

ChE 455/555: Thermodynamics

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Objective/Introduction

- Study different aspects of thermodynamics in electrochemical cells
 - Thermodynamics can tell if a reaction is favorable
 - Favorable thermodynamics reaction does not mean favorable kinetically
 - Complete analysis of cell includes:
 - Thermodynamics
 - Kinetics
 - Mass transport

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Outline

Cell thermodynamics
Temperature and Pressure effects
Nernst equation
Pourbaix Diagram
Equilibrium constant
Reversible heat transfer

- Cell thermodynamics
- Temperature and Pressure effects
- Nernst Equation
- Pourbaix Diagram
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- Reversible heat transfer

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Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="text-align: center;">Cell Thermodynamics</h2> <hr/> <ul style="list-style-type: none"> • Meaning of potential $\Delta G^0 = -nFE^0$ <p>n : number of electrons</p> <p>F: Faraday's constant (96485 C/eq)</p> <div style="border: 1px solid black; padding: 2px; width: fit-content; margin-left: auto; margin-right: auto;">?</div> <p>Previously we used the definition of potential</p> <div style="border: 1px solid black; padding: 2px; width: fit-content; margin-left: auto; margin-right: auto;">Is this true?</div>
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Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="text-align: center;">Cell Thermodynamics</h2> <hr/> <ul style="list-style-type: none"> • Need to relate thermodynamic (reversible) potential to state variables • Electrochemical cells are treated at constant T and P • Consider closed system (transport of material between system and surrounding is not permitted) at constant T and P to start relationships
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Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="text-align: center;">Cell thermodynamics</h2> <hr/> <ul style="list-style-type: none"> • Using first law of thermodynamics (closed system): $\Delta U = q - w \quad (1)$ <ul style="list-style-type: none"> • The work can be associated with <ul style="list-style-type: none"> – Mechanical changes – Other sources: magnetic, surface, or electric work
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<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> • Mechanical contribution to work $w_p = P\Delta V \quad (2)$ • Total work <ul style="list-style-type: none"> – Mechanical – Electrical (w_e) $w = w_p + w_e \quad (3)$ <p style="text-align: right;">ChE 455/555 7</p>

<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> • For a reversible change at constant temperature, the heat transferred is given by $q = T\Delta S \quad (4)$ <p>ΔS: change in entropy</p> <p style="text-align: right;">ChE 455/555 8</p>

<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> • The canonical state variable for a system operating at constant T and P is the Gibbs free energy: $\Delta G = \Delta H - T\Delta S \quad (5)$ • The enthalpy change is given by: $\Delta H = \Delta U + P\Delta V \quad (6)$ <p style="text-align: right;">ChE 455/555 9</p>

<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> Combining Eqs. 1 to 4: $\Delta U = T\Delta S - P\Delta V - w_e \quad (7)$ <p style="text-align: right;">ChE 455/555 10</p>

<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> Substituting Eqs. 6 and 7 into 5 yields: $\Delta G = -w_e \quad (8)$ <ul style="list-style-type: none"> Reversible work, therefore it is the maximum work that can be obtained from the system <p style="text-align: right;">ChE 455/555 11</p>

<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> The maximum electrical energy available in an external circuit is equal to the number of charges multiplied by the maximum potential difference (reversible work): $w_e = nFE \quad (9)$ <p>By using: F in C/eq and E in V, your work will give you J</p> <p style="text-align: right;">ChE 455/555 12</p>

<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> Equating Eqs. 8 and 9: $\Delta G = -nFE \quad (10)$ <ul style="list-style-type: none"> Equation 10 demonstrates that we can obtain thermodynamics information from electrochemical measurements, and vice-versa <p style="text-align: right;">ChE 455/555 13</p>

<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> Because Gibbs free energy is a state function (it does not depend on trajectory) we can manipulate equations mathematically to obtain reversible potentials, e.g.: $\Delta G_1 + \Delta G_2 = \Delta G_3 \quad (11)$ <p style="text-align: right;">ChE 455/555 14</p>

<p>Cell thermodynamics</p> <p>Temperature and Pressure effects</p> <p>Nernst equation</p> <p>Pourbaix Diagram</p> <p>Equilibrium constant</p> <p>Reversible heat transfer</p>	<h2>Cell thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> Substituting Eq. 10 into 11: $-n_1FE_1 - n_2FE_2 = -n_3FE_3 \quad (12)$ <ul style="list-style-type: none"> Dividing by F and simplifying: $n_1E_1 + n_2E_2 = n_3E_3 \quad (13)$ <p>Eq. 13 is ALWAYS TRUE</p> <p style="text-align: right;">ChE 455/555 15</p>

Thought

- We said before:
 - You can balance the stoichiometry of the equation by multiplying by any positive constant
 - This operation does not alter the potential of the cell (potential is an intensive quantity, unaffected by the number of electrons)

Is this true?
Did Dr. Botte
lie before?

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Answer to thought

- Yes it is true because we were multiplying the individual reactions to eliminate the electrons from the total reaction
- However, Eq. 13 is still true
- See demonstration on the board

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Thought

- What happen when the final reaction involve electrons?
 - Answer:
 1. Choose the electrode reactions from the standard electrode potentials table
 2. Reverse the sense of the reactions according to your system
 3. Reverse the sign of your standard potentials
 4. Add the potentials using Eq. 13 to obtain the potential of the total reaction

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Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Cell thermodynamics: conclusions</h2> <hr/>
	<ul style="list-style-type: none"> • Eq. 13 is always valid • If your overall reaction does not involve electrons you don't need to correct the potentials (use procedure explain before) • If your final reaction has electrons involved you need to strictly use Eq. 13. <p style="text-align: right; font-size: small;">ChE 455/555 19</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Standard Potential and Gibbs free energy</h2> <hr/>
	<ul style="list-style-type: none"> • According to Eq. 10 the standard potential of the cell can be calculated from the Gibbs free energy • Gibbs Free energy for a reaction: $\Delta G = \sum_i s_i \Delta G_{i,f} \quad (13.1)$ <p style="font-size: small;">Where: s: stoichiometric coefficient (positive for products and negative for reactants) ΔG: free energy of formation. Information tabulated see Thermochemical data handout</p> <p style="text-align: right; font-size: x-small;">ChE 455/555 20</p>

<h2>Exercise 1</h2> <hr/>
<ul style="list-style-type: none"> • Solve Problem 2 of the book (Ch 3, p. 46) <p style="text-align: right; font-size: x-small;">ChE 455/555 21</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Temperature Effect</h2> <hr/>
	<ul style="list-style-type: none"> We can calculate the reversible electrode potential at other temperature by calculating the Gibbs free energy at a specified T and using Eq. 10 <p style="text-align: right;">ChE 455/555 22</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Temperature Effect</h2> <hr/>
	<ul style="list-style-type: none"> Consider a reversible process were only mechanical work is permitted, then the first law is $dU = TdS - PdV \quad (14)$ <p style="text-align: right;">ChE 455/555 23</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Temperature Effect</h2> <hr/>
	<ul style="list-style-type: none"> The enthalpy is defined by $H = U + PV \quad (15)$ <ul style="list-style-type: none"> A differential change in enthalpy is given by $dH = dU + PdV + VdP \quad (16)$ <p style="text-align: right;">ChE 455/555 24</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Temperature Effect</h2> <hr/>
	<ul style="list-style-type: none"> The Gibbs free energy is defined by $G = H - TS \quad (17)$ A differential change in Gibbs free energy is given by $dG = dH - TdS - SdT \quad (18)$ <p style="text-align: right;">ChE 455/555 25</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Temperature Effect</h2> <hr/>
	<ul style="list-style-type: none"> Combining Eqs. 14, 16, and 18 $dG = VdP - SdT \quad (19)$ Consider a process from state 1 to state 2, we can write Eq. 19 for each state as: $\Delta G = G_2 - G_1 \quad (20)$ <p style="text-align: right;">ChE 455/555 26</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Temperature Effect</h2> <hr/>
	<ul style="list-style-type: none"> At constant pressure Eq. 19 becomes: $\left(\frac{\partial \Delta G}{\partial T}\right)_P = -\Delta S \quad (21)$ Substituting Eq. 10 into Eq. 21 yields: $\left(\frac{\partial E}{\partial T}\right)_P = \frac{\Delta S}{nF} \quad (22)$ <p style="text-align: right;">ChE 455/555 27</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Temperature Effect</h2> <hr/>
	<ul style="list-style-type: none"> Using Eq. 22 we can calculate the effect of temperature on the reversible potential. Over a small temperature range a constant entropy change of reaction is usually justifiable, then integrating Eq. 22: $E_2 - E_1 = \frac{\Delta S}{nF}(T_2 - T_1) \quad (23)$ <p style="text-align: center;">ChE 455/555 28</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>From thermodynamics</h2> <hr/>
	<ul style="list-style-type: none"> The enthalpy, entropy, or Gibbs free energy of a reaction is given by: $\Delta M_R = \sum_i s_i M_i$ <p>Where:</p> <p>M: property (H, S or G)</p> <p>S: stoichiometry coefficient (positive for products and negative for reactants)</p> <p style="text-align: center;">ChE 455/555 29</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Temperature Effect: Important considerations</h2> <hr/>
	<ul style="list-style-type: none"> It is very important to include the phases in the calculation, that is, make sure that you read the properties of the compounds at the T physical stage <p style="text-align: center;">ChE 455/555 30</p>

Exercise 2

- In a fuel cell the overall reaction is given by $H_2 + 1/2O_2 \rightarrow H_2O$
- Write the electrode reactions
- Estimate the standard cell potential
- Calculate the change in reversible potential with temperature (mV/K) near room temperature
- What is the reversible potential of the cell at 35 °C.

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Pressure Effect

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Pourbaix Diagram
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- The change in the reversible potential with pressure can also be calculated from Eq. 19 by taking its derivate at constant temperature:

$$\left(\frac{\partial \Delta G}{\partial P}\right)_T = \Delta V \quad (24)$$

Change of volume in the reaction

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Pressure Effect

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- Substituting Eq. 10 into Eq. 24 yields:

$$\left(\frac{\partial E}{\partial P}\right)_T = -\frac{\Delta V}{nF} \quad (25)$$

- At low pressure the ideal gas assumption is valid, then:

$$\Delta V = \frac{\Delta nRT}{P} \quad (26)$$

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Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="margin: 0;">Pressure Effect</h2> <hr style="width: 20%; margin: 5px auto;"/>
	<ul style="list-style-type: none"> • Substituting Eq. 26 into Eq. 25 and integrating: $E_2 - E_1 = -2.303 \frac{\Delta NRT}{nF} \log \left(\frac{P_2}{P_1} \right) \quad (27)$ <ul style="list-style-type: none"> • Eq. 27 can be used at low pressures where the ideal gas assumption is valid <p style="font-size: small; margin-top: 10px;">ChE 455/555 34</p>

<h2 style="margin: 0;">Exercise 3</h2> <hr style="width: 20%; margin: 5px auto;"/>
<ul style="list-style-type: none"> • Calculate the reversible potential of the cell described in Exercise 2 at 3 atm <p style="font-size: small; margin-top: 10px;">ChE 455/555 35</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="margin: 0;">Thermodynamics Properties</h2> <hr style="width: 20%; margin: 5px auto;"/>
	<ul style="list-style-type: none"> • Thermodynamics properties can be obtained by measuring electrochemical potential, e.g., enthalpy can be calculated by $\Delta H = -nFE + nFT \left(\frac{\partial E}{\partial T} \right)_P \quad (28)$ <ul style="list-style-type: none"> • Entropy change of a reaction can be calculated by using Eq. 22 <p style="font-size: small; margin-top: 10px;">ChE 455/555 36</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="margin: 0;">Nernst Equation</h2> <hr style="width: 20%; margin: 0 auto;"/>
	<ul style="list-style-type: none"> • Walther Nernst developed an equation that correlates the voltage of the cell with its properties • To calculate the reversible potential at conditions different to standard we can use the Eq: $E = E^0 - \frac{RT}{nF} \ln \prod a_i^{s_i} \quad (29)$ <p>Where "a" is the activity coefficient of species "i"</p> <p style="text-align: right; font-size: small;">ChE 455/555 37</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="margin: 0;">Nernst Equation</h2> <hr style="width: 20%; margin: 0 auto;"/>
	<ul style="list-style-type: none"> • As an approximation we can ignore activity coefficient corrections and use concentrations in place of activities: $E = E^0 - \frac{RT}{nF} \ln \prod c_i^{s_i} \quad (30)$ $\frac{RT}{F} = 0.0257V \text{ at } 298 \text{ K}$ <p style="text-align: right; font-size: small;">ChE 455/555 38</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="margin: 0;">Assumptions when using Nernst Equation</h2> <hr style="width: 20%; margin: 0 auto;"/>
	<ul style="list-style-type: none"> • Neglects: <ul style="list-style-type: none"> – Activity coefficients – Potential that arise from bringing two different liquid phases into contact <p style="text-align: right; font-size: small;">ChE 455/555 39</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2 style="margin: 0;">Nernst Eq. Important considerations</h2> <hr style="width: 50%; margin: 5px auto;"/>
	<ul style="list-style-type: none"> • It can't be used to make T and concentration corrections simultaneously. • In such a case: <ul style="list-style-type: none"> – Use Eq. 22 or Eq. 25 to make T and P corrections, respectively – Applied Nernst Eq. at the new T or P.
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	<ul style="list-style-type: none"> • Assume activity coefficient of 1 for the following cases <ul style="list-style-type: none"> – Substances in excess (e.g., solvents) – solids
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	<ul style="list-style-type: none"> • Consists of plotting the potential vs pH • This type of diagram is useful because it allows identifying phases in equilibrium providing critical information on the behavior of the system • It was proposed by <u>Marcel Pourbaix</u>
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Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Procedure to build Pourbaix diagram</h2>
	<ul style="list-style-type: none"> • Write down all the equations involved in the system • Use Nernst equation • Express all reactions as a function of potential, pH, or both • Plot Potential vs. pH <ul style="list-style-type: none"> – Eqs. independent of pH are plotted as a horizontal line – Eqs. independent of potential are plotted as a vertical line – Eqs. dependent on both (pH & potential) are plotted as an oblique line <p style="text-align: right; font-size: small;">ChE 455/555 43</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Pourbaix diagram other assumptions</h2>
	<ul style="list-style-type: none"> • Typical assumptions <ul style="list-style-type: none"> – Concentrations of ions 10^{-6} M – Gases at 1 atm <p style="text-align: right; font-size: small;">ChE 455/555 44</p>

<h2>Exercise 4</h2>
<ul style="list-style-type: none"> • Build the Pourbaix diagram for the lead-water system (also known as le <u>Planté</u> battery). Take into consideration the following reactions <ul style="list-style-type: none"> $2H^+ + 2e^- \rightleftharpoons H_2$ $1/2O_2 + 2H^+ + 2e^- \rightleftharpoons H_2O$ $Pb^{+2} + 2e^- \rightleftharpoons Pb$ $PbO + 2H^+ \rightleftharpoons Pb^{+2} + H_2O$ $K=5.012 \times 10^{12}$ $PbO + 2H^+ + 2e^- \rightleftharpoons Pb + H_2O$ $E^0 = 0.25$ $PbO_2 + 4H^+ + 2e^- \rightleftharpoons Pb^{+2} + 2H_2O$ $E^0 = 1.45$ $Pb_3O_4 + 2H^+ + 2e^- \rightleftharpoons 3PbO + H_2O$ $E^0 = 0.97$ $3PbO_2 + 4H^+ + 4e^- \rightleftharpoons Pb_3O_4 + 2H_2O$ $E^0 = 1.127$ <p style="text-align: right; font-size: small;">45</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Equilibrium Constant</h2> <hr/>
	<ul style="list-style-type: none"> When the reversible potential equals zero, means that the driving force for the electrochemical reaction is zero, which represents the stable equilibrium state for the cell $E = E^0 - \frac{RT}{nF} \ln K \quad (31)$ <p style="text-align: center;">ChE 455/555 46</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Equilibrium Constant</h2> <hr/>
	<ul style="list-style-type: none"> Since the potential is zero at equilibrium, we can determine the equilibrium constant from the standard open circuit potential: $\ln K = \frac{nFE^0}{RT} \quad (32)$ <p style="text-align: center;">ChE 455/555 47</p>

<h2>Exercise 5</h2> <hr/>
<ul style="list-style-type: none"> Calculate the equilibrium constant for the <u>Daniell</u> Cell Zn/Zn⁺²/Cu⁺²/Cu <p style="text-align: center;">ChE 455/555 48</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Heat Transfer</h2> <hr/>
	<ul style="list-style-type: none"> • Because the system works at constant T and Pressure, we expect to see some transfer of heat to the medium • Heat transfer includes: reversible and irreversible heat (always negative, loss to ambient) • The reversible heat is given by $q = T\Delta S \quad (33)$ <p>If $\Delta S > 0$ the process is endothermic If $\Delta S < 0$ the process is exothermic</p> <p style="text-align: center;">ChE 455/555 49</p>

Cell thermodynamics Temperature and Pressure effects Nernst equation Pourbaix Diagram Equilibrium constant Reversible heat transfer	<h2>Heat transfer</h2> <hr/>
	<ul style="list-style-type: none"> • For a reversible process in an open system, the energy balance is given by: $-w_e = \sum_i \left(m_i \bar{G}_i \right)_{out} - \sum_i \left(m_i \bar{G}_i \right)_{in}$ <p>\bar{G}_i : partial molar quantity (includes mixing energy)</p> <p>Most of the time mixing energy is low compared to energy of reaction, then the change can be calculated based on pure components</p> <p style="text-align: center;">ChE 455/555 50</p>

<h2>Summary</h2> <hr/>
<ul style="list-style-type: none"> • Meaning of potential (related to Gibbs free energy) • For temperature effects use the entropy change (be careful with the physical stage of the reactants and products) • For pressure change use the volume change (include only gases in the calculation) • Use assumptions in Nernst Equation <p style="text-align: center;">ChE 455/555 51</p>

Summary

- You can't correct for T, P and concentration effects simultaneously
 - Correct for T and P
 - Correct for concentration effect
- Know how to build and interpret Pourbaix diagram
- Calculate equilibrium constants
- Calculate heat transfer in open systems
- What's the meaning of a positive and negative entropy change?

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