

Joule-Heated Catalytic Reactors toward Decarbonization and Process Intensification: A Review

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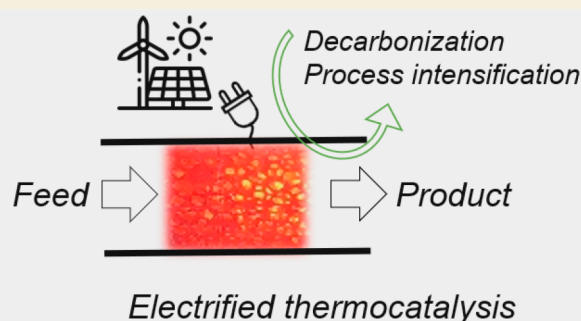
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ABSTRACT: The supply of the heat required for chemical processes via renewable electricity, i.e., process electrification, provides an alternative strategy for replacing conventional fossil fuel combustion. This approach enables fast, selective, and uniform heating, offers great potential for utilizing the excess renewable electric energy, and brings about an important chance for mitigating CO₂ emissions. In this work, we provide an overview of the state-of-the-art electricity-to-heat driven catalytic processes. The principle and fundamentals of Joule heating are provided and briefly compared to induction and microwave heating in view of electrifying catalytic processes. By this comparison, we assess that Joule heating can be regarded as the most promising method for process electrification, and its applications to methane reforming, cracking reactions, CO₂ valorization, and transient process operation are then reviewed. Advantages and disadvantages are critically addressed in terms of efficiency, potential for scale-up and possibility of retrofitting. The current challenges in the development of advanced electrified processes as well as the opportunities of next generation electrification techniques are discussed.

KEYWORDS: electrification, Joule heating, decarbonization, process intensification, methane reforming, cracking reactions, CO₂ valorization, transient operation



1. INTRODUCTION

To meet the demand of an increasing global population, unprecedented amounts of fossil fuels are used as a source of energy and feedstocks to produce chemicals, which releases significant amounts of greenhouse gas emissions and pollutants.^{1,2} It is estimated that energy consumption by the chemical industry reached 43 EJ/y in 2015, together with roughly 3.3 Gt CO₂eq emissions released to the environment.^{3,4} A significant share of fossil fuels is being burnt to sustain the heat of chemical reactions, which is responsible for approximately 0.7 Gt CO₂eq emissions.⁴ The energy intensive chemical sectors, such as ammonia, ethylene and methanol synthesis, contribute much to the overall production of CO₂ emissions. For example, the ammonia sector releases approximately 550 Mt CO₂eq (fuel combustion, decoking and utilities included) with an energy consumption of 4.5 EJ/y (2018),^{3,5,6} and the trend shows a constant growth as suggested by the IEA (International Energy Agency) outlook.⁷ The huge CO₂ emissions are associated with severe environmental challenges and their control to reduce the climate impact has become an urgent global need.^{8–10}

The share of renewable energy sources (RES) is continuously growing: it is estimated to reach 64–97% in Europe within 2050.¹¹ By exploiting sun and wind, most of the

installed renewable energy capacity is considered “non-programmable”, due to its intermittent nature. This actually represents a limitation in the maximum amount of “renewable electric power” that can be injected in the grid since precise control of energy supply and demand is needed to keep the electric grid under control. As previously discussed, the chemical sector is responsible for a great share of the energy consumption, therefore it may represent a relevant end-user if electrification strategies become widely employed.^{12–15} In this regard, with renewable electricity becoming more and more accessible, “Power-to-X” represents a promising way to supply the energy for chemical processes to address the challenge of decarbonization.^{2,16–25} This will lead to a transition to an electrified chemical industry, as suggested by Centi and Perathoner in their recent review paper.²⁶ The transition to renewable electricity-based chemical production requires novel

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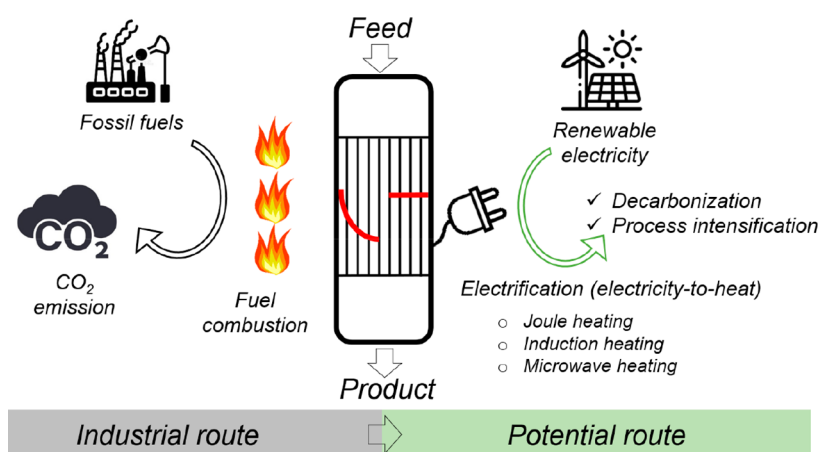


Figure 1. Concept of industrial route (fuel combustion) and potential route (electrification) to sustain the heat of endothermic chemical reactions. (The figure has been made by the authors by using the Microsoft Office Power Point application).

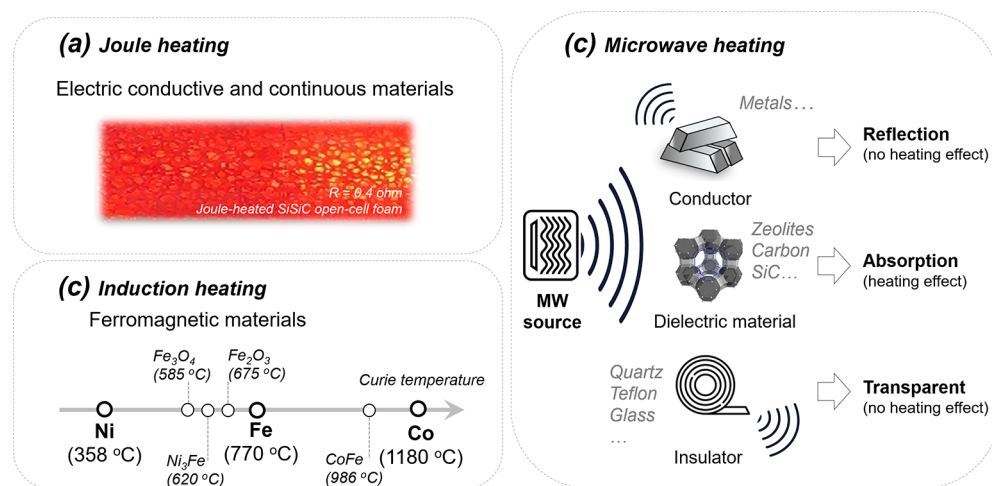


Figure 2. Material requirements of heating elements in designing electricity-to-heat driven catalytic reactors with (a) Joule heating, (b) induction heating, and (c) microwave heating. (The figure has been made by the authors using Microsoft Office Power Point application).

technical solutions as well as proper infrastructures in order to utilize the energy as efficiently as possible.¹⁸ As already well reviewed by Stankiewicz and Nigar,¹⁷ electricity can be applied to chemical processes based on the electron or charge transfer, such as in electrocatalytic, photochemical/photocatalytic, and plasma-based processes.^{27–30} However, these approaches involve nonthermal interactions with the catalyst active sites, and therefore, alongside thermal energy transfer, other complex phenomena may take place. These approaches are widely studied and may lead to a substantial contribution toward the reduction of CO₂ footprint of several chemical processes, such as direct water electrolysis for H₂ production.^{31,32} On the other hand, the electrification methods by conversion of electric energy to heat (direct heating) in catalytic reactors ensures higher technology readiness levels.²⁵ It should be pointed out that electrification is not only possible for bulk chemical production but also very attractive for other processes. For example, it has been already commercially applied in exhaust gas aftertreatment systems, where, however, the primary aim is to reduce the reaction light-off temperature.^{33–37}

Electricity can be converted into heat and transferred to thermally driven chemical reactors in several ways, such as microwave heating,^{38–42} induction heating,⁴³ as well as Joule

heating^{35,44–48} (also known as ohmic heating or resistance heating). As shown in Figure 1, those methods are promising to replace fossil fuel combustion heating and have been explored recently in several industrial processes for chemical syntheses.^{17,49–52} In addition to the reduction of CO₂ emissions by replacing fuel combustion, electricity-to-heat driven catalytic processes may enable significantly enhanced heat transfer as the catalysts could be located in intimate contact with the heating elements. This may enable process intensification with the design of compact reactors and reduce the intrinsic heat transfer limitations that affect some of the most widespread chemical processes. Moreover, unlike conventional externally heated reactors, where heat is uniformly distributed throughout the reactor, electrified reactors often selectively target the catalyst for heating, since in the vast majority of the applications either the catalyst support or the active phase is heated directly. Furthermore, such approaches favor the suppression of gas-phase side reactions, leading to enhanced product selectivity.^{53–57} Different from induction heating or microwave heating, which require the conversion of electrical energy into electromagnetic energy and are therefore affected by transformation losses, Joule heating is in principle the only approach that enables the direct transformation of electrical into thermal energy, so that a

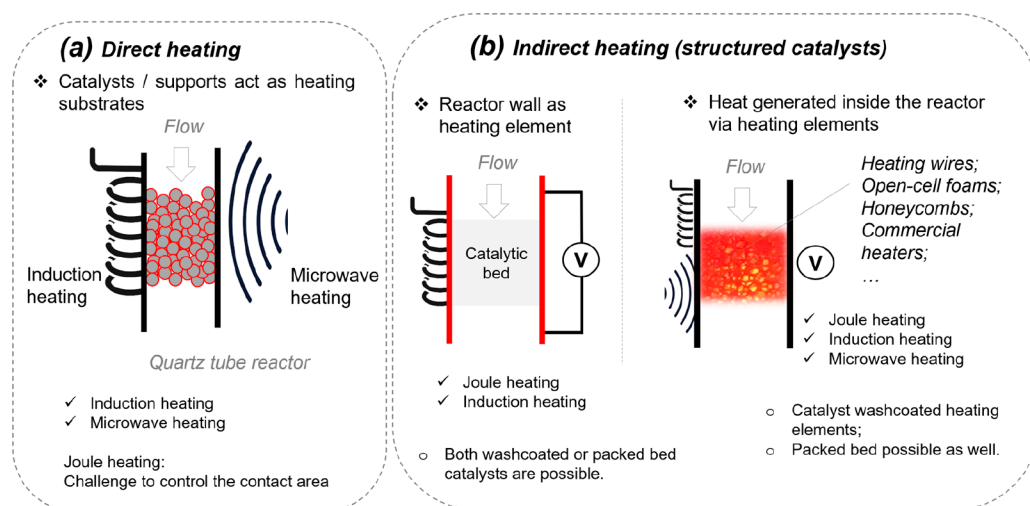


Figure 3. Heating strategies of the state-of-the-art electricity-to-heat driven catalytic reactors: (a) direct heating and (b) indirect heating with structured catalysts. (The figure has been made by the authors using Microsoft Office Power Point application).

Table 1. Comparison of Different Electricity-Driven Heating Methods in Terms of Principle, Material Requirements, Heating Strategies, Advantages, and Limitations in Catalytic Applications

	Joule heating	Induction heating	Microwave heating
Principle	Electric energy to thermal energy	Radio frequency energy to thermal energy	Electromagnetic energy to thermal energy
Material requirements	Conductive and continuous materials	Ferromagnetic materials (Ni, Co, Fe)	Dielectric materials
Heating strategies	Structured catalysts; reactor walls; embedded heating elements	Pellet catalysts; structured catalysts; reactor walls	Pellet catalysts; structured catalysts
Advantages	Existing experience in using electric heating for domestic and industrial applications; possibility of retrofitting	No need to insert electric circuits inside the reactor; decoupled system for control	No need to insert electric circuits inside the reactor; decoupled system for control
Limitations	Challenging with pellet catalysts	Not applicable to all materials (only ferromagnetic); need material transparent to electromagnetic waves; achieve field uniformity	Not applicable to all materials; need material transparent to electromagnetic waves; achieve field uniformity

theoretical energy efficiency (the ratio of enthalpy gain to the input electric energy) of 100% can be expected.⁵⁸

In view of exploiting readily available renewable electricity to address the challenges of decarbonization and process intensification, this work reviews the recent developments in electricity-to-heat driven catalytic processes, with a particular emphasis on the Joule heating method. The principle and fundamentals of Joule heating are discussed and compared to induction and microwave heating, with a focus on their potential for electrifying catalytic processes. The reported applications of Joule heating in methane reforming, cracking reactions, CO₂ valorization, and transient process operation are reviewed. Furthermore, the challenges and opportunities in developing advanced electrification processes are discussed.

2. FUNDAMENTALS OF ELECTRICITY-DRIVEN HEATING IN CATALYTIC PROCESSES

In catalytic processes, electricity-driven heating can be applied to heat the catalyst, the catalyst support, the reactor tubes, or a combination of those.^{59,60} It is also possible to locate heating elements outside the reactor shell: for example, BASF, SABIC, and Linde have recently developed a demonstration plant with large-scale electrically heated furnaces integrated into one of the existing steam crackers in Ludwigshafen, Germany.⁶¹ The periodic reactor replacement is easier if heating elements are located outside. On the other hand, however, excellent heat transfer could be expected if the heat is directly generated inside catalytic reactors, as the catalyst is in more intimate

contact with the heating sources. Herein, we mainly focus on methods where heat is directly generated inside the reactors.

This section aims to provide the reader with a brief description of the different electricity-driven heating methods, i.e. resistance heating, in comparison to induction and microwave heating. The material requirements (Figure 2) and general heating strategies (Figure 3) in heterogeneous catalysis are summarized. A short comparison of different electricity-driven heating methods in terms of principle, material requirements, heating strategies, advantages, and limitations in catalytic applications is provided in Table 1. These aspects will be discussed in detail in this section.

2.1. Joule Heating Fundamentals

Joule heating, also known as resistive or ohmic heating, is a process in which the electric energy is transformed into thermal energy when an electric current flows across an electrical conductor. In general, a voltage difference between two points of a conductor creates an electric field that accelerates charge carriers in the direction of the electric field, giving them kinetic energy. When the charged particles collide with ions in the conductor, the particles are scattered. Their direction of motion becomes random rather than aligned with the electric field, which results in an increase in the atomic vibrations. Thus, energy from the electric field is converted to thermal energy. The electric power (P) generated by the electrical conductor can be calculated based on the well-known Joule's law:

$$P = I^2R \quad (1)$$

where R is the resistance of the electrical conductor and I is the electric current flowing through the conductor.

A wide range of materials can be applied as resistance heating elements. In general, the materials should be highly temperature resistant. Although the general concept of Joule heating applies to various materials, the efficiency and dominant mechanisms can vary significantly depending on the material's electrical and thermal properties. Metals are most associated with efficient Joule heating, and ceramics are often chosen for their insulating properties. Carbon materials fall somewhere in between, depending on their specific properties. Ideally, if all the electric power is directly transferred to heat, the resistance heating can theoretically reach an energy efficiency of 100%.⁵⁸ In the design of the elements, it is imperative to bear in mind that all materials are characterized by a maximum dielectric voltage (DV, V/m): higher values may lead to spontaneous electric discharges in the material. Moreover, the maximum current density that can be carried by the element in (A/m²) should be carefully checked when designing such systems.

Joule heating is applicable when a given material, i.e., a resistor, is connected to an electric circuit. The Joule heating substrate should be physically continuous, therefore the direct heating of pellet catalysts by Joule effect is very challenging since it is practically difficult to control the size of the contact area of conductive particles.^{35,62–67} On the other hand, the Joule heating concept can be applicable to systems with structured catalysts,^{59,68,69} where the heating elements can be catalyst coated reactor walls,^{44,70,71} or catalyst activated heating elements such as heating wires,^{66,67} open-cell foams,^{72–74} or even some commercial heaters,^{75–78} as summarized in Figure 3.

In general, one of the complexities associated with such Joule-heated catalytic processes is the necessity of connecting the heating element to an external electric power source despite the high local temperatures, possibly high pressures, and corrosive environments. Joule heating has already found extensive use in both domestic and industrial applications, and the knowledge gained from these applications can be readily applied to the field of catalytic processes. Compared to conventional metal resistors, the electrically conductive ceramics appear to be promising substitutes that offer higher stability in high temperature or corrosive conditions and in general offer higher resistivities, which allows operating the processes with reasonable currents for the same heat duty.^{79,80} Moreover, advanced additive manufacturing, such as 3D printing, enables tuning the electrical resistivity of the ceramic composites in order to precisely control the temperature distribution inside the reactor, as reported by Klemm and co-workers (CHEMampere project).⁸⁰

2.2. Other Electricity-Driven Heating Technologies

2.2.1. Induction Heating.

Induction heating exploits the electromagnetic properties of the material, which in the presence of an alternating electromagnetic field creates eddy currents (circular electric currents generated in metals by variable magnetic fields) inside the material to be heated. To generate such a field, an induction coil (typically a long and thick copper rod wound around the object) is connected to a high-frequency and high voltage generator. The rapidly alternating magnetic field generated by the current that flows in the external conductor penetrates the object and can

generate eddy currents if the material is ferromagnetic. The frequency of the electric current used for induction heating depends on the object size, material type, coupling between the working coil and the heated object, and field penetration depth. An important feature of the induction heating process is that the heat is generated inside the object itself, and there is no need for any contact between the coil and the material to be heated. This method is widely employed in the process and metallurgical industry, in the food industry, and in crystallography, thanks to the possibility of avoiding contaminations between the heating element and the processed materials. This can be considered a great advantage for its potential application in chemical processes.

Induction heating was adopted as heating method for several heterogeneous catalytic processes,^{81–84} However, the catalyst susceptible to induction by hysteresis heating requires ferromagnetic components with a Curie temperature (i.e., the temperature above which those materials lose their permanent magnetic properties) similar to the reaction temperature. Only iron, cobalt, nickel, and their alloys are ferromagnetic above room temperature; some examples are shown in Figure 2b. This remains one of the biggest limitations for direct induction heating of catalysts. One possible solution could be that of mixing the catalysts with other ferromagnetic components which act as induction susceptors and afterward transfer the thermal energy to the catalytic sites, as reported, e.g., in the work of Mortensen and co-workers.^{85–88}

Concerning the reactor design, nonferromagnetic quartz or glass can be used as reactor materials at the lab scale, and the ferromagnetic materials inside the reactor can be directly heated up by induction heating.^{85–87,89–91} In addition, the stainless-steel tube reactor can as well be applied, and in this case the reactor tube is directly heated by induction heating, driving the catalytic reaction on the catalyst inside the reactor tubes.⁹² However, at the industrial scale, with the application of stainless-steel tubes, not only can the catalyst be heated up but also the tubes can be heated up by eddy currents. In this regard, it is suggested that thin tubes be applied in order to improve the energy efficiency.

During energy transfer to the target materials by the induction process, energy losses could occur via various pathways.⁸⁷ Even though the energy losses can be minimized by improving the design of the reactor, such as by using high radio frequency and long/narrow coils, to the best of our knowledge the energy efficiency reported so far for induction heating of catalytic processes remains low (below 25%).^{85–87,89–91} Considerations about the possible efficiency improvements when scaling-up are reported in a recent work by Almind and co-workers:⁵⁰ an energy efficiency up to 80% could be expected upon upscaling, according to their theoretical estimation.

2.2.2. Microwave Heating.

Microwave (MW) heating is based on the principle that a material absorbs electromagnetic energy in the form of microwaves and converts it into thermal energy. As a noncontact heating method, it is considered as an alternative to traditional heaters because of its advantages, namely, direct energy transfer to the target body.^{41,93} Due to the selective heating of solid absorbers, microwave-driven processes can effectively prevent undesired gas-phase reactions in heterogeneous catalytic reactors, leading to improved product selectivity.⁵⁴ This concept has been demonstrated by Santamaria and co-workers in multiple MW-driven reaction

systems, including the dehydrogenation of isobutane, propane, and the epoxidation of ethylene.^{54–57}

The choice of the proper materials for the catalyst and for the reactor is fundamental to a successful microwave-assisted heterogeneous catalytic process. The microwave absorber requires polar molecules to have an electric dipole aligned with the electromagnetic field. As shown in Figure 2c, based on their interaction behavior with microwaves, solid materials can be mainly divided into three categories:³⁸ (i) the conductors, such as metals, will reflect the microwaves, and thus, they cannot be directly heated with this method; (ii) the insulators, such as quartz, Teflon, and glass, are transparent to microwave and could be used as the windows to allow the microwaves entering the reactor; (iii) the dielectric materials, such as water, solvent, zeolites, and SiC, can absorb microwaves and thus can be directly heated by microwave heating.³⁸ It is also important to remark that the material properties also depend on the frequency of the wave; therefore, once the materials of choice for the application are fixed, it is possible to optimize the wavelength to maximize the heating efficiency. So far, no proof has been shown in the literature that microwaves could directly influence chemical reactions; i.e., microwaves do not interact with the molecules to enhance or steer chemical reactions.⁹⁴ Microwaves contribute to chemical reactions only by indirectly heating the system via microwave susceptor materials. The catalytic reaction enhancement that has been typically observed in the case of MW-heating systems is due to the temperature difference between the catalyst (which may be at higher temperature, since it is heated by waves) and the gas phase.

For heterogeneous catalytic processes, it is critical to design a proper reactor configuration that allows the microwave absorber to be directly exposed to the microwave source. In general, the MW generator is located outside the reactor. The glass- or quartz-based reactors,^{95–98} or the metallic reactors with transparent windows or a proper waveguide,⁹⁹ are commonly adopted for microwave reactors. In such cases, it is very challenging to operate the reactors in high pressure applications. Only few high-pressure microwave reactors were reported,^{100,101} where reactor tubes (SiC or metallic materials) worked as microwave susceptors, with reported operating temperatures only up to 300 °C. Reflection and transmission are the main sources of energy losses for microwave heating. Both reflection and transmission losses are in the form of microwave energy, which could be reabsorbed by the MW susceptor, whereas losses at the magnetron are converted into heat at low temperature. In reactors equipped with transparent windows, energy loss occurs when microwave radiation escapes through the windows or ports in the reactor cavity.

Different from the MW generator located outside of the reactor, generating the MW directly inside a metallic reactor could be one possible solution to prevent microwave escaping from the reactor either by reflection or transmission. But other challenges might arise: it is demanding for the MW generator to be directly exposed to high temperatures and possibly corrosive conditions from the gas feed. In this respect, a more complicated reactor design is required to avoid those issues.

2.3. Challenges for the Electrification of Catalytic Processes

The current electricity-to-heat driven reactor technologies are mainly in the proof-of-concept stage, facing many challenges.^{25,26,52,102} However, the very promising advantages,

such as the potential for decarbonization as well as for enabling fast and uniform heat distribution to address heat transfer limitations, make such technologies extremely attractive for both academia and industry. Moreover, such electricity-to-heat driven technologies are not limited to the reforming, cracking reactions and reverse water–gas shift reaction for CO₂ utilization,^{45,103} but are in general applicable to other endothermic processes.^{45,104} With renewable electricity becoming more accessible, those techniques will play a more and more important role in the near future and may become enabling technologies for novel processes. In this section, the technical challenges of those electrification approaches in catalytic processes are discussed. It should be mentioned that other hurdles, such as economic, organizational, and regulatory barriers, need to be overcome to fully utilize the potential of electrification. Intimate cooperation between the chemical industry, energy sector, equipment suppliers and governments is necessary to accelerate the innovation and implementation of electrification technologies.¹⁰⁵

2.3.1. Fluctuating Nature of Renewable Electricity. In addition to hydroelectric and geothermal energy as renewable energy sources, which can possibly ensure a continuous supply, other sources, such as solar and wind, suffer from the fluctuation issue. Currently, there is still a lack of efficient large-scale electricity storage solutions. Therefore, the direct electrification of catalytic processes suffers from the intermittent nature of the renewable electricity supply. This requires electrified reactors with high flexibility, which, i.e., can be operated in dynamic conditions with very fast changes of the input energy supply and a good thermal response along the catalytic bed and, besides, storage strategies for the feedstocks. In this regard, process intensification could represent a very promising way to address such challenges, as small systems offer high flexibility and low operative costs during startup and shutdown.^{106–109} It should be considered however that not only the reactor but also all the up- and downstream operations (e.g., separations) would need to be redesigned. Also the high level of heat integration/recovery makes running a process for just a few hours difficult.

On the other hand, operating the reactors in hybrid mode, i.e., combining electrification with conventional fuel combustion heating, can be a useful solution to mitigate the fluctuations and for the near-term future where only a limited share of renewables will be present in the electric grid. In our opinion, it is very demanding for induction heating to operate in combination with conventional fuel combustion as such an approach requires coils around the reactor, which is extremely challenging in the case of high temperature radiative heat transfer. Moreover, the presence of coils could lead to severe heat transfer limitations. Resistance heating, however, shows high potential for such a hybrid mode operation, for example in electrified methane steam reforming configurations based on the direct Joule heating of washcoated foam catalysts inside stainless-steel tubular reactors,⁷³ or with the use of conductive internals combined with embedded heating elements.⁶⁰ In such cases, the stainless-steel tube reactor can be heated up easily from outside by conventional heating thus achieving hybrid operation. In our perspective, it is also practically possible to integrate the systems with double beds, one heated conventionally and one heated electrically and operated cyclically, as a function of renewable energy availability, at the expense of greater capital cost. Such concepts are promising to solve the mismatch issue between continuous

Table 2. Summary of the Sources of Energy Losses from Different Electrification Methods, i.e., Resistance Heating, Induction Heating, and Microwave Heating

Electrification method	Source of energy loss	
	Can be recovered	Hardly recovered
Resistance heating	Heat dissipation from the reactor	Ohmic losses; transformation losses
Induction heating	Joule heating of the coil; heat dissipation from the reactor; Transformation losses	Induction heating of the insulation materials; Other types of losses such as from the energy transformation or from the connections
Microwave heating	MW transmission; Heat dissipation from the reactor; Transformation losses	MW reflection

chemical processes and the intermittent renewable energy supply. More research and development efforts should be devoted to further exploring such possibilities in the future. It should be mentioned that electric heating is the first choice in a normal operation, while a hybrid electricity supply, i.e., renewable electricity heating backed up by conventional fuel combustion, can be advised considering the intermittent nature of the renewable electricity supply.

2.3.2. Energy Loss and Recovery. For moving the direct electrification techniques to industrial applications, it is critical to identify the sources of possible energy losses and address the recovery of such losses. The sources of energy losses from different electrification methods are summarized in Table 2. For resistance heating, it is worth emphasizing that the system may suffer from different kinds of losses: (i) ohmic losses that are present in the electric circuit adopted to connect the heating element; (ii) thermal losses of the reactor/system that are not dependent on the heating element; and (iii) transformation losses if the voltage required is different from the voltage of the electric grid or if the frequency adopted is different from the grid frequency. The ohmic losses can be minimized by operating the system at high voltage and low current, depending on the thermal power demand and the element properties. The energy losses from contact resistance or the heat dissipation from the reactor are in the form of thermal energy, and the latter one can be more easily recovered by heat exchange, for example, to preheat the feed streams.

For induction heating and microwave heating, the electric energy should be first transformed into electromagnetic energy, with this process inevitably resulting in transformation energy losses, which are in a tiny fraction. The losses of this equipment generate low grade heat that may be recovered by, i.e., producing hot water, but this heat source is not useful if the main objective is providing high temperature heat. Reflection and transmission are the main sources of energy loss for microwave heating.¹¹⁰ Both losses are in the form of microwave energy. The transmitted part can be recovered by using another microwave absorption material located at the outlet of the MW window. However, this requires a nonconventional reactor design, since it cannot be recovered directly with a heat exchange or via steam generation. The reflected part is difficult to recover; however, it is possible to reduce such losses by a better design of the reactor configuration.¹¹⁰ There are three main sources of energy losses in induction heating: (i) energy loss through the insulation material which is in the form of radio frequency energy; (ii) energy loss by Joule heating of the coil, in this case the energy is in the form of thermal energy and can be possibly recovered by heat exchange; (iii) other types of losses such as from the energy transformation or from the connections, this part of energy is hardly recovered and sometimes cannot be

avoided; however, it can be minimized by improving the reactor design. In general, the energy losses due to the voltage transformation are inevitable in the overall system, even exploiting different heating methods than Joule heating. Moreover, renewable energy sources are generally located far from where chemicals are produced.¹⁸ The transportation of electric energy over distances will involve energy losses.

In terms of energy efficiency, defined as the ratio of enthalpy gain to the input electric energy, for Joule heated systems, as we reported in Zheng et al.,⁷² to our knowledge, the highest energy efficiency recorded experimentally at the laboratory scale exceeds 80%. In the experimental system, Joule losses in the wirings are expected to be <3%, and since a DC power generator was used, no transformation loss is present. Toward scaling up the concept, we expect that the energy efficiency of the reactor can be up to 95% with all losses being due to conduction in the insulation layer,^{72,111} whereas it is complex to estimate Joule losses in the wirings since the effective current employed and the reactor design will have an impact. In the case the grid voltage needs to be adjusted, the typical efficiency of an electric transformer exceeds 98%.

For induction heating, we refer to the excellent work of Almind et al.⁸⁷ that quantifies the energy loss at the laboratory scale and tries to extrapolate it in an industrial scenario. At the laboratory scale, a thermal efficiency of 23% was reported. Scale-up calculations performed by authors demonstrate the possibility to reach 80% overall efficiency since the insulation and the coil losses seem invariant with the scale considered.

In the case of microwave heated reactors, the big bottleneck in reaching high efficiency is the power loss from transformation, where efficiencies at the magnetron are still <75% in state-of-the-art equipment.¹¹² Recent works demonstrate that, for some applications, it is possible to absorb >90% of the power fed to the system.⁹⁵ In the case of low-temperature applications, it is possible to recover the energy in the form of sensible heat, but this is a clear limitation in the case of high-temperature applications.

2.3.3. Challenges with Scaling up and Retrofitting.

The transition from conventional fuel combustion to direct electrification of chemical productions requires novel technical solutions, as well as proper infrastructures in order to utilize the energy as efficiently as possible. The challenges for different electrification methods are summarized in Table 3. Metal-based reactors do not allow to transmit microwaves into the reactor, which makes them challenging for microwave heating applications at high temperatures, even high-pressure applications up to 300 °C reported.^{100,101} The glass- or quartz-based reactors or metallic-based reactors with transparent windows could be applied; however, they are hardly suitable for high pressure applications. In the case of induction heating of metallic reactors, not only the catalyst but also the reactor

Table 3. Summary of the Challenges for Different Electrification Methods, i.e., Resistance Heating, Induction Heating, and Microwave heating

Electrification method	Challenges
Resistance heating	Challenge with direct heating of pellet catalysts; reactor tube as heating element: mass transfer limitation; structured catalysts: current flows uniformly into the reactor
Induction heating	Catalyst as susceptor: not applicable to all materials (only ferromagnetic); challenge with quartz tube for high pressure applications; challenge with improving energy efficiency
Microwave heating	Reactor tube as heating element: limited reaction temperature; challenge with quartz tube for high pressure applications; challenge with improving energy efficiency

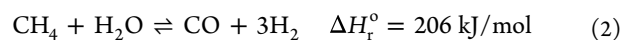
will be heated by the eddy currents mechanism at the typical temperatures required for the reactions discussed so far. Therefore, it is suggested to scale down the reactor size, using, e.g., small tubes, so that a higher portion of electromagnetic energy can be utilized to directly heat the catalyst, enabling a higher energy efficiency, and to design proper external cooling systems to not exceed operating temperatures of the pressure-shell materials. Another solution can be the design of a heat recovery system such as, for example, a bayonet-type reactor. Resistance heating is demanding for direct heating of pellet catalysts, which seems only feasible via heating elements.⁶³ The heating elements can be either dedicated elements inserted inside the reactor or just the reactor tube walls, as reported by Wismann and co-workers.^{44,70,71,113} For direct heating of the reactor wall, it is suggested to adopt reactor tubes with small diameter, since external mass transfer limitations will severely affect the process in the case of large tubes.⁷⁰ This leads to a possible final solution of adopting Joule-heated narrow-channel monolith catalysts for intensified processes.

Eventually, the transition to applications where the heat of reaction is provided by electrification may require the overall scheme of the conventional processes to be modified. For example, the purge gas from conventional methane steam reforming is recycled and burnt to supply the reaction heat. However, in the electrified methane steam reforming, the combustion section is removed, so the purge gas (rich in methane) can be recycled (after a shift converter, a CO₂ removal unit, and a pressure swing adsorption unit) to the feed section in order to allow further methane conversion, as suggested by Natrella and coworkers.¹¹⁴ In some process configurations where additional heat is required, these gases can be used to generate steam. However, detailed process analysis studies are required to identify the optimal use of these streams.

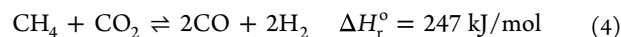
3. METHANE REFORMING DRIVEN BY JOULE HEATING

3.1. Methane Reforming: From Fuel Combustion to Electrification

As an efficient and clean approach for delivering high-quality energy services, the hydrogen-based energy system is widely regarded as a promising option toward a sustainable future.¹¹⁵ Hydrogen is also the basis of several large scale chemical and petrochemical processes, e.g., ammonia synthesis and hydrocracking reactions. Currently, methane steam reforming (MSR) accounts for more than half of the global hydrogen production.⁴⁴ In the reformer, hydrogen production is governed by methane reforming reaction (eq 2) and the water–gas shift reaction (WGS, eq 3).^{116,117}



Another process currently envisioned for H₂ production and CO₂ conversion is dry reforming (DR, eq 4). Such a process produces synthesis gas by reacting CH₄ and CO₂. The current challenge for methane dry reforming in industrial scale applications is the lack of proven commercial catalysts with sufficient high activity, stability, and resistance against carbon deposition.^{92,118}



Methane steam reforming is a highly endothermic reaction. The heat transfer limited industrial scale process is typically operated in multitubular reactors with very long and slender tubes (10–14 m long, ~10 cm diameter) at temperatures in the range of 800–1000 °C and pressures up to 35 bar.^{115,119} The tubes are loaded with catalyst pellets (typically Ni-based catalysts). The reactor is operated at high flow rates in order to maximize the heat transfer between the catalyst pellets and the reactor tubes. The heat of reaction is provided by fuel combustion; i.e., the tubes are externally heated by burning an additional amount of methane and, in some configurations, tail gases. The flue gases at the outlet of the reactor are then used to produce the steam required for the reaction and extra-steam for export in order to maximize heat recovery from flue gases. On one hand, the heat transfer limitations of the methane steam reforming process has been a subject of research for decades, especially when trying to operate the system at the small scale.^{49,50,120–123} On the other hand, the fuel combustion is responsible for roughly half of the CO₂ emission of this process.^{124–126} It is estimated that the industrial MSR process accounts for approximately 0.5% of global CO₂ emission.¹²⁷ Another relevant aspect is that a relatively high dilution makes

Table 4. Summary of Recent Developments for the Direct Joule Heating of Methane Reforming Reactions

No.	Reactor configuration	Conditions and outcome	Ref
1	FeCrAl-alloy tube washcoated with Ni-based catalyst	CH ₄ /H ₂ O/H ₂ = 30/60/10; 50 mbar; 102 L/h; X _{CH₄} = 87% at 700 °C	44, 70, 71, 113
2	SiC heating element (inside reactor) washcoated with Ni-based catalyst	CH ₄ /H ₂ O/Ar = 1/3/7; CH ₄ /CO ₂ = 1/1; GHSV = 182 h ⁻¹ ; X _{CH₄} = 70% at 790 °C (steam reforming); X _{CH₄} = 84% at 760 °C (dry reforming)	76
3	FeCrAl-alloy (inside reactor) coated with LaNi _{0.95} Ru _{0.05} O ₃ catalyst	CH ₄ /CO ₂ /N ₂ = 1/2/7; GHSV = 5470 h ⁻¹ ; X _{CH₄} = 29.4% at 900 °C	75
4	FeCrNi-alloy (inside reactor) coated with Ni-based catalyst	CH ₄ /H ₂ O/N ₂ = 1/3/2; GHSV = 157 000 cm ³ /h/g _{cat} ; X _{CH₄} = 97% at 700 °C	139
5	SiSiC foam (inside reactor) washcoated with Rh-based catalyst	CH ₄ /H ₂ O = 1/4.1; GHSV = 150000 cm ³ /h/g _{cat} ; X _{CH₄} = 96% at 700 °C	73

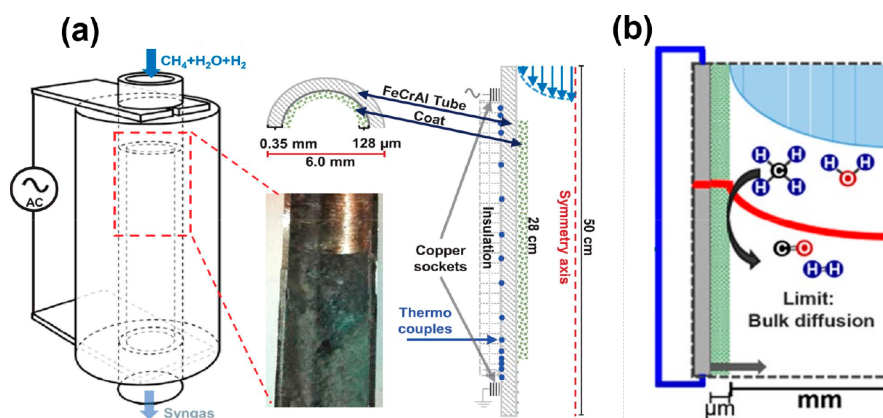


Figure 4. Electrified methane steam reforming proposed by Wismann and co-workers: (a) lab-scale reactor configuration,⁴⁴ (Adapted from ref 44. Copyright 2019 AAAS) and (b) reactor performance governed by diffusion⁷⁰ (Adapted with permission from ref 70. Copyright 2019 American Chemical Society).

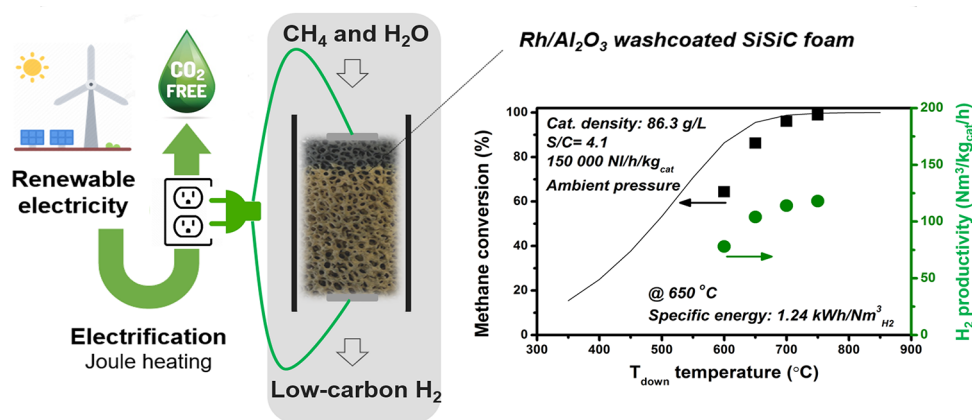


Figure 5. Direct Joule heating of the catalyst washcoated Si-infiltrated silicon carbide (SiSiC) foam for methane steam reforming.^{72,73} (Reprinted from refs 72 and 73. Copyright 2023 Elsevier and Copyright 2022 John Wiley and Sons, respectively).

the recovery of CO₂ from the flue gas (8–10% CO₂) more difficult and expensive than from the main process stream (approximately 45% CO₂, considering PSA off-gases), thus negatively affecting the economics of “blue hydrogen” production. In this regard, a novel strategy is urgently required for low-carbon hydrogen production via methane steam reforming.

Replacing the combustion heat in conventional fuel fired reformers with electrification is regarded as a very promising solution, also when considering the economically competitive prices of the renewable electricity compared with fossil fuels.^{128–130} Process simulation results from different studies show that the novel electrified methane steam reforming process is of great interest from energy, environmental and economic point of view.^{103,114,131–136} The specific power consumption per hydrogen production estimated for electrified methane steam reforming (approximately 1 kWh/Nm³ H₂ considering only electric input)¹¹⁴ is remarkably lower than that in water electrolyzers (3.8–4.5 kWh/Nm³ H₂).^{114,137} It should be noted that approximately 4% of the world’s hydrogen is derived from water electrolysis,¹³⁸ whereas conventional methane steam reforming contributes to over 70% of the global hydrogen production.⁴⁴ In this regard, the recent developments in direct Joule heating of methane reforming reactions are reviewed, as shown in Table 4. The adopted catalysts and heating elements are summarized.

3.2. Joule-Heated Methane Reforming

The direct electrification of methane steam reforming by Joule heating was already reported in 1992 by Spagnolo and co-workers,¹⁴⁰ who washcoated a Ni–Al₂O₃ catalyst onto a resistive metal screen. More recently, Wismann and co-workers proposed an innovative reactor concept with an electrically heated washcoated FeCrAl-alloy tube (OD = 6.0 mm, ID = 5.3 mm).^{44,70,71} The steel tube is connected to an AC power generator with an internal coating of Ni-based catalysts (thickness is about 128 μm), as shown in Figure 4. The FeCrAl-alloy tube is directly heated by the Joule effect. The heat source being in direct contact with the deposited catalyst ensures excellent heat transfer to the catalyst. The system was operated to reach an outlet temperature up to 900 °C and a methane conversion close to 87% was obtained at approximately 300000 h⁻¹ (based on catalyst volume), 50 mbar. The obtained methane conversion was lower compared to the equilibrium conversion, indicating that the system is working in a kinetic/external mass transport-controlled regime. The FeCrAl-alloy exhibits a relatively low resistivity, which indicates possibly the need of high currents flowing through the system. In a further numerical study,⁷⁰ the authors elucidate that the performance of the reformer with washcoated tubes is eventually controlled by external diffusion, i.e., gas–solid mass transfer, which calls for the adoption of tubes with small diameter. This possibly leads to the development of

reactor solutions based on either honeycomb monoliths or microchannel technologies, with the additional benefit of increasing the catalyst inventory at fixed catalyst washcoat thickness.

Recently, our group reported the electrification of methane steam reforming exploiting the direct Joule heating of 1%Rh/Al₂O₃ washcoated Si-infiltrated silicon carbide (SiSiC) foams ($d = 32$ mm, $L = 99$ mm).⁷³ Different from the tubular geometry proposed by Wismann and co-workers,^{44,70,71} the conductive foam offers largely enhanced gas–solid mass transfer coefficients,^{141,142} thus overcoming the bottleneck encountered by Wismann et al., whose solution suffers from external mass transport limitations associated with to laminar flow in narrow tubes and the need of having radial diffusion towards the tube walls.⁷⁰ In other words, in comparison with the direct electrification of a tubular geometry, the foams open opportunities to improve H₂ productivity with reduction of the reactor volume.⁶⁸ Figure 5 shows the schematic representation of the proposed eMSR reactor layout. The washcoated SiSiC foam was placed in a stainless-steel tube reactor (OD = 5 cm). To avoid electric contact, a ceramic tube is inserted between the foam and the stainless-steel tube. The foam is connected via electrical plates to a power generator. The foam-based eMSR system approached full methane conversion above 700 °C (temperature measured at the bottom of the foam), and methane conversions approaching equilibrium were obtained in a wide range of conditions. An energy efficiency of 81% was achieved on a washcoated foam with a catalyst density of 86.3 g/L (GHSV = 150000 NL/h/kg_{cat}, S/C = 4.1, ambient pressure), together with a specific energy demand as low as 1.24 kWh/Nm³_{H₂}, proving that minor energy dissipation occurred during high throughput tests.⁷² Preliminary scale up calculations suggest that it is possible to design highly efficient compact units in a single reactor with hydrogen productivity up to 200 Nm³/h.

Renda et al.,⁷⁶ reported the electrification of both steam reforming and dry reforming of methane with commercial heating elements made of silicon carbide ($d = 16$ mm, $L = 280$ mm), which were coated with a thin layer of Ni-based catalysts and loaded in a tubular reactor. Such an approach is promising to realize efficient heat transfer from the resistor to the catalyst. The systems were able to reach temperatures of >700 °C (measured at the outlet of the catalytic bed). The conversions for dry reforming were close to thermodynamic equilibrium; however, the steam reforming was working in the kinetic regime. The authors report results in terms of specific energy consumption of approximately 5 kWh/Nm³_{H₂}, that are far higher than the estimated values assuming no heat losses; therefore, the system proposed by Renda and co-workers exhibits a limited thermal efficiency, possibly due to the small dimensions of the setup.

A similar study was reported by Rieks and co-workers with FeCrAl alloy heating elements of a different shape, where significantly lower conversions were reported.⁷⁵ Notably, their geometries are not optimized for catalytic systems. In particular, the heating elements do not enable a high surface area for catalyst coating, and this may lead to both internal and external mass transfer limitations. Also, significant bypass phenomena are possible.

Zhang et al.¹⁴³ and Zhou et al.¹³⁹ reported the use of plate-type alumina supports impregnated with Ni for electrified methane steam reforming. The commercial plate-type AlCrNi alloy/Al clad base material consists of an Al layer thickness of

45 μm and a FeCrNi alloy layer thickness of 85 μm. The plate support was placed inside a plug flow reactor (inner diameter 10 mm): due to the existence of an FeCrNi alloy interlayer inside the plate support, its direct Joule heating was feasible. Compared to conventional oven heating, increased overall heat transfer coefficients were achieved for the electrified process. Even though Zhou et al.¹³⁹ reported a high methane conversion of 97% at 700 °C (surface temperature measured with a radiation thermometer), the employed plate support exhibited a very low hydrothermal stability, i.e., its surface area decreased to only 36% of the initial value after hydrothermal treatment at 700 °C for 50 h, which limits its industrialization.

In the open literature, patents claiming the application of directly electrified structured catalysts for MSR units are reported, although no data about the performances of these systems are available.¹⁴⁴ These structures can then be piled up, obtaining an array of structured materials up to a volume of 10 m³. A patent was recently filed by Pauletto for Joule heated structured ceramic catalysts, where the honeycomb catalyst includes wire resistances. They report the application of this reactor to methane steam reforming.⁷⁷

4. OTHER ENDOTHERMIC REACTIONS DRIVEN BY THE JOULE EFFECT

In general, direct Joule heating can be used to supply the heat required for reactions; therefore, it is in principle applicable to different endothermic processes. In addition to the above-mentioned methane reforming reactions, here we summarize the reported applications of Joule heating to other endothermic processes, such as cracking and CO₂ activation reactions.

4.1. Joule Heated Cracking Processes

Catalytic cracking or hydrocracking reactions are key processes for the production of light fractions from crude oil.^{145,146} As a result of their endothermic nature, a significant energy input is required to sustain those reactions, which are typically carried out at temperatures lower than that of methane steam reforming. In large-scale plants, the energy required by the reaction is supplied by cracking furnaces, where an enormous amount of fuel is burnt, being responsible for roughly 90% of the CO₂ emissions from such processes.^{147,148} It is estimated that steam cracking processes emit approximately 300 million tons of CO₂ to the environment per year.¹⁴⁷ As already discussed, electrification is a promising approach to providing energy to catalytic systems. Electrified cracking reactions offer considerable advantages over the traditional thermal heating solutions, such as more compact equipment and pollution-free operation due the removal of fuel combustion.^{149,150} Moreover, as a result of rapid and selective heating, the direct electrification offers the possibility to enhance heat transfer, therefore to minimize the coke formation on the catalyst surface, one of the limiting factors in such processes.^{145,147} It should be mentioned that such an electrification concept is not only advantageous for cracking of crude oil, but also applicable to other cracking reactions such as, biomass upgrading where energy is required to sustain the reaction.^{151–165}

As for electrical resistance heating for hydrocarbon cracking reactions, the following examples refer to noncatalytic processes with the Joule heated elements placed inside the reactor to heat up the feed components.^{166–168} Shekunova et al.¹⁶⁷ reported the use of resistance heated tungsten, molybdenum or nichrome wires inside a tubular quartz reactor

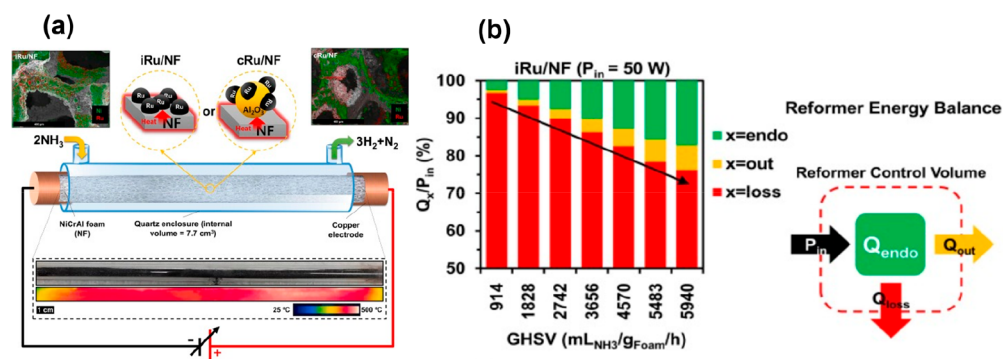


Figure 6. Direct Joule heating of a Ni-based foam for ammonia cracking reaction. (a) Reactor configuration and (b) fractional energy distribution in the reactor. Washcoated (cRu/NF) or impregnated (iRu/NF) on the foam. Experimental conditions: GHSV range of 1000–11000 mL/g_{Foam}/h, and temperature range of 450–580 °C, at varying power input.⁷⁴ (Adapted with permission from ref 74. Copyright 2021 Elsevier).

for C1–C4 hydrocarbons cracking to olefins. The heating elements were in contact with the flowing gas stream, and a temperature up to 600 °C could be reached for the cracking reaction. Porsin and co-workers investigated the methane pyrolysis reaction on a Joule heated tungsten wire, located in a quartz tube reactor and in direct contact with the flowing feed gas.¹⁶⁶ With resistance heating, a methane conversion of only 80% was achieved with temperatures up to 2000 °C. This indicates that the proposed reactor configuration is limited by methane mass transfer to the heated wire (S-shaped tungsten coil). In addition to those electricity-to-heat strategies, it is worth to mention that Coolbrook recently developed a revolutionary roto dynamic reactor (RDR),^{169,170} which exploits the electric energy transforming it into heat via rotating blades. When such a reactor was applied to cracking reactions, a higher ethylene yield was reported in comparison to that of conventional technologies.

In addition to hydrocarbon cracking, Badakhsh et al.⁷⁴ studied the ammonia cracking reaction by direct resistance heating of a NiCrAl foam (NF), which was activated by Ru-based catalysts. The catalytic tests were carried out in a small-scale prototype reactor (reactor volume = 7.7 cm³), with operating GHSV range of 1000–11000 mL/g_{Foam}/h, and the temperature range of 450–580 °C. As shown in Figure 6a, the NiCrAl foam was connected to a DC power supply and could be heated by the Joule effect, with the heat being thus transferred directly to the Ru-based catalyst for the ammonia cracking reaction. The catalytic components were either washcoated (cRu/NF, Ru/Al₂O₃ coated on the foam) or impregnated (iRu/NF, Ru impregnated on the foam). Interestingly, the bare Ni-based foam already exhibited a very good catalytic activity with an input power of 65 W. The impregnated foam was systematically studied in a wide range of input powers and space velocities. As a result of a small reactor volume, the authors reported a reforming efficiency of around 70% (calculated as the ratio of the outlet hydrogen enthalpy to the sum of inlet ammonia enthalpy and input power) with an input power of 72 W, and a very high reactor power density of 128 W/cm³ (based on the catalyst volume instead of the reactor or foam volume). However, an energy efficiency of ~30% was achieved if computed as the ratio of the enthalpy gain to the input power. In addition, they reported a linear decrease in energy loss with increasing space velocity, as shown in Figure 6b. This indicates that such a reactor configuration is advantageous when working at high space velocities in terms of the energy efficiency. The same trend was noticed also in our

SiSiC foam based eMSR system electrified by resistance heating.⁷³

Our team at Politecnico di Milano is currently working on Joule-heated ammonia cracking,¹⁷¹ with the same reactor concept already reported for methane reforming.^{72,73} Instead of being washcoated on the structure, the catalyst is packed in the foam cavities: the change in the reactor configuration is due to the relatively slow reaction kinetics that call for a large catalyst inventory.¹⁷¹

4.2. CO₂ Activation in Electrically Heated Reactors

To meet the demands of an increasing global population, unprecedented amounts of CO₂ emissions are produced and show a continuously increasing trend. Only reducing the CO₂ emissions may not be enough to limit growing environmental concerns. As another option, CO₂ can be captured and utilized for chemicals production. Electrochemical reduction of CO₂ (CO₂R) enables the direct utilization of CO₂ with electric energy. However, as the only technology approaching commercialization, the solid oxide electrolyzer requires a specific energy demand as high as 6–8 kWh/Nm³CO₂.^{111,172} On the other hand, CO₂ conversion processes into synthesis gas, such as dry reforming of methane or reverse water–gas shift (RWGS), may play a strategic role for the future sustainable production of chemicals and energy carriers.^{26,141,173} On the other hand, fuel combustion to supply the heat of the associated endothermic reactions would result in unwanted CO₂ emissions, thus frustrating the overall objective. Joule-heated CO₂ utilization processes based on renewable electricity may represent the technological solution to reduce the carbon footprint of chemical products in the near future, as suggested by process, environmental and economic calculations.^{78,95}

In this context, Wismann and co-workers reported for the first time the electrified RWGS process,⁴⁵ based on the same direct Joule heating of Ni-catalyst washcoated tube reactor configuration discussed in Figure 4. By employing an industrially relevant condition: H₂/CO₂ feed ratio of 2.25 at 10 bar, a syngas with H₂/CO ratio of 2 was obtained at 1050 °C, which was suitable for downstream Fischer–Tropsch synthesis. No carbon formation was observed, which, according to the authors, could be ascribed to the efficient heating of the system. Optimized working conditions also allow for almost complete selectivity to CO.

Recently, our group studied the direct electrification of the CO₂ reforming of methane (eCRM) and of the reverse water–

Table 5. Summary of Recent Developments for Direct Joule Heating in Fast Heating Applications

No.	Reaction	Catalysts/heating elements	Motivation for fast heating	Comments (Fast heating)	Ref
1	Methane pyrolysis	No catalyst; a flexible carbon heater	Avoid coke formation	PHQ method: 0.02 s on, 1.08 s off, up to 2000 K; fast heating avoids coke formation	46
2	Ammonia synthesis	Ammonia synthesis catalysts; a flexible carbon heater	Avoid catalyst sintering	Same PHQ method as Nr. 1; fast heating avoids catalyst sintering	47
3	Dry reforming of methane	Rapid pulse Joule heating (approximately 14000 °C/min) of a PtNi/SiO ₂ coated carbon fiber paper	Avoid coke formation and catalyst sintering	Fast dynamic Joule heating suppresses coke formation and catalyst sintering	53
4	Methane steam reforming	Ni-catalyst coated FeCrAl tube reactor	Fast start-up and shutdown to address the intermittent nature of renewable energy	Cold start and cyclical operation experiments	71
6	CO ₂ methanation	Ni foam coated with Ni–Al catalysts	To address the intermittent nature of renewable energy	Fast heating to 200–300 °C	175, 176
7	Propylene epoxidation	FeCrAl-alloy belt; coated catalysts	Fast catalyst regeneration	Fast heating (2 min from 50 to 300 °C)	177
8	Total oxidation of HCHO	Ag-Co ₃ O ₄ @3DCM	Fase response for VOC elimination	7 times faster heating/cooling rate than conventional heating	178

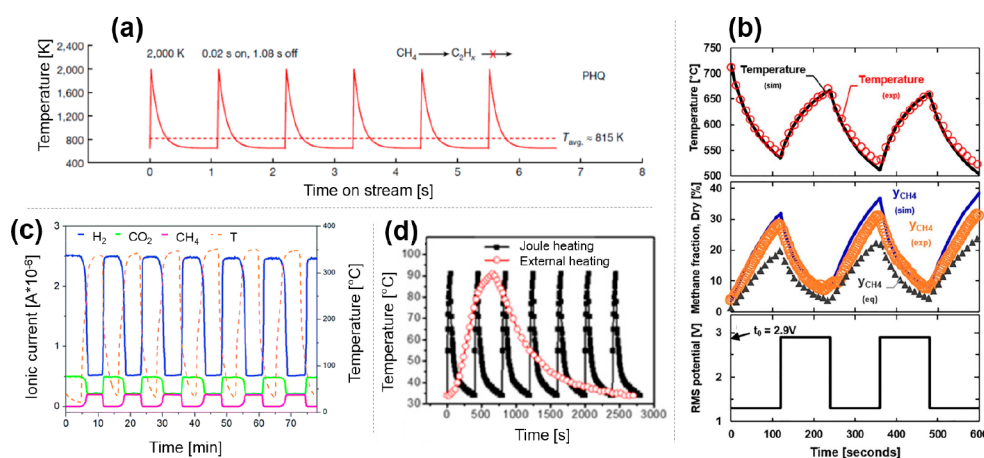


Figure 7. Fast heating with Joule-heated reactors: (a) Temperature profile of programmable heating and quenching (PHQ, 0.02 s on, 1.08 s off, up to 2000 K) method for methane pyrolysis reaction.⁴⁶ (Adapted with permission from ref 46. Copyright 2022 Springer Nature) (b) Transient response of temperature and methane concentration during periodical changes of input voltage in direct electrification of a reactor tube for methane steam reforming.⁷¹ (Adapted with permission under a Creative Commons license [CC-BY] from ref 71. Copyright 2021 Elsevier) (c) Temperature and online mass spectrum of successive on/off catalytic CO₂ methanation cycles via direct resistance heating of a Ni foam.¹⁷⁶ (Adapted with permission from ref 176. Copyright 1996 Royal Society of Chemistry) (d) Start-up and shut-down response during formaldehyde total oxidation driven by direct Joule heating of a washcoated carbon monolith and by a conventional external heater.¹⁷⁸ (Adapted with permission under a Creative Commons license [CC-BY] from ref 178. Copyright 2020 Elsevier).

gas shift (eRWGS) processes in washcoated structured reactors.¹¹¹ Similar to the reactor configuration discussed in Figure 5, the catalytically activated open-cell foams both provide optimal heat and mass transfer properties and serve as Joule heated substrates for the catalytic conversion of CO₂ via the reaction with methane or hydrogen. CO₂ conversions approaching equilibrium were measured across a wide range of conditions for both reactions. Such a reactor concept is proven to ensure remarkably low specific energy demand for CO₂ valorization, reaching approximately 0.7 kWh/Nm³CO₂ for eRWGS process in an optimized process configuration (considering an overall adiabaticity of 95% and a recovery of 90% sensible heat). If the feed H₂ is sourced from water electrolysis (3.8 kWh/Nm³H₂),^{114,137} it is possible to achieve an overall specific energy consumption of 4.5 kWh/Nm³CO₂ for CO₂ valorization, which is lower compared to the solid oxide electrolyzer for CO₂ reduction to CO (6–8 kWh/Nm³CO₂¹⁵²). Furthermore, the proposed system utilizing Joule-heated Rh/Al₂O₃-coated SiSiC foam demonstrated excellent catalytic and electrical stability for over 75 h. By replacing fuel combustion

with Joule heating driven by renewable electricity, electrified CO₂ valorization processes provide an important approach for dealing with the intermittent nature of renewable sources by storing the energy in chemicals with a low carbon footprint.

Meloni and co-workers studied SiC-based structured catalysts (both foam-based and monolith-based) for electrified (both Joule heating and microwave heating) dry reforming.¹⁷⁴ They reported that foam-based catalysts are more efficient when driven by the Joule effect, in which case, the energy demand (2.6 kWh/Nm³H₂) was about a fourth that in the case of microwave heating.

5. ENABLING FAST HEATING WITH JOULE-HEATED REACTORS

Driven by renewable energy, the direct electrification of catalytic processes through the Joule effect not only eliminates CO₂ emissions from fuel combustion but also provides the required heat for highly endothermic processes, as discussed previously for methane steam reforming (Section 3.2), cracking processes (Section 4.1), and CO₂ activation reactions

(Section 4.2). Furthermore, it enables fast and selective heating of the catalytic bed, making it particularly advantageous for applications where rapid heating is required.^{179–183} On the other hand, the direct electrification of catalytic processes suffers from the intermittent nature of the renewable electricity supply, such as from sun and wind. This calls for high operational flexibility of the electrified reactors to sustain fast dynamics of the input energy supply. Herein, recent works on the direct electrification of fast heating applications are reviewed, as summarized in Table 5. Note that the heating process is essentially specific for the applied systems, which is correlated to several factors, such as the applied reactions, the size, material, and thermal mass of the reactors, the applied catalysts, as well as the insulation approaches.

Dong and co-workers⁴⁶ reported a novel programmable heating and quenching (PHQ) method for efficient chemical synthesis thanks to the fast heating capability of Joule heating. In such temperature-modulated heating, high-temperature operation provides required activation energy and enables high conversion, while low-temperature duration inhibits undesired side reactions or avoids the catalyst stability issue associated with high temperatures. They applied this concept first to the noncatalytic model reaction of methane pyrolysis, which suffers from significant coke formation at 1273 K in conventional continuous heating as the carbon chain grows with operating time. However, a significant decrease in coke formation was noticed with PHQ heating by using a flexible carbon heater (0.02 s on, 1.08 s off, up to 2000 K, in comparison to conventional heating (data from ref 184), as shown in Figure 7a. Moreover, an average of only 815 K from PHQ heating indicates that such a technology is more energy saving (1273K for conventional heating). The authors applied the same concept to another model system: ammonia synthesis with Ru nanoparticles supported on a carbon felt heater; the catalysts are thermally stable with this PHQ method thanks to the low temperature operation. In summary, such a programmable heating and quenching method is very promising as it could lead to process intensification and decarbonization by exploiting renewable electricity; moreover, the flexibility associated with appropriate turning of temperature levels, duration of the heating phase and quenching time, and the residence time allow to on-purpose control the dynamics of the reaction and improve the reactor performance.¹⁸⁵ Similarly, Yu et al.⁵³ reported the rapid pulse Joule heating (approximately 14000 °C/min) of a PtNi/SiO₂ coated carbon fiber paper for dry reforming reaction. The authors demonstrated that the catalyst in fast dynamic operation exhibited enhanced catalytic performances with excellent stability due to the short time spent at low and high peak temperatures, which suppressed coke formation and catalyst sintering.

Wismann and co-workers investigated the transient operation of a Joule heated Ni-catalyst coated FeCrAl tube reactor for methane steam reforming.⁷¹ The reactor configuration is similar to that already discussed in Figure 4. They investigated the cold start properties of the reactor, which starts at 15% of the steady state power (275 °C) to avoid water condensation. The reactor showed a very fast temperature and conversion response in the first 2 min, while above 600 °C the tendency significantly changed because of the strongly endothermic MSR reaction. Within 12 min the reactor reached 95% of the thermal steady state, while it took around 30 min to reach the 95% conversion steady state. In addition, they also investigated the transient response of the temperature and conversion upon

periodical changes of the input power, Figure 7b. The reactor was operated by periodically changing the input voltage from 2.9 V (a steady state corresponding to 80% methane conversion) to 1.3 V every 2 min with a linear function. The system exhibited very good temperature and conversion responses to the input power. However, the system did not reach the steady state as the peak temperatures decreased with time. This was explained by the thermal inertia of the thick insulation layer.

As previously discussed, Joule heating can be used to provide the necessary heat for the endothermic processes. However, it has also been reported to light-off exothermic reactions and enable rapid transient operation. The following examples illustrate this phenomenon.

Dou and co-workers investigated the direct electrification of a Ni-catalyst washcoated Ni foam for the exothermic CO₂ methanation reaction.^{175,176} A Ni foam (4 mm × 70 mm × 1.5 mm, 100 PPI (pores per inch)) was placed inside a quartz tube reactor (inner diameter 8 mm, length 200 mm), where the temperature was recorded using an infrared thermometer via a ZnSe window. They reported a foam temperature of 300 °C with an input power of 10 W, which allows fast heating of the reactor during transient operation. As shown in Figure 7(c), the proposed reactor exhibited very fast heating and cooling: heating from 50 to 350 °C could be accomplished in about 2 min. As a result, the catalytic activity exhibited a very fast response to the temperature as well with intermittent hydrogen production. This is highly relevant considering the intermittent nature of renewable electricity.

Wang et al.¹⁷⁸ reported the application of resistance heating to the formaldehyde total oxidation. In their study, a Ag/Co₃O₄ catalyst was coated on a carbon monolith and used as a heating substrate. They showed that with 5 W input power the catalyst could reach a core temperature of 125 °C with direct Joule heating. However, with conventional oven heating, the core temperature was only 90 °C even with a larger input power of 37.5 W. In this regard, the resistance heating is more energy efficient as a result of selective heating on the target material. More interestingly, the resistance heating shows a faster temperature response, resulting in a seven times faster heating–cooling cycle from 35 to 90 °C in comparison to conventional heating (Figure 7d). In addition, the authors also investigated the resistance heating system with 100 h at steady state test as well as 100 cycles from 35 to 90 °C. Almost full conversion obtained from those experiments confirmed the excellent stability of the proposed electrified system.

For propylene epoxidation, the Au/TiO₂ catalyst showed a fast activity loss due to possible deposits on the catalyst surface such as possible further propylene oxide transformation products. While the catalyst could be regenerated, the regeneration temperature, which is around 280 °C, is relatively high compared to the reaction temperature of 70 °C. In this regard, Yuan et al.¹⁷⁷ proposed a FeCrAl-alloy microreactor coated with Au/TiO₂ catalyst. The FeCrAl-alloy in a belt shape (0.1 mm thick, 20 mm wide, 3 m, resistance of 2.13 Ω) was folded to obtain 12 channels and placed inside the reactor. The FeCrAl-alloy reactor was connected to the power supply and directly heated by resistance heating. Fast heat transfer was achieved, as the catalyst was directly coated onto the heating substrate. As a result, the catalyst could reach 280 °C (from 50 °C) in less than 2 min, enabling rapid regeneration (maintained at 280 °C for 20 min). The catalyst showed good activity after rapid regeneration.

6. CONCLUSIONS

With renewable electricity becoming more and more accessible, electricity-to-heat driven reactors will play a crucial role in addressing the decarbonization issue in chemical processes. Electricity-driven heating methods, including Joule, induction, and microwave heating, enable fast, selective, and uniform heating, which makes them advantageous in applications where fast heat transfer is required. In this review, we have discussed the different heating methods from their fundamentals to their advantages and disadvantages in terms of their application to catalytic processes. Moreover, we reviewed the applications of Joule heating in endothermic catalytic processes like methane reforming, cracking reactions, CO₂ activation and other applications where transient operation is required. The current electricity-to-heat driven reactor technologies are mainly in the proof-of-concept stage: accordingly, challenges coexist with opportunities. Studies concerning the design of energy-responsive catalysts and reactors, controlling the heat distribution, and improving the energy efficiency are necessary to push such technologies forward.

In our opinion, among the three electrification approaches, resistance heating represents the most promising method for industrialization. First, such an approach is more straightforward compared to the other two methods in terms of materials, reactor design, and temperature flexibility. Moreover, the existing knowledge and experience from the direct heating of furnaces can be transferred directly to the development of Joule heated catalytic reactors. However, there are still challenges: in order to minimize contact resistances and improve the energy efficiency, more efficient power to reactor connection methods should be developed.

Although such technologies are in their early-stage development, we expect a rapid rise of research works dedicated to the study of electrified reactors from both academia and industry. These studies will enable a better understanding of the concepts as well as the development and optimization of a new family of electrified reactors due to the combination of detailed experimental, modeling, and computational fluid dynamics (CFD) studies at the reactor scale with the industrial and experimental practice.

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Notes

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