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Chemical Applications, and Quantitative Calculations of Entropy Decrease in Chemistry

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ABSTRACT

First, based on basic problems in modern chemistry, some chemical applications are discussed. Next, we propose possible entropy decrease in isolated system is existence of internal interactions. Third, we research self-assembly and catalysis, etc., should be entropy decrease in chemistry. Final, we calculate quantitatively entropy decrease by the known formulas for some ordering phenomena. In chemistry entropy change will be very important tests and applications.

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Introduction

In modern chemistry the molecular structures include the valence bond theory, the molecular hybrid orbital theory and the supramolecule, etc [1,2]. The foundation of modern theoretical chemistry is quantum mechanics and statistical mechanics. Further, modern chemistry may use the Density Functional Theory (DFT) and is related with environment, material, information, energy and life, etc [3].

Special quantum chemistry can be combined with general quantum theory. The approximate exchange-correlation functional is reduced to the fifth order [4]. Stone discussed that intermolecular potential uses self-assembled membranes [5]. Klein, et al., searched large-scale molecular dynamics simulations of self-assembling systems and coarse grain [6]. Kroes studied frontiers in surface scattering simulation [7]. The chemical kinetics of molecules on metal surfaces is central to understanding catalytic processes. Non-elastic scattering of molecules on solid surfaces can accumulate or dissipate collision energy through internal vibrations [7]. The 2007 Nobel Prize in Chemistry was awarded to Ertl, who elucidated the mechanism of the Haber-Bosch surface catalytic process. Quantum dynamics of chemical reaction is researched. These are combination of quantum mechanics and molecular mechanics.

Qiu, et al., used the highly sensitive H atom Rydberg tagging time-of-flight method to observation of Feshbach resonances in the F+H2→HF+H reaction [8]. In this paper, we discuss some chemical applications, and propose possible entropy decrease in isolated system calculate quantitatively entropy decrease by the known formulas for some ordering phenomena.

Some Chemical Applications and Quantum Chemistry

Theoretically, Dirac equation and Schrödinger equation can describe all chemical processes, or provided some appropriate chemical conditions. This constitutes quantum chemistry, whose base is Schrödinger equation and its potentials. Quantum chemistry corresponds to stochastic statistics. One type of potential corresponds to electromagnetic interactions and Coulomb potential. When extended to d-dimensional systems, it relates to many-body systems and phase space. The equilibrium potential is V=c, whose special case is V=0, which connects to group theory and reveals new duality. A notable example of quantum chemistry is photosynthesis. The ultra-rapid ($\approx 10^{-15}$ sec.) conversion process of light energy remains unclear. It involves potentially the photoelectric effect and nonlinear tunneling.

In chemistry the four types of interactions are ionic, covalent, electronic, and molecular bonds, and share both attractive and repulsive forces. Consequently, certain aspects of particles can be applied to the composition of molecules and atoms. SU(2), u, d plus e (two quarks and a lepton), i.e., the three particles (p-n-e), are closely related to chemistry and macrophysics.

Gavai discussed the chemical potential on the lattice [9]. It is related with the molecular topology, in which the molecular structures become the chemical graph, whose key is the topological index.

Stochastic chemistry includes that it can be applied to chemical reactions, etc. For this several fundamental principles should first be established. The renormalization group method should also be applicable to chemistry with numerous degrees of freedom. In chemistry, general relativity and the equivalence principle may be tested. It is comparing chemical properties between gravitational fields and inertial forces, including their potential interactions with space chemistry experiments and equivalence groups.

Thermochemistry and Possible Entropy Decrease

Recently, the universality of the second law of the thermodynamics

is queried from various regions. In chemical reactions changes of entropy may increase or decrease. Since the second law of thermodynamics is based on an isolated system and statistical independence, when various internal complex mechanism and interactions exist among various subsystems of an isolated system, under some conditions entropy decrease, a state with smaller entropy (for example, self-organized structure) may appear. In these cases, the second law of thermodynamics should be different [10-15].

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 an isolated system [16]. 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. We discussed the adsorption, fluctuations, self-assembly and the coarse-grained method, etc. 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 [17].

We discussed fluctuations magnified due to internal interactions, etc., so that the equal- probability does not hold. In this case, the entropy with time would be defined as [12]

$$S(t) = -k\sum_{r} P_r(t)\,\mathrm{h}\; P_r(t) \qquad (1)$$

From this a possible entropy decrease is calculated in an internal condensed process. Internal interactions, which bring about inapplicability of the statistical independence, cause possibly entropy decrease in an isolated system. The possibility is researched for attractive process, internal energy, system entropy and nonlinear interactions, etc. An isolated system may form a self-organized structure.

We discussed entropy decrease under some cases [10-17], and one in which is calculated in an internal condensed process [13]. We proposed that ordering is the formation of structure through the self-organization from a disordered state, in which entropy must decrease [14].

Negative temperature is based on the Kelvin scale and the condition dU>0 and dS<0, we find that the negative temperature derives necessarily entropy decrease [12]. This is a fallacy, and contradiction with usual meaning of temperature and with some basic concepts of physics and mathematics [12]. In chemical reactions there are various internal interactions, so that some ordering processes with entropy decrease are possible on an isolated system. Further, we discussed the hysteresis as limit cycle, oscillation, catalyst and so on [16,17].

For any isolated system with internal interactions, we proposed that the total entropy should be extended to [10]

$$dS = dS^a + dS^i, \qquad (2)$$

where dS^a is an additive part of entropy and is always positive, and dS^i is an interacting part of entropy and can be positive or negative. Based on the Eq.(2), the sufficient and necessary condition of entropy decrease in isolated system will be expressed quantitatively [11]:

$$0 > dS^i > -dS^a, \quad \text{i.e.,} \quad |dS^i| > dS^a \quad (\text{for negative } dS^i). \qquad (3)$$

In usual cases, the condition corresponds to some stronger internal attractive interactions in isolated systems.

When we add entropy decrease, a complete formulation on entropy change should be the symmetrical structure:

$$Entropy \rightarrow \begin{cases} increase. \\ decrease \rightarrow \begin{cases} \mathrm{dS} = \mathrm{d_i}S + \mathrm{d_e}S. \\ dS = dS^a + dS^i. \end{cases} \end{cases} \qquad (4)$$

Here entropy decrease may be the dissipative structure for an open system, or be the internal interactions for an isolated system. In chemical reactions there are various internal interactions, so that some ordering processes with entropy decrease are possible on an isolated system. In general, any chemical reaction can take place in two opposite directions. The oscillatory of the non-markovian quantum relaxation may extend to various oscillations. The Belousov-Zhabotinski (BZ) reaction shows a period change automatically, at least a certain time.

We proposed a possibility of entropy decrease due to fluctuation magnified and internal interactions in some isolated systems [10-15]. The opposite exothermic and endothermic reactions, the acid and alkaline reactions, etc., cannot all be entropy decrease. We researched possible entropy decrease in various phase transformations, and searched membrane and enzyme in chemistry and biology, in which membranes is namely Maxwell demon. A special growth of diamonds from low pressure is discussed [18]. Moreover, various known calculated results on entropy decrease are listed.

The entropy decrease formula, analogous to the dissipative structure formula, can be applied to chemistry. Chemical cyclic reactions may exhibit entropy stability without necessarily being entropy cycles. Chemical oscillations such as BZ reactions have already challenged the thermodynamic principle that chemical processes tend unidirectionally toward equilibrium. There have various bidirectional natures of chemical reactions.

For macromolecule, the free energy of the ideal chain must obtain [19]:

$$dS = S(N,R) - S(N,0) = -\frac{3}{2}k\frac{R^2}{Nb^2}. \qquad (5)$$

So dS<0. The maximum number of conformations corresponds to the zero-end vector, and the number of conformations decreases with the increase of the end vector, leading to a decrease in the polymer's entropy and an increase in free energy. The free energy F(N, R) of the ideal chain increases with the square of the end vector R, indicating that the entropy elasticity of the ideal chain conforms to Hooke's law.

Various solvents exist in chemistry. When real chains are in the absence of heat or in good solvents, expansion occurs due to effective repulsive energy between units. The deformation of molecular chains during expansion leads to entropy decrease. Thus, the equilibrium between repulsive energy and entropy decrease

determine the conformation of real chains. Based on this principle, Flory theory can simply calculate the contributions of energy and entropy to free energy. The total Flory free energy of real chains is the sum of interaction energy and entropy contributions:

$$F = F_{\text{int}} + F_{ent} \approx kT(v\frac{N^2}{R^3} + \frac{R^2}{Nb^2}) \text{ . (6)}$$

The net attraction exists in the bad solvent, and the minimum value of free energy is located at R=0. The entropy and energy decrease with the decrease of R.

Hydrophobic substances typically exhibit higher surface tension as they repel water molecules, causing the liquid surface to contract. In contrast, hydrophilic substances prefer to interact with water, thereby reducing the liquid's surface tension. These properties are extensively studied and applied in biology, chemistry, physics, and related fields. When hydrophobic and hydrophilic substances react within the same system, their responses cannot both result in entropy increase; if one undergoes entropy increase, the other must experience entropy decrease.
We introduced θ temperature:

$$\theta = -\frac{1}{kb^3}\int_0^\infty U(r)r^2dr\,. \qquad (7)$$

Below the critical temperature θ, the polymer chains contract in poor solvents (v<0), whereas above this temperature θ, they expand in good solvents (v>0). Near the critical temperature

$v \approx b^3(\frac{T-\theta}{T})$, good solvents typically exhibit T>> θ, though their temperature θ becomes difficult to observe due to crystallization at low temperatures. Similarly, θ solvents cannot be heated significantly above this temperature θ, as they begin to boil at higher temperatures.

Self-assembly, Catalysis, and Entropy Decrease
Self-assembly is a phenomenon widely observed in nature, particularly in living systems, where ordered structures are formed through interactions between assembly units (molecules, nanostructures) [20].

Some liquids tend to form clusters under surface tension, a phenomenon commonly observed in daily life. These aggregated clusters hold potential applications. In the review by L. Mahadevan and D. Vella, such behavior is influenced by chemical interactions, force equilibrium, buoyancy, surface tension, and rotational torque, with the intensity and state of these effects being controllable.

Beyond the thermodynamic branch instability point, we may develop a novel tissue structure that links coherent spatiotemporal behavior with internal system dynamics (e.g., chemical reactions and convection). The formation and maintenance of self-organizing systems are results from nonlinear coupling between non-equilibrium constraints and molecular-level regulatory processes [21].

We proposed possible entropy decrease, which exists in transformations of internal energy, complex systems, and some new systems with lower entropy, nonlinear interactions, etc. Self-organization and self-assembly must be entropy decrease. Generally, much structures of Nature all are various self-assembly and self-organizations. We researched some possible tests and predictions on entropy decrease in isolated systems. They include various self-organizations and self-assembly, some critical phenomena, physical surfaces and films, chemical reactions and catalysts, various biological membranes and enzymes, and new synthetic life, etc. They exist possibly in some microscopic regions, nonlinear phenomena, many evolutional processes and complex systems, etc. As long as we break through the bondage of the second law of thermodynamics, the rich and complex world is full of examples of entropy decrease [22].

The discussion of entropy in chemical oscillations and BZ reactions suggests that neither may represent entropy increase. Dissipative structures can be incorporated into these processes. Entropy decrease should occur during the natural generation of chemical systems. Chemical oscillations like BZ reactions contradict the thermodynamic principle that chemical systems tend unidirectionally toward equilibrium.

In chemistry the free energy is:

$$\Delta F = \sum_{i=1}^{2} N_i\mu_i = Q - ST\,, \qquad (8)$$

$$\text{so} \quad S = (Q - N\mu)/T\,. \qquad (9)$$

Chemical potential flows from high to low. Their relation with the concentration is the free energy. If other quantities are fixed, entropy S will change periodically along to concentration N.

Pauling molecular complementarity theory suggests that small crystals like potassium hydrogen tartrate may spontaneously form in substances such as jam, potentially resulting in entropy decrease. Any spontaneous crystal formation likely involves entropy decrease. In 1952 A. Turing proposed Chemistry of Morphogenesis Theory, which posits that diffusion under specific conditions can disrupt the spatial uniformity of chemical reactions. When some reactants diffuse faster than others, the faster-diffusing species may inhibit the reaction, while the slower-diffusing species exhibit self-catalytic properties. This dynamic could spontaneously generate spatial morphologies from homogeneous phases, potentially occurring in isolated systems.

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. Moreover, enzyme as biological catalyst and membrane, and general entropy decrease in biology are researched. The entropy change should be a testable science [23].

Catalyzers become the way and the rate of chemical reactions. Some metal ligand polymers may be catalyzers. Catalysts are known to lower reaction barriers and alter the rate of chemical reactions. Most catalytic effects are surface-driven, which leads to discussions about catalysts and entropy decrease. Their interactions should be linked to entropy decrease. The addition or removal of catalysts invariably involves entropy increase or decrease

processes, as seen in phase-transfer catalysis. Furthermore, chemical reactions such as oxidation, reduction, and enzymatic reactions can be considered isolated systems. Enzymes, analogous to Maxwell's demon, exhibit specificity and selectivity.

Catalyst nanoparticles exhibit fluctuations and bistability [24]. Chemical stability and bistability as two kinetically stable regions coexist. Size effects and structural factors also play roles [25]. The mechanism of action is a complex process.

In chemistry the oxidation and reduction reactions are two opposite processes, and energy absorption and release are also opposite processes, and they may not always result in entropy increase. These phenomena should be associated with entropy increase or entropy decrease. Moreover, the different structures of matter, chemistry, and biomacromolecules lead to variations of energy and entropy.

Some Quantitative Calculations of Entropy Decrease in Chemistry and Microstructure

Homogeneous thermodynamics.

A mixture divided into different phases is termed heterogeneous, such as oil-water mixtures. They can separate spontaneously. Crystallization should result in entropy decrease. When two solvents are mixed and crystallize under isolation, entropy is decrease.

In the network model of gel and network, the entropy change of the chain caused by deformation is [26]:

$$dS = S(N,R) - S(N,R_0) = -\frac{3}{2}k\frac{(\lambda_x^2-1)R_{x0}^2 + (\lambda_y^2-1)R_{y0}^2 + (\lambda_z^2-1)R_{z0}^2}{Nb^2}. \quad (10)$$

As long as λ >0, then dS<0. The entropy change caused by network deformation is the central problem in rubber elasticity:

$$\Delta S_{net} = -\frac{nk}{2}(\lambda_x^2 + \lambda_y^2 + \lambda_z^2 - 3). \quad (11)$$

The single-axis deformation $\lambda_x = \lambda, \lambda_y = \lambda_z = 1/\sqrt{\lambda}$ replace (11), so

$$\Delta S_{net} = -\frac{nk}{2}(\lambda^2 + \frac{2}{\lambda} - 3). \quad (12)$$

When λ =2, so $\Delta S_{net} = -nk$ <0.

Melting Entropy

Usually, gas entropy>liquid entropy>solid entropy [27,28]. Such the coacervation from the supersaturated gas to the fluid should be a process of entropy decrease. For the entropy of hydrated ions when ions transition from the gas phase to the aqueous phase, their standard molar entropy decreases significantly; the entropy decrease is more pronounced for smaller ions or those with higher charges.

By Sackur-Tetrode equation it is derived:
S=(3/2)Rln *Mr* (mole mass)+108.745J/Kmol. (13)

Nanomaterial and the host-guest fabric possess big adsorption. They as an isolated system first are two matters, and are a metter after adsorption. The Langmuir-Blodgett films material is also a membrane. For these new matter entropy decrease is possible.
When gaseous ions dissolve into a solution, they interact with adjacent polar water molecules, and the dissolved electrolyte exerts a complex influence on the structure of water [29].

$Na^+(g) + Cl^-(g) = Na^+(ag) + Cl^-(ag)$, standard creation entropy ΔS_m =-186J/Kmol. (14)

$Li^+(g) + F^-(g) = Li^+(ag) + F^-(ag)$, standard creation entropy ΔS_m =-281J/Kmol. (15)

$Ca^{2+}(g) + S^{2-}(g) = Ca^{2+}(ag) + S^{2-}(ag)$, standard creation entropy ΔS_m =-374J/Kmol. (16)

Here (*ag*) is solution.

For non-aqueous solvents, such as liquid ammonia; entropy is: 2 x 130.57+205.03→2 x 188.715(gas phase), 2x 69.91(liquid phase), i.e., 466.17>377.43 >>139.82.

For the Melting Entropy, it Encompasses Three Scenarios

- Gas dissolution in liquids
- Mixing of two liquids (similar to gas-gas mixing)
- More generally, chemical reactions (a special case is BZ reaction)

Periodic changes indicate entropy increase or decrease, while regular patterns suggest order, thus entropy should decrease. It is known that ion entropy decreases with radius reduction, particularly with increasing charge. Therefore, when dissolved ions become smaller or carry higher charges, the dissolution entropy becomes more negative [29]. When anions and cations generated during dissolution carry three charges, they exhibit extremely negative dissolution entropy. Even single-charge anions show very negative dissolution entropy [29]. This phenomenon is especially pronounced in ionic salt dissolution. Notably, dissolution entropy can be positive, such as KCl, KBr, KI, KNO_3, K_2SO_4; or be negative, such as MCO_3-150J/Kmol (hard dissolve), $LaPO_4$, below -400. One of the two dissolution processes should be an entropy decrease.

Solid Entropy

For solid entropy, in 1952 Latimer proposed S=(3/2)Rln A_r -5.9J/Kmol. For crystal enthalpy of sublimation, it changes ΔS_L=+112.6(a+c)J/Kmol. It is positive, but it condenses into negative.

In Reaction Table 3[29], ΔS_m is generally positive, such as $Ce^{3+} + SO_4^{2-}$, 130J/Kmol; but $Ce^{3+} + ClO_4^-$ is negative -130J/Kmol. While

$$CeClO_4^{2+}(ag) + SO_4^{2-}(ag) \rightarrow CeSO_4^+(ag) + ClO_4^-(ag), \quad \Delta S_m = 260 J/Kmol. \quad (17)$$

The cause of chelation effect can be mainly attributed to entropy change.
For nonmetal

$$2CI_2 + 2H_2O = 4H^+ + 4CI^- + O_2, \quad \Delta_r G_m = -49.9 kJ(mol)^{-1}. \quad (18)$$

$$2Br_2 + 2H_2O = 4H^+ + 4Br^- + O_2, \quad \Delta_r G_m = +62.9 kJ(mol)^{-1}. \quad (19)$$

$$2I_2 + 2H_2O = 4H^+ + 4I^- + O_2, \quad \Delta_r G_m = +267.6 kJ(mol)^{-1}. \quad (20)$$

For HF, HCI, HBr and HI, $T\Delta_r S_m = -29 kJ(mol)^{-1}$, -13, +4.0 and +4.0. Because T>0, so $\Delta_r S_m$ <0 and $\Delta_r S_m$ >0.

On the strength of inorganic acids in aqueous solutions [29], the series of hydrogen halides HX (where X = F, Cl, Br, and I).

$$HX(ag) \rightarrow H^+(ag) + X^-(ag). \quad (21)$$

During the dissociation of HX, HI exhibits entropy increase, while the other three species show entropy decrease, with HF demonstrating the most significant entropy decrease and the most negative ΔS_m. This phenomenon is primarily attributed to the smallest radius of F^- (ag), and the hydrogen bonding interactions between water molecules in the solvent, which restrict its translational entropy and constrain the translational and rotational entropy of the hydrated molecule. Typically, the first-order dissociation of an inorganic acid corresponds to an entropy decrease of -85J/Kmol, the second-order dissociation to -125J/Kmol, and the third-order dissociation to an even more negative entropy change. Additionally, in Appendix 5 of [29], the entropy of the general substance S is generally positive, but many cases show negative values. These include ag, all 3+ and 3-, some 2, and one F^- (ag).

In chemical thermodynamics, the entropy of formation is a variant with pressure. In general, any chemical reaction can take place in two opposite directions. The oscillatory of the non-markovian quantum relaxation [30] may extend to various oscillations. BZ reaction shows a period change automatically, at least a certain time.

Discussion and Summary

In the self-organization processes of chemistry and biology, certain isolated systems can spontaneously evolve toward ordered states. These phenomena demonstrate that nature does not always tend toward disorder, and certain ordering processes are accompanied by entropy decrease. Our conclusion is that chemical reactions are complex processes. When isolated systems contain various internal interactions and fluctuations: 1). We should examine whether all intermediate changes from initiation to termination constitute continuous entropy increase processes. 2). The principle of entropy increase may not always hold true for these processes. 3). The entire universe does not always tend toward disorder. At present entropy increase as a new world view [31] already becomes a conviction and a scientific alienation.

For more complex organic chemistry, we should explore its possible entropy decrease. A single carbon atom can only form one molecular structure; ten carbon atoms may generate 75 molecular configurations; 20 carbon atoms could produce 366,319 molecular variants; and 40 carbon atoms form 62 trillion possible configurations. The combination of long hydrocarbon chains with oxygen atoms produces alcohols, aldehydes, and ketones. Moreover, molecular diversity increases exponentially with structural complexity [32]. Additionally, we will investigate potential entropy decrease in chlorophyll under solar energy exposure, and examine correlations between entropy decrease and chemical bonds as well as magnetization orientation. Beyond Coulomb interactions, exchange energies and association energies (related to van der Waals forces and dispersion forces) also play significant roles.

Dark matter and dark energy are basic focus in astronomy, astrophysics, cosmology and total science. Based on the known phenomena of dark energy and dark matter, since 2007 we proposed the negative matter unified dark matter and dark energy [32-37]. This is repulsion between positive matter and negative matter, both form two different regions of topological separation, which forms invisible dark matter, and repulsion is dark energy. It agrees on Occam's Razor. It is not only the simplest, and may be calculable, observable and testable, and may be mechanism of inflation. Perhaps the current astronomical data can already prove the existence of negative matter, such as some amplifying Einstein rings. If the negative matter is confirmed by some observatories, it will open up a new world. Chemistry of negative matter should be the same, only whose mass is negative. It will compose atoms by negative mass, and form molecules, which consequently exhibit similar chemical compositions and reactions.

In a word, many experiments and theories show that the second law of the thermodynamics should be developed. Entropy decrease is possible when there are internal interactions in an isolated system [10-15]. In chemistry entropy change will be very important tests and applications.

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