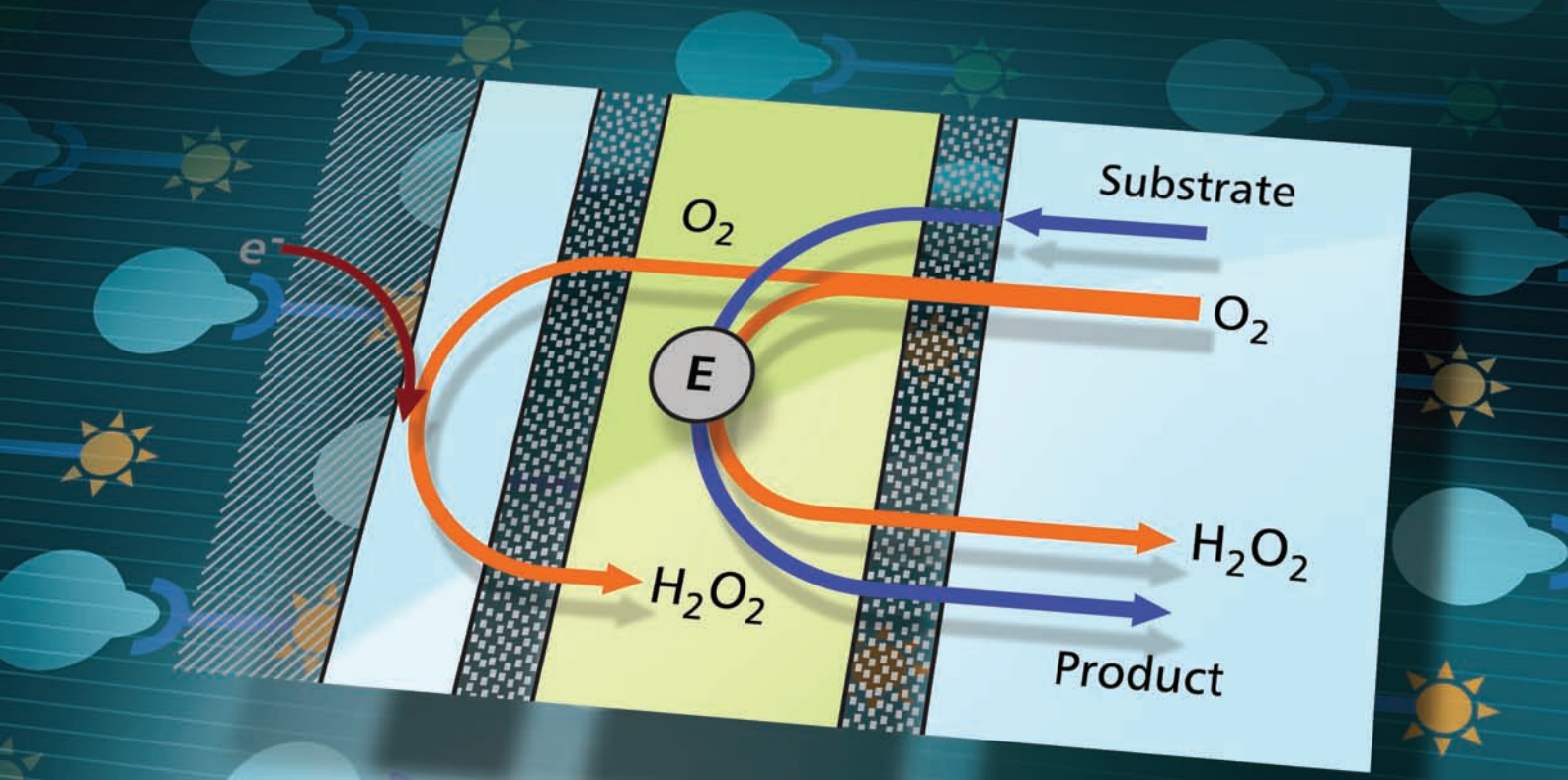
Chemical Sensors and Biosensors

Fundamentals and Applications



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Fundamentals and Applications

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Preface

As suggested by Marshal McLuhan, media (in the more general meaning of the term) act as extensions of the functions of the human body [1]. In the same way that the microphone acts as an extension of the ear, chemical sensors can be considered to be extensions of the organs of chemical perception that are the nose and the tongue.

The development of chemical sensors responds to the increasing demand of chemical data that characterize various systems of interest. Such a system can be the human body itself, whose physiological state can be assessed unequivocally by physical, chemical and biochemical parameters. The quality of the ambient and natural environment is characterized by measuring the content of noxious chemical species. No less important is the automatic control of certain industrial processes that depend on specific chemical parameters.

In general, standard analytical methods (e.g., chromatography, spectrometry and electrophoresis) can provide the same kind of information as that produced by chemical sensors. The advantage of the chemical sensor approach results from the fact that they are specialized, small size, portable and inexpensive devices that are suitable for *in situ* analysis and real-time monitoring of chemical parameters. Worthy of mention is the capability of dedicated chemical sensors to identify pathogen micro-organisms and viruses via characteristic compounds that are parts of the structure of the target species.

"There's plenty of room at the bottom" said Richard Feynman in a seminal lecture in 1959, that anticipated the advent of nanotechnology. This sentence can be paraphrased as follows: "There's plenty of new opportunities at the bottom". This applies well to the development of chemical sensors. Indeed, the most important trend in this area is the application of nanomaterials, either as substitutes for classical materials and reagents or in the implementation of completely new sensing and transduction methods. Of outstanding importance is the size compatibility of nanomaterials with biopolymer molecules, which allows fabrication of bionanocomposites with promising potential for application in the design of chemical sensors. New fabrication technologies, mostly inspired by microelectronic technology and nanotechnology, are expected to lead to an increase in the degree of integration in chemical-sensor arrays, thus prompting advances in production and application of artificial nose/tongue devices. Integration of chemical sensors with microfluidic systems is another promising trend since microfluidic systems allow extremely small sample volumes to be processed and analyzed automatically.

New books on chemical sensors are published regularly, but most of them are collective volumes profiling particular kinds of chemical sensor and particular applications of chemical sensors. A comprehensive overview of chemical sensors in one single book is needed for two reasons. First, such a book would serve as a useful teaching aid for use in courses covering the subject of chemical sensors. Secondly, an indepth introduction to the field of chemical sensors for scientists and engineers new to this subject would be advantageous. There are currently on the market a series of volumes that are intended to respond to the above aims. However, as the field progresses, a new book that covers recent advances is always welcome.

The development of a chemical sensor is very often a matter of material synthesis and processing. Synthetic materials (both inorganic and organic), materials of biological origin (proteins, nucleic acids, micro-organism and living cells), as well as biomimetic synthetic materials are widely used in the development of chemical sensors. Of equal importance is the fabrication technology, because the final goal in chemical-sensor research is the production of a marketable product. That is why the first eight chapters in this text introduce the main kinds of material used in the development of the chemical sensors, as well as typical processes and technologies involved in fabrication of chemical sensors. The next fourteen chapters present various classes of chemical sensors organized according to the transduction method. The final chapter is devoted to chemical sensors based on highly organized biological material such as micro-organisms and living cells.

This book has been designed mostly as an instruction manual in chemical sensors, with a particular attention on balancing classical topics with contemporary trends. Clearly, owing to its extent, the contents of this book cannot be covered in a normal course of lectures. However, the course instructor can select topics that fit the class level and the particular interest of the attending students. Moreover, the curriculum can be personalized by encouraging each student to explore more deeply into certain advanced topics. In addition, a study of chemical sensors is an enlightening excursion through various scientific and technological areas, thereby contributing substantially to the development of the student's scientific knowledge.

Additionally, this book will be useful to any scientist who needs an introduction into the field of chemical-sensor science and technology. As this is an interdisciplinary field, this book will be of interest to engineers, chemists, biochemists, microbiologists and physicists endeavoring to start up research work in the field of chemical sensors.

Nothing done by humans can be perfect, but, at least, it could be perfectible. Hence, any critical comment or suggestion is welcome.

1. McLuhan, M. (2003) *Understanding Media: The Extensions of Man*, Gingko Press, Corte Madera, Calif.

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Finally, I am grateful, in writing this book, to all those scientists who have contributed to the advance of chemical sensor science and technology. Many of these scientists are cited in the book, but, owing to space limitations, much valuable work in this area could not be included or cited.

List of Symbols

Roman Symbols

Symbol	Meaning	Section Reference
A	(a) surface area	4.2.2; 13.3.1
	(b) absorbance	18.3.1
	(c) amplitude of an electromagnetic wave	18.6.2
$_{AC}$	subscript pertaining to alternating current	
a	(a) thermodynamic activity	10.2.1
	(b) sensor sensitivity	1.5
	(c) molar absorptivity	18.3.1
	(d) the exponent in the expression of CPE impedance	17.2.1
b	thickness of a light-absorbing layer	18.3.1
bipy	2,2′-bipyridine	
C	capacitance	11.1.3
C_{dl}	capacitance of the electric double layer	17.2.2
C_f	proportionality constant in the Sauerbrey equation	21.2.3
C_0	static capacitance in the equivalent circuit of a TSM oscillator	21.2.2
C_1	capacitance at the motional branch of the equivalent circuit of a TSM piezoelectric oscillator	21.2.2
c	(a) analyte concentration	1.5
	(b) concentration of the enzyme–substrate complex	3.6.1
	(c) concentration of the antibody–antigen complex	6.4
	(d) light velocity in vacuum	18.1
c^*	concentration of an excited-state species	18.3.6
c_A	concentration of the species A	10.4.2
c_{AR}	concentration of the analyte–receptor combination	18.4.1
c_B	concentration of the species B	10.4.2
$c_{O,b}$	concentration of the oxidized form of a redox couple in the bulk solution	11.3.1
$c_{O,i}$	concentration of the oxidized form of a redox couple at the electrode/electrolyte interface	11.3.1
c_Q	concentration of a fluorescence quencher	18.3.7
c_R	receptor concentration	18.4.1
$c_{R,t}$	total concentration of the receptor	18.4.1
$c_{R,b}$	concentration of the reduced form of a redox couple in the bulk solution	11.3.1
$c_{R,i}$	concentration of the reduced form of a redox couple at the electrode/electrolyte interface	11.3.1
D	diffusion coefficient	4.2.2; 13.3.1
Da	Damköhler number for internal diffusion in an immobilized enzyme layer	4.3.1
Da_M	mediator Damköhler number	15.2.2
Da_S	substrate Damköhler number	15.2.2
$_{DC}$	subscript pertaining to direct current	
D_M	diffusion coefficient of a redox mediator	15.2.1
$D_{P,e}$	diffusion coefficient of the product within an immobilized enzyme layer	4.3.1
$D_{P,m}$	diffusion coefficient of the product in the external membrane of an enzymatic sensor	4.2.2
$D_{S,e}$	diffusion coefficient of the substrate within an immobilized enzyme layer	4.3.1
$D_{S,m}$	diffusion coefficient of the substrate in the external membrane of an enzymatic sensor	4.2.2
E	enzyme	3.6.1
E	(a) energy	11.1.2
	(b) electrode potential	10.2.2; 13.3.1
	(b) Young modulus	22.1.2

ΔE	difference between the actual electrode potential and the formal electrode potential	13.6.1
E_{cell}	cell voltage	10.2.2
E_{AC}	sine wave alternating potential	13.7.7
E_{F}	Fermi energy	11.1.2
EMF	electromotive force	10.2.2
E_{ph}	photon energy	18.1
E_{pzc}	potential of zero-charge	13.5.2
E_{O}	oxidase enzyme in the oxidized form	14.2.1
E_{r}	reference electrode potential	10.4.1
E_{R}	oxidase enzyme in the reduced form	14.2.1
ES	enzyme–substrate complex	3.6.1
$E_{1/2}$	half-wave potential	13.3.2
E^0	standard electrode potential	10.2.2
E_{f}^0	formal electrode potential	10.2.2; 13.3.1
E_0	prelogarithm constant in the response equation of a potentiometric ion sensor	10.4.1
e	(a) enzyme concentration	3.6.1
	(b) elementary charge	11.1.3
	(c) the base of natural logarithm	17.1
e_O	concentration of the oxidized form of an oxidase enzyme in an immobilized enzyme layer	15.1.1
e_{R}	concentration of the reduced form of an oxidase enzyme in an immobilized enzyme layer	15.1.1
e_{t}	total enzyme concentration	3.6.1
e_{y}	measurement error of the sensor response	1.5
e^-	electron	
F	(a) Faraday constant	
	(b) power of a fluorescence light beam	18.3.5
F_0	fluorescence power in the absence of a quencher	18.3.7
f	(a) *F/RT*	13.6.1
	(b) frequency	13.7.7; 21.2.2
f_{e}	enzyme loading factor	4.3.1
Δf_{L}	change in the resonance frequency due to liquid loading on a TSM piezoelectric resonator	21.2.5
Δf_{m}	change in the resonance frequency due to mass loading	21.2.3
f_{O}	(a) m_{O}/m_t	15.1.1
	(b) $m_{\text{O},0}/m_t$	15.2.3
$\tilde{f}$	complex frequency of a TSM resonator	21.2.7
f_0	resonant frequency of an oscillator	21.2.1; 22.1.3
G	Gibbs free energy	10.2.1
ΔG	Gibbs free energy change in a chemical process	10.2.2
ΔG^*	activation energy of a chemical reaction	13.6.1
ΔG^0	standard Gibbs free energy change in a chemical process	10.2.2
GOD_{ox}	glucose oxidase, oxidized form	3.5.1
GOD_{red}	glucose oxidase, reduced form	3.5.1
ΔH	heat of reaction	Chapter 9
ΔH_r^0	standard enthalpy of reaction	Chapter 9
h	(a) Plank's constant	18.1
	(b) microcantilever thickness	22.1.1
I	(a) ionic strength	10.2.1
	(b) electric current	17.1
I_{AC}	sine-wave alternating current	17.1
I_{D}	drain current of a metal-insulator-semiconductor field effect transistor	11.1.4
I_{DC}	DC current	17.1
I_{m}	AC current amplitude	17.1
i	electrolytic current	13.3.1
i_{a}	anodic current	13.3.1
$i_{\text{a,d}}$	limiting, diffusion-controlled anodic current	13.3.2
i_{C}	capacitive current	13.5.3
i_{c}	cathodic current	13.3.1
$i_{\text{c,d}}$	limiting, diffusion-controlled cathodic current	13.3.1

i_f	Faradaic current	13.7.7
i_l	limiting current at an mediator-based amperometric enzyme sensor	15.1.3
i^*	the particular value of the limiting current recorded at $\alpha \gg 1$ and for $S = 1$	15.1.3
i_0	exchange current	13.6.4
J	diffusion flux	
J_d	limiting flux under first order kinetics and external diffusion control	15.1.2
J_l	flux limiting value	15.1.2
$J_{l,0}$	flux limiting value under zero-order kinetics	15.1.2
$J_{l,1}$	flux limiting value under first-order kinetics	15.1.2
J_M	mediator flux	15.2.1
J_P	product flux in an enzymatic sensor	4.2.1
J_S	substrate flux in an enzymatic sensor	4.2.1
$J_{P,m}$	product flux in the membrane of an enzymatic sensor	4.4.1
$J_{S,m}$	substrate flux in the membrane of an enzymatic sensor	4.4.1
J^*	the particular value of J_d for $s = K_M$	15.1.3
j	(a) current density	13.6.2
	(b) imaginary unit ($\sqrt{-1}$)	17.1; 21.2.2
j_a	anodic current density	13.6.2
$j_{a,d}$	limiting (diffusional) anodic current density	13.6.3
j_c	cathodic current density	13.6.2
$j_{c,d}$	limiting (diffusional) cathodic current density	13.6.3
j_0	exchange-current density	13.6.4
K_a	affinity constant	6.2.2
K_d	dissociation constant	18.4.1
K_e	equilibrium constant for the analyte–receptor interaction	18.4.1
K_{ex}	ion-exchange constant for a glass membrane	10.6.2
K_{exch}	ion-exchange constant for an ion-exchanger liquid membrane	10.8.3
K_M	Michaelis–Menten constant	3.6.1
K_p	partition coefficient	10.3.2
$K_{p,M}$	partition coefficient of the ion M	10.9.4
K_s	solubility constant of a sparingly soluble salt	10.5.2
k	spring constant of the microcantilever material	22.1.3
k_a	surface normalized pseudo-first-order rate constant for an enzymatic sensor	4.2.4
$k^{pot}_{A,B}$	potentiometric selectivity coefficient relative to ions A and B	10.4.2
k_B	Boltzmann constant	11.1.2
k_{cat}	turnover number of an enzyme	3.6.3
k_d	decay rate constant of an excited state species	18.3.6
k_e	(a) pseudo-first-order reaction rate for an enzyme-catalyzed reaction	6.2.2
	(b) excitation rate constant	18.3.7
k_H	proportionality coefficient in the Henry isotherm	11.3.5
k_m	mass-transfer coefficient	13.3.1
k'_M	reaction rate for enzyme regeneration by reaction with a redox mediator	15.1.1
$k^{pot}_{M,N}$	potentiometric selectivity coefficient relative to ions M and N	10.6.2
$k_{m,P}$	mass-transfer coefficient of the product	4.2.2
$k_{m,S}$	mass-transfer coefficient of the substrate	4.2.2
$k_{P,m}$	mass-transfer coefficients of the product in the membrane of an enzymatic sensor	4.4.1
k_s	standard rate constant of an electrochemical reaction	13.6.1
$k_{S,m}$	mass-transfer coefficients of the substrate in the membrane of an enzymatic sensor	4.4.1; 15.1.1
k_{SV}	Stern–Volmer constant	18.3.7
k_1	forward rate constant of the first step in the Michaelis–Menten mechanism	3.6.1
k_{-1}	backward rate constant of the first step in the Michaelis–Menten mechanism	3.6.1
k_2	rate constant for the second step in the Michaelis–Menten mechanism	3.6.1
L	(a) luminophores species	18.3.7
	(b) analyte-analog	18.4.2
L	electrical conductance	17.8.1
L^*	excited luminophores species	18.3.7
L_l	inductance at the motional branch of the equivalent circuit of a TSM piezoelectric oscillator	21.2.2
l	(a) distance between the plates of a capacitor	13.5.3
	(b) distance between the electrodes of an idealized conductometric cell	17.8.1
	(c) microcantilever length	22.1.1

l_e	thickness of an immobilized enzyme layer	4.2.1; 15.1.2
l_m	thickness of the external membrane in an enzymatic sensor	4.2.1
M_O	oxidized form of a redox mediator	14.2.1
M_R	reduced form of a redox mediator	14.2.1
m	(a) activity of an unspecified M^+ ion	10.5.2
	(b) mass	22.1.3
Δm	mass variation	21.2.3; 22.1.3
m^*	effective mass of a vibrating microcantilever	22.1.3
m_{aq}	activity of an unspecified M^+ ion in solution	10.3.2
m_m	activity of an unspecified M^+ ion in an ion-selective membrane	10.3.2
m_O	concentration of the oxidized form of a redox mediator	15.1.1
$m_{O,0}$	concentration of the oxidized mediator at the electrode surface	15.2.3
m_Q	mass of the vibrating zone of a TSM piezoelectric oscillator	21.2.3
m_R	concentration of the reduced form of a redox mediator	15.1.1
$m_{R,0}$	concentration of the reduced mediator form at the electrode surface	15.2.3
m_t	$m_O + m_R$	15.1.1
m_1	activity of an unspecified M^+ ion within the left-hand solution of an ion-selective membrane cell	10.5.2
m_2	activity of an unspecified M^+ ion within right-hand solution of an ion-selective membrane cell	10.5.2
N_O	number of moles of oxidized form of a redox couple	13.3.1
N_R	number of moles of reduced form of a redox couple	13.3.1
n	(a) number of moles	10.2.1
	(b) number of electrons in an electrochemical reaction	10.2.2; 13.3.1
	(c) activity of an unspecified N^+ ion	10.5.2
	(d) refractive index	18.2.1
	(e) overtone order	21.2.1
n_0	refractive index of the medium from which a light beam comes to an optical fiber	18.2.1
n_1	refractive index of the waveguide core	18.2.1
n_2	refractive index of the waveguide cladding	18.2.1
n_{eff}	effective refractive index	18.6.2
Ox	oxidized form of a redox couple	10.2.2; 13.3.1
$_O$	subscript denoting quantities pertaining to the oxidized form of a redox couple	10.2.2; 13.3.1
P	reaction product	3.6.1
P	power of the transmitted light beam	18.3.1
P_e	dimensionless concentration of the reaction product in an immobilized enzyme layer (p_e/K_M)	4.2.5
P_0	power of the reference light beam	18.3.1
p	(a) concentration of a reaction product	4.2.1
	(b) partial pressure	10.2.2
p_e	concentration of the reaction product within an immobilized enzyme layer	4.2.1
$p_{e,0}$	concentration of the product at the transducer/immobilized enzyme layer interface	4.3.1
p_{CO_2}	partial pressure of carbon dioxide	10.17.4
pH	the negative logarithm (base 10) of hydrogen ion activity	
p_{H_2}	partial pressure of hydrogen	11.3.1
p_{O_2}	partial pressure of oxygen	10.17.2
$p_{m,i}$	product concentration at the membrane-enzyme layer interface	4.4.1
Q	(a) electrical charge	
	(b) quality factor of a resonator	21.2.7; 22.1.3
QY	fluorescence quantum yield	18.3.4
R	recognition receptor	
R	(a) ideal gas constant	
	(b) electrical resistance	9.1.1; 17.2.2
	(c) reflected light power	18.3.2
$_R$	subscript pertaining to the reduced form of a redox couple	10.2.2; 13.3.1
R_{Air}	electrical resistance of a resistive gas sensor in contact with pure air	12.2.2
Red	reduced form of a redox couple	10.2.2; 13.3.1
R_{et}	electron-transfer resistance	13.6.5; 17.2.3
R_{Gas}	electrical resistance of a resistive gas sensor in contact with analyte-containing gas	12.2.2
R_{ref}	resistance of a resistive gas sensor in contact with a reference gas	12.2.2

RH	relative humidity	17.9.1
R_S	electrical resistance of a resistive gas sensor	12.2.2
R_s	resistance of an electrolyte solution	13.2; 17.2.1; 17.2.3
R_1	resistance at the motional branch of the equivalent circuit of a TSM piezoelectric oscillator	21.2.2
r	(a) radius	
	(b) receptor concentration	6.4
r_h	relative humidity	17.9.1
r_T	the turnover number of the substrate conversion relative to that of the enzyme reoxidation	15.1.2
r_0	total receptor concentration	6.4
S	enzyme substrate	3.6.1
S	dimensionless concentration of the substrate (s/K_M)	15.1.3
S_e	dimensionless concentration of the substrate within an enzymatic layer under external diffusion control (s_e/K_M)	4.2.5
s	substrate concentration	3.6.1
s_e	substrate concentration in an enzymatic layer	4.2.1; 15.1.1
$s_{e,0}$	substrate concentration at the transducer/immobilized enzyme layer interface	4.3.1
$s_{m,i}$	substrate concentration at the membrane/enzyme layer interface	4.4.1
T	(a) absolute temperature	
	(b) transmittance	18.3.1
T_d	dew point	17.9.1
T_{xy}	shear stress	21.2.4
t	time	
Δt	change in the thickness of a TSM piezoelectric oscillator	21.2.3
t_Q	thickness of a TSM piezoelectric oscillator	21.2.1
t_r	response time	4.3.2
u	ion mobility	10.3.1
V	(a) voltage	
	(b) volume of an enzyme layer	4.2.2
V_{AC}	sine wave alternating voltage	7.1
V_D	drain voltage for a metal-insulator-semiconductor field effect transistor	11.1.4
V_{DC}	DC voltage	17.1
V_G	gate voltage for a metal-insulator-semiconductor field effect transistor	11.1.4
V_{FB}	flat-band voltage	11.1.3
V_m	amplitude of sine-wave alternating voltage	17.1
V_T	threshold voltage of a metal-insulator-semiconductor device	11.1.3
V_T^*	threshold voltage of an electrolyte-insulator-semiconductor device	11.2.1
v	(a) velocity	21.2.4
	(b) potential scan rate	13.7.4
	(c) reaction rate	3.6.1
v'	reaction rate within an immobilized enzyme layer	4.2.2
v_a	surface-normalized reaction rate in an enzymatic sensor	4.2.2
v_e	reaction rate of an electrochemical reaction	13.3.1
$v_{e,a}$	velocity of an anodic reaction	13.6.1
$v_{e,c}$	velocity of a cathodic reaction	13.6.1
v_M	reaction rate of enzyme regeneration	15.1.1
v_m	maximum reaction rate of an enzyme-catalyzed reaction	3.6.1
v_S	reaction rate for the formation of an enzyme–substrate complex	15.1.1
v_{tr}	propagation velocity of a transverse wave	21.2.3
v_V	volume reaction rate within an immobilized enzyme layer	4.2.2
v_x	velocity along the x-axis	21.2.7
v_C	reaction rate of the substrate conversion in an enzyme–substrate complex	15.1.1
w	microcantilever width	22.1.1
X_C	capacitive reactance ($1/\omega C$)	21.2.2
X_L	inductive reactance (ωL)	21.2.2
x	distance	
Δx	microcantilever deflection	22.1.2
Y	admittance	17.1

Y_t	total admittance	17.1
y	response signal of a sensor	1.5
z	ion charge	10.2.1
Z	(a) electrical impedance	17.1
	(b) acoustic impedance	21.2.7
$\|Z\|$	impedance modulus	17.1
Z'	real part of the acoustic impedance of a TSM piezoelectric oscillator	21.2.2
Z''	imaginary part of the acoustic impedance of a TSM piezoelectric oscillator	21.2.2
Z_C	capacitive impedance	17.2.1
Z_F	Faradaic impedance	17.2.3
Z_{im}	imaginary part of electrical impedance	17.1
Z_m	motional impedance	21.2.7
Z_{m1}	motional impedance of an unloaded TSM piezoelectric resonator	21.2.2
Z_{m2}	motional impedance produced by loading a TSM piezoelectric resonator	21.2.7
$Z_{m,t}$	total motional impedance	21.2.7
Z_{re}	real part of electrical impedance	17.1
Z_s	mechanical impedance of a TSM resonator	21.2.7
Z_t	total impedance	17.1
Z_W	Warburg impedance	17.2.1

Greek Symbols

Symbol	Meaning	Section References
α	(a) substrate modulus for an enzymatic sensor under external diffusion control	4.2.4
	(b) transfer coefficient of a cathodic electrochemical reaction	13.3.3
β	(a) Biot number	4.4.1
	(b) transfer coefficient of an anodic electrochemical reaction	13.6.1
Γ	surface concentration	5.2
Γ_{max}	maximum surface concentration	5.2
γ	(a) activity coefficient	10.2.1
	(b) enzyme reoxidation capacity relative to the substrate conversion capacity in the absence of any diffusion limitation	15.2.2
δ	(a) thickness of the Nernst diffusion layer	13.3.1
	(b) charge fraction transferred in the interaction of a polar molecule with a semiconductor	11.3.5
	(c) Debye length	12.1.7
δ_{dl}	thickness of the electrical double layer	17.2.2
δ_S	partition coefficient of the substrate	4.4.1
δ_P	partition coefficient of the product	4.4.1
ε_d	dielectric constant	13.5.2
ε_{dl}	dielectric constant within the electrical double layer	17.2.2
η	(a) Da_M/Da_S	15.2.2
	(b) overvoltage (difference between the actual electrode potential and the equilibrium potential)	13.6.5
	(c) dynamic viscosity	21.2.4
η_L	dynamic viscosity of a liquid	21.2.5
θ	surface coverage degree	5.2
θ_e	internal lag factor	4.4.1
θ_c	critical incidence angle	18.2.1
θ_m	external lag factor	4.4.1
θ_1	incidence angle	18.2.1
θ_2	refraction angle	18.2.1
Λ	molar conductivity	17.8.1
Λ_i	molar conductivity of an ion i	17.8.1