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NONLINEAR CHEMISTRY, AND POSSIBLE ENTROPY DECREASE IN PROCESS ENGINEERING

BY

***Yi-Fang Chang**

*Department of Physics, Yunnan University, Kunming, 650091, China

Abstract: First, based on some basic problems in modern chemistry, the nonlinear chemistry is discussed. Next, we propose entropy decrease in isolated system of chemistry, and calculate quantitatively entropy decrease by the known formulas for some ordering phenomena. Third, we research possible entropy decrease in process engineering, as two examples Dujiangyan (都江堰) and Yuenyang Terrace (元阳梯田) in China are exemplary models of sustainable development. Final, chemistry and process engineering do not always tend toward disorder, entropy decrease is possible.

Key words: chemistry, nonlinearity, entropy decrease, calculation, process engineering.

1. Introduction

The foundation of modern theoretical chemistry is quantum mechanics and statistical mechanics. Stone researched that intermolecular potential uses self-assembled membranes [1], and quantum dynamics of chemical reaction. Klein, et al., studied molecular self-assembly and coarse grain [2]. 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 [3].

Based on engineering thermodynamics, we researched that entropy as a new world view may be a key guideline for engineering and design, and we proposed a concrete form: it is a continual optimization process, and corresponds to entropy of each stage constant decrease. From this we discuss “negative temperature” fallacy [4], which will derive impossible efficiency. Further, the knowledge economics and social sustainable development are searched, which are determined by engineering, design and entropy decrease [5]. In this paper, we research the nonlinear chemistry, and possible entropy decrease in chemistry and process engineering.

2. Nonlinear Chemistry, Quantum Chemistry and Their Developments

In *The Princeton Companion to Mathematics (III)* there have chemistry and mathematics [6]. The phase law of chemical systems is P+F=C+2, where P represents the number of phases, F denotes the number of degrees of freedom, and C indicates the number of components. It mirrors the relationship between vertices, edges, and faces in polyhedra. This principle forms the basis of chemical topology, with both concepts being mutually corresponding and scalable. Various isomers can be analyzed using generating functions and permutation groups for synthetic studies; molecular Mobius strips may also be formed. Macromolecules include DNA, which involves biochemical concepts such as winding numbers, twisting numbers, and writhing numbers derived from knot theory. This book discussed also the Belousov-Zhabotinski reaction and nonlinear chemical kinetics.

Based on the nonlinear mathematics [7], which included thee nonlinear geometry, analyze and algebra, we discussed the general nonlinear methods and various nonlinear physics, and searched the general nonlinear sciences, in instance, nonlinear astronomy, biology, Earth science, and social sciences, etc. In particular, we researched chaos and fractal and their applications [8].

Chemistry studies typically many-body systems, which are inherently nonlinear, particularly in the context of many-particle interactions [9,10]. For many-electron systems, Hartree-Fock Hamilton operator is:

$$H_0=\sum_{i=1}^{n} f(i). \qquad (1)$$

Here *f(i)* is operator of electron i. Hartree-Fock-Roothaan matrix equation is:

$$\sum_{\gamma} F_{\mu\gamma} C_{\gamma i} = \varepsilon_i \sum_{\gamma} S_{\mu\gamma} C_{\gamma i} \text{ (i=1,2,…k).} \qquad (2)$$

This must be nonlinear. Gray and Scott introduced non-linear phenomena in chemical kinetics using simple model schemes, which includes oscillations and instabilities, etc [11].

Chemical reactions with nonlinear kinetic behavior can give rise to a remarkable set of spatiotemporal phenomena. These include periodic and chaotic changes in concentration, traveling waves of chemical reactivity, and stationary spatial (Turing) patterns. Now the existence and the relevance of these phenomena are searched, in characterizing, designing, modeling, and understanding them. Several nonlinear dynamical phenomena in chemical systems provide simpler analogues of behaviors found in biological systems [12].

Saguésa, et al., presented some of the key results of nonlinear chemical dynamics, and discussed the range of phenomenology from chemical oscillation to chaos to waves and pattern formation, as well as experimental methods, mechanistic considerations, theoretical techniques, and the results of coupling and external forcing [13].

The mathematical models on chemical reactions dynamics have Lotka-Volterra model, Brusselator, Belousov-Zhabotinskii reaction [14,15] and so on. They may exhibit the equilibrium point, the oscillation, the limit cycle, chaos, etc.

In the non-equilibrium state, both the chemical clock (concentration periodic oscillation) and biological clock of chemical reactions correspond to nonlinear theory, exhibiting the regression property of solitons. Various natural clocks also demonstrate nonlinearity.

The nonlinear chemistry includes the nonlinear chemical phenomena, the chemical chaos [16,17] and fractal structures, the complex networks [18-20], etc. We must develop various chemical theories, for instance, chemical bonds exhibit nonlinear behavior, and nonlinear chemical kinetics, etc. Chaos can be utilized in the study of biological systems as a whole, macromolecules, and similar contexts.

Theoretically, Dirac equations and Schrödinger equation can describe all chemical processes, or provided some appropriate chemical conditions. This constitutes quantum chemistry, whose base is Schrödinger equation and potential energy. Quantum chemistry corresponds to stochastic statistics. The potential energy encompasses 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.

Nonlinear interactions may derive two results: 1) Various structures, as dissipation structures, self-organization, cooperation, soliton, fractal, hypercycle and so on. 2) Various changes, as phase transformation, bifurcation, chaos, catastrophe and so on. Both must be combined, world may form various complex nonlinear structures. Chaos is very important in dynamical systems [21,22].

Stochastic chemistry includes that it can be applied to chemical reactions, and 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.

3. Entropy Decrease in Chemistry, and Some Quantitative Calculations

So far, general people have believed that entropy must increase in isolated systems. But, its preconditions are: (1) Various internal interactions in the system must be neglected, i.e., it has the statistical independence and the additivity of entropy. (2) They must be thermal equilibrium systems. We proposed that if interactions exist among various subsystems of an isolated system, in which entropy decrease is possible [23,24]. They include physics, chemistry [25-28], astronomy, geoscience, biology, and social sciences, etc. An isolated system may form a self-organized structure with lower entropy for these cases of attractive processes, internal energy, system entropy, nonlinear interactions, etc.

For any isolated system with internal interactions, we proposed that the total entropy should be extended to [24]

$$dS = dS^a + dS^i, \quad (3)$$

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.(3), the sufficient and necessary condition of entropy decrease in isolated system will be expressed quantitatively:

$$0 > dS^i > -dS^a, \text{ i.e., } |dS^i| > dS^a \text{ (for negative } dS^i\text{)}. \quad (4)$$

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

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.

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

$$Entropy \to \begin{cases} increase. \\ decrease \to \begin{cases} \mathrm{dS} = \mathrm{d_i}S + \mathrm{d_e}S. \\ dS = dS^a + dS^i. \end{cases} \end{cases} \quad (5)$$

Here decrease of entropy may be the dissipative structure for an open system, or be the internal interactions for an isolated system.

Generally, the entropy increase law possesses relativity and approximation, because it must be an isolated system, but which always dependent of some conditions. Complete exact isolated system does not exist in essence, since the gravitational interaction cannot be screened completely. Change of entropy must consider relations, structures, time and evolution, etc.

In various phases and phase transitions, the most suitable for entropy increase is only the special gas diffusion process, and the nebula may be condensed into stars, liquid has also the capillary phenomenon. When internal interactions exist, entropy decrease is possible. Entropy increase or entropy decrease should be symmetrical and beautiful. This can derive the complexity of natural evolution and social phenomena. If the world is always entropy increasing, it is not only pessimistic, but also lack of basic beauty and complexity.

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 [24]

$$S(t) = -k\sum_r P_r(t)\ln P_r(t). \quad (6)$$

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 decreases 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.

The chemical reactions are very complex, and include oscillation, condensation, catalyst and self-organization, etc. In these cases 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, a state with smaller entropy (for example, self-organized structure) may appear. In these cases, the statistics and the second law of thermodynamics should be different [23-28]. A negative temperature is based on the Kelvin scale and the condition dU>0 and dS<0, we found that the negative temperature derives necessarily entropy decrease. This is a fallacy, and contradiction with usual meaning of temperature and with some basic concepts of physics and mathematics [4]. We proposed quantitatively the sufficient and necessary condition of entropy decrease in isolated system. If fluctuations are magnified due to internal interactions in the system, entropy will decrease possibly. 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 [25].

We calculated some quantitative results of entropy decrease in chemistry, which includes homogeneous thermodynamics, melting entropy, solid entropy and so on [29]. An example is that 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 [30].

$Na^{+}(g)+Cl^{-}(g)=Na^{+}(ag)+Cl^{-}(ag)$, standard creation entropy ΔS_m=-186J/Kmol. (7)

$Li^{+}(g)+F^{-}(g)=Li^{+}(ag)+F^{-}(ag)$, standard creation entropy ΔS_m=-281J/Kmol. (8)

$Ca^{2+}(g)+S^{2-}(g)=Ca^{2+}(ag)+S^{2-}(ag)$, standard creation entropy ΔS_m=-374J/Kmol. (9)

Here (*ag*) is solution.

Some possible entropy decreases are calculated quantitatively [24,29]. We proposed quantitatively a total formula of the entropy change for the universal evolution of any natural and social systems. 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 [23-29].

Strogatz discussed systematically that nonlinear dynamics and chaos and their applications in physics, biology, chemistry and engineering [31]. The analysis for many experiments and theories shows that the second law of the thermodynamics should be developed. We researched some possible tests for entropy decrease in isolated systems for physics, chemistry, biology and so on. It is confirmed by many stable states in Nature.

Further, we should research entropy decrease in more complex organic chemistry. A single carbon atom can only form one type of molecule; ten carbon atoms can yield 75 distinct molecules; twenty carbon atoms can generate 366,319 molecules; and forty carbon atoms result in 62 trillion possible configurations. When long hydrocarbon chains combine with oxygen atoms, they produce alcohols, aldehydes, and ketones [32]. Moreover, the number of molecular varieties increases more rapidly with increasing molecular size.

For entropy decrease and chlorophyll action under the solar energy, the relations between entropy decrease and chemical bonds as well as magnetization direction must be studied. In addition to Coulomb energy, there are also exchange energy and correlation energy (related to van der Waals forces and dispersion forces).

4. Possible Entropy Decrease in Process Engineering

Self-assembly is a phenomenon widely observed in nature, particularly in chemical and living systems, where ordered structures are formed through interactions between assembly units (molecules, nanostructures) [33].

The concept of entropy increase has been pushed to its extreme as a worldview [34], which assume that entropy always increase regardless of underlying conditions. At any stage of a complex system's evolution, it can function not only as an isolated system but also exhibit entropy decrease. For instance, the management process inherently demands entropy decrease [35].

The entropy of the Universe increases in all natural processes, and isolated systems tend to greater disorder [34]. Although the total entropy for whole system is positive and increase possibly, but, so long as different entropy states for any systems exist, entropy should decrease in transformation process from a higher entropy state to a lower entropy state (in Fig. 1 from A to B), for example, from chaos to order, from war to peace and so on [36,37]. If this system is isolated, it will correct and develop the second law of thermodynamics.

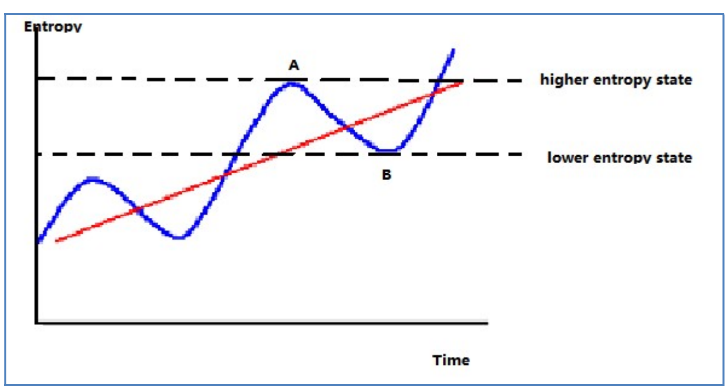

Fig.1. Transformation processes between states with higher entropy and lower entropy

According to current theory, all processes tend spontaneously toward disorder; therefore, any process engineering inherently involves management. Management is defined as the process of administering and controlling the organization, and its nature, type, structure, and size. It is an act of creating and maintaining such a system in which the members of the organization can work together, and achieve the objectives efficiently and effectively. Management acts as a guide to a group of people working in the organization and coordinating their efforts, towards the attainment of the common objective [38]. This is a classical internal interaction in isolated systems.

Characteristics of management include seven aspects, in which management forms supernatural force as the internal attraction to create an orderly party. If internal interactions exist in isolated systems, entropy decrease will be possible for this system. From this possibility we discuss management as a typical mode, which includes much internal adjustment and control, and through them to achieve order, more efficient, competitive, and better survive and develop. A key of management is to change the natural tendency disorganized with entropy increase and build the best system with entropy decrease. It is the thermodynamic meaning of management. Further, this theoretical mode may be extended to ecology, biology, sociology, engineering, etc. The naturally formed and artificially intervened ecosystems are through interaction and regulation within the systems to reach the dynamic balance, order and entropy decrease. These systems have constant evolutions and go optimization processes with order and entropy decrease. In many social systems and the human society there are usually some no equilibrium dynamic processes, which are accompanied by order and entropy decrease [35].

Chinese traditional agriculture has formed a recycling economy. The crop rotation and interplanting may preserve fertility of soil, and realize the agricultural sustainable development. hinese traditional country forms a complete recycling system on farm-mulberry-herd-fish. Chinese traditional house used grass as roof and land as wall are easy for regression to nature. Our clothing is cotton, our house is a structure of land-tree-grass, they may be used as manure, and recur to land. Such a complete recycle is formed by land→farm produces→human→wastes→land, and it is a basis of Chinese existence and development through thousands years [39]. This may provide a paradigm as reference for the sustainable development of world.

Fallen flowers are not merciless and useless
They transform manures, and enrich flowers.

It is a poetic recycle.

In China Dujiangyan (都江堰) and Yuenyang Terrace **(元阳梯田)** as two World Cultural Heritages are all exemplary models of sustainable development.

Dujiangyan is located in the west of Chengdu Plain on the Minjiang River. This was originally a region with severe water and drought disaster. At the last years of Qin Zhao King (about 256 BC), Li Bing (李冰) and his son organized on the basis of predecessors to construct Dujiangyan, which is a large-scale comprehensive water conservancy engineering integrating flood control, irrigation and shipping (Fig.2).

Fig.2 Dujiangyan(都江堰)

Dujiangyan is mainly composed of three main projects: fish mouth (鱼嘴), flying sand weir (飞沙堰) and Aquarius mouth (宝瓶口). The three organically cooperate, and restrict each other, coordinated operation, water diversion and irrigation, flood diversion and disaster reduction. The fish mouth is a stone ridge made of stones in the Minjiang River. It is a water-divided construction project that divides the water flow of the Minjiang River into two. The east is inner river (内江) for irrigation canal; the west is outer river (外江) as the current of Minjiang River. During construction, the inner river riverbed is lower than outer river. In autumn and winter, 60% of the water flows into inner river and 40% into the outer river; during spring and summer, 60% of the water and 40% into outer and inner rivers. It can achieve the purpose of drainage.

The function of flying sand weir is mainly used for sand discharge. In summer, the Minjiang River roses, fish mouth flooded, it became the second diversion. This is used the curve circulation and concave flushing and convex silt principle, a small amount of sediment water flows into inner river, and a large number of sand and gravel water flows into outer river. The water into the inner river will be clearer than that of outer river. And when the rainy season water flow reaches a certain amount, the excess water will flow from the weir to outer river.

The most important role of Aquarius mouth is "drainage". It is controlling inlet flow, which may ensure that the water into the Chengdu Plain is appropriate. Inner river enters the irrigation system covered above the western Sichuan plain.

Historically, the constructions of Dujiangyan take local materials. The building materials are mainly bamboo, wood and pebbles, which are widely used in water conservancy construction practices such as water diversion, embankment protection, rescue and blocking, forming the unique traditional hydraulic technology of Dujiangyan. For more than 2000 years, Dujiangyan management has formed a unified standard format (Fig.3).

Fig.3 "Three-word classics" on water control of Dujiangyan

Dujiangyan is not only a water conservancy hub and an engineering system, but also a cultural embodiment on the unity of nature and man. Dujiangyan applies mountain terrain, and persists in the guidance rather than hinder confrontation, and adopts the traditional weir engineering technology, and avoids the destructive transformation for environment. This is based on the natural construction way, and makes unified engineering and natural environment, and become true-good-beauty as one of the human wisdom crystallization.

According to current conditions, taking the advantage of the situation and taking measures are the practical principle of water control culture in Dujiangyan. Dujiangyan follows the law of water from the Minjiang River, and the law of water and sand movement, and the law of river evolution, and grasps the natural geography and seasonal characteristics. The water system in the irrigation area is interwoven and interconnected, and shows the harmonious coexistence of man and water across time and space.

The water control concept and ecological wisdom of Dujiangyan follow the “harmonious coexistence between man and nature”, and this is the successful application of water conservancy engineering and management technology, and also the vivid practice on the wisdom of the unified man and nature. In 2020 *Water Conservancy in China* (Nov. 3) some experts published many results for the commemorate 2275th anniversary of Dujiangyan.

Dujiangyan is a world cultural heritage. Built in 256 BC, it is the oldest, only retained and grand water conservancy project characterized by dam-free water diversion in the world. Now there are many cultural relics near Dujiangyan, and form a beautiful tourist resort.

Yuenyang Terrace(元阳梯田) located in the south of Ailao Mountain in Yuanyang County of Yunnan. This is a wonder of terraced farming civilization created by people of all nations, mainly Hani, using special geography and climate. It is on a large scale and magnificent momentum. Terraced fields change with the terrain and take measures according to local conditions, while the slope is large, small slope and small fields, and even in the stone gap under the ditch. Therefore, the terraced fields are several mu (亩) and only large dustpan, and often thousands of mu on one slope. The landscape forms the changing symphony of heaven and earth, covering about 16,603 hectares from the southern section of Ailao Mountain along the Red River. Over the last 1400 years, a complex farming system has formed to transport water from hilltops covered virgin forests to layers of terraces. As a world cultural heritage, it is a masterpiece left by generations for more than 1,400 years. It is an idyllic landscape painting carefully carved on the Earth.

From the high mountain area above 2,000 meters above sea level to the river and dam area of 600-144 meters, the seven nations live roughly according to the sea levels, they construct and maintain together this human miracle. Among them, the Hani people live the upper mountain. They have cultivated terraced fields to plant rice since the Sui and Tang Dynasties and dug 4,653 backbone ditches on the mountain, solving the —— water conservancy problem on the lifeblood of terraced rice making. The highest series of terraces reaches 3000 levels.

Hani has formed a set of scientific and reasonable methods in terraced farming, water system and fertilization management.

This is a miracle created by the people of all nations mainly Hani for thousands of years. It is a magnificent and beautiful picture of the different four seasons written on the Earth with nature and human harmony (Fig.4).

Fig.8 Terrace landscape

The magnificent Yuanyang terraces not only solve the ecological problem of paddy fields, but also a model of artificial wetland, but also solve the world agricultural problem of reduced soil fertility, and entropy does not increase for thousands of years, and sustainable development.

Dujiangyan and Yuanyang Terrace are the sustainable engineering. They are two classical modes of sustainable development beyond 2,200 and 1,400 years [40].

According to the recycling economics and the sustainable development, society should keep a lower entropy state, so it can be sustainable development. Daly researched beyond growth from the economics of sustainable development [41]. This should be a harmonious development of society-economy-environment.

The sustainable development is related to the ecological economics [42]. New economics is based on human and the earth, and proposes that real wealth should be measured by increasing happiness and environmental sustainability, not just the accumulation of goods and consumption [43]. Thackara researched the next economy as designing tomorrow's world from soil, land and water to food, clothing, housing, health and so on [44]. New growth means that ecological diversity and watersheds are healthier. The most important thing is to have the ability of ecological thinking, a kind of life model as a whole ability to connect with each other. It corresponds to the unity of man and nature in Chinese traditional culture.

We proposed the Chinese cultural-social ecology, which may resolve the human crises. Its aim is that human and nature are completely harmonious, and reach the highest state of the unity of nature and human [45].

These process engineering cannot be always increase entropy. For the circular economy and any cyclic process, entropy cannot increase always.

5. Discussion and Conclusion

At present entropy increase as a new world view [34] already becomes a conviction and a scientific alienation. The change of entropy should be a testable science. At least some extensions of the second law of thermodynamics are not universal. As long as we break through the bondage of the second law of thermodynamics, so the rich and complex world is full of examples of entropy decrease.

In a word, the analysis for many experiments and theories shows that the second law of the thermodynamics should be developed. Entropy decrease is possible when there are internal interactions in an isolated system [23,24,46-48]. A major category of these interactions is nonlinear, which can lead to solitons, chaos, and fractals. 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 reduction. 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. Chemistry and process engineering do not always tend toward disorder; there are inevitably internal interactions, and entropy decrease should also occur.

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