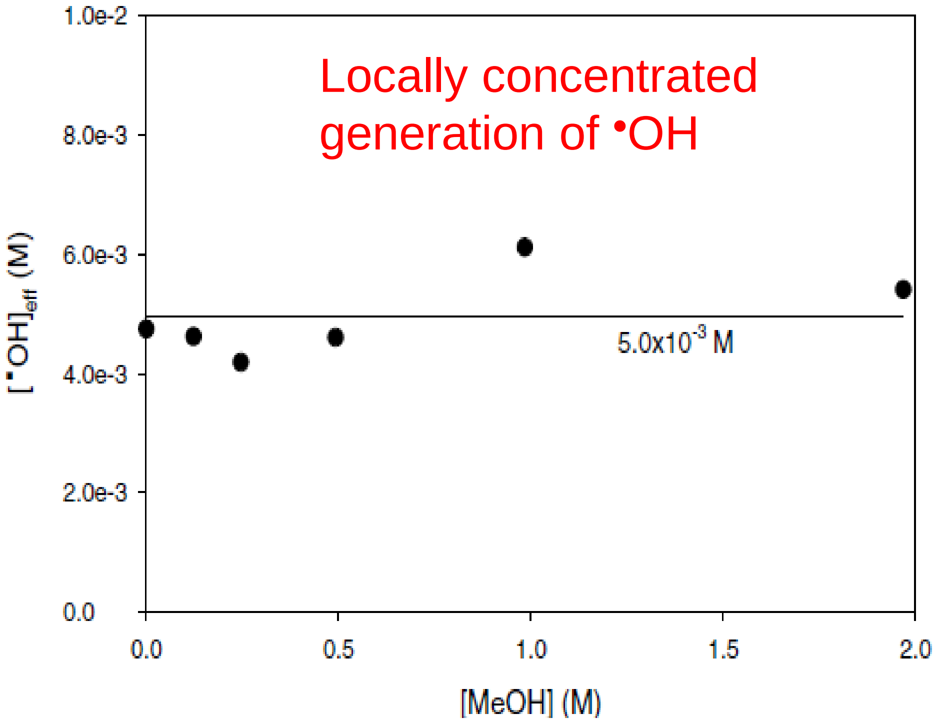
Sun et al., 1997 (J. Electrostatics)

Degradation of Contaminants by SCD System

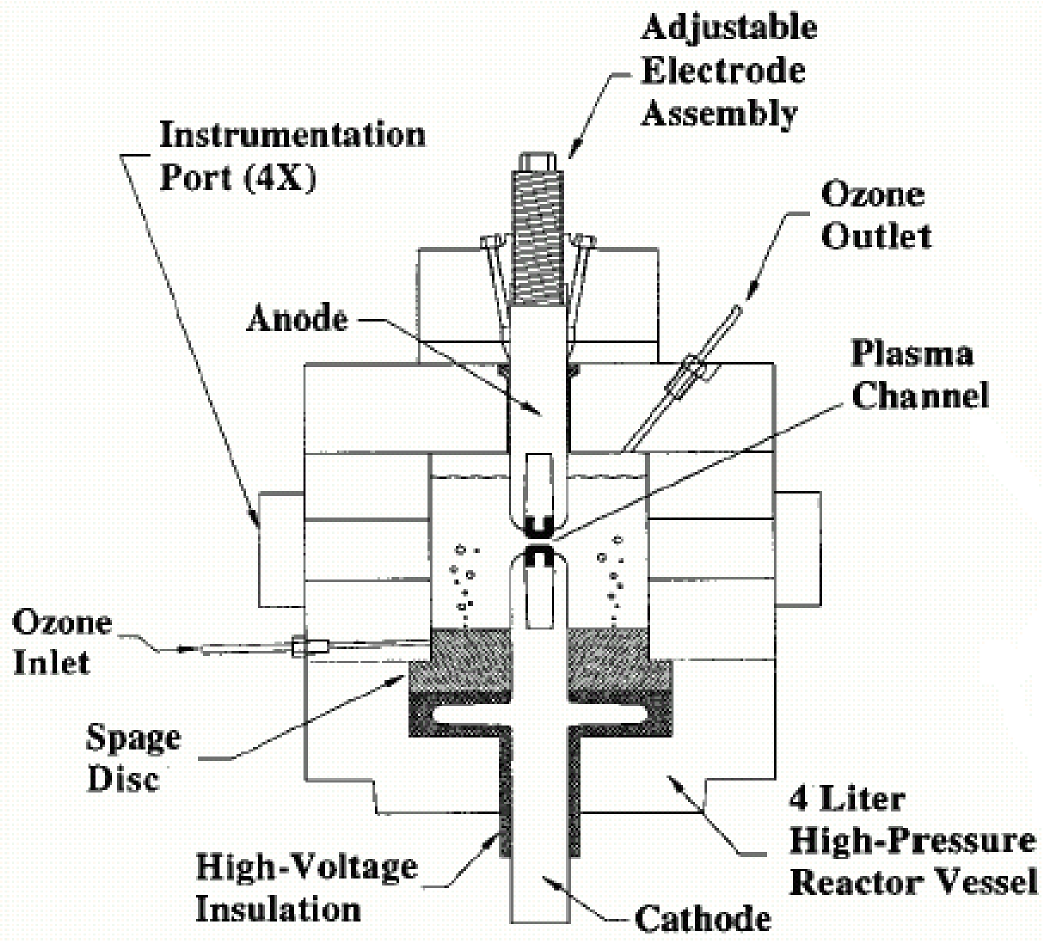
Lee et al., 2006 (Chemosphere)

Oxidative degradation of dimethylsulfoxide (DMSO) by the streamer corona discharge process




Locally concentrated generation of •OH

Degradation of Contaminants by EHD System



Enhancement by $h\nu/O_3$

$$O_3 + h\nu \rightarrow O(^1D) + O_2$$

$$O(^1D) + H_2O \rightarrow 2{}^{\bullet}OH$$

Lang et al., 1998 (Environ. Sci. Technol.)

Degradation of Contaminants by EHD System

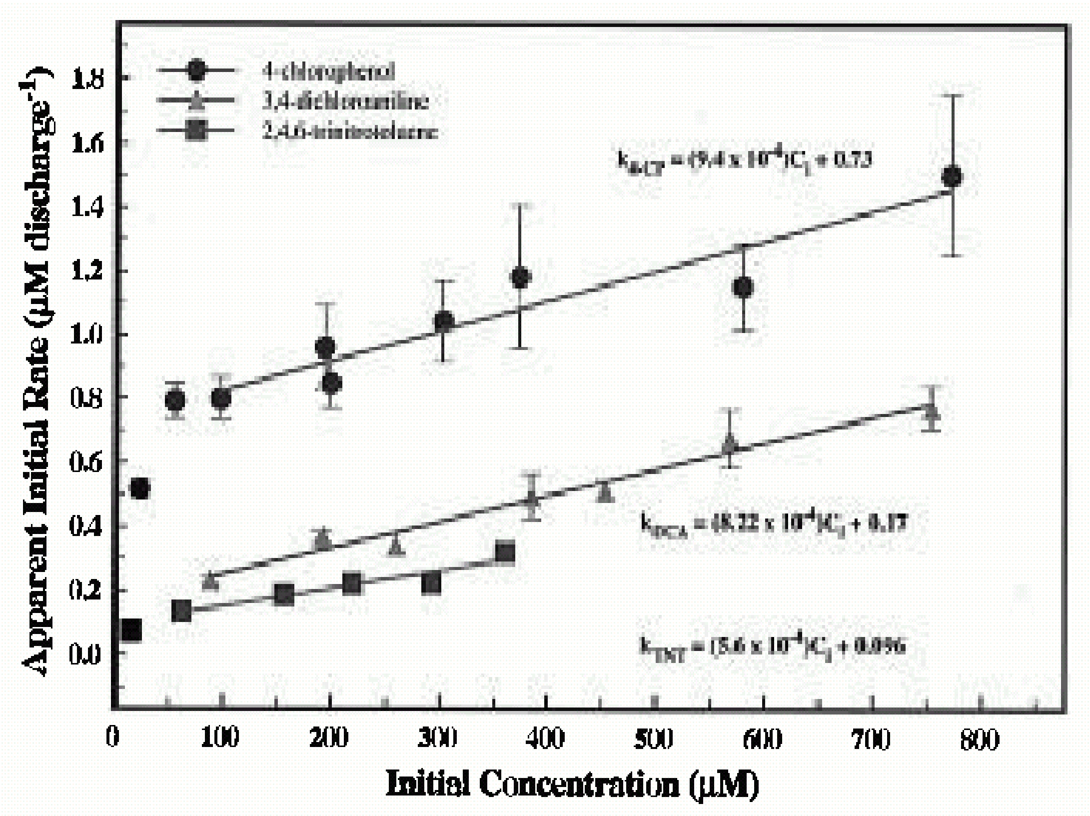
Two reactive regions in the EHD system

1. Plasma channel (~ 3 ml) : Oxidation in a extreme plasma
   Multiple factors causing the contaminant degradation such as pyrolysis, oxidation by radicals, super critical water oxidation…
   First-order decay of the contaminant (k_1)

2. Bulk solution : Degradation mainly by the UV photolysis
   Zero-order decay of the contaminant (k_0)

Degradation kinetics : $$\frac{\mathrm{d}C}{\mathrm{d}N} = -k_1 C_\mathrm{i} - k_0 = -k_\mathrm{A}$$

Degradation of Contaminants by EHD System



Willberg et al., 1996

$$\frac{dC}{dN} = -k_1 C_i - k_0 = -k_A$$

4-chlorophenol :

$(9.4 \times 10^{-4})C_i + 0.73$

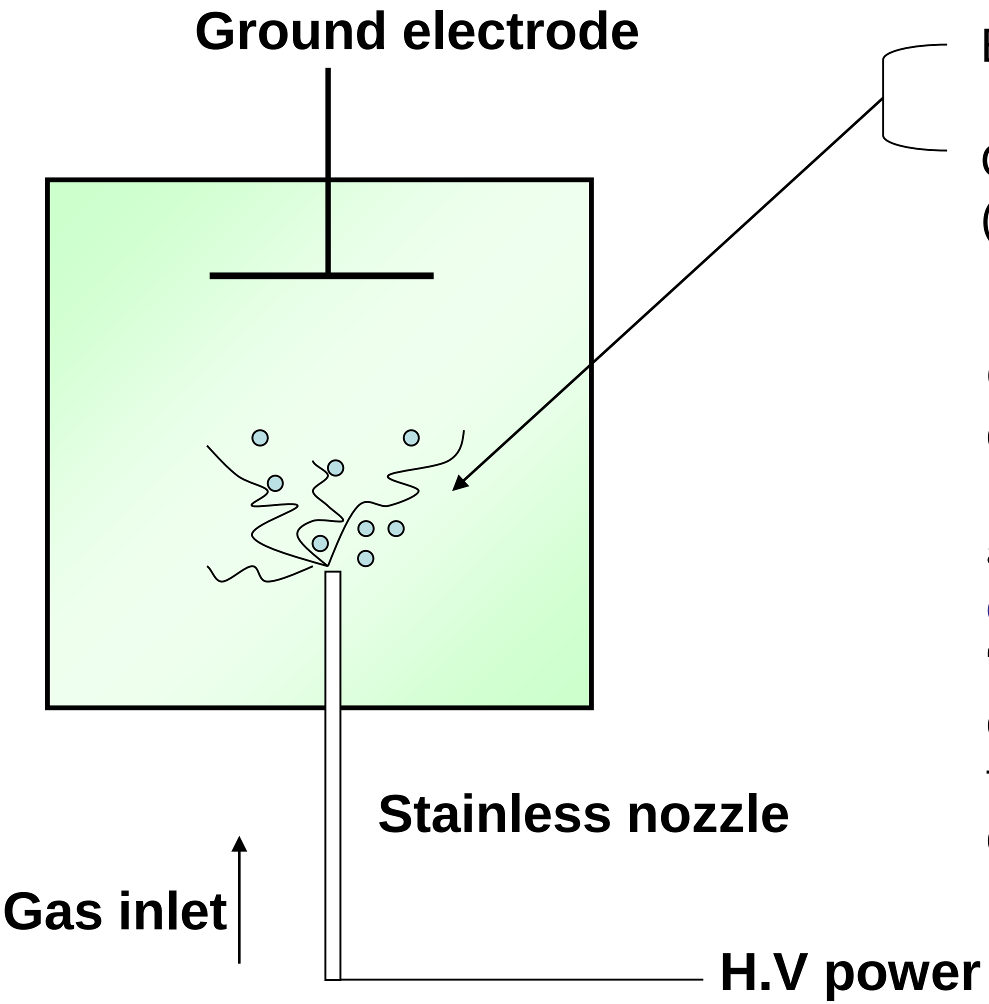
3,4-dichloroaniline

$(8.2 \times 10^{-4})C_i + 0.17$

2,4,6-trinitrotoluene (TNT)

$(5.6 \times 10^{-4})C_i + 0.096$

Electrohydrodynamic Spraying of O_3 or O_2



Ground electrode

Stainless nozzle

Gas inlet

H.V power

Electrical discharge

O_3 micro bubble formation
(Enhancement of mass transfer)

Gas bubbles may be broken up as an electric field passes from a fluid with **large dielectric constant** (water, ε=80) to a fluid with **small dielectric constant** (air, ε=1) In effect the water "**squeezes**" the gas bubbles, electrically generated force exert on the fluid with the smaller dielectric constant

Qualitative Comparison to Energy Efficiency of Plasma Technologies

- **Three categories of plasma technologies : remote, indirect, and direct** (Emelko et al., 2003)

(1) Remote plasma technologies
e.g. ozone process

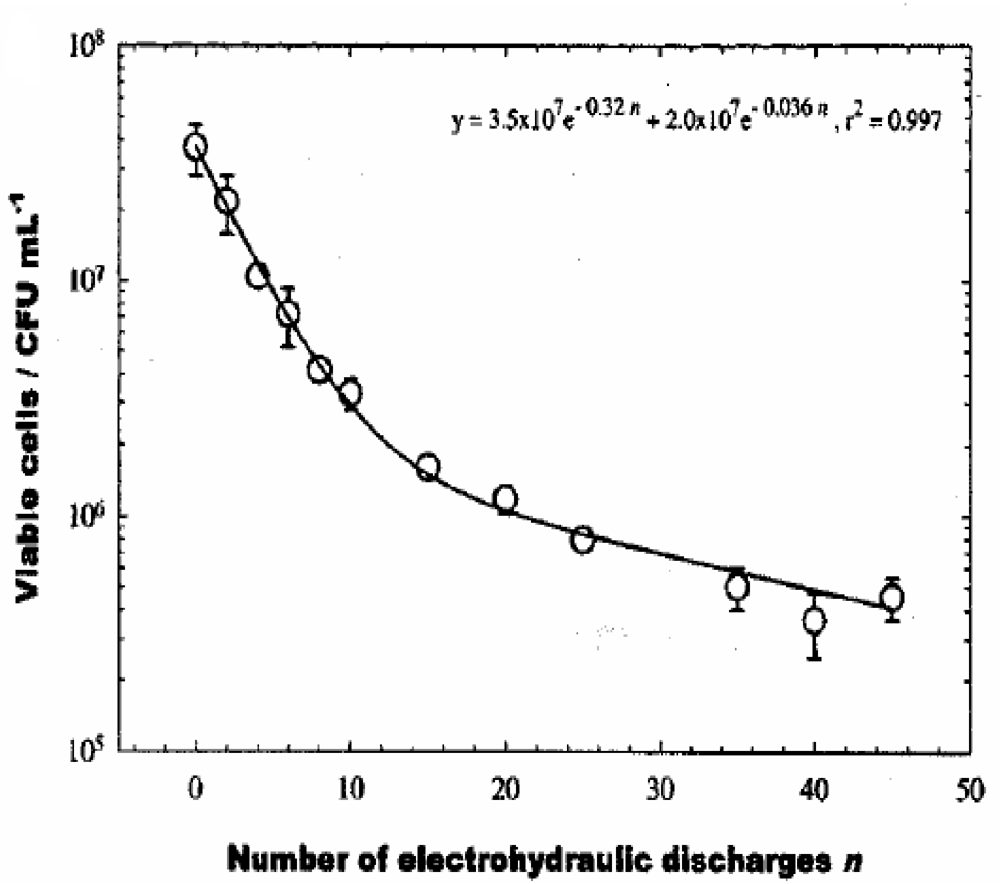
(2) Indirect plasma technologies
e.g. UV, electron beam

(3) Direct plasma technologies
(electrical discharge process)

Potentially more efficient ?

Disinfection Using Electrical Discharge

Disinfection of *Escherichia coli* by electrohydraulic discharge
(Ching et al., 2001, Environ. Sci. Technol.)



Conditions:

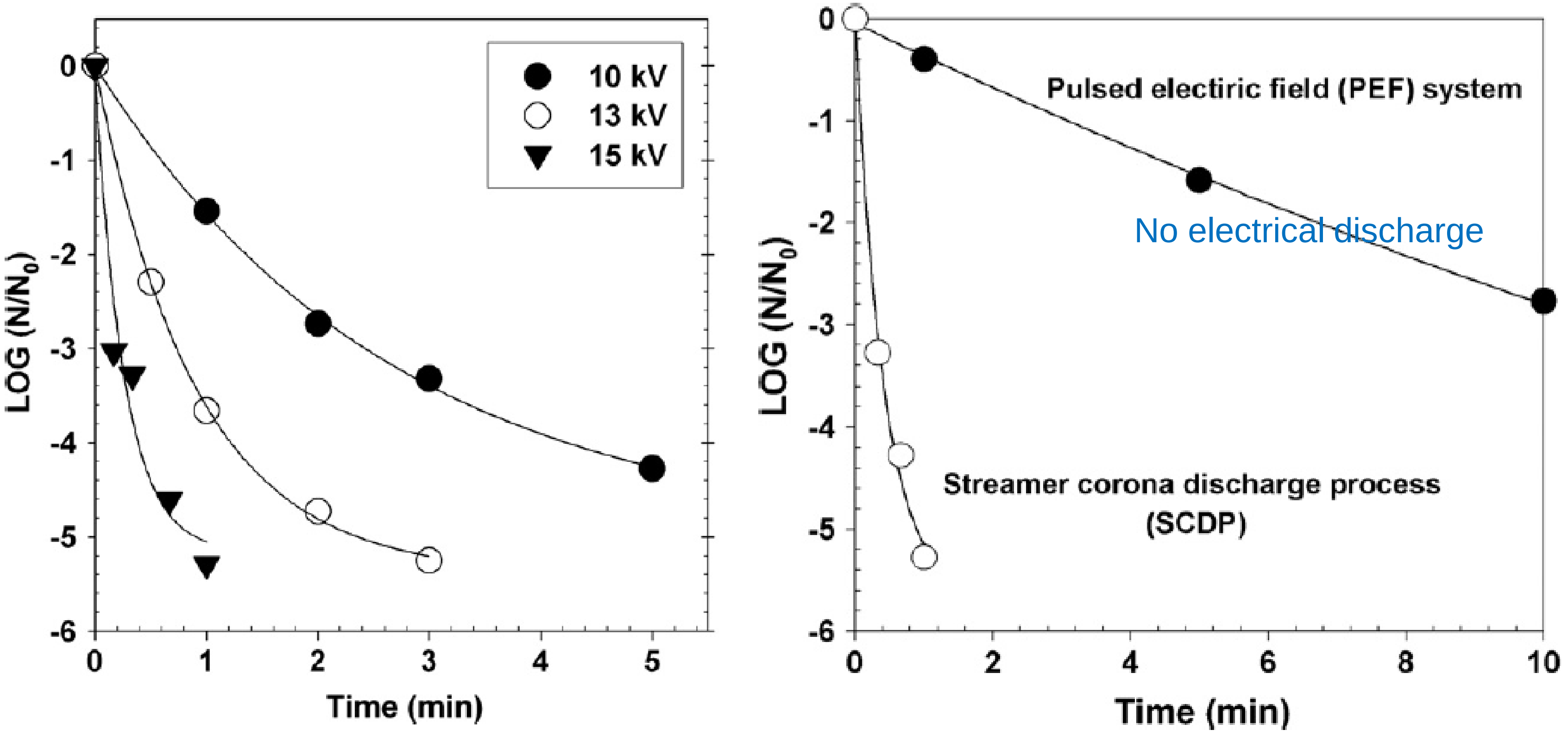
Electrohydraulic discharge mode
7 kJ / unit pulse
Vol. : 3 L

E. coli (ATCC 25922)

Disinfection Using Electrical Discharge

Disinfection of MS2 coliphage by streamer corona discharge

(Lee et al., 2011, Chemosphere)



No electrical discharge

1000 pulses per minute

Disinfection Using Electrical Discharge

Germicidal factors by electrical discharge

- Reactive chemical species

  $^{\bullet}OH$, $O_2^{-\bullet}$, O, O_3 etc.

- Pressure shockwave

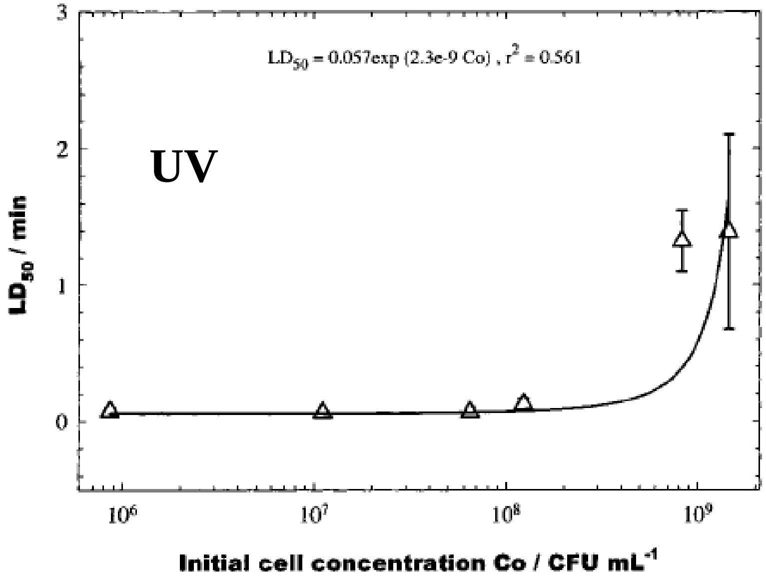
  Pressure gradient with a short rise time

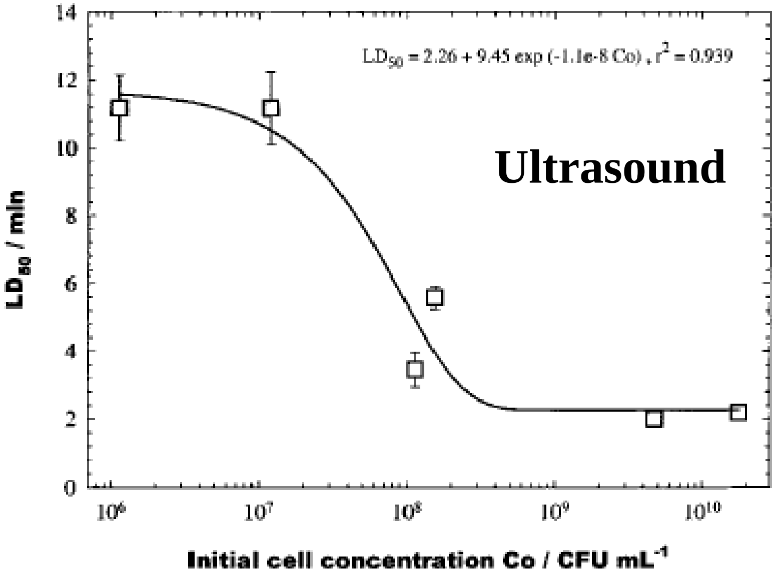
- UV radiation

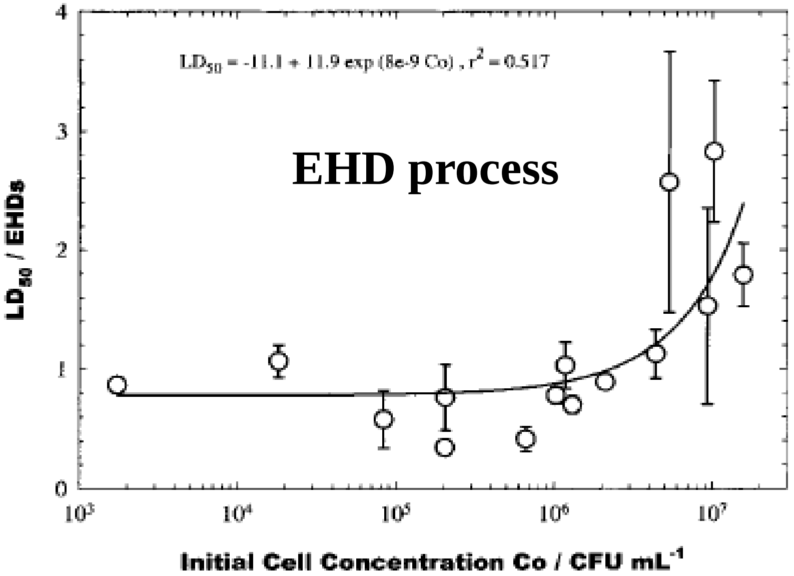
  Pulsed UV of high intensity

The major mechanism for the microbial inactivation depends on the electrical discharge mode?

Disinfection Using Electrical Discharge



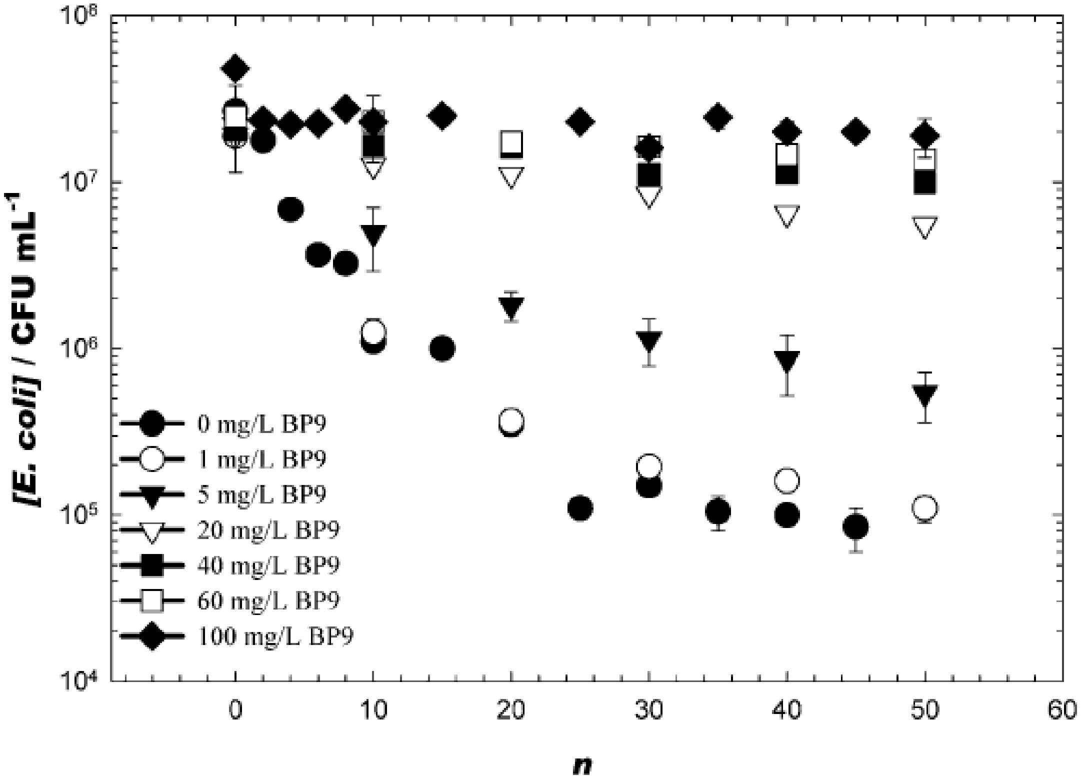




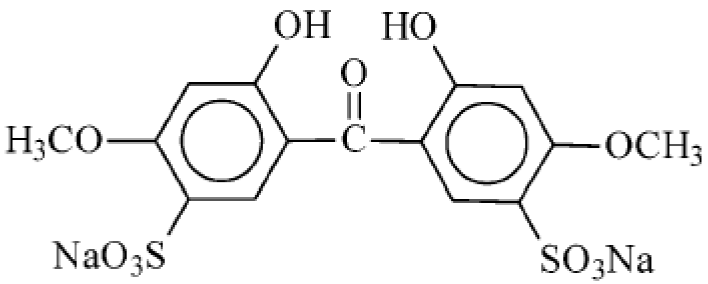
Disinfection Using Electrical Discharge

Soluble sunscreens fully protect *E. Coli* from disinfection by electrohydraulic discharge

(Ching et al., 2003, Environ. Sci. Technol.)

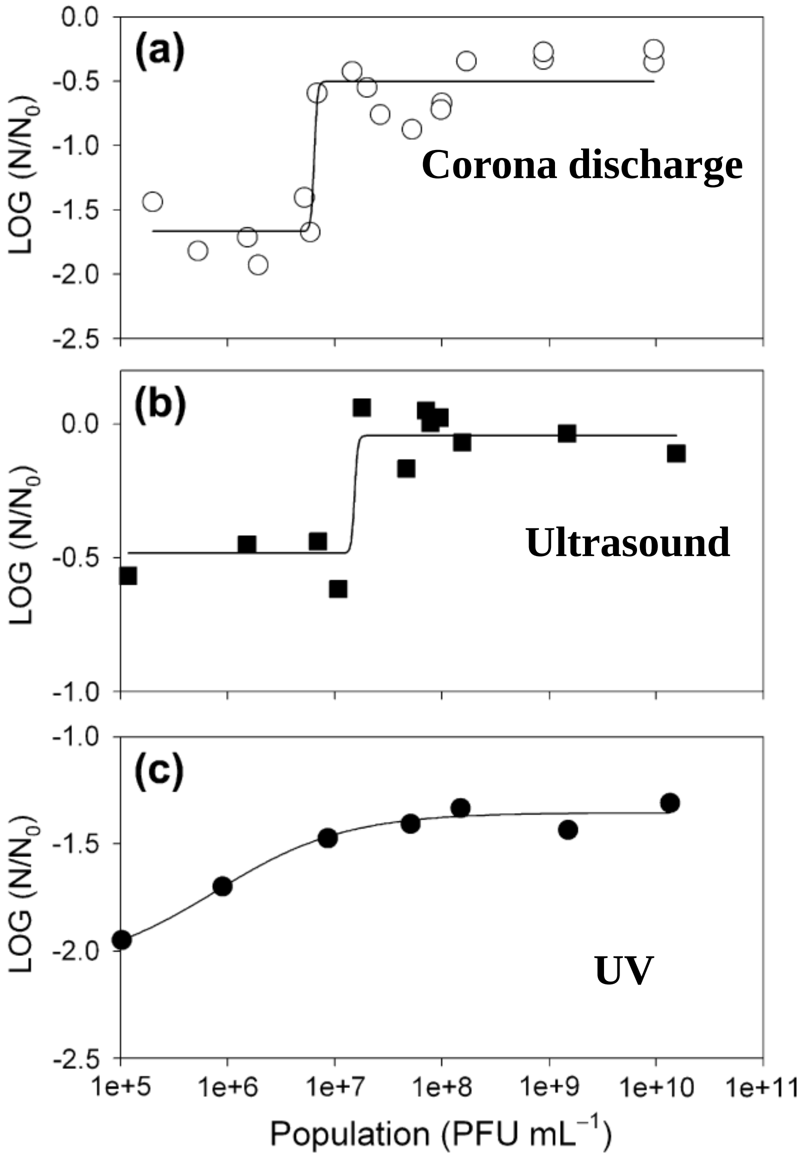


Effect of Soluble Sunscreens



Benzophenone-9

Disinfection Using Electrical Discharge



Viruses are inactivated
by different disinfection mechanism

⇒ Pressure shock wave?

Lee et al., 2011 (Chemosphere)

Water Treatment Using Electrical Discharge

- **Advantages**

  - No production of harmful disinfection byproducts
  - No limitations to the water (or wastewater) conditions
  - Convenient due to the direct usage of the electrical power
  - Inactivation of microorganisms concurrent with the degradation of organic pollutants

- **Issues**

  - Energy efficiency
  - Cost of facilities