

Possible Entropy Decrease in Physical Chemistry

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Abstract: We proposed a possibility of entropy decrease due to fluctuation magnified and internal interactions in some isolated systems. Next, possible entropy decrease in various chemical reactions is researched. The opposite exothermic and endothermic reactions, the acid and alkaline reactions, etc., cannot all be entropy decrease. Third, we study possible entropy decrease in various phase transformations. Fourth, we search membrane and enzyme in chemistry and biology, in which membranes is namely Maxwell demon. Final, a special growth of diamonds from low pressure is discussed. Moreover, various known calculated results on entropy decrease are listed.

Keywords: Entropy; Physical Chemistry; Reaction; Phase Transformations; Membrane; Enzyme

1. Introduction

So far it is generally believed that entropy 'never' decreases in any 'natural' process when the system is in isolation.^[1] The entropy is a measure of the disorder and of the lack of information content.^[2]

But, chemists have long argued that the entropy increase corresponds to disorder, because it cannot be universally concluded whether the generator C-D is ordered or invalidated from the reactant A-B. The original single-phase super cooled liquid isolated system spontaneously solidifies into part of the crystal coexisting with the liquid at the solidification point temperature, which should tend to order. Such whether the entropy increase should be studied. In this paper, we further research possible entropy decrease in physical chemistry, which is mainly based on two classic tutorials.^[3,4]

2. Possible Entropy Decrease in Isolated System

The basis of thermodynamics is the statistics, in which a basic principle is statistical independence: The state of one subsystem does not affect the probabilities of various states of the other subsystems.^[1] We proposed that if various internal complex mechanism and interactions cannot be neglected, fluctuations and its magnified exist among various subsystems of an isolated system,

under some conditions entropy decrease,^[5,6] which includes physics,^[7-10] biology,^[11,12] astronomy^[13,14] and social sciences.^[15,16]

The chemical reactions are very complex, and include oscillation, condensation, catalyst and self-organization, etc., in which there are various internal interactions, and some ordering processes will be entropy decrease in some isolated systems.^[17] Catalyst as substance remained constant in the reaction may regard as an isolated system. When internal interaction exists, for example, the kinetic energy is transformed to the potential energy, order of this isolated system increases, and entropy decrease. The adsorption, fluctuations, self-assembly and the coarse-grained method etc, are discussed. Further, entropy decrease of macroscopic thermodynamics may be probably obtained in chemistry from the microscopic atomic, molecular and nano-theories, and entropy decrease in thermodynamics of microstructure is calculated quantitatively. We researched enzyme as biological catalyst and membrane, and general entropy decrease in biology. The change of entropy should be a testable science.^[18]

3. Possible Entropy Decrease in Various Chemical Reactions

Gibbs equation is:

$$dU = TdS - PdV + \sum_i \mu_i dn_i. \quad (1)$$

The electrochemical potential μ_i is related to U and electric potential $\phi = \pm Q / r$. Eq. (1) becomes:

$$dS = [dU + PdV - \sum_i \mu_i dn_i + \phi dQ] / T. \quad (2)$$

For the isolated systems U and V are invariance, there is chemical potential and electric potential, $dS < 0$ is possible. If the volume is smaller $dV < 0$, which correspond to the formation of stars and galaxy,^[14] and $dS < 0$. The formula (2) develops to a general chemical potential $\mu_i = \text{repulsive force} + \text{attraction}$, which is in favour with $dS < 0$.

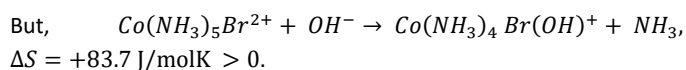
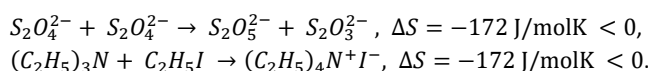
The known standard electrode potential of half-battery reaction in aqueous solution may be positive or negative. For $H_2(g) + 2AgCl(s) = 2Ag(s) + 2HCl(aq)$, $\Delta S = -93.71 \text{ J/molK}$.^[3]

Electrochemical reactions convert the chemical energy into electric energy. This can form isolated systems. They and the general chemical reactions are often reversible. Entropy decrease is possible for various different electrodes and batteries.

It is known

$$\Delta G = \Delta H - T\Delta S, \quad (3)$$

here ΔS is entropy of activation, $\Delta S = -137.6 \text{ J/molK} < 0$.^[4] For reactions in solution



Both cannot all be entropy increase.

Photosynthesis can be generally expressed as



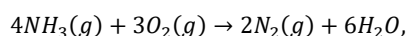
and $\Delta G = 477.0 \text{ kJ/mol} > 0$, $\Delta S < 0$. But, it has light into, so is not an isolated system, but a closed system.

For macromolecules various non-covalent interactions in protein molecules are static force, hydrogen bonds, hydrophobic bonds, VdW force.^[19]

The hydrophobic group CH_4 is $\Delta S = -175.3 \text{ J/molK} < 0$ at 298 K; the benzene is transferred to water at 291K, $\Delta S = -58.6 \text{ J/Kmol} < 0$.^[4] When the hydrophobic side chains are exposed to water as the helix unfolds, its result around a non-polar group creates an ordered water structure with entropy decrease. This effect may exceed generating unregulated coils to entropy increase. The total entropy change of the denaturation may actually be negative.

Elongated rubber, r increases, the polymer may have a reduced number of conformations. This result is similar to gas compression with entropy decreases. Most solid $S > 0$; some liquids $S < 0$.

In standard thermodynamics functions of reaction (5 chapter),^[3] the known



$$\Delta S = 2(45.77) + 6(16.71) - 4(45.97) - 3(49.003) = -139.1 \text{ cal/molK} < 0.$$

Matter states have different levels: 1. Gas, 2. Liquid. Sediment and other non-solution body are automatically separated. Ideal solution $\Delta S_{mixg} > 0$ corresponds to the ideal gas. Further, they are the non-ideal solution, the electrolyte solution. The interaction is stronger. 3. Solid.

One may argue that the mixing entropy ΔS_{mixg} of the solution is always positive at constant temperature, because it is intuitively felt that the solution is more disordered than its own independent pure components. Indeed, the contribution of ΔS_{mixg} for the volume increase of various components is always positive. But the contribution made by changes in molecular interactions can be positive or negative, such as 0.5 mol H_2O and 0.5 mol $(C_2H_5)_2NH$ mixed under 49C and 1atm, and the experiment derives $\Delta S_{mixg} = -8.8 \text{ J/K} < 0$.^[3] This can be explained by the stronger hydrogen bond between amine and water than the average strength of the hydrogen bond in the pure component. Of course, when mixing large amount of exothermic, usually think that the environmental entropy is bigger than ΔS_{mixg} , so the total entropy is positive. In chemical reactions, if entropy increases for the exothermic reactions, the endothermic reactions should be entropy decrease.

The mol entropy of gas is often higher than the entropy of liquid, and the mol entropy of liquid is often higher than the mol entropy of solid. The mol entropy often increases with the number of atoms in the molecule.

In the standard state $S_{298}^0 \text{ cal/molK}$ most are positive, but $CO_3^{2-}(aq)$ is -13.6; $Cu^{2+}(aq)$ is -23.8; $Fe^{3+}(aq)$ is -75.5; $OH^-(aq)$ is -2.57. Four are all aq .^[3]

The entropy of gas is significantly bigger than that of liquid or solid, and substances with similar molecular sizes have similar entropy. Therefore, for reactions containing only gas, pure liquid and pure solid, the symbol of ΔS depends on the change in the total mol number of the gas, which is negative $\Delta S < 0$, which is positive $\Delta S > 0$, which is zero ΔS is very small.^[3]

For $2H_2(g) + O_2(g) \rightarrow 2H_2O(l)$, it is -3,

$$\Delta S = 2(16.71) - 2(31.208) - 49.003 = -78 \text{ cal/molK} < 0.$$

For $2H_2S(g) + 3O_2(g) \rightarrow 2H_2O(l) + 2SO_2(g)$,

$$\Delta S = 2(16.71 + 59.30) - 2(49.16) - 3(49.003) = -93.31 \text{ cal/molK}.$$

For real gases (8 chapter)^[3] the compressibility factor $Z(P, T) = PV/RT$. Figure 8.1 in Levine I.N.^[3] for CH_4 , $Z - P$ are rises or falls.

In thermodynamics there are definitions, strict formulas, and different approximate formulas. Van der Waals equation

$$P = \frac{RT}{V-b} - \frac{a}{V^2}. \quad (5)$$

Redlich-Kwong equation [20]

$$\left[P + \frac{a}{V(V+b)T^{1/2}} \right] (V - b) = RT. \quad (6)$$

Shah and Thodos thought that it is the best two-parameter equation [21].

Virial state equation:

$$PV = RT \left[1 + \frac{b_1(T)}{V} + \frac{b_2(T)}{V^2} + \frac{b_3(T)}{V^3} + \dots \right]. \quad (7)$$

$$PV = RT[1 + a_1P + a_2P^2 + a_3P^3 + \dots]. \quad (8)$$

We may derive these corresponding entropy formulas.

There is interest in the difference between the thermodynamic properties of U, H, A, S, G, etc., on actual and ideal gases at a given temperature and pressure.^[22]

$$S_{id} - S = R \left[\left(a_1 + T \frac{da_1}{dT} \right) P + \frac{1}{2} \left(a_2 + T \frac{da_2}{dT} \right) P^2 + \dots \right]. \quad (9)$$

In solutions (9 chapter)^[3] the mol entropy S_i ,

$$\mu_i \equiv G_i = H_i - TS_i. \quad (10)$$

If $\Delta G_{m \times g}$, $\Delta H_{m \times g}$ are known, we may obtain $\Delta S_{m \times g}$ by

$$\Delta G_{m \times g} = \Delta H_{m \times g} - T\Delta S_{m \times g} \quad (11)$$

Figure 9.1 in Levine I.N^[3] V are rises or falls, entropy should also correspond to rises or falls.

For the non-ideal solutions (10 chapter)^[3] the activity coefficient $a_i = \gamma_i x_i$ determines the deviation from the ideal, and the corresponding chemical potential is:

$$\mu_i = \mu_i^0 + RT \ln \gamma_i x_i. \quad (12)$$

Figure 10.1(b) in Levine I.N^[3] G, H, S are all rises or falls for C_3H_6O , in which $T > 0$, so $S = (-1200 \text{ J/mol})/T < 0$, and is exothermic.

The pure state of the electrolyte is composed of ions. The crystals of NaCl, $CuSO_4$, MgS are composed of positive and negative ions. The corresponding charge is attraction or repulsion. The balance of the ion pairs generated in $Ca(NO_3)_2$ solution is:^[3]



$$dG = -SdT + VdP + n_A\mu_A + n_i\mu_i. \quad (14)$$

Gibbs-Duhem equation of electrolyte is:

$$n_A\mu_A + n_i\mu_i = 0 \quad (\text{For invariant } T, P). \quad (15)$$

Figure 10.3 in Levine I.N^[3] γ_i of HCl and $Mg(NO_3)_2$ are rises or falls. For electrolyte $\Delta S_{f,298}^0 [HCl(aq)] = -28.68 \text{ cal/molK} < 0$. But,

For $H^+(aq) + OH^-(aq) \rightarrow H_2O(l)$, $\Delta S_{f,298}^0 = 19.28 \text{ cal/molK} > 0$.

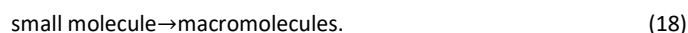
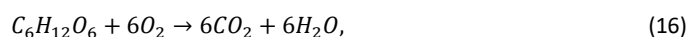
For $CO_3^{2-}(aq) + 2H^+(aq) \rightarrow H_2O(l) + CO_2(g)$, $\Delta S_{f,298}^0 = 81.4 \text{ cal/molK}$.

Some ions in aqueous solution, S_i^0 decrease when the absolute value of the charge increases, and S_i^0 also decrease when the ion size decreases. Because the Coulomb law is proportional to $|Q|/r^2$, so the strong electric field of small ions with high charge produces high hydration, which is larger, so the water order around the ion is also larger, and the entropy of solution is smaller. Because hydration produces strong order, some processes of salt dissolved in water have $\Delta S_{m \times g} < 0$.^[3]

In reaction equilibrium in non-ideal Systems (11 Chapter),^[3] many ion reactions in solution are acid or alkaline reactions. Are both entropy increases? The water molecules are amphotropic.

For $Ag_2SO_4(s)$, $SO_4^{2-}(aq)$, and $Ag^+(aq)$, $\Delta G_{f,298}^0 = -147.2, -177.3$, and 18.4 Kcal/mol , which may be positive or negative.^[3]

Biochemical coupling reactions are very important in biology, such as:^[3]



In biological competence the adenosine triphosphate (ATP) plays an important role, which is synthesis from adenosine diphosphate (ADP) and (H_3PO_4). It converts the energy of food into the chemical energy of cells, i.e., the oxidative phosphorylation, and the understanding of biological energy transference by the chemiosmotic theory of biosynthesis.



Hydrolysis of ATP to the action of ADP and P_i , $\Delta G^0 < 0$, which is favourable in thermodynamics. In life, this hydrolysis is coupled to thermodynamically unfavourable processes like the synthesis of biological macromolecules (such as amino acids, proteins, RNA and DNA) by small molecules. Chemical species transfer through active transmission from regions with low chemical potential to regions with high chemical potential. The oxidation of glucose and $\Delta G^0 < 0$ allows unfavorable reaction from ADP to ATP in thermodynamics. If $\Delta G^0 > 0$, $dS < 0$. This is the chemical interaction. In biochemical coupling, an enzyme catalyst allows reactions without two common species as $A + B \rightleftharpoons C + D$, $F + G \rightleftharpoons H$ to occur simultaneously $A + B + F + G \rightleftharpoons C + D + H$.

Figure 12.13 in Levine I.N^[3] the solid-liquid $T - x_A$ phase diagrams of $Cu - Au$ and $C_{10}H_{14}NOH$ have all rises or falls, and both are opposite direction. Figure 12.12 in Levine I.N^[3] the solid-liquid phase diagrams of $NaNO_3 - H_2O$ and $Li_2SO_4 - H_2O$ all have minima, and both sides may not be entropy increases. At this time, when placed in the Dova bottle, it is basically an isolated system.

Opposite adhesion and cohesion exist in the capillary. The surface layer on liquid has hydrophilic or hydrophobic effects. They cannot all be entropy increase.

For closed system

$$dU = TdS - PdV + \gamma dA. \quad (20)$$

For the isolated systems of two-phases U and V are unchanged, $\gamma > 0$ and $dA > 0$, so $dS < 0$.

In statistical mechanics (21 chapter),^[3] assume no interaction between the molecules in the system. This corresponds to the ideal state. The fluctuations are usually very small.

$$dS = d(U/T) + kd \ln Z. \quad (21)$$

In the isolated system N is necessarily invariance. Z is a partition functions, $S = U/T + k \ln Z$.

For translation, rotation, vibration and electronic states,

$$S = S_{tr} + S_{rot} + S_{vib} + S_{el}. \quad (22)$$

But this is no interaction. Further, for the ideal bimatom and polyatom gas the specific S are derived.^[3]

4. Possible Entropy Decrease in various Phase Transformations

In one-component phase equilibrium and surfaces (7 Chapter)^[3] the corresponding Clapeyron equation is:

$$\frac{dP}{dT} = \frac{\Delta S}{\Delta V} = \frac{\Delta H}{T \Delta V'} \quad (23)$$

$$\text{i.e. } \Delta S = \frac{dP}{dT} \Delta V. \quad (24)$$

V Increases $\Delta S > 0$; V decreases $\Delta S < 0$. Gravity, ions, strong interactions, etc., are volume decrease.

When liquid transforms to gas, and solid to liquid, general V becomes bigger, $dP/dV > 0$, But, Ga, Bi, in particular H_2O dissolved, V become smaller.^[3] If $dP/dV > 0$, $\Delta S < 0$.

The paramagnetism transforms to the ferromagnetism under a magnetic field, which is the electron spin orientation from disorder into order. This must be the entropy decrease. We should study the entropy of both subjects.

In electrochemical systems (13 chapter),^[3] the chemo sorption are bond formed and opposite bond breaks. Such ΔH can be positive or negative. Chemo sorption of gas on the solid $\Delta S < 0$, and is very large. Colloids include sedimentation, emulsion, gels, and have opposite hydrophilic and hydrophobic colloids. Both cannot all entropy increase.

The intermolecular attractions include hydrogen bonds, dipole-dipole, dipole-induced dipole, London dispersion energy, and the latter three are collectively called van der Waals force.^[3] They are all the 6th power of distance r. It may be simplified merger into the Lennard-Jones 6-12 potential energy:

$$E = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right]. \quad (25)$$

For this we discussed possible entropy decrease.^[5]

$$\ln Z = \ln Z_{id} + \ln Z_{con} - N \ln V. \quad (26)$$

Here Z_{con} is the configurationally partition function.

$$S = S_{id} - nR \left[\frac{1}{V} (b_1 + T \frac{db_1}{dT}) + \frac{1}{2V^2} (b_2 + T \frac{db_2}{dT}) + \dots \right] < S_{id}. \quad (27)$$

The formation of potential differences all leads to matter separation, such as Cu (-e) and Zn (+e).

We research various phase transitions that must be spontaneous isolated systems, such as overcooled, overheated spontaneous phase transitions.

5. Membrane and Enzyme in Chemistry and Biology

It is known:^[4]

$$\Delta S = nF \left(\frac{\partial \epsilon}{\partial T} \right)_P. \quad (28)$$

If $\left(\frac{\partial \epsilon}{\partial T} \right)_P < 0$, $dS < 0$. For half-cell reaction $\epsilon > 0$ or $\epsilon < 0$. Semi-reactions of certain organisms have $\epsilon > 0$ or $\epsilon < 0$. Membrane potential $\epsilon > 0$ or $\epsilon < 0$. It can be reversed when stimulated, but may not be an isolated system.

The catalytic action of enzyme is:^[4]



Here E and S are enzyme and substrate, ES is the enzyme-substrate complex, and P is the producer.

Each cell of an organism is surrounded by a bio-membrane that sensitively regulates the chemical flow between the cells and the environment. The formation of ATP by ADP is performed by enzyme.

Osmosis and membranes are very important in biology. In multicomponent phase equilibrium (12 chapter),^[3] both processes of the osmosis and the reverse osmosis phenomenon cannot all be entropy increase. The semi-permeable membrane has already been used for seawater desalination. It is an opposite direction of spontaneous flow, and active cells can transport chemical species from regions of low chemical potential through membranes to regions of high chemical potential. This is the active transport linked to the enzyme.

Maxwell demon is the well-known model of violating the second law of thermodynamics.^[23-25] The membrane is namely the Maxwell demon of biology.^[11,12]

Reaction kinetics discusses various reactions, such as catalysts and negative catalysts. The enzymatic catalysis occurs in biological tissues. For example,



here E is catalyst. Two-direction reaction cannot all be entropy increases.

In theories of reaction rates (22 chapter),^[3] for $CH_3Br + Cl^- \rightarrow CH_3Cl + Br^-$, $\Delta S^{0++} = -18.0 \text{ cal/molK}$.

6. Growth of Diamonds from Low Pressure

In physical chemistry it is known that the 1 mol liquid benzene solidifies at $T = 268.2\text{ K}$ and standard pressure, and is exothermic antipyretic process of 9874 J . The benzene melting point is known 278.7 K , the standard soluble heat is $H = 9916\text{ J/mol}$, the isopiestic heat capacity of liquid benzene is $C_p(l) = 126.8\text{ J/molK}$, and the isopiestic heat capacity of solid benzene is $C_p(s) = 122.6\text{ J/molK}$. This is an irreversible process of spontaneous isothermal solidification of super cooled liquids. The entropy change of the system is negative value $S = -35.42\text{ J/K} < 0$. While the exothermic process is not isolated system, must consider the environment. Environment is absorption heat, which is the entropy increase as $S = H/T = 9874\text{ J}/268.2\text{ K} = 36.82\text{ J/K}$.^[26] So the total entropy increases by $S = 1.40\text{ J/K} > 0$. But this is more ordered.

Polycrystalline phenomena of solids include the well-known graphite and diamond. It is known that diamond is smaller entropy than graphite, and are 2.4389 J/molK and 5.6940 J/molK at standard pressure and standard temperature (298.15 K), respectively, and the difference is 3.2551 J/molK , so the diamond is more ordered. In the low pressure the growth of diamond from graphite is a closed system, in which growth of diamond at low pressure gas phase and corrosion of graphite occur simultaneously. This is spontaneously ordered in a closed system. The initial super-equilibrium atomic hydrogen (SAH or H^*) in the closed system is formed by plasma excitation generated by microwave or radio frequency (RF) induction. In the process of hydrogen atoms corrosion of graphite, there is a spontaneous process of interaction, and the formation of diamond. Here graphite is stable phase; diamond is sub stable phase, so it is repeatedly considered a thermodynamic paradox, and violating the second law of thermodynamics, is called alchemy. Wang, et al., proposed the thermodynamic coupling model that can be used to explain the formation of diamond in low pressure.^[27] Coupling is the internal interaction. A transition occurs when the concentration of H^* reaches a certain critical value, and is high enough, graphite activation rises to a low pressure sub stable state of carbon, while diamond turns to a low pressure stable state. The role of H^* is to corrode graphite, referring the carbon atoms in graphite to a higher energy level, equivalent to a pump, i.e., interaction. The conversion of graphite into diamond is more orderly, and is necessarily an entropy decrease.

It is notable that SAH (H^*) at $T = 800\text{ K}$ $S = 179.36$ is bigger than $S = 179.00$ of hydrogen molecules; while at $T = 900\text{ K}$ $S = 181.82$ is smaller than $S = 182.51$ of hydrogen molecules.^[27] There is an inflection point.

The generation of a low-pressure diamond can be approximated as an isolated system.

In 1968-1976, B.V. Deryagin, B.V. Spitsyn, D.V. Fedoseev, et al., obtained experiments on growth of diamond by activating low pressure gas phase. In 1982-1987, K.M. Sato, N. Setaka, S. Matsumoto, et al., obtained the same results.^[28-31]

Even according to the Wang model^[26] entropy is not always increase, at least in the local process of low-pressure graphite change into diamond, $dG = 6.96\text{ kJ}$ must be the entropy decrease $dS < 0$.

7. Discussion

There are the critical temperatures T_c , the critical pressure P_c , the critical volume V_c , we predict that there is possibly the critical entropy S_c , which changes from entropy increase to entropy decrease.

Further, we should research general molecular-atomic thermodynamics, nuclear thermodynamics corresponding to strong interaction, quantum-particle thermodynamics, and continue to explore various thermodynamic quantities P, H, S , etc., and their formulations. We provided that there is entropy decrease in the evolutions of celestial objects,^[13,14] and should study the thermodynamics in high-energy astrophysics, such as neutron stars and black holes.

In a word, thermodynamics begins with the study of heat engines and heat. When energy expands to chemical energy, biological energy, nuclear energy, electromagnetic energy, gravitational energy and various microscopic, cosmic and complex phenomena, there is no need to be bound by the laws of thermodynamics, and change it to a belief.

Conflicts of Interest

The authors declare no conflict of interest.

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