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# Reverse traveling microwave reactor – Modelling and design considerations

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## HIGHLIGHTS

- A novel heterogeneous catalytic microwave reactor concept is introduced.
- The microwave incident direction is reversed periodically to avoid temperature differences along the load.
- The novel reactor provides new insights to the microwave-assisted catalytic process scale-up.
- A new reactor's loading pattern is introduced to keep the impedance matching along the reactor.
- Simulation results show that the temperature distribution homogeneity improves.

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## ABSTRACT

Microwave heating presents a potentially green alternative for energy supply to chemical and catalytic reactors as it can be based on the electricity from renewable sources. The Reverse Traveling Microwave Reactor (RTMR) is a novel heterogeneous catalytic reactor concept, based on the coaxial waveguide structure. The reactor has two microwave ports on both ends, and microwave irradiation is periodically switched between those ports to minimize the temperature gradients along the catalyst bed. In the current paper, COMSOL MULTIPHYSICS® simulation environment has been used to develop a 3D multiphysics model of the RTMR. Based on the model, operational characteristics of the reactor including electric field distribution and transient temperature profiles have been studied. Simulation results show that periodically reversed microwave irradiation improves the homogeneity of the temperature distribution inside the catalyst bed. The study provides new insights into the design and scale-up of microwave-assisted catalytic flow processes.



## 1. Introduction

Microwave chemistry applications have been investigated for more than three decades, due to the several advantages that microwave energy offers in terms of reaction speeding up, volumetric heating, and improving product yields and selectivity (Polaert et al., 2017; Oliver Kappe, 2008; Adnadjević et al., 2017; Cherbański, 2011). Compared to traditional heating methods, the microwave-assisted heterogeneous catalyzed reaction systems perform better in terms of reaction rate and selectivity, increasing reaction rate and selectivity (Horikoshi and Serpone, 2014; Bond et al., 1993; Xu et al., 2016; Díaz-Ortiz et al., 2019; Zhang et al., 2003). Microwave irradiation was investigated as an alternative to traditional heating for temperature swing adsorption processes in Candice Ellison et al. research. By measuring CO2 desorption from zeolite 13X at various temperatures, microwave and conventional heating efficiencies during sorbent regeneration were evaluated. Their findings show that microwave regeneration has been found to improve adsorption/desorption cycling efficiency while also theoretically lowering the energy penalty associated with temperature swing (Ellison et al., 2021). Regarding the energy and CO2 emissions perspective, Ahmadreza Amini et al. showed that a 60–80 percent reduction in energy consumption could be achieved, and the ability for microwave heating could reduce CO2 emissions by 3–5 times (Amini et al., 2021). According to Dina Ewis et al., microwave synthesized adsorbents have improved adsorption for various water pollutants due to a better porous structure than conventionally synthesized ones (Ewis and Hameed, 2021). Priyanka Pareek et al. described an environmentally friendly method for microwave irradiation-based graphene oxide modification that improves the adsorptive capacity for dye removal from treated wastewater (Pareek et al., 2021). The benefits

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of microwave heating in several operations were investigated in a recent literature by Palma et al. (2020). They emphasized how microwave-assisted processes allowed for a significantly increased conversion to traditional heating. Despite evident intensification effects of microwave irradiation, its reliable scale-up for industrial applications remains as the main troublesome challenge for all researchers in this domain (Stankiewicz, 2006).

Currently, three major types of microwave applicators are being used in chemical laboratories, namely, mono-mode, multi-mode, and traveling-wave. Mono- and multi-mode microwave applicators, also known as microwave cavities, are dominant types. However, they generally suffer from non-uniform heating and hot spots formation, due to the existence of a standing microwave pattern (Stankiewicz et al., 2019). Moreover, these cavities are strongly dependent on the supplied microwave frequency and sample size, particularly mono-mode cavities. These limitations prevent the use of mono- and multi-mode cavities for critically controlled chemical reactions (Stankiewicz et al., 2019). In contrast to mono- and multi-mode cavities, the traveling-wave applicators are capable of avoiding the standing-wave formation and consequently providing a highly uniform heating profile (Sturm et al., 2016). Frequency variation have no effects on the performance of the traveling-wave microwave applicators (Mehdizadeh, 2015). The main design principle for traveling microwave reactor is to avoid microwave reflections through the whole structure to ensure that no standing-wave pattern is generated along the assembly (Eghbal Sarabi et al., 2020).

In our previous paper (Eghbal Sarabi et al., 2020), the critical aspects of designing a coaxial traveling-wave microwave reactor, TMR, have been investigated, and consequently, a solution for process scale-up has been proposed. Simulation results showed that there is no standing-wave generated along with the structure. Furthermore, microwave-susceptible material (catalyst) loading patterns in the TMR has been introduced to keep the impedance matching and minimize the microwave reflections. Simulation results demonstrated that for the proposed method, the catalyst could interact more efficiently with microwave energy and produce a fair homogeneous heating profile. The primary source of limitation of scaling-up the traveling-wave microwave reactor is due to the penetration depth of the microwave. For non-magnetic dielectric materials, microwave penetration depth, $D_p$, is defined as the distance from material's surface at which microwave power drops to 1/e (=37%) from the strength at surface. This value in a microwave-susceptible material is a good indicator of the material to convert microwave energy to heat (Peng et al., 2010) and gives an idea of the heat distribution within the material. When the magnetic susceptibility is high (materials with higher loss factor $\varepsilon_r''$), the material could absorb the microwave energy faster while its penetration depth is low (Sun et al., 2016). Specifically, if the length of the catalyst bed is larger than the penetration depth, a temperature difference along the catalyst bed would be observed during the microwave heating process since the forepart of the catalyst, which is closer to the inlet port of microwave, absorbs more energy and accordingly dissipates more heat in that region.

In this paper, we further studied the uniformity of the temperature distribution inside a microwave-susceptible material load in a packed-bed configuration. This load can be either catalytically active for a specific reaction or catalyst support. However, uniform heating of the load needs to be achieved for a better control of the chemical reactions, specifically for the large-scale considerations. Herein, a novel microwave reactor concept is introduced based on the coaxial wave-guide structure. This reactor has two microwave ports on both sides, and microwave irradiation is periodically switched between these two ports to provide a more uniform heating profile and minimize the temperature gradients along the load. In other words, the microwave incident direction is reversed in order to avoid temperature differences along the load.

## 2. Reverse traveling microwave reactor design

The operational principles of a simple "one-way" traveling-wave reactor have been discussed in (Eghbal Sarabi et al., 2020). For the fully loaded reactor with a catalyst sample, which is microwave absorbent, large temperature gradients along the axial direction of the TMR is observed. This could be due to either impedance mismatching along the waveguide or the limited capacity of the material to absorb microwave energy, i.e., its penetration depth. These two would result in a non-uniform heating profile and hot spots formation. To increase the efficiency of the reactor and avoid the first problem caused by sudden impedance changes, a new loading pattern has been introduced. However, to overcome the second issue a novel microwave reactor concept is now proposed, based on the coaxial wave-guide structure.

This reactor has two microwave ports on both sides and microwave irradiation is periodically switched between these two ports to provide a more uniform heating profile and minimize the temperature gradients along the catalyst bed. Once a temperature difference along the catalyst bed is observed, the microwave incident direction can be reversed. To this extent, we can reduce the effect of the microwave penetration depth by half.

The essential factors which affect the traveling-wave microwave reactor (TMR) performance are the waveguide type, the reactor dimensions, and the microwave reflections. TMR can be implemented in various configurations, e.g., rectangular or coaxial types. Coaxial traveling-wave microwave reactor concepts has been studied over the recent years (Sturm et al., 2016; Mehdizadeh, 2015; Gentili, 2009; Mehdizadeh, 2009; Longo and Ricci, 2007; Mitani et al., 2016; Durka, 2013). Coaxial waveguide could potentially provide several advantages in terms of having no cut-off frequency for TEM propagation mode (Kouzaev, 2013; Kapranov and Kouzaev, 2019). The reactor dimensions should be determined based on restrictions regarding electromagnetic wave propagation. Therefore, the diameter of the outer conductor cannot exceed half of the electromagnetic wavelength. Accordingly, for the typical frequency of 2.45 GHz, the upper limit of the diameter is approximately 61 mm (Durka, 2013). Further, the reflections of the structure need to be minimized to ensure that no standing wave is generated along the waveguide. Moreover, a smaller microwave reflection leads to faster heating and less energy losses since more microwave energy penetrates into the sample. In order to minimize the electromagnetic reflections in the coaxial structure, the impedance matching between the different sections of the waveguide plays an important role. In that sense, the characteristic impedance of the waveguide should be kept equal to the characteristic impedance of the microwave source (50 Ω).

To this end, the first consideration is to adjust the parameters to achieve an impedance matching between different parts of the structure to diminish the microwave field reflections. And the second is adjusting the cut-off frequency beyond the working frequency to assure that the reactor works in TEM mode, so that the hotspot can be avoided along the reactor. Combining the results of these constraints, the diameter of the outer and the inner conductors are determined as 51 mm and 22 mm, respectively.

Additionally, in order to couple two different size coaxial waveguides, i.e., the microwave excitation port and the reactor, a conical-shape coaxial transition part needs to be positioned in between. The size of the port was determined based on the connector standard (Type N Connectors, xxxx). According to the previous discussions, the length of the transition part does not have a significant effect on the microwave power reflections, so 50 mm

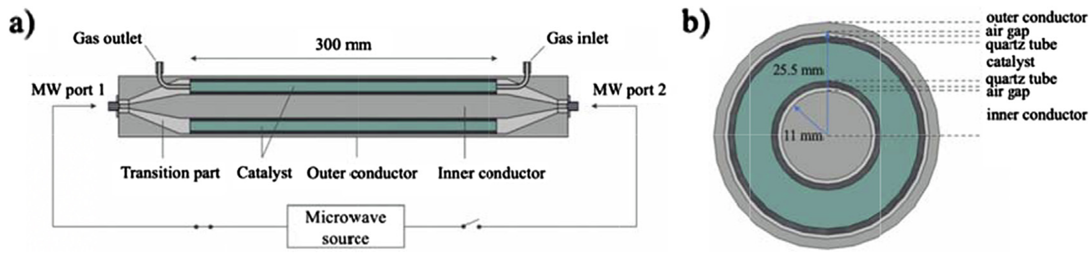


**Fig. 1.** Cross-sectional schematic of reverse traveling-wave microwave reactor, a) axial, b) radial (at the middle of the bed).

is chosen for the length of the transition part, which contains air (εr = 1).

Comparing the original traveling-wave microwave reactor, the reverse traveling-wave microwave reactor should be able to receive the microwave energy from both sides. For this purpose, the input microwave port and the conical-shape transition part are mirrored at the end of the reactor. The cross-sectional view of the proposed microwave reactor along the axial direction is presented in Fig. 1. The dimensions of the coaxial waveguide are determined as described in the previous section, and the length of the catalyst zone is 300 mm. Microwave radiations can be introduced from both ends of the reactor, while one port is ON, the other port needs to be switched OFF.

## 2.1. Mathematical model of RTMR

The electromagnetic waves and heat transfer physics are fully coupled in the reactor's 3D model using COMSOL MULTIPHYSICS® simulation environment. The numerical problem is solved using the finite element method (FEM), and the governing equations used for each physic module are described in this section. The current study is simulation-based. However the model used in the simulations has been successfully validated in our earlier work (González et al., 2021)

During the model development, several assumptions have been made:

i. The materials are isotropic media.
ii. The permeability of all the materials used is assigned as free space ($\mu_r$ = 1), since there is no magnetic material.
iii. The metal walls of the reactor were assigned as impedance boundary condition (IBC) to account for the presence of the electric surface current in them (Nigar et al., 2019).
iv. The temperature-dependent dielectric properties of catalyst material (PtC) were obtained from our previous study (Gangurde et al., 2017), and a constant value is used for the temperatures above 850 °C.
v. What about porosity of the material?
vi. No fluid dynamics?
vii. A continuity condition was used at the interface of two different domains 1 and 2: $-n\cdot(-k_1\nabla T_1) - n\cdot(-k_2\nabla T_2) = 0$ (Nigar et al., 2019).

### 2.1.1. Electromagnetic waves

To determine the electric field distribution in the coaxial waveguide, the wave equation derived from Maxwell's equations is solved (Muley et al., 2016; COMSOL RF, 2017) in the domains between the waveguide, i.e., the air domain, catalyst and quartz tubes:

$$\nabla^2\mathbf{E} + \omega^2\varepsilon\mu\mathbf{E} = 0 \tag{1}$$

where **E** is the electric field vector (V/m), $\mu$ and $\varepsilon$ stands for the permeability and relative permittivity of the materials and $\omega = 2\pi f$ is the angular frequency (rad/s).

When the electric field distribution in the packed-bed is obtained, the power dissipated in the material can be calculated using the following equation (Nigar et al., 2019):

$$Q_{MW} = \pi f \varepsilon_0 \varepsilon_r'' \mathbf{E}\cdot\mathbf{E}^* \tag{2}$$

where $Q_{MW}$ represents the dissipated power density in the material (W/m$^3$), $f$ is the incident microwave frequency (Hz), $\varepsilon_0$ is the permittivity of free space, $\varepsilon_r''$ is the dielectric loss of the material, $\mathbf{E}^*$ is the complex conjugate of **E**.

### 2.1.2. Heat transfer in solids

This module is applied to all the domains (catalyst, quartz tubes, inner and outer conductors) of the reactor. In this physic interface, the following transient equation is solved (Heat Transfer Module User Guide, 2015):

$$\rho C_p \frac{\partial T}{\partial t} + \nabla\cdot(-k\nabla T) = Q_{MW} \tag{3}$$

where $\rho$ is the material density (kg/m$^3$), $C_p$ is the specific heat capacity (J/(kg·K)), $T$ is the temperature (K), $k$ stands for the thermal conductivity (W/(m·K)).

The first term on the left side is the rate of heat accumulation, and the second term is the conductive contribution to heat transfer. On the right-hand side of Eq. (3), the dissipated power obtained from the *Electromagnetic Waves* module is defined as the heat source for the reactor. On the other hand, the temperature calculated from the *Heat Transfer* interface is used to evaluate the temperature-dependent properties required in the *Electromagnetic Waves* interface. This way, these two physics are fully coupled in the model.

The simulated result of the normalized electric field distribution inside the reactor without a catalyst bed is presented in Fig. 2. The simulation results reveal that the reflected power of the overall structure is less than 1% at the working frequency of 2.45 GHz,

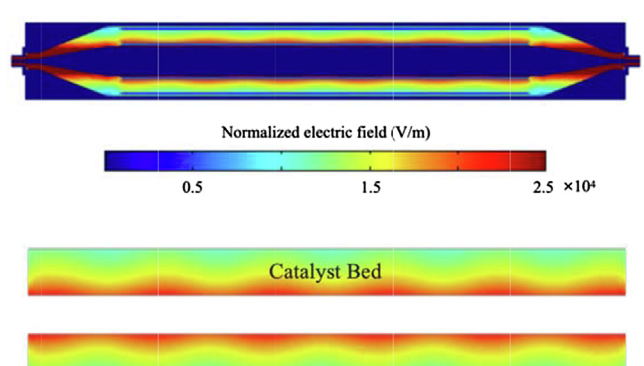


**Fig. 2.** Normalized electric field distribution inside the unloaded reactor (microwave energy introduced from left port): a) the whole configuration, b) catalyst section.

which proves that no standing-wave is generated along the structure and the microwave field is ideally traveling along the reactor.

## 3. Heating profile control

All the optimizations, has been done so far, concerned with the empty reactor. Once we load the reactor with catalyst as a microwave absorbing material, microwave energy would be absorbed differently depending on the material dielectric properties. This may occur due to so many reasons. e.g., not having the impedance matching along the reactor, which results in hot zone formation and ununiformed correlated heating profile. However, in the reverse traveling microwave reactor, loaded with catalyst, we could improve the heating homogeneity by different factors. The most effective parameters which could significantly influence the heating distribution, are investigated below.

### *3.1. Microwave power switching frequency*

One of the items, which enables us to control the heating pattern, is the microwave power switching frequency. Being able to introduce the microwave power to the catalyst bed from two ports, we can reduce the effect of the microwave penetration depth. This way, the temperature uniformity inside the packed bed would be improved. Furthermore, it is important to determine how often the microwave irradiation needs to be switched during the process to achieve a more uniform heating of the catalyst.

To illustrate the effect of the switching frequency in the loaded reactor, the maximum and minimum temperatures within the packed bed with different switching frequencies were extracted and compared. In this study, platinum on carbon (Pt/C) catalyst is used due to its excellent microwave absorbing ability. The temperature-dependent dielectric properties of catalyst are provided by (Gangurde et al., 2017). The electromagnetic waves and heat transfer physics are fully coupled in the reactor's 3D model using COMSOL MULTIPHYSICS® simulation environment. The numerical problem is solved using the finite element method (FEM) and the microwave power input is 400 W. According to Fig. 3(a), if the microwave is not reversed during the process (i.e., previous design (Eghbal Sarabi et al., 2020) TMR), the maximum-minimum temperature difference along the bed is 324 °C, after 60 min heating with 400 W MW power. On the other hand, as illustrated in Fig. 3(b)-(c), where the microwave power switching is applied, the temperature profile is distributed much more uniformly along the catalyst bed. The microwave irradiation is switched between the reactor's two ports at regular intervals using the same power as before. The switching frequency is defined as the time elapsed between adjacent switches. In this case, the heating of the catalyst bed is symmetric, with the minimum temperature at the middle. The temperature difference along the bed is 156 °C and 75 °C for the switching frequencies of 5 min and 30 s, respectively. The maximum and minimum temperatures of the packed bed with the corresponding switching frequencies are presented in Fig. 4. The results show that a lower switching frequency results in a lower temperature difference. This is because the MW penetration depth has a reduced effect. As a result of shortening the switching time, the temperature variation at the ends of the catalyst bed would be smaller, allowing for better homogeneity along the packed bed. A temperature difference of less than 75 °C is expected for a switching frequency of less than 30 s, and the packed bed's increasing rate of temperature is expected to be more stable. However, for analyzing the temperature distribution within the bed, the heat transfer to the bed and the reactor structures should be taken into account, especially for switching frequencies approaching zero, which generates less heat before each switch.

Changing of the maximum and minimum temperature of the catalyst over time for the switching frequency of 30 s is plotted

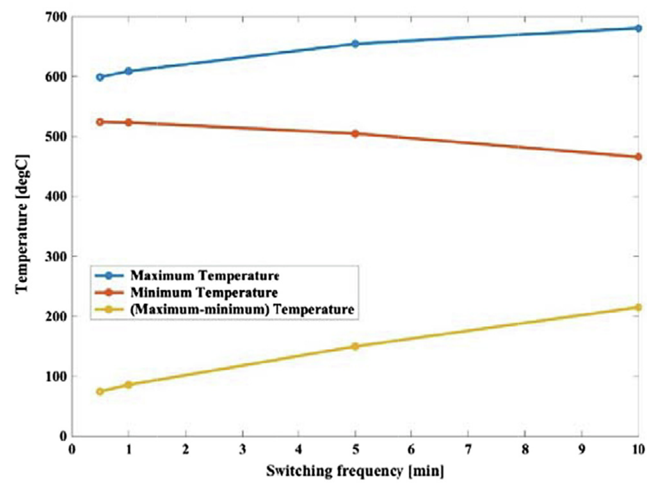


**Fig. 4.** Effect of microwave switching frequencies on the maximum and minimum temperature of the packed bed.

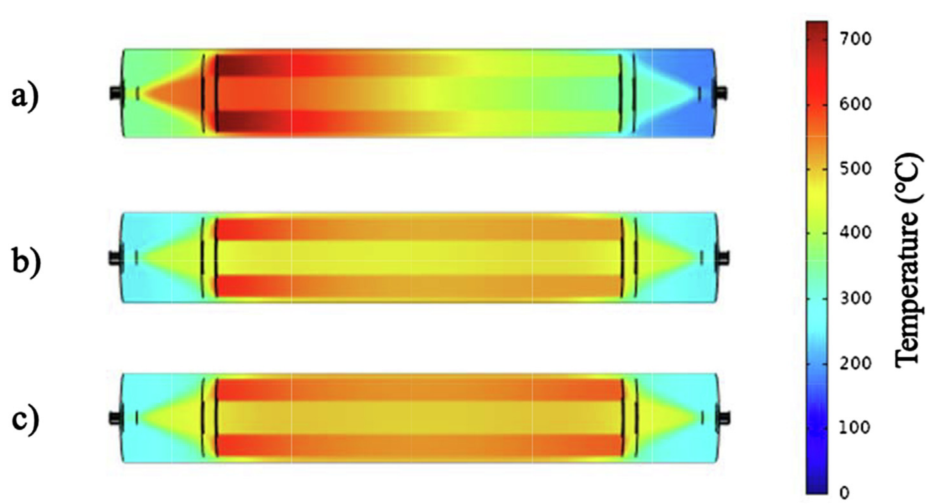


**Fig. 3.** The catalyst heating profile for different switching frequencies: (a) No switching (b) 5 min (c) 30 s.

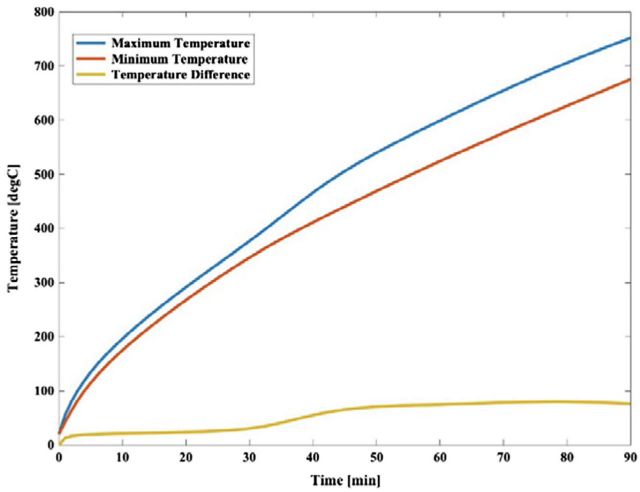


**Fig. 5.** The maximum and minimum temperature for half-minute switching frequency over time.

in Fig. 5. The temperature keeps increasing over time, but the temperature difference along the packed-bed is stabled around 75 °C after 45 min of heating. The temperature difference has decreased slightly after eighty minutes. This is due to the model's use of temperature-dependent dielectric properties of Pt/C. When the temperature reaches around 650 °C, the material's increasing rate of loss tangent decreases. According to the simulations, the temperature difference takes nearly the same amount of time to stabilize for different switching frequencies, such as 30 s, 1 min, and 5 min. The material properties determine the heating pattern, but the value of the temperature difference varies depending on the switching frequency.

### *3.2. Effects of the catalyst geometry (impedance matching)*

According to the simulation results, when the microwave power is switched between the two ports, the center of the bed is heated only by heat conduction due to the limited microwave penetration depth. To facilitate the microwave penetration deeper into the catalyst bed, the impedance matching between the different sections of the waveguide should be retained for the reactor loaded with the catalyst. For the fully loaded RTMR, around 22.3% microwave power is reflected under high temperatures due to the difference between the dielectric properties of catalyst (PtC) and air. However, microwave reflection presents a crucial issue influencing the TMR performance. A smaller reflection results in more microwave energy penetration into the sample and thus, a more uniform heating profile. To keep the impedance matching all along the reactor, loaded with a catalyst sample as an absorbing media, the catalyst mass should increase progressively. Otherwise, it would behave such as a mirror at the intersection on the way of the microwave and reflect a significant amount of the incident microwave. This would result in creating a standing-wave pattern inside the waveguide, which leads to a non-uniform heating profile and hot spots formation. Moreover, for the case where catalyst uniformly filled in the reactor, the microwave field intensity and therefore the heating rate would attenuate exponentially along the reactor.

However, as reverse traveling microwave reactor has the microwave inputs from both sides, the catalyst mass should increase symmetrically from both port intersections, see Fig. 6. The two end conical-shaped with different cone length is studied to show the effect of impedance matching on the catalyst heating pattern. According to the simulation results, the least MW reflections would be achieved in the case of the whole two-end conical sample (conical length of 150 mm from each sides).

The temperature distribution of normally and gradually loaded RTMR after 60 min heating can be found in Fig. 7. In both cases, the microwave switching frequency between the ports is considered to 30 s and microwave input is 400 W. As can be seen, the temperature profile is distributed more smoothly when the catalyst load is gradually increased, and a higher maximum temperature can be reached with the same microwave power input (877 °C for conical-shaped bed and 600 °C for fully-loaded bed).

The maximum and minimum temperature for the two different catalyst loading patterns along the heating time is extracted and compared in Fig. 8. According to the results, the temperature difference of both type of bed is stabilized after 50 min heating.

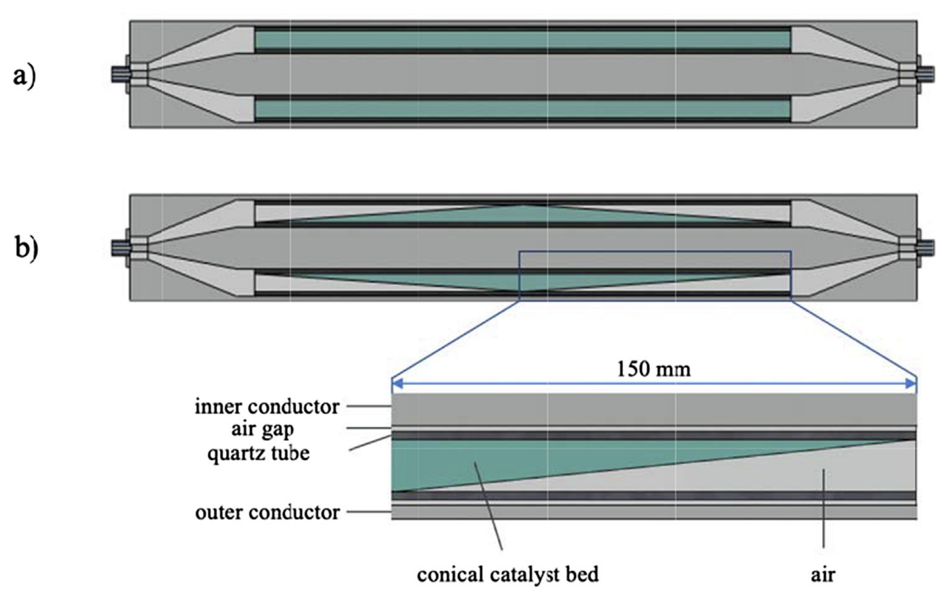


**Fig. 6.** Schematic diagram of different catalyst loading pattern: (a) Fully loading, (b) Conical-shaped laoding (Gradually increasing the catalyst mass from both sides of the reactor.)

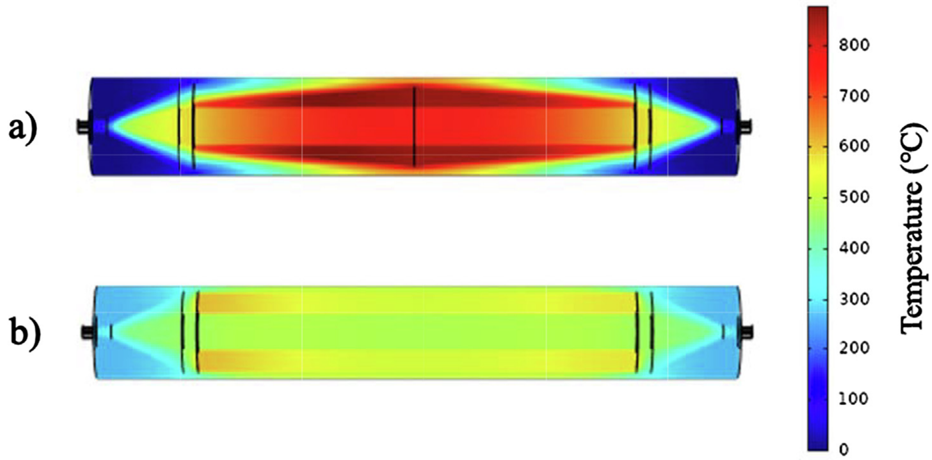


**Fig. 7.** Heating profile of the reactor for different loading patterns: (a) conical shaped bed, 30 s (b) fully loaded bed with 30 s switching frequency.

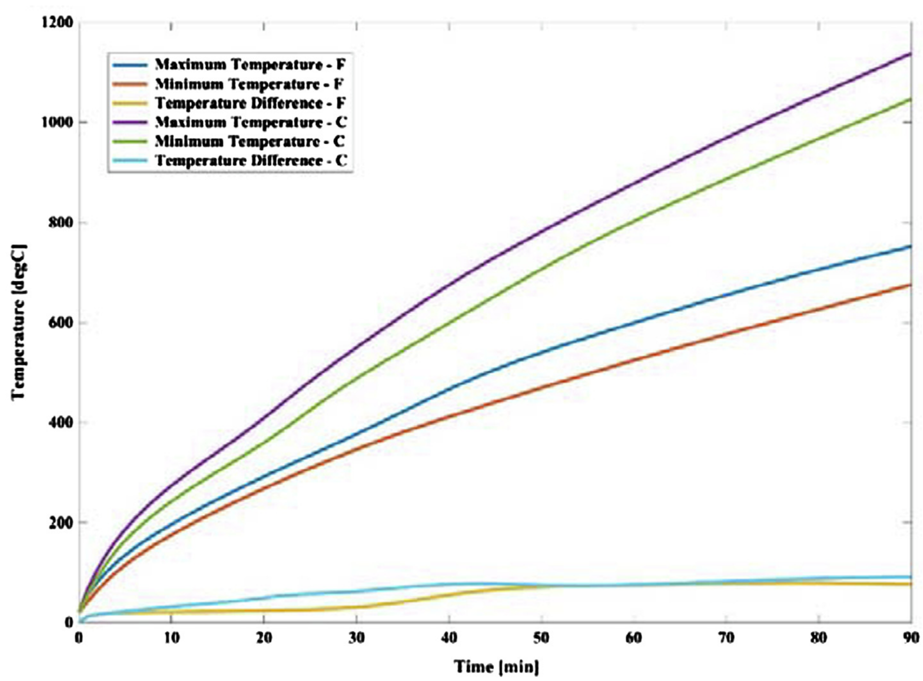


**Fig. 8.** The maximum and minimum temperature for 30-second switching frequency over time: F = fully loaded bed, C = conical shaped bed.

However, with a similar temperature difference, the temperature of conical-shaped bed is much higher compared to the normal bed.

In fact, the microwave reflections for the conical-shaped loading pattern is less than 0.8%. Compared to the fully loaded pattern, the reflection is reduced significantly. Thus, more microwave energy can be converted into heat and a higher average temperature achieved, which indicates a higher microwave utilization efficiency. However, by changing the shape of the bed, some volume that could have been available to the catalyst is lost, potentially reducing production capacity. Nevertheless, this could be easily compensated by elongating the reactor volume axially.

## 4. Conclusion

The Reverse Traveling-wave Microwave Reactor (RMTR) eliminates or reduces the operational problems seen in the simple "one-way" traveling-wave units. It presents a potential solution to the scale-up of microwave-driven catalytic processes. This current simulation-based study shows the effectivity of the microwave switching frequency on the homogeneity of the temperature distribution inside the catalyst bed. Furthermore, a new catalyst loading pattern is introduced in order to facilitate the microwave energy to penetrate deeper into the catalyst sample. This way, the impedance matching along the loaded reactor is kept and subsequently, the catalyst bed heating uniformity is improved. The results also demonstrate a more efficient coupling between the catalyst sample and the microwave energy for the proposed conical loading pattern.

## CRediT authorship contribution statement

**Farnaz Eghbal Sarabi:** Conceptualization, Methodology, Investigation, Validation, Software, Writing - original draft, Writing - review & editing. **Jiawen Liu:** Validation, Software. **Andrzej I. Stan-**

**kiewicz:** Writing - review & editing, Supervision. **Hakan Nigar:** Writing - review & editing.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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