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# Prediction of future breakthroughs in materials synthesis and manufacturing techniques: a new perspective of synthesis dynamics theory

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The continuous development of different kinds of materials plays a significant role in social productivity. However, the lack of a complete synthesis kinetic theory has resulted in the absence of scientific guidance for the emergence of advanced manufacturing technologies, limiting the research and production of new types of materials. The present work aims at obtaining the basic form of the diffusion flux-driving force equation through the concept of ion diffusion so as to establish a synthesis kinetic theory. Using this theory, the scientific principles of existing synthesis technologies are summarized, and the key directions that future manufacturing technologies need to break through are proposed as well. Based on a comprehensive analysis of this theory, the feasible directions are discussed, providing strong support for the early realization of targeted design and manufacturing of new materials with specific functions.

## New concepts

Herein, this study aims to establish a synthesis theory based on the most important processes of materials synthesis and ion diffusion. In particular, a basic form of the diffusion flux-driving force equation was obtained through mathematical deduction for the first time. This equation indicates the two main coefficients that affect ion diffusion flux and the synthesis parameters that affect these coefficients. By adjusting the relevant parameters, the synthesis path of the material and the structure–performance relationship can thus be altered. Using this theory, the principles of existing synthesis technologies were summarized, and three key breakthrough directions for future material manufacturing were proposed. In addition, some constructive approaches, including the design of atomic-level mixed precursor synthesis methods, non-equilibrium synthesis technologies with index-controlled coefficients, and advanced characterization tools with high spatiotemporal resolution for extreme environments, were suggested. It is believed that the continuous efforts to improve the synthesis kinetic theory underlying ion diffusion will provide strong guidance for the emergence of new generation material manufacturing technologies and state-of-the art materials, thereby leading to further innovations in materials science.

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## 1. Introduction

Throughout the various industrial revolutions in human history, the continuous liberation of productivity has led to a rapid increase in production efficiency.[1,2] In particular, new materials provide a strong basis for further technological innovations.[3,4] Meanwhile, the development of advanced materials requires the combination of scientific theories and manufacturing technologies.[5] However, the slow progress of the synthesis theory has caused a lack of corresponding theoretical guidance for manufacturing, hindering the design of new materials.[6] The current manufacturing techniques are still focused on conventional methods, such as hydrothermal synthesis and high-temperature sintering, which exhibit extremely high energy consumption and low efficiency, resulting in prolonged synthesis times.[7] In addition, excessive input of energy significantly reduces the kinetics reaction rate as well as the reaction activity of the material. Therefore, overcoming the shortcomings of existing manufacturing technologies and achieving the rapid and accurate preparation of new materials has become an urgent task.[8]

Nowadays, some limitations can be avoided *via* ultrafast synthesis technology due to excellent energy controllability, outstanding synthesis universality, and the ability to enhance the ion diffusion flux by specific geometrical means.[9–11] The technology allows one to generate a large energy input in an extremely short period of time, resulting in a geometrical growth of ion diffusion flux inside the material. This ensures effective bypass in the intermediate synthesis steps, reducing the formation energy barrier of the material and thus enabling its rapid mass production.[12–14] However, regardless of a breakthrough in ultrafast synthesis technology, there are many bottlenecks that limit its practical application. To date, the main route to implement ultrafast synthesis has consisted of a rapid variation in the macroscopic temperature. An abrupt change in temperature induces the conversion of the kinetic energy of ion diffusion into the potential energy. Once the potential energy of the vibration exceeds the potential well

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depth, the ions will break away from the crystal structure, causing large-scale transition and the formation of numerous vacancies in the material. The excessive vacancy content will affect the stability of its structure, thus making it difficult to maintain the reaction stability. In addition, it is very limited to rely solely on changing the temperature to achieve a rapid increase in the diffusion flux. The reason for this is that there is still no complete synthesis theory to guide the specific parameter control. Therefore, it is of decisive significance to establish a mathematically sound synthesis theory for the further development of new generation synthesis technologies and advanced materials.

In recent years, various manufacturing technologies have been proposed to avoid the shortcomings of traditional methods and open up new prospects for the design of state-of-the-art synthesis routes.[15–17] However, the reasons why these limitations could be overcome are still unclear because of the lack of an appropriate synthesis theory. It is noteworthy that without scientific foresight guidance, the development of new technologies becomes directionless. In view of the above, this study aims to establish a synthesis theory based on the most important processes of materials synthesis and ion diffusion. In particular, a basic form of the diffusion flux-driving force equation was obtained through mathematical deduction. This equation indicates the two main coefficients that affect ion diffusion flux and the synthesis parameters that affect these coefficients. By adjusting the relevant parameters, the synthesis path of the material and the structure–performance relationship can thus be altered. Using this theory, the principles of existing synthesis technologies were summarized, and three key breakthrough directions for future material manufacturing were proposed. In addition, constructive approaches have been proposed along with a list of scientific and technological challenges that remain unresolved and need to be addressed. This provides a clear direction for the future development of the field. It is believed that the continuous efforts to improve the synthesis kinetic theory underlying ion diffusion will provide strong guidance for the emergence of new generation material manufacturing technologies and state-of-the-art materials, thereby leading to further innovations in materials science.

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## 2. Theoretical establishment based on ion diffusion

The current synthesis technologies consist of regulating the temperature transfer rate.[18–20] Fig. 1 displays various ultrafast heating routes demonstrating strong competitiveness in the synthesis of multicomponent high-entropy materials in comparison with traditional synthesis methods.[21–23] However, there is still a lot of room for improvement in order to ensure that these technologies can be applied on an industrial scale. Recent studies have shown that higher energy input, smaller target space and shorter heating time need to work together so as to achieve faster heating rates. This means that the current

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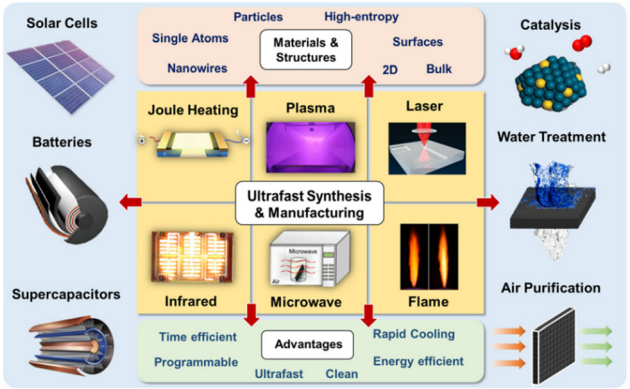


Fig. 1 Ultrafast synthesis and manufacturing methods and their applications.[26]

ultrafast heating methods are typically used for material preparation in situations where the space is limited and the volume is very small (usually less than $cm^3$). As a result, the manufacturing of target products through this technology is still in its early stages, providing per-batch quantities from milligrams to grams, which is far from meeting the demands of large-scale industrial applications. Therefore, the development of related equipment is of utmost importance. The stringent requirements for atmosphere, pressure, peak temperature, and heating/cooling rates inevitably involve various reaction chambers, sensors, and precise controllers operating at high temperatures or pressures, which deserve significant investment.[24,25] Indeed, adjusting the temperature transfer rate enables one to realize the precise and rapid synthesis of complex materials, but in global aspects of the synthesis technology and the impact on the resulting material, the temperature transfer rate is only a part of the whole process. Furthermore, the intrinsic reasons explaining how ultrafast technology affects the synthesis process have not yet been clearly understood. Until now, it has been suggested that the synthesis process mainly depends on ion diffusion. Therefore, exploring the influencing factors of ion diffusion in the synthesis would provide new prospects for breakthroughs in manufacturing technologies. For this purpose, the classic Fick's law was first introduced to describe the main parameters of ion diffusion in materials synthesis and to establish a complete mathematical framework based on ion diffusion kinetics that affects the synthesis course.

The relationship between the chemical potential ($\mu$) and the relative activity ($a_i$) can be expressed as follows:

$$\mu = \mu^* + RT\ln a_i = \mu^* + RT\ln x_i\gamma_i \quad (1)$$

where $x_i$ is the ion content, $\gamma_i$ is the activity factor, $R$ is the ideal gas constant, and $T$ is the temperature. For the complex multivariate systems with high entropy, the ion content can be found from the equation below:

$$x_i = \frac{c_i}{\sum_{i=1}^{n} c_i} \quad (2)$$

where $c_i$ is the ion concentration, $\sum_{i=1}^{n} c_i = \text{constant} = C$. Hence, the following relationship can be derived:

$$\frac{dx_i}{dx} = \frac{1}{C}\frac{dC(x)}{dx} \quad (3)$$

According to Fick's law:

$$J = -D\frac{dC(x)}{dx} \quad (4)$$

Thus, eqn (1) can be differentiated as follows:

$$\frac{d\mu_i}{dx} = \frac{RTd\ln(x_i\gamma_i)}{dx} = \frac{RTd\ln a_i}{dx} = \frac{RTd\ln C(x)}{dx} \quad (5)$$

Since $\frac{d\mu_i}{dx} = \frac{RT}{C}\frac{dC(x)}{dx}$, it can be found that:

$$\frac{dC(x)}{dx} = \frac{C}{RT}\frac{d\mu_i}{dx} \quad (6)$$

Substituting eqn (6) into eqn (4) results in:

$$J = \frac{DC}{RT}\frac{d\mu_i}{dx} \quad (7)$$

According to the definition of Gibbs free energy ($G$):

$$G = \sum_{i=1}^{n} x_i G_i \quad (8)$$

and knowing that $\mu = \frac{\Delta G}{\Delta N} = \frac{dG}{dN}$, $T = \frac{dE}{dS}$, the following relationship can be obtained:

$$\mu = -T\frac{dS}{dN} \quad (9)$$

By substituting eqn (9) into eqn (7), the diffusion flux-driving force equation can be written as:

$$J = -\frac{DC}{R}\frac{\partial^2 S}{\partial N\partial x} \quad (10)$$

where $J$ is the diffusion flux, $D$ is the diffusion coefficient, $C$ is the molar concentration, $N$ is the number of particles in the system, $x$ is the generalized coordinate value, is the quantity related to the free path, and $S$ is the entropy representing the degree of ionic mixing.

It is worth mentioning that this formula is formally consistent with the general form of the diffusion equation ($J = AQ$, where $A$ is a constant and $Q$ is the driving force of ion diffusion). Therefore, since $C$ in eqn (10) represents molarity, its value can be considered constant. Hence the true ion concentration is determined by $\frac{\partial S}{\partial N}$, and then $\frac{\partial^2 S}{\partial N\partial x}$ represents the ion concentration gradient, the degree to which ions are distributed in space.

The diffusion coefficient $D = D_0 e^{\frac{-E_a}{RT}}$ can be further substituted into the diffusion flux-driving force equation, and $T = K_T t$ ($K_T$ is the temperature transmission rate, and $t$ is the time) can be defined to get the expansion form of this equation as follows:

$$J = -\frac{D_0 C}{R}\frac{\partial^2 S}{\partial N \partial x} e^{\frac{-E_a}{RT}} = -\frac{D_0 C}{R}\frac{\partial^2 S}{\partial N \partial x} e^{\frac{-E_a}{RK_T t}} \quad (11)$$

According to eqn (11), there are two main parameters that affect the ion diffusion flux. The first one is the linear control coefficient $\frac{\partial^2 S}{\partial N \partial x}$, which reflects the mixing degree of the precursor and refers to the intrinsic influencing factors of the material. The greater the mixing degree of ions in the precursor, the stronger the driving force for ion diffusion. The second one is the exponential control coefficient $e^{\frac{-E_a}{RK_T t}}$, which is influenced by the following synthesis parameters: the activation energy required to overcome the ion diffusion barrier and the temperature transfer rate $K_T$ that determines the speed of temperature change. Adjusting the exponential control coefficient, especially the external temperature transfer rate, is a very effective means to increase the ion diffusion flux, which can enable more reaction kinetics steps to be crossed during the synthesis process, thereby shortening the kinetic barrier.[27] However, the current synthesis technologies mainly focus on the temperature transfer rate aspect of the exponential control coefficient, ignoring the contributions from other influencing factors, which limits the opportunities of modern materials synthesis to some extent. In addition, the blind development of ultrafast synthesis methods causes bottlenecks in the final products. For example, the large dramatic changes in temperature at the macroscopic level hinder the atomic diffusion inside the material; therefore, a part of the kinetic energy used for diffusion is converted into the potential energy of the atoms themselves. When the self-potential energy is too high, it makes atoms break through the kinetic barrier, resulting in a large number of vacancies and affecting the structural stability of the material. Therefore, future synthesis directions should take into consideration the influence of various factors on the final phase of the product so as to elaborate more efficient synthesis tools.

## 3. Synthesis kinetic theory for guiding future materials manufacturing

Traditional manufacturing technologies exhibit very slow energy transfer, resulting in the low ion diffusion flux and extending the synthesis time to a large extent. This also leads to the extremely low Gibbs free energy and a very stable atomic arrangement of the target product, bringing it into a thermodynamically stable state. Such passivated structure usually exhibits low reaction activity, preventing the development of theoretically predicted phases with excellent performance. Therefore, enhancing the ion flux during the synthesis has become the focus of some synthesis technologies, providing new technical means for solving the above problems. However, because of the incomplete synthesis theory, the scientific principles of those technologies have not been fully explained, which hampers the birth of new generation synthesis tools. Therefore, the concepts of the control coefficients in the proposed ion diffusion flux-driving force equation were afterward discussed in terms of various synthesis parameters so as to define the future synthesis directions. These discussions would provide new ideas for the design of state-of-the-art manufacturing technologies.

### 3.1 Synthesis kinetic theory to explain new manufacturing technology

To better understand the ion diffusion flux-driving force equation, a *J*–*t* curve model based on eqn (11) was created so as to elucidate the influence of relevant parameters on different types of synthesis technologies (Fig. 2). When the exponential control coefficient exerts a small effect on the ion diffusion flux, the latter changes slowly within a unit time interval. Therefore, the linear control coefficient has a greater influence on the ion diffusion flux (Fig. 2(a)), and there is a linear relationship between the diffusion flux and time. This can be regarded as a heating process dominated by thermodynamics laws. The situation mainly corresponds to the extremely slow energy transfer that is typical of traditional synthesis methods. Because of the slow injection of energy, the material needs to pass through various steps to form the final phase, which results in a significant impact of the linear control coefficient (*i.e.*, the mixing degree of the precursor) on the ion diffusion flux. Therefore, enhancing the mixing degree of the initial phase would further increase the ion diffusion flux in conventional synthesis approaches.

It is worth noting that diffusion is a temperature-dependent fundamental process that serves as the foundation for synthesis reactions. The exponential control coefficient represents the microscopic movement of atoms during the heating. In solid materials, the diffusion is relatively slow, and its rate increases significantly with the rise in temperature. When the

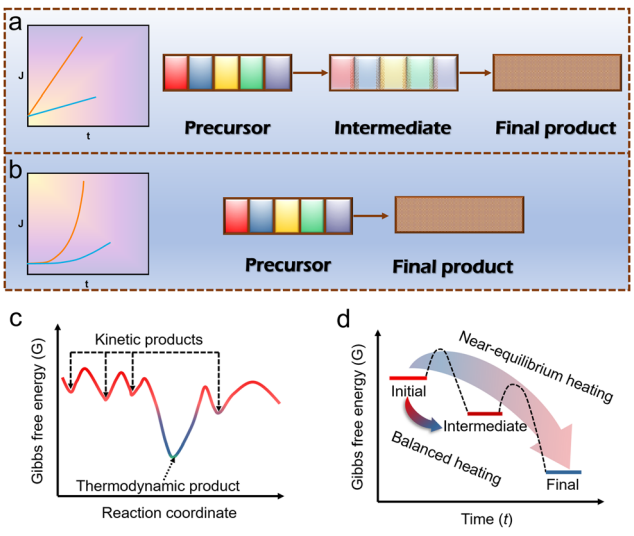


Fig. 2 Synthesis controlled by (a) thermodynamics and (b) dynamics laws. (c) Comparison of Gibbs free energy of products formed *via* different kinetic pathways. (d) Comparison of Gibbs energy pathways for products formed conforming to various kinetic modes.

exponential control coefficient has a larger effect on the ion diffusion flux, its influence dominates. At this time, there is an exponential relationship between the diffusion flux and time, that is, the diffusion flux drastically increases in an extremely short period of time, and ions are uniformly distributed throughout the free path to form the final phase (Fig. 2(b)). Thus, the synthesis process is dominated by the kinetics laws, which take place in advanced synthesis technologies generating a huge energy injection within a very limited time interval. Thanks to the instantaneous high-energy injection, the material overcomes many kinetic barriers during the synthesis, bypassing the low-temperature stage of the non-dense surface diffusion. Therefore, adjusting the exponential control coefficient would ensure the rapid increase of ion diffusion flux, which is also the key adjustment direction of newly developed synthesis technologies.

Utilizing thermodynamics models in the analysis of the characteristics of products and reaction pathways generated under different kinetic modes is essential to get a comprehensive understanding of materials synthesis. Substances that are produced under controllable thermodynamic conditions exhibit the lowest Gibbs energy and highly stable atomic arrangements, thereby falling into the category of thermodynamically controlled products. Conversely, when a reaction is governed by kinetic control, the resulting substances typically exist in a thermodynamically metastable state, highlighting the dominance of kinetic factors (Fig. 2(c)). This observation underscores the potential to achieve different thermodynamic states by manipulating the reaction pathway and time scale, thereby offering a novel and versatile approach to modulating the functional properties of the final product. Moreover, unlike the ability to tune the linear control coefficients, the adjustment of exponential control coefficients leads to a swift amplification of the ion diffusion flux. In the synthesis process, this effect manifests itself by a step that circumvents the kinetic barrier, enabling the direct formation of the target product from the precursor without the intermediate generation of phases (Fig. 2(d)).

### 3.2 New manufacturing technology based on linear control coefficient

According to the above conclusion, the linear control coefficients mainly affect the traditional conventional materials synthesis technologies dominated by thermodynamics laws. In traditional high-temperature manufacturing processes, the linear control coefficients become predominant due to the relatively insignificant variations in exponential control coefficients over time. As a result, the diffusion flux-driving force equation degenerates into a linear form. Therefore, when the precursor materials are highly mixed, there is an increase in the initial ion mixing entropy in the above equation, this will lead to a significant reduction in the barriers that need to be overcome during the synthesis process. This, in turn, promotes favorable changes in the setting of macroscopic synthesis parameters, among which are a substantial decrease in synthesis temperature and a significant reduction in synthesis time. These improvements are attributed to the effective enhancement of linear control coefficients, which boosts the ion

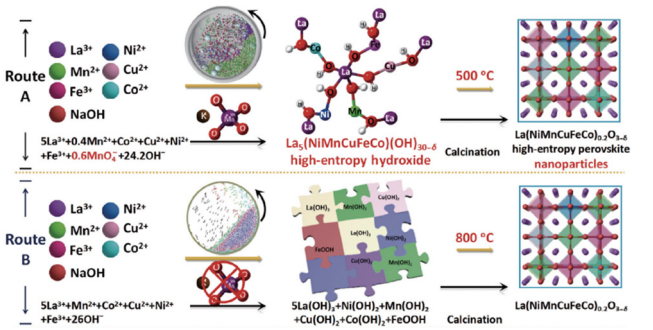


Fig. 3 Illustration of the synthesis route of the high-entropy $LaMnO_3$.[30]

diffusion flux during the synthesis. For example, when using the macroscopic precursor mixing represented by high-temperature solid-phase methods, the synthesis temperature of the high-entropy perovskite material must be above 1200 °C, and several tens of hours are required to produce a single homogenous phase.[28] When the degree of precursor mixing is reduced to the molecular level using the template method, the overall spatial ion distribution gradient $(\frac{\partial^2 S}{\partial N \partial x})$ increases. This significantly enhances the contribution of the linear control coefficient to the ion diffusion flux. According to experimental results, the synthesis temperature for similar high-entropy perovskites can be lowered to 700–800 °C, and the synthesis time can be shortened to 3–5 hours.[29] Recently, Zhang *et al.* have further decreased the degree of precursor mixing to the atomic level, thereby achieving thorough mixing within the lattice (Fig. 3). Such an advanced level of mixing significantly increases the ion mixing entropy, $S$, in the linear control coefficient. As a result, it diminishes the difficulty of ion migration during the synthesis process. Consequently, the synthesis temperature for high-entropy perovskites is as low as 400 °C, and the synthesis time is limited to 2 hours.[30] It is evident that the linear control coefficient influenced by the degree of precursor mixing plays a crucial role in enhancing ion diffusion flux for materials synthesis techniques with slow input energy characteristics. This can significantly reduce the required synthesis temperature and synthesis time. Therefore, realizing a higher level of ion mixing will have a decisive impact on the revitalization and reutilization of traditional synthesis techniques in the future.

### 3.3 New manufacturing technology based on the exponential control coefficient

**3.3.1 Regulation of the $E_a$ value.** With the aim of rapid enhancement of the ion diffusion flux, adjusting the exponential control coefficient is undoubtedly the most effective means, and the exponential growth of the ion diffusion flux will endow the final product with the kinetics-dominated characteristics. According to a thorough analysis, there are two synthesis parameters that affect the exponential control coefficient, but very few reports explain how to adjust the activation energy to achieve the above purpose. Currently, the regulation of the

 

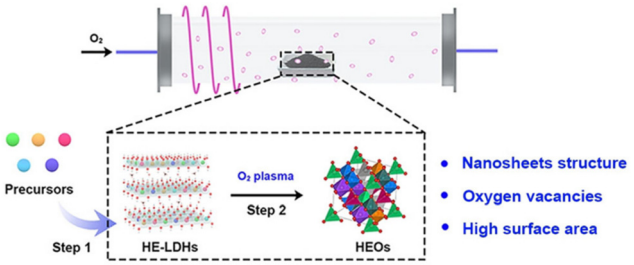


Fig. 4 Low-temperature plasma strategy for high-entropy oxides.[31]

exponential control coefficient is focused on the continuous improvement of $K_T$, but this measure is obviously limited by its impact on the synthesis technology field. As mentioned earlier, a significant change in temperature at the macroscopic scale will exert many negative effects on the structural stability and physicochemical properties of the final product. Therefore, besides reducing the adverse effects caused by the rapid variation in temperature, it is also necessary to ensure that the dynamic control characteristics of high-energy injection remain unchanged. Excitingly, the effective modulation of the activation energy opens up new prospects for regulating exponential control coefficients. A thorough understanding and quantification of $E_a$ will better promote the development of state-of-the-art technologies. According to the relevant definition, $E_a$ represents the energy required for 1 mole of atoms to undergo diffusion. In other words, it signifies the energy barrier that atoms must overcome when transitioning from their original crystal structure, while undergoing the binding forces exerted by surrounding atoms. In this regard, the quantification of $E_a$ can be effectively addressed by utilizing the Maxwell–Boltzmann distribution.

Through an interpretation of the mechanisms underlying the effects of $E_a$, it has been assumed that effective modulation of $E_a$ can be realized by reducing the energy required for breaking internal chemical bonds in materials. The modulation consists of adjusting the phonon states of materials to optimize the level of lattice vibrations during the synthesis process. This insight provides a guiding principle for material synthesis, that is, the ability to input energy without significantly altering the temperature. In other words, it opens up possibilities for the development of material synthesis techniques that allow for high-energy input in low-temperature environments. Achieving this goal would help to prevent the inauspicious consequences to the target product caused by the drastic change in macroscopic temperature, and also realize effective regulation of the exponential control coefficient to promote the enhancement of ion diffusion flux. For instance, the aforementioned goal can be successfully accomplished through a low-temperature plasma etching strategy recently developed by Wang *et al.* In this technique, energy transfer is realized through inelastic collisions between high-energy electrons and oxygen molecules. As a result, the chemical bonds of the precursor get disturbed, overcoming the potential barriers created by the binding forces of the surrounding atoms, and the $E_a$ value is thus effectively reduced (Fig. 4).[31] Therefore, the design of future materials synthesis technologies targeting the exponential control coefficient should take into consideration the activation energy aspect. In particular, it is important to utilize non-destructive methods to weaken the bonding within the material so as to allow for the rapid movement and participation of ions in the reaction. The aim is to minimize the damage to the physicochemical properties of the final material phase caused by the instantaneous input of a large amount of energy.

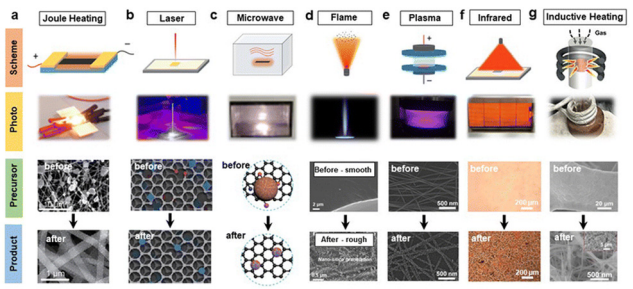


Fig. 5 Introduction of typical ultrafast manufacturing technologies. Each column represents one method.[26] (a) Joule heating synthesis of nanoparticles.[37] (b) Laser synthesis of single atoms on graphene.[38] (c) Microwave synthesis of nanoparticles.[39] (d) Flame treatment for poly(dimethylsiloxane) surface modification.[40] (e) Oxygen plasma treatment for Ag nanowire oxidation.[41] (f) Flash infrared assisted fabrication of perovskite thin films.[42] (g) Rapid inductive heating synthesis of electrocatalysts on Ni foam.[43]

**3.3.2 Regulation of $K_T$.** Another important influencing factor for the exponential control coefficient is the temperature transfer rate $K_T$ that makes part of various synthesis technologies,[32–34] among which are laser-assisted and, microwave-assisted methods, Joule heating, infrared and plasma synthesis approaches, flame-assisted synthesis and so on (Fig. 5). These techniques rely on the instantaneous release of a huge amount of energy, providing simultaneously high power and high temperatures during the synthesis. Hence, the synthesis process can be regarded as non-equilibrium heating driven by kinetics laws, and the products mostly have unstable atomic arrangements and local Gibbs free energy minima. The final phase formed belongs to the kinetic product. The ions inside this structure are not completely diffused, so their kinetic energy is interrupted by the energy input. At the same time, the original kinetic energy is converted into potential energy of its own vibration, which endows the material with extremely high surface energy, that is high reactivity. However, this characteristic has two sides. When the vibration amplitude is too large, the ions will jump out of their potential well, which will lead to poor structural stability and inability to stably output reaction energy in the long-term synthesis. Therefore, in the future, when developing manufacturing technologies aimed at enhancing the exponential control coefficient by accelerating $K_T$, a precise energy input should be achieved, that is the instantaneous temperature increase at the microscopic level. For example, Dai *et al.* have developed a mechanochemical approach that utilizes mechanical energy and frictional heating of precursors containing metal oxides and halides to

accelerate ion diffusion within the material. This method enables localized heating within the material, inducing nearly instantaneous atomic diffusion. As a result, the thermodynamic metastable phases can be formed with significant changes in $K_T$ within a very short period of time.[35] In addition, the recently emerged sonochemical fabrication technique ensures higher specificity and better control over molecular and chemical processes. It underlies the phenomenon of acoustic cavitation, where bubbles form and subsequently implode within the reaction solvent. This process generates instantaneous ultrahigh pressure, ultrahigh temperature, and rapid changes in $K_T$. As a result, sonochemistry is being increasingly applied in the synthesis of various novel materials.[36] In summary, these technologies leverage the ability to realize instantaneous high temperatures within a small localized area along with a rapid change in $K_T$. This opens up new prospects for overcoming the limitations of existing technologies that are associated with macroscopic temperature variations resulting from the instantaneous input of a large amount of energy.

### 3.4 The relationship between synthetic kinetics and crystal growth

In the synthesis of functional materials, achieving controllable synthesis parameters according to pre-designed sizes, compositions, phases, and morphologies is crucial for product quality. In this context, the influence of crystal growth is significant. Several representative works have demonstrated the tremendous potential of kinetics-driven synthesis techniques in controlling the reaction time scales and pathways, thereby enabling the synthesis of specific functional materials. For instance, utilizing Joule heating technology to reduce the synthesis duration from hours to milliseconds allows for the controllable preparation of single-crystal Pt foils, Pt nanoclusters, and Pt single-atom catalysts. However, the limited heating time kinetically constrains the nucleation and growth of Pt or the diffusion of thermally activated Pt atoms. This aspect can be described by the relationship between grain size ($d$) and temperature ($T$) as follows:[44]

$$d^n - d_0^n = k_0 e^{-\frac{E_a}{RT}} t \quad (12)$$

In this equation, $d_0$ represents the initial grain size, $n$ stands for the grain growth exponent, and $k_0$ is a constant. During the grain coarsening, the parameter $n$ usually takes the value of 2 or 3. A case of $n = 2$ is associated with normal grain growth without complementary factors such as secondary phases, impurities, or pores that may impede grain boundary mobility. In contrast, when $n$ is equal to 3, the grain growth process is typically accompanied by the dragging of secondary phases, impurities, or pores along the grain boundaries. Interestingly, it has been found that the exponent control coefficient plays a crucial role in this process. This is because the effective adjustment triggers the grain growth within a normal and controllable range, leading to the desired quality of target products.

## 4. Summary and outlook

The thermodynamic research on synthesis processes focuses on establishing whether a reaction can occur, so that the material state to be analyzed is limited to the initial and final states of the chemical reaction without considering time. In other words, thermodynamics restrains the upper limit of the reaction and the quasi-static process of the reaction, which makes the existing synthesis thermodynamic theory unable to provide strong guidance for the development of advanced synthesis technologies and the preparation of new materials. Fortunately, being a theory that describes chemical dynamic processes, synthetic kinetics focuses on time and needs to consider the intermediate processes during synthesis. This will be a powerful supplement to synthesis thermodynamics.

The complexity of these processes also brings considerable difficulties to the establishment of relevant theories. However, it is undeniable that developing a synthesis kinetics theory with a complete mathematical basis is very beneficial for the flourishing elaboration of synthesis science and technology, making it possible to achieve the long-awaited oriented design and manufacture of advanced materials. However, the lack of effective guidance from scientific theories has led to an unbalanced situation in manufacturing technologies. If the synthesis kinetics theory is not fully explained, it will become a bottleneck restricting the breakthrough in this field. Given the aforementioned circumstances, a summary has been provided along with a list of scientific and technological challenges that remain unresolved and need to be addressed.

### 4.1 Developing precursor synthesis methods for atomic-level mixing

Through the analysis of the diffusion flux-driving force equation, it can be seen that the degree of mixing of the precursors has a significant impact on the energy input required for the thermodynamically controlled material synthesis process. A review of works aimed at developing the material synthesis methods reveals that as the degree of precursor mixing gradually increases, the required energy input also decreases significantly. However, the currently available degree of mixing is only limited to the molecular level; therefore, in order to further reduce the energy input in the future, it is necessary to develop precursor synthesis methods with atomic-level mixing. For example, precursors with the same crystal structure as the target product seem to be suitable for realizing the optimal mixing inside the crystal structure. In addition, the design of manufacturing methods that can directly construct the material structures at the atomic scale will maximize the achievement of the above goals. For example, in recent years, synthesis technologies represented by atomic layer deposition have brought new hope to accomplish this purpose as soon as possible.

### 4.2 Developing new non-equilibrium synthesis technology with adjusted index-controlled coefficients

For the kinetics-controlled synthesis processes, regulating the activation energy $E_a$ and temperature transfer rate $K_T$ are two

 

important ways to further increase the ion diffusion flux. However, the mainstream research directions are aimed at constantly increasing $K_T$ to record-breaking values, while neglecting the technical aspects for $E_a$. This not only causes a series of problems with the target products due to the rapid rise and fall of temperature, but also an unbalanced development in the field of synthesis. The design of advanced non-equilibrium technologies through the adjustment of the exponential control coefficient plays a key role in upgrading synthesis technologies. Therefore, the synthesis technologies based on the tuning of $E_a$ to enhance diffusion flux should conform to the principle of energy input without significantly changing the temperature. While maintaining the instantaneous characteristic of huge energy input, the large-scale change of macroscopic temperature should be avoided as much as possible, and the irreversible effect caused by instantaneous energy input on the material should be reduced to a large extent. As for the temperature transfer rate $K_T$, precise energy input should be achieved at higher spatiotemporal resolution. In the future, molecular-scale material synthesis technologies with instantaneous temperature changes can be developed, and the recently proposed mechanochemical method can serve as a suitable basis. In addition, the spatiotemporal input scale of existing ultrafast synthesis technologies can be strengthened through the design of attosecond laser facilities to implement a faster and more precise energy input. In summary, the introduction of non-equilibrium technologies with tunable exponential control coefficients as a scientific principle would open up new prospects for the production of high-performance materials.

### 4.3 Feasibility analysis of targeted manufacturing for specific functional materials

To pursue the ambitious goals of targeted manufacturing of specific functional materials, recognizing the challenges facing the relevant area of industry and finding viable solutions is of great significance. The flourishing development of computational materials sciences brings new hope to the advancement of materials science, enabling us to provide strong evidence for experimental conclusions and results through the high computational power of computers. However, traditional theoretical calculations mainly focus on the initial and final states of reactions, aiming to define the conditions required for the reaction to occur, which essentially falls within the realm of synthesis thermodynamics. This makes it challenging to predict and explain phenomena that are difficult to directly observe in synthesis through theoretical calculations, thus lacking support for synthesis kinetics theory. Machine learning as an essential high-throughput computational tool in computer sciences presents new opportunities for accurately describing the physicochemical parameters during synthesis and those of target products. This field has attracted widespread attention from researchers as a powerful tool for effective simulation of actual synthesis processes, providing guidance for adjusting synthesis parameters and thereby promoting the targeted development of advanced manufacturing technologies. Additionally, a shared database of thermodynamic parameters at different states of synthesis can be established. Based on that, the physical and chemical properties of the target product can be pre-simulated under different synthesis conditions, enabling the selective screening of the most favorable synthesis parameters and achieving the research and development path that would integrate the design, manufacturing and application of multidimensional functional materials. This will greatly enhance the efficiency of discovery of new functional materials and provide ample data support for the completeness of synthesis kinetics theory.

Furthermore, in terms of future breakthrough directions for synthesis technology, kinetics-controlled synthesis processes have demonstrated overwhelming advantages over thermodynamics-controlled mechanisms. Therefore, the design of innovative non-equilibrium synthesis technologies is a necessary measure for future materials. When conducting synthesis under extreme environments, it is essential to explore the phase state in extremely short time scales. This will deepen the understanding of synthesis processes in non-equilibrium reactions and allow for more accurate revisions and supplements to synthesis kinetics theory. For example, advanced characterization tools that withstand high temperature, high pressure and corrosion can be developed to address these issues. Additionally, it is recommended that these methods possess high spatiotemporal resolution (in ps, fs, nm, or atomic units) to explore the essence of synthesis kinetics and provide more specific guidance on the final physicochemical properties of the target product during the synthesis. This will optimize synthesis kinetics theory and broaden its applicability range.

### 4.4 Establishing effective research paradigms to accelerate the birth of synthesis theory

From the perspective of research background and development trends, the biggest challenge in the targeted design and manufacturing of novel materials with specific functions lies in the lagging development of synthesis science compared to synthesis approaches. Because of the lack of effective scientific paradigms, a comprehensive and mathematically grounded synthesis kinetics theory has not yet emerged. Therefore, it is essential to seek physical phenomena that are more relevant to actual synthesis conditions, as well as to abstract approximate models based on widely accepted assumptions and compare them with experimental data for validation. The results should be continuously fed back to the models and refined to ensure that the conclusions provided are more aligned with reality. Undoubtedly, establishing a comprehensive and mathematically sound synthesis kinetics theory holds strategic importance for the innovation of manufacturing technologies and the continuous exploration of functional materials. We can anticipate that the continuous improvement of this theory will realize the long-standing dreams of researchers in targeted design and manufacturing of novel materials with specific functions. With such theory, various disciplines and fields will further merge, bringing new potential to the development of advanced material manufacturing technologies.

## Author contributions

Z. S. Meng: conceptualization, data curation, formal analysis, investigation, methodology, project administration, resources,

supervision, validation, and visualization; Z. J. Xu: software and roles; Z. Y. Du: writing – original draft; T. Deng: methodology; D. Wang: investigation; Y. Zeng: formal analysis; S. S. Yu: investigation; X. Y. Hu: project administration and resources; H. W. Tian: writing – review & editing and funding acquisition.

## Conflicts of interest

The authors declare no conflicts of interest.

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