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Electrothermal synthesis of commodity chemicals

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Electrothermal synthesis of commodity chemicals has received notable interest in recent decades as renewable electricity becomes more available and environmental challenges are increasingly recognized. Representative electrothermal approaches, such as Joule heating, microwaves, induction heating and plasma, have rapidly evolved from operating in millimeter-sized micro-reactors toward modular and even industrial-scale systems. Meanwhile, new chemical engineering concepts, such as dynamic and programmable operation for non-equilibrium chemical reactions using nanosecond- to millisecond-long energy pulsing, spatial and temporal heating by electrifying various reactor components (for example, the reactor walls, catalyst bed or reactant in porous media), and field-enhanced reactions and catalysis, have been discovered to improve synthesis outcomes. Despite the rapid progress of this field, there remain many knowledge gaps and technical hurdles. Here we review the critical engineering advances, analyze the unaddressed challenges and discuss the potential directions for the electrothermal synthesis of commodity chemicals toward its broader implementation for future chemical manufacturing.

The industrial production of commodity chemicals plays a major role in global energy consumption and greenhouse gas emissions[1]. This is because the conventional heating process used for thermochemical synthesis is typically powered by the combustion of natural gas or fossil fuels, which is energy-intensive, inefficient and environmentally harmful because of the heavy emission of carbon dioxide (CO_2), nitrogen oxides, sulfur dioxide and particulate matter, among others. In addition to these major challenges, the performance of thermochemical synthesis using conventional reactors requires further improvement to meet the growing productivity, sustainability and energy efficiency demands[2].

To this end, the use of electrified heating with renewable electricity in lieu of process heat from fossil fuel combustion has gained traction, offering an important opportunity to decarbonize the chemical industry and produce useful chemicals with potentially improved reaction outcomes. Over the past decade, a range of electrothermal approaches (including Joule heating[2–19], induction heating[20–24], microwaves[25–36] and plasma[37–48]) have been studied at the laboratory scale for synthesizing a range of commodity chemicals, including hydrogen (H_2), syngas, carbon monoxide (CO), C_2, aromatic and other hydrocarbons, oxygenates (such as methanol (CH_3OH)), plastic monomers (such as ethylene (C_2H_4) and propylene (C_3H_6)), ammonia (NH_3), and hydrogen cyanide (HCN), among others (Fig. 1). In addition to improving process performance, the new operation protocols using the aforementioned electrothermal methods have resulted in many reaction outcomes that are unattainable via traditional reactors. For example, resistive heating (also known as Joule heating) enables a variety of reactor designs that can be used to achieve either close-to-equilibrium conversion[4] or far-from-equilibrium product selectivity[2]. Meanwhile, studies on non-resistive heating approaches have also led to numerous conceptual innovations and engineering advances, such as field-induced reactions and catalysis[49], breaking of the activation–adsorption scaling relationship via plasma-assisted molecular vibrational excitation[39],

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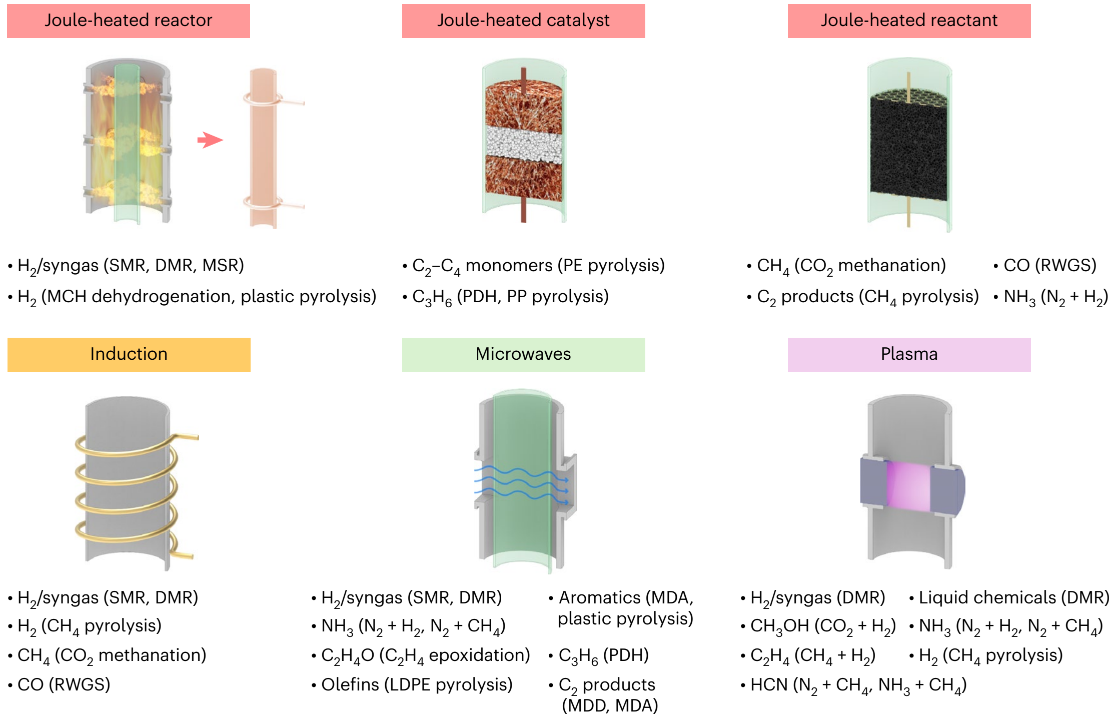

Fig. 1 | Electrothermal chemical syntheses using resistive heating (Joule heating), induction heating, microwaves and plasma. The text in the bracket refers to the reactions and/or processes used to achieve the aforementioned commodity chemicals. Note that this schematic includes only commodity chemicals. SMR, steam methane reforming; DMR, dry methane reforming; MSR, methanol steam reforming; MCH, methylcyclohexane; PE, polyethylene; PDH, propane dehydrogenation; PP, polypropylene; CH_4, methane; RWGS, reverse water–gas shift (reaction); C_2H_4O, ethylene oxide; LDPE, low-density polyethylene; MDA, methane dehydroaromatization; MDD, methane direct dehydrodimerization.

and dynamic operations[48,50]. In this Perspective, we provide an analysis of recent engineering advances and unaddressed challenges of various electrothermal synthesis approaches for making commodity chemicals. We recognize that there is also a large body of literature that focuses on using electrified heating for the synthesis of fine chemicals, which does not fall within the scope of this Perspective. At the end of the this article, we will also discuss emerging opportunities and broader impacts presented by the electrothermal techniques for the future chemical industry.

Resistive heating for electrothermal synthesis

Among various electrothermal chemical synthesis systems, resistively heated reactors sometimes require fewer modifications to the conventional reactor design and existing industrial infrastructure. In addition, the resistive heating approach (that is, Joule heating) offers a wide range of design and/or operation options for the heater (material and structure), reactor, scale-up strategy and process parameters. As such, substantial progress has been made lately on resistively heated reactors and the corresponding concepts for electrothermal chemical synthesis.

Selection of the heater material

To conduct resistive heating, the rational design and selection of heater materials is crucial. First, the heater must have suitable electrical conductivity—neither too high nor too low—so that enough heating energy can be generated with a reasonably low electrical power input to ensure good efficiency[19] ($E = i^2 \times R \times t$, where E is the energy, i is the current flowing through the resistor, R is the resistance and t is the time of electrification). In addition to the electrical properties, the thermal properties (such as heat capacity, thermal conductivity and phase-transition temperature), chemical stability (for example, stability toward oxidation, reduction and corrosion) and mechanical properties (such as creep and hydrogen embrittlement) must be taken into consideration. Note that the criterion of dimensional accuracy (via the machining or assembly of raw heater materials) is of equal importance, without which non-uniform heat generation could occur to deteriorate the reactor performance. This is because, although Joule heating is known to offer improved temperature homogeneity if designed properly[4], dimensional inaccuracy and misalignment of the heater can easily cause undesirable spatial variation of the temperature, thereby undermining the reaction outcome. Historically, metal-based resistive heaters have been the dominant choice in traditional furnaces and ovens due to their overall suitable material properties. Leveraging the existing knowledge and material scope, metal-based resistive heaters have been explored for more than three decades in electrothermal chemical reactors. Starting from the pioneering work of Spagnolo and co-workers[3], a range of metal-based resistive heaters have been tested. A notable example involving rational selection of the metal heater is the wall-electrified steam methane reactor developed by Chorkendorff and colleagues[4]. Compared with conventional side-fired thermochemical reactors, this wall-electrified set-up reduces CO_2 emissions, enables a smaller total reactor volume by removing fuel burners and improves the temperature homogeneity through more direct heat transfer. A key enabling factor of these benefits lies in the use of an iron–chromium–aluminum (FeCrAl) alloy as the reactor wall material, whose suitable temperature–resistance relationship under the studied conditions ensures finer control over the reactor operation. Owing to this key feature, the FeCrAl alloy in different heater forms (for example, coils placed inside the reactor chamber) was adopted in several other electrothermal processes[6,51,52].

Whereas metal-based Joule heaters have a longer history of development, other heater materials have recently been discovered and used for their advantageous electrical, thermal and/or physico-chemical properties. For example, Tour, Hu, Vlachos and colleagues have pioneered the use of carbon materials as resistive heating elements for electrothermal synthesis[2,5,11,19]. Notably, while most metal-based heaters often rely on convection and conduction for heat transfer, a major part

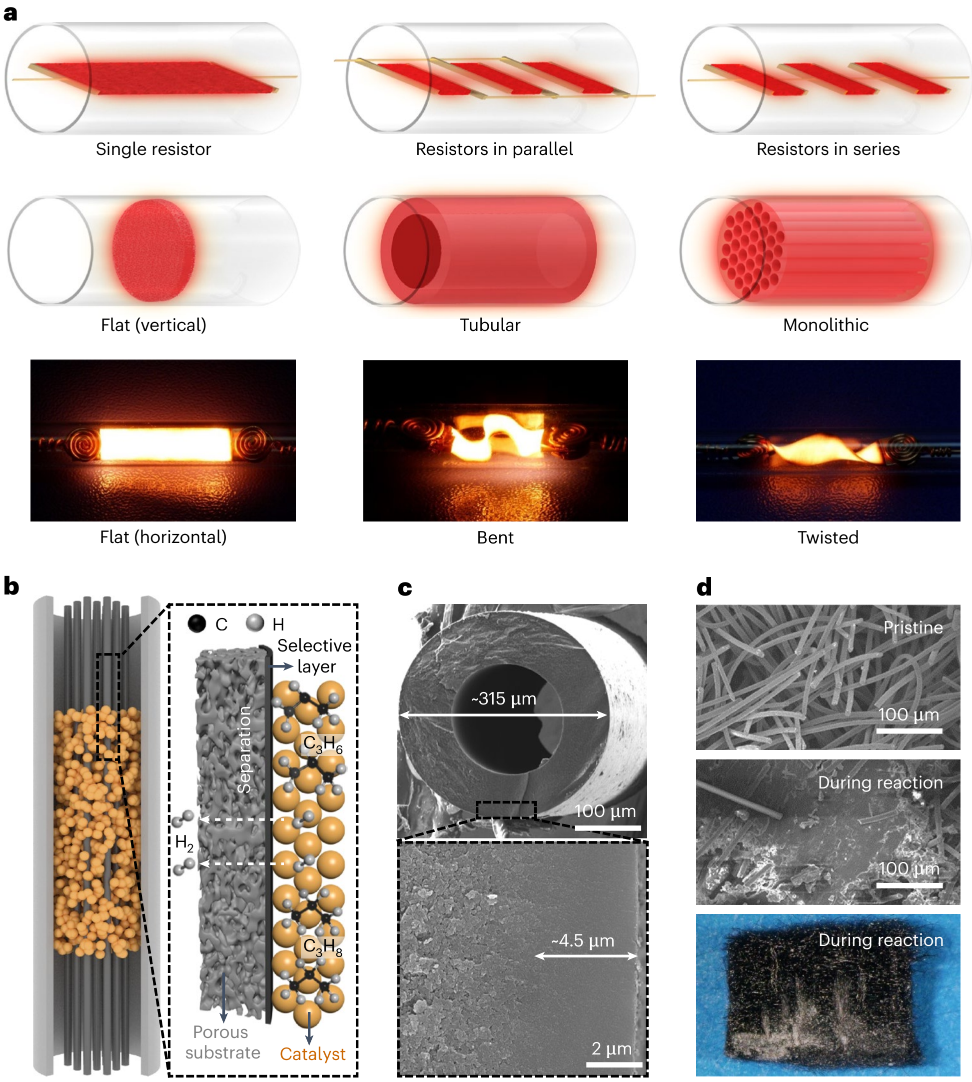

Fig. 2 | Model resistive heater designs. a, Representative alignments (single resistor, resistors in parallel and resistors in series), macrostructures (flat vertical, tubular and monolithic) and configurations (flat horizontal, bent and twisted) of resistive heaters. **b**, An electrifiable carbon hollow-fiber micro-reactor with graded porous walls for simultaneous PDH reaction and H_2 separation. Propane (C_3H_8) is converted to C_3H_6 over a zeolite catalyst. Meanwhile, the H_2 co-product is separated from the hydrocarbons. **c**, Scanning electron microscope images showing the graded porous walls of the material described in **b**. **d**, Plastic pyrolysis using a porous carbon Joule heater, which enables plastic melt infiltration, as shown in the scanning electron microscope images (top and middle) and optical image (bottom). This process enables efficient heat transfer as well as synchronized temperature profiles of the heater and reactant. Panels adapted with permission from: **b**,**c**, ref. 17, Elsevier; **d**, ref. 5, Springer Nature Limited.

of the energy generated from Joule-heated carbon can be transferred through radiation in a contactless mode, enabling high efficiency and flexible reactor designs. From a physico-chemical property standpoint, carbon materials, especially the porous ones, also possess three distinct advantages compared with metal-based heaters. First, porous carbon materials often have a low specific heat capacity, and thus a small thermal mass. In addition, carbon materials have a high emissivity ($\varepsilon_{\text{carbon}}$ is approximately 0.8–0.9 for visible wavelengths; by comparison, $\varepsilon_{\text{tungsten}}$ is around 0.4–0.5) and hence a high radiation power density. These characteristics together empower porous carbon Joule heaters with rapid rates of heating (when turning on the electrical signal) and cooling (when turning off the electrical signal), even up to 100,000 K s^{-1} within certain temperature ranges[2,19]. The rapid heating has a profound impact on the reaction kinetics according to the Arrhenius equation, whereas the fast cooling enables the use of transient heating durations to accurately control the reaction progress by limiting secondary or side reactions. This feature has not yet been realized with metal heaters due to their large thermal mass. Second, the porous scaffold enables the gas- and/or liquid-phase reactants to travel through and closely interact with the large surface area of the carbon heater material, establishing a markedly improved heat-transfer efficiency. The pores can be made either uniform throughout the heating element or fabricated with a volume gradient along certain directions to actively guide heat and mass transport. Last, carbon materials are typically rich in defects, enabling them to anchor various heterogeneous catalysts[11].

One potential drawback of carbon-based heaters is their limited chemical stability under oxidative (such as air, O_2 and CO_2) and strongly reductive (such as H_2 and hydrocarbons at high pressures) reaction conditions. Similarly, metal-based heaters are not stable against oxidants. To satisfy oxidative reaction environments and other extreme conditions, ceramic heaters can be applied as an alternative candidate. For example, Tronconi and co-workers led the development of a series of ceramic-based materials for resistive heating[8,10,12], where the high chemical and thermal stability of silicon carbide renders its utility in many thermochemical transformations such as SMR[8,12], DMR[10] and the RWGS reactions[10]. Note that many ceramic heating elements can also be fabricated into porous scaffolds (such as monoliths)[33] or hierarchical structures to mimic the effect of porous carbon heaters, albeit without as rapid temperature tunability due to the relatively higher specific heat capacity and thus larger thermal mass. In addition, ceramic heaters often lack the mechanical flexibility and cost-effectiveness of carbon-based heaters, but these can potentially be addressed through compositing with flexible carbon or metal materials. Because of their merits, ceramic heaters have gained notable commercial interest for industrial applications[53].

Heat transfer, mass transport and design of the heater structure

Electrothermal reactors possess different design considerations and therefore unique advantages for hosting thermochemical processes compared with conventional combustion-powered reactors. For example, in conventional reactors, fuel burners are often used to conduct heat into a single reactor vessel or a multiple reactor tube array, transferring heat from the outside to the inside through the reactor wall. Due to the limited thermal conductivity of the reactor wall and catalyst bed, this mode of heat transfer creates large temperature inhomogeneity inside the reaction medium, which is undesirable for homogeneous gas-phase processes (where temperature uniformity is desired) and heterogeneous catalytic processes (where localized heating is more efficient). By comparison, many resistively heated chemical reactors place heating elements inside the reactor chamber, by which heat is transferred from the heater directly to the catalyst, catalyst bed and/or reactant. This heat-transfer mode enables highly selective and efficient energy delivery to the key reactor components that directly participate in the chemical reactions while minimizing the heat loss to the unparticipating reactor components.

In addition to a favorable heat-transfer mode, the resistively heated reactors can also benefit from a broad range of heater designs (Fig. 2). Many studies have used model heater morphologies such as wires, sheets and plates for fundamental exploration at the laboratory scale[3,6]. Although these structures come without optimized heat-transfer and mass-transport characteristics, they are more suitable for characterizing the reaction outcomes due to better control and thus desired reproducibility of the set-up. These structures are also beneficial for setting up diagnostic tools to measure spatial and temporal temperature profiles along with the evolution of intermediates for mechanistic understanding. On the basis of the simplest configurations (such as wires, sheets and plates), a number of heater structures and alignments can be derived[54]. For example, connecting multiple pieces of heating element can offer zone heating to tailor the complex reactivity of chain and tandem reactions. The heating elements can be connected in series or parallel, offering different means of controlling the spatial temperature distribution and temporal heating pattern. For another example, column and tubular heaters can be derived from the plate morphology via elongation and the introduction of holes and channels[17]. In addition, assembling multiple columns or tubular heaters in an array or honeycomb structures can be applied to further improve the scale of synthesis[54]. Other than increasing the size and number of heating elements, many resistive heaters possess structural tunability[7] and flexibility[2]. For instance, by bending or twisting flat carbon paper heaters, the interactions between gas-phase reactants and heaters can be dramatically enhanced to boost the conversion efficiency. Such structural flexibility may even enable dynamic modulations of the heater shape and morphology in operando to affect the reaction outcome.

Note that the heat-transfer mode and design of the heater structure can be tailored together to benefit specific reaction schemes using resistive heating. For homogeneous gas-phase reactions, three-dimensional porous heaters are desirable to achieve optimal temperature uniformity for gas-phase reactants. To improve the contribution from radiation toward a high heat-transfer efficiency and instant temperature tunability, high-emissivity heater materials such as carbon and certain types of steel are preferred. Similar to gas-phase reactions, the conversion of solid- or liquid-phase feedstock also requires a high surface area of the heater to enhance heat transfer via conduction. On the other hand, for heterogeneous catalytic reactions, heat can be selectively exerted onto the local environment near the catalyst surface through resistive heating. This scenario eliminates the need to heat up the entire reactor chamber for potential energy savings. The overall efficiency can be improved further by facilitating heat conduction to the catalyst while minimizing radiation loss to the gas phase via a careful selection of materials, which also helps to reduce unwanted homogeneous reaction pathways.

In addition to the macrostructural heater designs and alignments (Fig. 2a), the microstructure of resistive heaters can be tailored to manipulate mass-transport behaviors, heat-transfer properties, reaction kinetics and reactant/product separation. For example, Fig. 2b,c showcases an electrifiable carbon hollow-fiber micro-reactor used for non-oxidative PDH[17], whose wall is composed of a thick, porous inner layer with submicrometer- and micrometer-sized pores for high H_2 permeability, as well as a thin, dense outer layer with nano-sized pores for operando H_2/hydrocarbon separation. Whereas the original study applied dehydrogenation catalysts on the outer layer, the inner layer could also be decorated with catalysts for reactants feeding from the inside of the channel. As another example, Fig. 3d depicts a porous carbon Joule heater used for the pyrolysis of plastics[5]. The large pore volume of the carbon-fiber network and its strong hydrocarbon affinity facilitates the wicking and infiltration of the polyolefin plastic melt to achieve a close contact with the fibers, leading to a high heat-transfer efficiency and a synchronized temperature profile of the heater and reactant during pulsed Joule heating at millisecond-level timescales.

Scale-up strategies and process innovations

Transitioning from traditional combustion-powered reactors to resistively heated reactors faces formidable scale-up challenges. Owing to the presence and critical role played by resistive heaters, scaling up resistively heated processes needs to consider the design of the reactor as well as the heater[55]. As shown in Fig. 3a, two common scaling strategies can potentially be implemented for improving the electrothermal synthesis throughput: (1) sizing up by increasing the length or diameter (that is, the surface area) of the heater along with the reaction chamber; and (2) numbering up by increasing the number of heaters and/or reaction chambers. The first strategy (sizing up) resembles a common approach used for traditional combustion-powered reactors at larger scales, which helps to mitigate the risks and introduces fewer changes to the existing reactor designs. Nevertheless, the reactor volume cannot be infinitely increased due to heat-transfer limitations. In addition, increasing the length of the reactor requires the length of the heater to be increased in electrothermal reactors, which affects the capital and operational costs more than it does to traditional thermochemical systems (for example, the longer the heater, the higher the fabrication cost, the lower the dimensional accuracy and the poorer the durability and mechanical performance). By comparison, the second strategy (numbering up) enables a higher theoretical throughput without encountering severe heat-transfer or mass-transport limitations during scale-up. In addition, numbering up the electrified modules offers greater flexibility of operation by digitally accommodating the system to a fluctuated power supply (for example, by turning on or off the heaters in part), potentially enabling the direct utilization of intermittent electricity for chemical synthesis at distributed locations. The challenge for the second strategy, however, is the higher cost of the controlling and operation units. In a typical practice, both strategies will probably need to be adopted for improving the production rate and yield, power-to-chemical efficiency, economic benefit and environmental impact (Fig. 3b–d)[4,5]. Note that a unique benefit of scaling up electrothermal chemical reactors is the ability to achieve exceptionally

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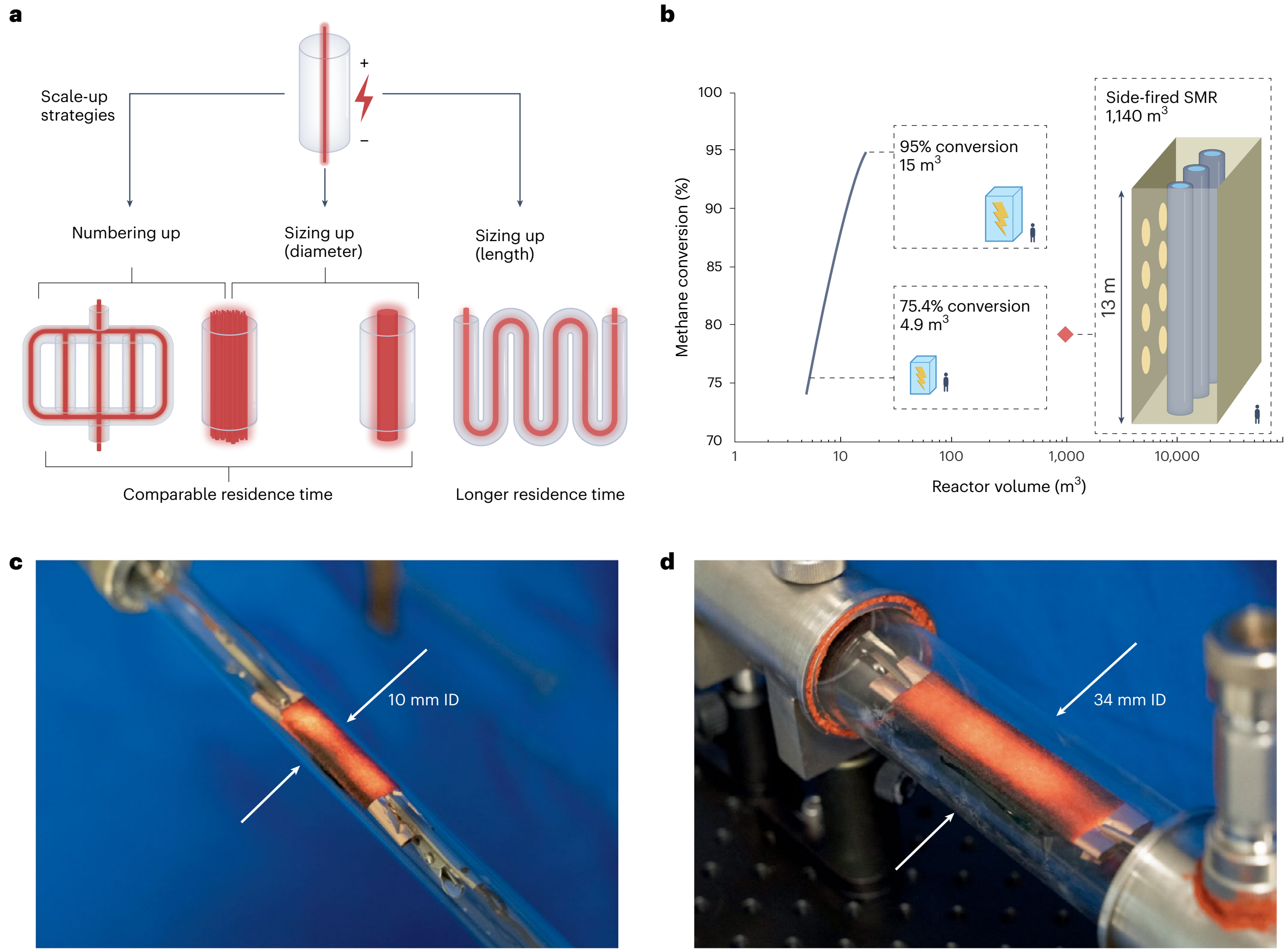


Fig. 3 | Scale-up strategies and system design considerations for resistively heated reactors. a, Scale-up strategies for resistively heated reactors. **b**, Scale-up calculation based on the wall-electrified reactor for SMR. **c**,**d**, Laboratory exploration of scaling factors on the Joule-heated process of plastic pyrolysis using smaller (**c**) and larger (**d**) systems. ID, inner diameter. Panels adapted with permission from: **b**, ref. 4, AAAS; **c**,**d**, ref. 5, Springer Nature Limited.

compact systems, which has been demonstrated using both resistive and non-resistive heating approaches[4,22,33].

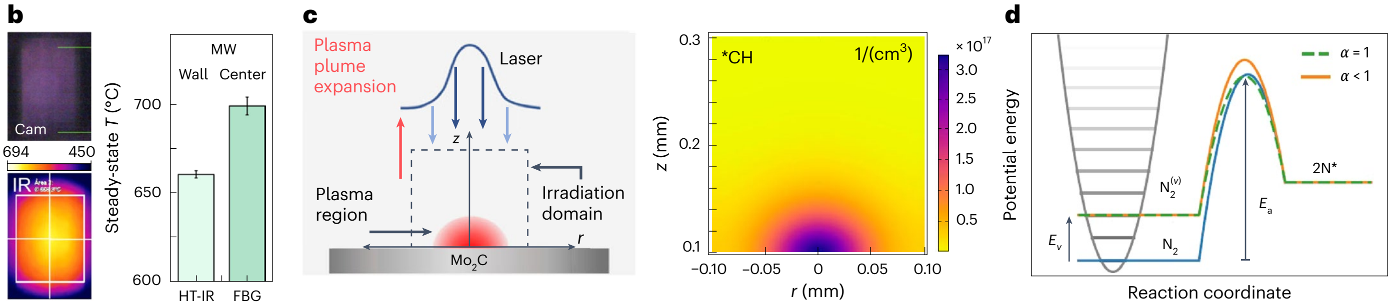
The tunable resistive heater, reactor and system designs have led to a number of reaction engineering innovations for electrothermal chemical synthesis. Interestingly, distinctive synthetic features can be obtained by varying the reactor set-up and process parameters. One representative example is the use of Joule heating to modulate the reaction kinetics and thereby the chemical equilibrium. The wall-electrified reactor developed by Chorkendorff and colleagues offers a uniform supply of heat to the reactant and catalyst bed with nearly constant heat flux, thereby driving the reaction close to the chemical equilibrium throughout the catalytic zone[4]. By contrast, the groups of Hu, Vlachos and others have reported far-from-equilibrium (also referred to as non-equilibrium) reactions and products using pulsed Joule heating with transient pulse heating durations. These approaches enabled manipulation of the reaction pathways (as demonstrated in methane pyrolysis and plastic depolymerization)[2,5,15], catalyst agglomeration and sintering (demonstrated via NH_3 synthesis)[2], catalyst regeneration (as shown in DMR)[11] and unconventional surface adsorption and desorption processes (as demonstrated via CO_2 hydrogenation)[13]. In addition to manipulating the reaction kinetics and chemical equilibrium, another important feature introduced by resistively heated reactors is the spatiotemporal temperature tunability. Hu and co-workers showed that a spatiotemporally controlled temperature program can be used to decouple and thus regulate a complex melting, wicking, vaporization and reaction process during the conversion of solid plastic wastes to their monomers, resulting in high product selectivity and yield[5]. Note that whereas the temperature gradient has shown great promise in controlling the reaction progress and thus achieving high product selectivity in conventional thermochemical reactors[56], temporal tunability has so far been realized more often in electrothermal systems.

Non-resistive heating for electrothermal synthesis

In parallel with the efforts on developing resistively heated reactors, there have been numerous technological breakthroughs and engineering advances made to systems using non-resistive heating in the past few decades, where microwaves, induction heating and plasma have led the way. While sharing some level of similarities with resistive heating, these non-resistive electrothermal methods possess unique features (Fig. 4a). For example, nanosecond-pulsed dielectric barrier discharge plasma is known for the non-equilibrium generation of unconventional intermediate species (such as excited molecules, radicals, ions and electrons) via electron impact and vibrational energy transfer[48]. Comparable timescale tunability could be achieved using microwaves through design of the cavity and electronics for dynamic, field-enhanced and non-equilibrium reaction and catalysis. Note that tuning the energy input at ultrashort timescales (for example,

a

Method	Material selectivity	Heat transfer	Length scale	On/off timescale	Energy scale
Joule heating	Moderately conductive materials	Surficial	Resistor size	Millisecond	Moderate
Microwaves	Dielectric materials	Volumetric	Penetration depth	Nanosecond	Moderate
Induction	Moderately conductive materials	Surficial	Penetration depth	(Sub)second	Moderate
Plasma	Not selective to specific materials	Volumetric/surficial	Electrode distance	Nanosecond	High



Spatial temperature Local intermediates Vibrational excitation

Fig. 4 | Non-resistive heating for electrothermal chemical synthesis. **a**, Comparison of various electrothermal synthesis methods on their material selectivity, heat-transfer mode and pathway, length scale based on where and how heat and/or energy are generated, the timescale in response to the on/off electronic control, and the scale of the energy input. This table is only intended to showcase the complementary nature of various electrothermal methods using qualitative information based on literature reports. **b**, Localized heating can be achieved using microwaves to reduce waste heat on the reactor wall, limiting the formation of by-products while saving energy in the PDH reaction. Cam, digital camera; IR, infrared camera; MW, microwave; HT-IR, high-temperature infrared camera; FBG, fiber Bragg grating. **c**, In the DMR reaction set-up (left), the critical *CH intermediate in the pulsed laser-induced plasma is concentrated at the catalyst surface, as shown in the simulated spatial distribution (right), where the color scale denotes the spatial density distribution of *CH. **d**, The N_2 molecules can be vibrationally excited (to $N_2^{(v)}$) by non-thermal plasma to break the activation–adsorption scaling relationship in NH_3 synthesis. E_v, energy of vibration; E_a, activation energy; α, efficiency factor. Panels reproduced with permission from: **b**, ref. 33, AAAS; **c**, ref. 45 under a Creative Commons license CC BY 4.0; **d**, ref. 39, Springer Nature Limited.

nanoseconds or lower) could be used to manipulate the formation and lifetime of conventionally short-lived intermediate species, which is difficult to achieve by resistive[2] or induction heating[50] due to material property constraints. On the other hand, modulating the transient energy input via plasma and/or microwaves could be costly and challenging to scale up. For another example, it was recently demonstrated that pairs of carbon-fiber-tip electrodes can be used to generate stable, atmospheric-pressure plasma with a temperature of up to 8,000 K (close to the surface temperature of the Sun)[57], substantially higher than the maximum temperatures of other electrothermal approaches that are limited by the conductor or susceptor materials. In general, a higher temperature leads to an exponentially increased reaction rate and improved reactivity; however, the product selectivity and energy efficiency may be sacrificed in the meantime due to expedited competing reactions and severe heat loss to the surrounding environment.

Similar to resistive heating, microwave heating is also highly selective because thermal energy is mostly converted from the adsorbed electromagnetic energy via a microwave-susceptible material. The ability to absorb electromagnetic energy largely differs by material, offering the opportunity to achieve selective heating and local thermal effects. One application of the selective heating feature is the suppression of undesired gas-phase chemistries while facilitating heterogeneous catalysis pathways[29,33,35,36] (Fig. 4b). In addition, the localized heating potentially offers better energy efficiency compared with conventional reactor designs powered by combustion. From a fundamental viewpoint, the mechanistic role of microwaves in chemical synthesis has been subject to heated discussion[58]. Whereas the heat generated through microwaves has been widely recognized to play a key role in promoting chemical reactions, it has been argued that the enhanced performance observed in microwave reactors could be attributed to the electromagnetic field exerted on the reactants and intermediates, which potentially leads to reduced activation energy and/or an improved reaction rate. Resolving such debate may rely on decoupling the temperature and electromagnetic field modulations, which can in turn be used to program microwave reactors for unconventional reactivities. To scale up microwave reactors, similar protocols of resistively heated systems (Fig. 3a) may be adopted while taking into consideration the microwave penetration depth, which is determined by the microwave frequency and material properties. In addition, it is important to note that increasing the throughput of microwave-assisted synthesis requires scale-up of the microwave generator, power input, control unit and reaction chamber, sometimes making the energy efficiency and capital cost less favorable for practical implementation. Further research is needed to develop more cost- and energy-efficient settings.

Induction heating is contactless, like microwave heating, but it is also pseudo-resistive, similar to Joule heating. Induction heating relies on an alternating electromagnetic field to induce an eddy current that flows inside a conductor for heat generation. During induction heating, modulating the power frequency and material properties can result in an adjustable heat penetration depth. This phenomenon is sometimes referred to as the skin effect, which may have a ubiquitous impact on electrothermal chemical synthesis. Compared with Joule heating that often uses direct current for electrifying the entire heating element, the skin effect generated by the alternating electromagnetic field potentially induces even more efficient heating[20], such as on a ferromagnetic catalyst surface[21], thereby improving the instant tunability for electrified catalytic processes. Compared with other electrothermal chemical synthesis approaches, inductively heated chemical reactors sometimes require not only high-frequency, high-voltage power generators but

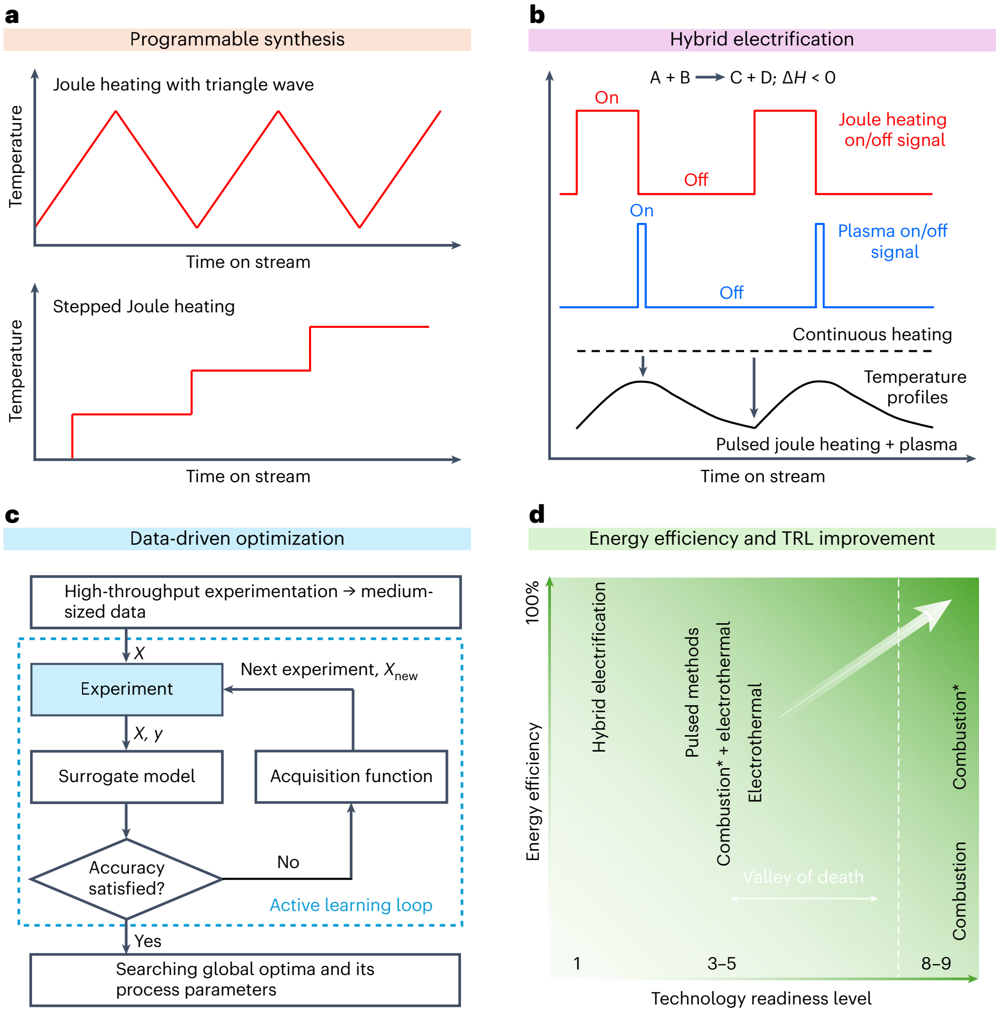


Fig. 5 | Potential future directions for electrothermal chemical synthesis. **a**, Programmable synthesis, which involves digital control over temperature or other formats of energy input with temporal tunability. **b**, Hybrid electrification, which integrates two or more electrothermal approaches (for example, adding plasma to a Joule-heated system) for fine control of the energy input along the reaction coordinate. Δ*H*, change in enthalpy. **c**, Data-driven optimization via active learning, which can be combined with high-throughput and/or autonomous experimentation for high efficiency and a desirable reaction outcome. *X*, independent variables; *y*, objective response. **d**, Energy efficiency and technology readiness level (TRL) improvement for various electrothermal chemical synthesis technologies toward practical implementation. Note that the absolute positions of various technologies are not meant to be quantitative, but rather a qualitative reflection of our assessment of literature reports. The plot suggests a general comparison of various technologies and future goals of electrothermal methods for the synthesis of commodity chemicals. 'Combustion*' refers to 'Combustion' operations plus extensive heat recovery and process integration[64]. The vertical dashed line schematically shows the transition from pre-industrial exploration to industrial implementation.

also thick coils wrapped around the outside of tubular-shaped vessels[22,50], which could incur obstacles for reactor volume reduction, cost reduction and improvements in energy efficiency. However, it is important to note that the scale-up of inductively heated chemical reactors may greatly benefit from the well-developed induction furnaces used for material processing at decent scales. Combining with recent progress made on efficiency improvement and rational reactor design, the industrial implementation of inductively powered chemical manufacturing may become possible in the near future.

Different from other electrothermal approaches, plasma-assisted reactions can involve energetic electrons, ions, electronically and vibrationally excited molecules and thermally generated radicals[45]. These species can be generated homogeneously in the gas phase or locally on the catalyst surface. Taking advantage of the spatial generation of intermediates[45] (Fig. 4c) and energy perturbation[39] (Fig. 4d), plasmas can be used to enable new reaction pathways[59], achieve high reactivity[44], reduce the reaction temperature[60] or break the activation–adsorption scaling relationship[39]. Plasma-assisted reactors feature a variety of configurations such as dielectric barrier discharge, spark, corona, arc, microwave and glow discharge, which can be selected for the desirable energy input scale and electron number densities. Along with the unique characteristics of plasma-assisted reactions are the underexplored non-equilibrium reaction and catalysis mechanisms. From a fundamental viewpoint, an in-depth understanding of the energy transfer between electrons, excited states and neutral molecules remains lacking, hindering the design of a more efficient plasma-assisted process and the development of a geared catalytic material. From an engineering viewpoint, to efficiently implement plasma in thermochemical synthesis, it is crucial to accurately modulate the reaction temperature and the electron number density for well-controlled reaction kinetics and pathways. Furthermore, it sometimes remains challenging to maintain volumetric plasma for stable and continuous chemical synthesis under certain reaction conditions, such as elevated pressures and relatively low temperatures. These features make the scale-up of plasma-assisted chemical manufacturing processes more challenging compared with other electrothermal methods. In addition, plasma-assisted processes are known to feature relatively high energy costs and low energy efficiencies. These issues create challenges for implementing plasma for the large-scale manufacture of commodity chemicals. To address these issues, hybrid approaches that integrate plasma with other electrothermal methods may offer a reduced energy intensity while improving the control over the reaction temperature and pathway.

Outlook

Compared with conventional thermochemical operations, electrified approaches often involve distinctive heater and reactor configurations, heat-transfer modes, mass-transport characteristics, more tunable but rather complex process parameters, different kinetic features (that is, near- or far-from-equilibrium reactions) under dynamic conditions and new catalyst design considerations. Although such complexity creates challenges for optimizing the synthesis outcome and achieving system scale-up, it also bestows electrothermal chemical synthesis with numerous new opportunities that are unattainable via conventional methods.

Programmable reactions and dynamic catalysis

Most traditional thermochemical reactors lack instantaneous temperature tunability because of their large thermal inertia. By contrast, electrothermal reactors possess the ability to dynamically modulate the reaction temperature with a fine time–temperature resolution and unique programmability (Fig. 5a). For example, using pulsed Joule heating, a chain reaction that involves multiple consecutive intermediate processes can be halted in between by tuning the pulse heating duration and applying fast temperature quenching. This concept has been demonstrated in several pyrolysis reaction schemes[2,5]. For another example, using stepped heating, a mixture of substrates or intermediates with different reaction-temperature thresholds, physical-transformation temperatures and/or kinetics features can potentially be sequentially and selectively converted to valuable products[54]. This function may find applications in the handling of mixed feedstocks that is commonly seen in biomass and waste streams. In addition to programming the temporal temperature profile, one can also program the spatial temperature distribution via a gradient[5,56] or multilayer resistive heater design[17], constructing heterostructures made of materials with different microwave susceptibilities[32], taking advantage of the skin effect and ferromagnetic susceptibility during induction[20,21], among others. Other than programming the reaction temperature, the catalysis process can be programmed during chemical synthesis in a dynamic fashion[61,62]. For instance, the change of electric field in plasma can be used to manipulate the energy level and lifetime of surface species on catalysts[41]. As another example, a fast temperature swing in periodic resistive or induction heating can potentially lead to dynamic changes in the surface properties of a catalyst, such as strain and local atomic arrangements, thereby affecting the reactivity and/or selectivity[63].

Hybrid electrification

Although various electrothermal techniques are being developed and advanced, there has yet to be a hybrid approach for electrothermal chemical synthesis. Figure 4a compares the key characteristics of the four representative electrothermal synthesis approaches, showcasing complementary features that can be used for integration toward improved reaction outcomes. For example, recent studies have demonstrated that plasma can substantially lower the thermochemical reaction temperature and improve the reaction rate[59,60]. However, plasma-powered processes are known to feature relatively low product selectivity due to the presence of a variety of metastable species and the lack of process temperature control. By comparison, several reported electrothermal set-ups have enabled the accurate modulation of the spatial and temporal temperature distributions, and thus are capable of achieving excellent product selectivity[2,33,50]. As such, these processes can potentially be coupled with plasma to achieve high selectivities, fast rates and reduced operation temperatures (for energy saving). These benefits through hybridization may be particularly useful in exothermic processes. For example, when plasma is coupled with Joule heating, the plasma can be used to lower the operation temperature (through molecular vibrational excitation) compared with conventional steady-state operations. Meanwhile, the fast cooling by Joule heating can be used to shift the chemical equilibrium toward achieving a higher reactant conversion and product yield (Fig. 5b). Note that coupling two or more electrothermal methods may cause interruptions to each other's operation (such as induction and microwaves), which may result in undermined controllability and reaction performance. Moreover, the materials chosen for the reactor wall, catalyst, catalyst support, heating element, susceptor and other reactor components need to be considered as a whole to maximize the coupling effect.

Data-driven optimization and high-throughput experimentation

Electrified techniques typically offer larger amounts of variables and process parameters compared with conventional combustion-powered operations. Taking pulsed Joule heating as an example, the groups of Hu, Vlachos and co-workers demonstrated the tunability and programmability of Joule heating for a range of electrothermal chemical syntheses[2,5,11,13,15]. The heating rate, cooling rate, heating duration, cooling duration, peak temperature, trough temperature and pulse frequency can all be programmed through electrical and electronic control. Similar to Joule heating, non-resistive electrothermal synthesis methods are also capable of tuning the energy level, reaction temperature, energy input timescale and other parameters. To make the situation even more complex, integrating two or more electrothermal synthesis approaches can make the tunable process-parameter space expand exponentially. With the much larger number of variables for electrothermal synthesis compared with conventional operations, it is not practical any more to simply rely on trial-and-error for screening optimal reaction protocols. Fortunately, such a challenge could be tackled through data-driven optimization with machine learning algorithms (such as active learning) to identify global optima of the reaction metrics with a small- or medium-sized dataset[2]. In the meantime, to improve the accuracy of the learning model, it is also beneficial to develop high-throughput experimentation platforms to enlarge the dataset. The high-throughput experimentation protocol and set-up will probably benefit from the fast ramping rate and compactness of electrothermal reactors. In addition to synthesis, we envision that data-driven optimization coupled with high-throughput experimentation will take effect in multi-scale modeling, operando characterization and catalyst development in future electrothermal applications (Fig. 5c).

Energy efficiency and improvement of the technology readiness level

Conventional reactors powered by combustion typically operate at an energy efficiency of <50% but can achieve close to 95% via extensive heat recovery and process integration[64]. By comparison, it has been shown that volumetrically heated microwave reactors can achieve an energy efficiency of >90% (>98% with heat recirculation in a scaled setting)[65]. A recent study by Fan and colleagues developed an approach through frequency modulation to achieve a markedly improved energy efficiency of more than 85% for inductively heated reactors[20]. In parallel, an industrially established process of the Invista company[66] has also demonstrated a high efficiency of 89% through innovative inductively heated reactor designs. Whereas reactors powered by plasma are known to have relatively low energy efficiencies, and Joule-heated reactors are yet to be assessed for their energy efficiency at large scales, there exist great opportunities to enhance the energy efficiency as well as the reactor performance through hybrid electrification (Fig. 5b). Despite these promises, most electrothermal chemical synthesis technologies are at a relatively early stage. Many studies that reported a promising reactor performance were conducted at a small laboratory scale, a portion of which assessed the energy efficiency and reaction outcome as a function of the reactor size, studied experimentally and/or computationally[4,5,64,65]. It is important to note that the scale–performance relationship can serve as a critical criterion for evaluating the technology readiness level for each approach at an early stage. Owing to the extensive efforts from both academia and industry, most electrothermal methods can now reach technology readiness levels beyond 3 or 4, meaning that they are in the transition from proof-of-concept and laboratory-scale validation to testing and implementation in a relevant environment. It is expected that small businesses will probably play an invaluable role in carrying out various approaches across the technology 'valley of death' toward higher technology readiness levels and improved energy efficiency (Fig. 5d), for which concerted efforts and contributions from academia, large businesses and policymakers are equally, if not more, essential.

Bypassing electricity for decarbonization and efficiency improvement

Along with active research into electrothermal chemical synthesis, it is necessary to reflect on the concept of direct renewable-to-chemical conversion that bypasses the step of transforming renewable energy to electricity for generating process heat. For example, using light to directly synthesize chemicals via photothermal reactions[67] or to drive thermochemical reactors via concentrated solar power[68] could offer improvements in decarbonization capability and energy efficiency while reducing the energy loss during multi-step conversions (Fig. 5d). They can also enable process intensification while attaining the co-benefits of many aforementioned electrothermal processes, such as the broad temperature range and instant timescale tunability. In conventional photothermal synthesis, catalytic approaches have already received broad interest owing to the integration of both thermal and light-induced effects. In particular, photothermal catalysis could potentially enable milder reaction conditions (for example, reduced reaction temperatures), tunable product selectivity, enhanced catalyst stability, facile catalyst treatment and dynamic operation[69]. It is important to recognize that utilizing valuable photons merely for heat generation is not considered an efficient use of energy unless it can be coupled with the photo-generated charges (for example, electron–hole pairs) when a semiconductor and a metal co-catalyst are present[70]. For applying concentrated solar power to thermochemical synthesis, future efforts may first need to address the integration of its surficial light-to-heat conversion mode with the relatively more scalable volumetric chemical reaction systems through receiver and reactor designs. In addition, the ability of accommodating the reactor to an intermittent solar resource will play a pivotal role in determining the scalability and applicability of the process for chemical manufacturing. Furthermore, the solar resources may not geographically align with the available chemical feedstock supplies. Heat-storage materials may be needed in such scenarios while the economic and energy efficiency factors should be systematically weighed.

Summary

Research interest in electrothermal chemical synthesis is growing rapidly, yet the pace at which the chemical industry widely embraces process electrification is comparatively slow due to the scalability challenges of electrothermal reactors, the high cost of electricity, the unsatisfactory durability of some reactor components and/or the large capital investment needed for electrothermal systems. In the foreseeable future, it is likely that electrothermal technologies will initially become a complementary piece to the existing infrastructure of the chemical industry for distributed chemical synthesis, before potentially replacing traditional reactors if the challenges can be well managed. Note that electricity is often considered to be more economically valuable than heat. As such, simply using renewable electricity to replace combustion for process heat generation is considered to be an undesirable form of energy transduction, unless predominant advantages of chemical synthesis performance over conventional methods can be obtained at meaningful scales. Future efforts should focus on rationally developing electrothermal operations with balanced economic and environmental benefits, which will help to build confidence for the chemical industry and stakeholders to better adopt these technologies toward greater sustainability and productivity.

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The authors declare no competing interests.

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