Satoru Yamamoto

Reaction Kinetics Based on Time-Energy Uncertainty Principle

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ISBN 978-981-19-9672-6 ISBN 978-981-19-9673-3 (eBook)
https://doi.org/10.1007/978-981-19-9673-3

Translation from the Japanese language edition: "Atarashii Hannosokudoron no Kokoromi" by Satoru Yamamoto, © The author 1979. Published by Showado. All Rights Reserved.

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The registered company address is: 152 Beach Road, #21-01/04 Gateway East, Singapore 189721, Singapore

Translators' Preface

To illustrate Arrhenius' empirical formula for rate of chemical reactions, several theories including collision-reaction models have been proposed, but the most currently accepted of these is Eyring's so-called absolute reaction kinetics. We ourselves tried to understand the content when we were students, but we were not completely convinced.

Eyring hypothesized a transition state in order to theoretically derive Arrhenius equation. That is, suppose the existence of an activated complex, $[AB]^{\ddagger}$, in a transition state when A and B react to produce C, and consider the following process:

$A + B \rightleftarrows [AB]^{\ddagger} \rightarrow C$.

The assumptions are as follows:

(1) A, B, and $[AB]^{\ddagger}$ are assumed in pseudo-equilibrium even when the reactants (A, B) and products (C) are not in equilibrium with each other.
(2) $[AB]^{\ddagger}$ reacts irreversibly to become C.

It is assumed that A, B, and $[AB]^{\ddagger}$behaves in classical mechanical manner, and the reaction does not proceed unless the reactants overcome the energy barrier of the transition state, which is the state of energy maximum along the reaction path, by thermal activation. Although various methods have been devised to create model structures of the activated complex, it is imaginary or hypothetical and is not subject to observation, which is different from reaction intermediates with definite lifetime.

In 1979, S. Yamamoto wrote this book which stated that Arrhenius formula can be explained by using the uncertainty principle.

The uncertainty principle is expressed by

$\Delta p \cdot \Delta x \sim h/2\pi$

$\Delta E \cdot \Delta t \sim h/2\pi$

However, there is still controversy over what is the physical meaning of ΔE and Δt [1, 2].

Yamamoto proposed the interpretation that ΔE is the fluctuation of the energy of the system, and that Δt represents the lifetime of that system.

If ΔE is zero and the system has a definite energy value, the lifetime of the system, Δt, is infinite; that means the system is steady and stable. If the system in question is described by the superposition of two or more quantum mechanical eigenstates, then ΔE has a nonzero finite value, which makes Δt also has a nonzero value (finite lifetime). This interpretation was applied not only to so-called chemical reactions but also to changes of states such as phase transformation.

When this book was published in Japan, this interpretation has sparked much controversy in the Japan materials science community. However, without final conclusions reached, the debate subsided perhaps because the materials science community in Japan was piling up practical demands such as the development of new exotic materials, and there was no time to thoroughly discuss basic issues at that time. However, the issues proposed by Yamamoto still have the importance, and we thought that it is important to make his theory widely known to the world.

According to his theory, the reacting system is in a quantum-mechanically uncertain state, and the lifetime of the state is determined by the magnitude of energy fluctuation. This principle can be applied not only to thermal effect by increasing temperature of the system but also to other effects of such as light irradiation and application of magnetic field. It should be noted that not only so-called chemical reaction but also physical changes of materials including solidification, phase transformation in metals and alloys, crystal growth, etc. can fall within the scope of the theory since they are also reflecting the changes in chemical bonds.

In the end of this preface, we must state that "Abstract" and "key words" for each chapter are written or selected by translators for ease of readers' understanding. Furthermore, references are listed at the end of each chapter in the original Japanese version, but many of them are not directly cited. Therefore, in this English translation, the list is given in the end of each chapter as "Further readings" without citation numbers, but those references where the citation is obvious are written as "[3]," "[3, p.3]," or "[3, Chapter 5]".

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August 2022

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Preface to the English Version

I know a variety of rate equations have been proposed in the past. I have recognized those equations had not fit with experimental data especially by using electrical resistivity which is continuously changed with time. Therefore, an experiment was chosen that could precisely measure the time variation and examine in detail whether it fits the kinetic theory. It turned out that the rate equations of the past did not agree with the experimental data.

Consequently, I recognized we must reconsider in a fundamental way. In the past, I have noticed that people make up changes in things only in their minds. Let us assume that this thinking as human-being-centered principle. In contrast to this principle, countermethod of thinking would be called material-centered principle. In the following, I would explain this way of thinking more precisely.

Material-centered principle is the standpoint of view that is willing to recognize the nature as it is. The most important problem is how to answer what is correct. In the standpoint of material-centered principle, correct or incorrect would be determined whether the predictions are consistent with experimental data. It would be helpful to understand material-centered principle by presenting one more example of material science.

In material science a concept of "vacancy" is often used. I found this concept of atomic vacancy problematic upon careful consideration. Water quenching from solution treatment temperatures would have no effect on the number of atoms in the material. Then the difference between furnace-cooled and water-quenched would be the spacing between the atoms.

Then, comparing the water-quenched and furnace-cooled cases, the spacing between atoms would be larger in the water-quenched case. Therefore, in the classical human view, the larger the space between atoms, the greater the diffusion. However, bcc metals with large space per unit cell, such as Cr, Mo, and W, diffuse more slowly than fcc metals with the densest structures with small space, such as Cu and Al, even though the space are larger. Cu and Al metals, which have smaller space between atoms, diffuse more easily. This is a good example of how and what we think in our minds does not match what is actually occurring in matter.

Under the above consideration, the way of thinking of material-centered principle turns out to be accordance to quantum mechanics. In quantum mechanics, there is an uncertainty principle of time–energy, where the wave function deals with the properties of particles and the properties of waves at the same time, and electrons can only be described probabilistically. Since quantum mechanics is a theoretical system that uses these concepts, it is better to assume that physics is quantum mechanics and philosophy is materialism, which greatly expands our view of the material. This viewpoint will help to greatly expand material science.

Kyoto, Japan
January 2022

Satoru Yamamoto

Preface to the Original Japanese Version

The most important thing for a researcher is to have the right attitude toward research. What kind of research results a researcher achieves depends on the objective situation in which he or she finds himself or herself and on his or her attitude toward that situation. Of particular importance in objective situations are past research results and current dominant ideas. We are easily dominated by past research results and current dominant ideas. We cannot expect progress and development of research from a blind attitude that is easily influenced by existing authoritative results and ideas. A critical spirit that clarifies the limitations and difficulties of existing results is necessary for the progress and development of research. To criticize does not mean to unilaterally destroy and abandon past achievements by pointing out their limitations and difficulties. It is to identify what should be inherited from the past and what should not. This critical inheritance and development of past results is the proper attitude for researchers to take.

In this book, we attempted to critically inherit and develop the past results of reaction kinetics. In Chap. 1, the limitations and difficulties of the past reaction kinetics are clarified, and a viewpoint for overcoming them is established. In Chap. 2, we analyze the limitations and difficulties of current reaction kinetics. A new attempt to overcome the fundamental difficulties of the current reaction kinetics and to take a step forward is developed from Chaps. 3 to 6.

Chapter 1 describes a scientific and epistemological nature of our reaction kinetics. This book emphasizes the historical and hierarchical viewpoints, and from these viewpoints, the logic of reaction kinetics and its background in the natural sciences, especially classical mechanics, quantum mechanics, thermodynamics, and statistical physics, and their interrelationships, are discussed. Therefore, the contents of Chap. 1 will be of interest not only to readers interested in reaction kinetics, but also to readers who are interested in philosophy of science and epistemology.

In Chap. 2, the fundamental difficulties of the current reaction kinetics including nucleation-growth theory in phase transformations are clarified from the viewpoint described in Chap. 1. The reader will thus realize how fundamental difficulties exist in the current reaction kinetics, which at first glance appears to be a complete system.

In Chaps. 3 and 4, a new attempt to overcome the fundamental difficulties of the current reaction kinetics is developed. It is based on the uncertainty relation between time and energy. In consideration of the readers who are not familiar with such a concept, the contents of these chapters are explained in detail. Therefore, due to the limited number of pages, I had to limit my discussion of specific applications to simple and typical ones, which were described in Chap. 5. The applications to a wider range of more complex reactions will be discussed at another time. In Chapter 6, the basic features of new reaction kinetics are summarized.

This book is intended mainly for graduate students, young researchers, and teachers in science and engineering, but it can be understood by undergraduate students as well as liberal arts students if they read it in order from Chap. 1. Experts would be able to understand the contents of this book by starting from Chaps. 3–6 and referring to Chaps. 1 and 2 as necessary.

In this book, I have tried to discuss the reaction kinetics from a consistent standpoint. Unfortunately, in Japan, there are not enough opportunities to discuss one's standpoint on any subject sufficiently thoroughly to expose each position. The critical inheritance of reaction kinetics and its development described in this book is only an attempt, and as the author, I would be more than happy if the issues raised in this flawed book could trigger discussions in seminars, study groups, reading groups, etc., on reaction kinetics, the fundamentals of science in general, and issues of scientific cognition.

Ichijoji, Kyoto, Japan
December 1979

Satoru Yamamoto

Contents

About the Author and His Bibliography

Dr. Satoru Yamamoto was born in 1941. He received doctoral degree from Kyoto University in 1970 and continued his research for 35 years at the same institution. He has achieved notable results in variety of research fields such as Graphitization of Cementite (10), Spheroidization of Graphite (20), Martensitic Transformation (3), Aging Precipitation (6), Reaction Kinetics (5), Molecular Orbital Calculation and Alloy Theory (10), Microstructural Control of Alloys and Alloy Design (8), High Abrasive-resistant Spherical V-C Carbides (9), and Others (7). The number of published articles in scientific journal is shown in parentheses. Representative articles written in English associated with this book and books written by Dr. Yamamoto are introduced here (H. Yoshida).

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Part I
Basis for Construction of Our Reaction Kinetics

Chapter 1
What is Reaction Kinetics as a Scientific Cognition?—Toward Innovating Conventional Reaction Kinetics

Abstract Reaction kinetics is a theory of changes over time in natural phenomena, and the theory begins with human understanding of the origins of the world (nature). Since the nature consists of various layers of hierarchy and changes over time, human understanding of nature must also be based on various concepts from these two aspects of hierarch and historicity. In this chapter, the characteristics of quantum mechanics, classical mechanics, and thermodynamics are first described, and the relationship between the three disciplines is derived from statistical averaging operations, either classical or quantum, with statistical physics as a mediator, and quantum mechanical averaging operations as the more fundamental. In addition, concepts such as non-equilibrium, non-stationarity, irreversibility, uncertainty, discontinuity, and so on are important to reaction kinetics and are discussed with each counterconcept. These conceptual considerations suggest the logic in this chapter that the time–energy uncertainty principle of quantum mechanics, which explicitly contains time, is necessary to theoretically construct a full-fledged theory of our reaction kinetics.

Keywords Reaction kinetics · Hierarchical-historical movement · Uncertainty principle · Quantum mechanics · Classical mechanics · Thermodynamics

1.1 Introduction

The purpose of this book is to clarify underlying problems in the current kinetics as a whole, to identify the way how reaction kinetics should proceed in the future, and to show a new attempt along this perspective. What is reaction kinetics? "The answer is in the question."—"Zen Buddhism" saying. It means that questions never appear suddenly without any relation to the answer and also answers are never found by decomposing the questions from outside of them. When you ask a question, the answer is already involved in the question to some extent. In fact, it must be said that how we ask questions and what we ask are inextricably linked to the kind of answers we are seeking. What questions will be addressed and how will they be answered are quite dependent on what to think about reaction kinetics, what and how to ask about reaction kinetics.

S. Yamamoto, *Reaction Kinetics Based on Time-Energy Uncertainty Principle*,
https://doi.org/10.1007/978-981-19-9673-3_1

In the present chapter, therefore, we would like to deeply ask from the broadest possible perspective what reaction kinetics is. Even if this way is a bit of a detour, the harvest will be rich and fruitful, and let us be willing to take a detour. You may be able to hope that the discussion in this chapter will be useful as a viewpoint for the critique of the absolute reaction kinetics in Chap. 2 and as a guideline for the construction of a new reaction kinetics in Chap. 3.

1.2 Hierarchical and Historical Development of World—Our Cognition

The world in which we mankind live is expanding with history and will continue to grow. Such a spread of the world is not just a spread but has two aspects: hierarchical (spatial) and historical (temporal) structures. At first, the world of mankind was limited to a human-level world (macro-world) that could be directly seen and touched by humans. With the development of history, however, mankind has come to know the existence of molecules and atoms and has actively exploited the phenomena related to these molecules and atoms. It is now known further that molecules and atoms are composed of more basic particles such as protons, neutrons, and electrons. It may be revealed in the future that even elementary particles such as electrons have an internal structure. Such hierarchical spread of the world extends not only to the direction of the "micro" and "submicro", but also to the "macro" and "supermacro" such as the earth, star clusters, galaxies, galaxy clusters, and supergalaxies.

The reason why the world of mankind (human being) expands in this hierarchical way is that the world (nature) itself has such a hierarchical structure. The world (nature) movement has a historical (temporal) aspect in addition to such a hierarchical (spatial) aspect. That is, modern science has revealed that since the creation of the universe, it has historically generated, developed, and evolved from elementary particles to atomic nuclei, atoms, molecules, inorganic substances, organic substances, plants, animals, and then mankind. In this way, the world (nature) that includes us mankind is in a hierarchical and historical movement. Since the world is undergoing a hierarchical and historical movement, the spread of our world has been and will continue to be hierarchical and historical.

Our world expands hierarchically and historically. But the expansion of our world does not mean that the world will expand without our action of anything about it. To expand the world of mankind means to expand the world in which mankind works and to make the world (nature) work for us. In order for mankind to expand the world of its activities and to make non-our world our world, we must know the laws of movement in each hierarchical level and in each historical process of the world. In other words, there exists the problem of cognizing the hierarchical and historical laws of movement of the world behind the expansion of our world. Then, how has mankind cognized the hierarchical and historical laws of movement in this world? What do we mean when we cognize things? In the following, we will consider the problem of cognition of the things.

When we try to cognize the world (nature), the specific content of that cognition is naturally defined by the object of cognition. Since the world (nature), the object of cognition, is in hierarchical and historical movement, its cognition must be hierarchical and historical. Because of the hierarchical nature of world movements, our cognition must necessarily be structural. Therefore, it is necessary to disassemble a composite or a structure into constituent elements, and conversely to construct and assemble a composite or a structure from those elements. The above process of thinking decomposes a substance into its constituent, i.e., atoms, and conversely examines the structure of how the atoms combine to form a substance. By repeating such structural pursuits, complex materials are broken down into simple ones, which are further broken down into simpler ones.

In this way, cognition of multiple hierarchical structures is obtained. And finally, if it is possible to reach elementary particles that cannot be further decomposed, all other substances will be constituted by such elementary particles. On the other hand, the historicity of the world argues that it is necessary to cognize the development of the world, or the motions, changes, and reactions of matter. Such a movement of substances is understood as a time variation of its state, the future is understood by the past and the result by the cause with linking a later state in time to an earlier state. The results of cognition are then summarized in a form of the law of motion and the law of causality. Thus, our cognition about the world increases its hierarchical and historical breadth and depth, due to the hierarchical and historical development of the world.

1.3 Scientific Cognition—Axioms of Objectivity and Criteria of Truthfulness

In the previous section, the readers would recognize such viewpoints that the world (nature), which is the object of cognition, is performing a hierarchical and historical movement, and therefore, that the cognition must be a hierarchical and historical. However, it is not always allowed to use any kind of cognition as long as cognition of the world is hierarchical and historical. Actually, we are aiming for a scientific cognition of the hierarchical and historical movements of the world. Comparing other fields of science, such as religion, philosophy, and ethics, to the realm of science, what makes science the very "science" is that it is based on the "axioms of objectivity" and "criteria of truthfulness" that science presupposes.

It is well known that science puts the logical consistency and the possibility of experimental verification as a criterion of its truthfulness. However, this criterion of truthfulness alone did not make the cognition scientific. This is evident in the historical context where modern science could not be established by pre-Galilei or pre-Descartes way of cognition. Certainly, it is not that pre-Galilei and pre-Descartes scholar ignored reason, logic, or experimentation, or they did not think of organizing and matching them. However, "science" in the sense we understand today could not be constructed based on these alone. Galilei and Descartes denied the conventional teleology that interpreting phenomena from the viewpoint of their intrinsic purpose allows us to arrive at a cognition of truth. According to their postulate, there is not a "purpose" or a god in the universe, and nature is an objective being. By accepting this postulate of objectivity and abolishing Aristotle's "Physics" (natural philosophy) and cosmology, the foundation for cognizing modern science has been laid.

1.4 Progress and Innovation in Scientific Cognition

Looking at the innovation in scientific cognition about the hierarchical and historical movement of nature, we can recognize some characteristics. The first is a change in the method of scientific logic from induction to deduction. In each field of science, at their early days, the method of induction is often relied upon in combination with the scientific idea of respect for facts. In contrast, the use of deductive methods was relatively rare. A typical example of the deductive theoretical system is Lagrange's analytical mechanics. His theory of mechanics was an example of a theoretical system based on the belief that simply developing formulas would give all the equations needed to solve the problem.

The wealth of observations accumulated by inductive methods was then organized into a logical system of scientific theory based on a few well-established laws of motion. In electromagnetics, such a change has been completed from Faraday's inductive theory to Maxwell's deductive one. Such a revolution has started in the field of chemistry since the emergence of quantum mechanics. Quantum mechanics was founded between 1925 and 1926, and its effects have begun to emerge gradually. Textbooks of chemistry have been completely rewritten from inductive way to deductive one after this gradual change.

The second characteristic of this progress in scientific cognition is the shift from qualitative to quantitative description. Scientific cognition always begins with the discovery of new phenomena or new qualities. First, a qualitative observation is made. However, rather than staying at a level of a qualitative description of phenomena, quantitative measurements are gradually being made, and eventually, leading to the form of quantitative laws. In this way, when scientific cognition progresses from inductive way to deductive one and from qualitative way to quantitative one, the content of that cognition comes to be expressed by mathematical equations of motion. At this point, scientific cognition has dramatically increased its power from the passive interpretation of nature to the positive prophecy and reaches a stage where it can be incredibly powerful.

In addition to the progress of scientific cognition mentioned thus far, we must point out that our historical experience is not only mere progress of cognition, but also what can be called cognitive innovation. Einstein's theory of relativity is the first revolution in scientific cognition brought about by the various discoveries in the twentieth century. The revolution in scientific cognition is caused by not only the theory of relativity but also quantum mechanics. Here, let us consider not the contents of the theory of relativity or quantum mechanics, but the meaning of their appearance affected on the scientific cognition.

For example, the idea of relativity was at first strongly opposed by physicists and philosophers. Physicists have approached this new hypothesis with a critical spirit. In order for the new idea to be accepted in physics, it had to pass the following three-step verification. First, the new idea must not disturb what has been successful in the past work, but must inherit the past idea as a result, and must not overturns the explanation of the observations that have been used to support the theories so far. Second, it must explain the new evidence in a convincing way that makes old ideas questionable and suggests new ones. Third, it must anticipate new phenomena or new relationships that were not known or were not clearly understood at the time when it was conceived.

The forms of scientific progress and innovation mentioned above can be considered as follows. The theoretical system of science L is accompanied by a range of experience A to which it applies. If we apply this L to a wider range of experience than A, we may find a discrepancy between experience and theory in the application. And a new theory L' is found that eliminates this discrepancy. This new theory L' is also accompanied by its range in application of experience A'. A' contains A. Such process is the evolution of theory that the transition from L to L' is repeated one after another [1, p. 5].

1.5 Reconstruction of World in Terms of Our Concepts

Cognition of things has two aspects: the object of cognition and the subject of cognition. Until the previous section, the problem of cognition was mainly considered from the side of the object to be cognized. In this section, let us consider the problem of cognition from the human side as the subject of cognition.

Cognition can be defined as the conceptual reconstruction of hierarchical and historical movements of the world, the object of cognition, by humans, the subject of cognition. Therefore, the hierarchy and history of the nature as the object is reflected in the conceptual cognition system of humans and has its counterpart. The hierarchical aspect of the nature is reflected in the conceptual system of mankind as follows.

The nature has a hierarchical structure. That is, there is such a multi-layered hierarchical structure in the nature that a complex substance is composed of simple substances, while this simpler substance is composed of further simpler substances and so on. By repeating this hierarchical cognition, we finally reach elementary particles. Reflecting this hierarchical structure of the outside world, there is also a hierarchical structure in our conceptual system. Some concepts are complex, and others are simple. Complex concepts are described by simple concepts, and simple concepts are described by simpler concepts, and so on. There exists “elementary concept” for materialistic elementary particles. And all other complex concepts are