Traveling Microwave Reactor: Design Challenges and Solutions

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Traveling Microwave Reactor: Design Challenges and Solutions

Traveling Microwave Reactor: Design Challenges and Solutions

Proefschrift

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T.H.E.S.I.S =
True Happiness Ended Since It Started.

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Summary

With the increasing importance of renewable electricity as a primary source of energy on the planet, the importance of electricity–based technologies in process industries is expected to rise as well. Microwave heating is a well–known electricity–based industrial technology that is used in a variety of commercial applications.

This work addresses challenges and explores opportunities for industrial–scale utilization of microwave heating in heterogeneous catalytic gas-phase reactors. Through critical analysis of microwave applicators, the thesis highlights the limitations of traditional cavity–based reactors and underscores the potential of traveling–wave systems for achieving uniform heating profiles in heterogeneous catalytic flow reactors. The contribution of the thesis lies primarily in the design and optimization of traveling microwave reactors (TMRs). Challenges associated with catalyst heating profiles and process scale–up are addressed by introducing a coaxial waveguide structure and a tailored catalyst loading pattern. The TMR model demonstrates its effectiveness in accurately predicting temperature profiles and reaction dynamics along the reactor through simulation studies and experimental validation. Furthermore, the thesis introduces the Reverse Traveling Microwave Reactor (RTMR) as a novel reactor concept aiming to minimize temperature gradients along the catalyst bed by periodic reversal of microwave irradiation. Simulation–based studies showcase the RTMR's potential in achieving temperature uniformity within the catalyst bed, offering new insights into reactor design and scale–up considerations for microwave–assisted catalytic flow processes.

Samenvatting

Met het toenemende belang van hernieuwbare elektriciteit als primaire energiebron op onze planeet, wordt verwacht dat de rol van elektriciteitsgebaseerde technologieën in procesindustrieën ook zal toenemen. Magnetronverwarming is een bekende elektriciteitsgebaseerde industriële technologie die in verschillende commerciële toepassingen wordt gebruikt.

Dit werk behandelt uitdagingen en verkent kansen voor industriële toepassing van magnetronverwarming in heterogene katalytische gasfase– reactoren. Door een kritische analyse van magnetronapplicatoren belicht het proefschrift de beperkingen van traditionele op holtes gebaseerde reactoren en onderstreept het de potentie van reissysteemgolven om uniforme verwarmingsprofielen te bereiken in heterogene katalytische stromingsreactoren. De bijdrage van het proefschrift ligt voornamelijk in het ontwerp en de optimalisatie van reizende magnetronreactoren (TMR's). Uitdagingen met betrekking tot de verwarmingsprofielen van katalysatoren en procesopschaling worden aangepakt door de introductie van een coaxiale golfgeleiderstructuur en een op maat gemaakte katalysatorlaadpatroon. Het TMR-model toont zijn effectiviteit aan in het nauwkeurig voorspellen van temperatuurprofielen en reactiedynamiek langs de reactor door middel van simulatiestudies en experimentele validatie.

Verder introduceert het proefschrift de Omgekeerde Reizende Magnetronreactor (RTMR) als een nieuw reactorconcept dat erop gericht is temperatuurgradiënten langs het katalysatorbed te minimaliseren door periodieke omkering van de magnetronstraling. Op simulatie gebaseerde studies laten zien dat de RTMR potentie heeft om temperatuuruniformiteit binnen het katalysatorbed te bereiken, wat nieuwe inzichten biedt in reactorontwerp en opschalingsaspecten voor magnetronondersteunde katalytische stromingsprocessen.

Preface

I saw a book
whose words were all made of crystal,
a paper made of spring ...

Sohrab Sepehri

Dedicates to,

my dear mom and my sister, Sanaz, for their endless love and support, even from long distance,

my forever best friend, Maysam, for his patience and encouragement,

the light of my life, Radin, for the joy he gives me every single day, and

my beloved hero, who is resting in heaven, watching us, and being the most proud.

Farnaz Eghbal Sarabi
Eindhoven, Winter 2024

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Introduction

Life is an apple,
One should bite it unpeeled

Sohrab Sepehri

This chapter provides a brief overview of microwave heating technology and its applications. The current status of the utilization of microwaves, as a selective and locally controlled heating method, in heterogeneous catalytic flow reactors, is discussed. In this context, the scope of the thesis along with the research questions are presented at the end of the chapter.

Parts of this chapter have been published in *The Chemical Record* **19** (2019) [1].

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1.1. Introduction

Electricity–based technologies are expected to become more significant in process industries as renewable electricity becomes a more important source of energy on Earth. Microwave is an electromagnetic wave situated between the radio wave and infrared in the electromagnetic spectrum. This is translated in a range of wavelengths from 1 m to 1 mm, or frequencies from 300 MHz to 300 GHz, respectively [2]. Whereas, industrial microwave heating is performed at frequencies of 915 MHz and 2.45 GHz [3].

Microwave heating is an established electricity–based industrial technology, which is commercially used for several purposes, including pasteurization, sintering, drying, and thawing. Unlike conventional heating methods that rely on conduction or convection, microwave heating directly couples electromagnetic radiation with the molecular structure of reactants, resulting in rapid and uniform heating throughout the sample.

For over thirty years, researchers have studied carrying out chemical reactions under microwave irradiation, initially in homogeneous liquid–phase systems and then in heterogeneous gas-phase catalytic processes. This unique heating mechanism offers numerous advantages, including shorter reaction times, higher yields, improved product purity, and the ability to perform reactions under milder conditions [4–12].

Three major types of microwave applicators can be considered for carrying out solid–catalysed gas–phase reactions. These are the mono–-mode, the multi–-mode, and the traveling–-wave systems. Mono– and multi–mode microwave applicators, also known as microwave cavities, are the dominant types. However, they generally suffer from non–uniform heating and hot spot formation, due to non–uniform energy fields [13]. 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 [13]. 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 [14]. Frequency variation has no effects on the performance of the traveling–wave microwave applicators [15]. 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 [16].

Despite the promising outcomes of numerous laboratory–scale investigations, microwaves have never been used commercially in heterogeneous catalysis [17]. This is primarily due to the complexity of the interactions between microwaves and solid catalysts, as well as to several important design factors influencing the performance of a continuously operated microwave-assisted flow reactor.

1.2. Interaction of Microwave with Matter and Microwave Heating Principles

When exposed to microwave radiation, the orientation of dipoles causes magnetic energy to be converted to heat through ionic conduction and friction. The molecules of the heated object align with the electric field of the microwaves. The dielectric loss (also known as dipolar polarization or dipolar reorientation) is the dominant factor at high frequencies, i.e., microwave frequencies [18]. In dielectric materials, complex permittivity describes the material's interaction with the electric field of the exposed electromagnetic field (EMF). The complex permittivity is made up of the dielectric constant, ϵ', and the dielectric loss, ϵ''. It is usually represented as a dimensionless function with normalization to the free space permittivity ($\epsilon_0 = 8.85 \times 10^{-12} F/m, \epsilon = \epsilon_0 \epsilon_r$), which is equivalent to the equation below:

$$\epsilon = \epsilon' - j\epsilon'' \quad = \epsilon_0(\epsilon_r{}' - j\epsilon_r{}'') \tag{1.1}$$

where $j = \sqrt{-1}$ is the complex operator. Generally, the real part is associated with the ability of the material to propagate electromagnetic wave, whereas the imaginary part is the loss factor, which shows the ability of a material to dissipate microwave energy [15, 19].

Similarly, the measure of the ability of a material to support the formation of a magnetic field within it is expressed as permeability.

$$\mu = \mu' - j\mu'' \quad = \mu_0(\mu_r{}' - j\mu_r{}'') \tag{1.2}$$

where $\mu_0 = 4\pi \times 10^{-7}$ is the free space permeability, $\mu_r{}'$ and $\mu_r{}''$ represent the ability of transmitting and dissipating magnetic energy, respectively [15, 19].

The majority of microwave–absorbing catalysts in heterogeneous catalysis are non-magnetic materials, with the exception of some metal oxides (e.g., iron, nickel, and cobalt) that heat via magnetic losses. As a result, magnetic losses can generally be neglected [20]. Non-magnetic microwave–absorbing materials are also known as dielectric materials, such as zeolites, which are an important category of heterogeneous catalysts [21]. Some zeolites can be quickly and efficiently heated to a glowing temperature [22]. The main two physical heating mechanisms in microwave heating are based on dielectric and conductive losses.

The material composition, structure, temperature, and microwave frequency all have a strong influence on these loss mechanisms and their contribution [23]. Due to the complexity of the structures and chemical compositions involved, it is challenging to determine the relative contributions of dielectric and conduction losses. Ohgushi [22, 24, 25] and Legras' group [26] investigated the microwave heating mechanism of LindeType A (LTA) and Faujasite (FAU) zeolites in terms of different compensated mobile cations (e. g., Na, K, Ca), Si/Al ratios, and hydration degrees, i.e., water uptake. A recent study [27] used a molecular dynamic simulation for cation movements to theoretically explain the heating mechanism of zeolites under microwave irradiation.

When solid matter is introduced into an electromagnetic field (EMF), such as a microwave field, it either transmits, reflects, or absorbs the exposed wave. Figure 1.1 depicts the interactions of microwaves with various mediums and EMF distributions. Transparent materials are those that allow microwaves to pass through. Due to the extreme low interaction with the EMF, the microwave propagates within these materials with little (or even no) attenuation, as shown in Figure 1.1.a. As a result, most transparent materials (for instance, quartz, borosilicate glass, and PTFE) are used as vessels in many microwave applications. Conductive materials (e.g., aluminum, silver, copper), also identified as opaque materials, reflect microwaves (Figure 1.1.b.) and are essential parts in the construction of transmission lines (e.g., wave-guide, coaxial cable). Since absorbing materials (Figure 1.1.c.) have polar features, they can absorb microwave energy through polarization and convert it to heat [28–30].

However, in microwave-assisted catalytic systems, the dielectric properties of the catalysts are the deciding factors. In those systems, the catalyst serves a dual purpose, converting both electromagnetic energy and reactants. Dielectric catalysts are suitable for specific reactions, depending on the operating temperature required. In general, less energy-intensive pro-

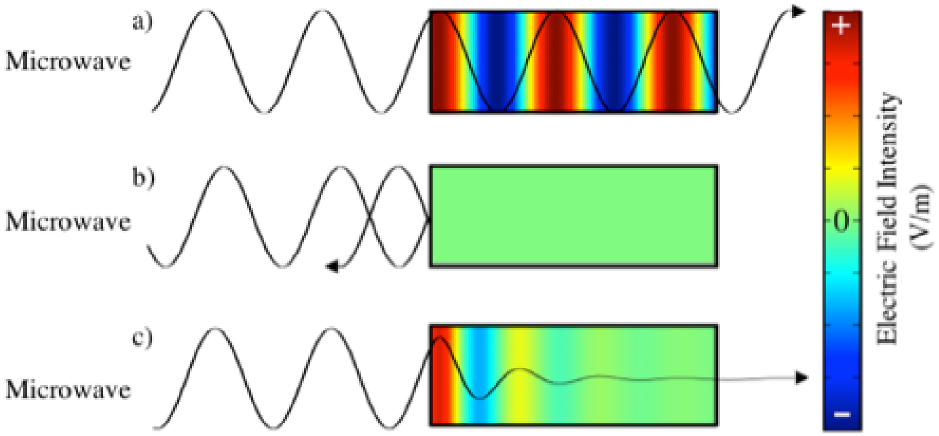

Figure 1.1: Interactions between microwaves and different materials and EMF distributions: A) transparent, B) reflective, and C) absorbing medium. The simulations were performed on COMSOL Multiphysics (v.5.3a) simulation environment using a Radio Frequency module.

cesses have a greater potential for use in microwave-assisted systems with moderate dielectric materials. Endothermic reactions, which require high temperatures (>300°C), necessitate the use of a catalyst with adequate dielectric properties.

In the void spaces inside the catalyst bed, dielectric reactants and products may contribute to electromagnetic heat conversion. The dielectric constant of liquid reactants, such as water, is higher than that of gaseous reactants. As a result, microwave results differ depending on whether the catalytic mechanism is gas-solid or liquid-solid. Table 1.1 summarizes the dielectric properties of the common materials and reactants.

Because polarization depends on particle density, which is low in gases, all gases have dielectric constants similar to air,~1, as shown in Table1.1. Therefore, in gas-solid interactions, the dielectric characteristics of the catalytic bed material control the amount of heat generated. In general, gas–solid reactions take place at high temperatures, making microwave–assisted systems for such reactions a difficult material to develop. The effects of the microwave in the catalytic bed on liquid–solid reactions are dependent on the dielectric discriminating between the solid catalyst and the flowing re-

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Table 1.1: Dielectric properties of common materials and reactants at 2.45 and 2.5 GHz and ambient temperature

Materials	**Dielectric Properties** $(\epsilon_r{}' - j\epsilon_r{}'')$	**Ref.**
Air	1-j 0	[3]
Benzene	2.3-j n/a	[31]
Nitrogen	1-j 0	-
Water	77-j 13	[3]
Methane	13-j n/a	-
Carbon	26-j 10	[32]
Methanol	32.7-j 30	[31]
Quartz	3.78-j 0.001	[3]
Ethanol	24.6-j 1.5	[31]
Teflon	2.04-j ~0	[3]

n/a indicates that the imaginary part (loss factor) is not available.

actant (s).

1.3. Microwave Applications in Heterogeneous Gas–Solid Catalysis: Advantages and Challenges

Microwave energy's application in heterogeneous gas–solid catalysis has been studied over the past few decades. Researchers have consistently observed promising improvements in reaction efficiency, selectivity, and kinetics compared to conventional heating methods [31, 33–36]. Early studies by Wan et al. was a turning point in microwave–assisted catalysis [37]. Their groundbreaking research on methane conversion to acetylene over activated carbon catalysts demonstrated significant enhancements in reaction kinetics and selectivity [38]. This finding prompted further investigations into various catalytic reactions, revealing similar trends of improved performance under microwave irradiation [39–44].

Subsequent studies by different research groups corroborated these findings, showing enhanced conversions and selectivities in various gas–solid catalytic reactions when subjected to microwave irradiation. For instance, Zhang [45, 46], Cha [47–49], and Wang's groups [50] explored different reactions, such as sulfur dioxide reduction, NOx decomposition, and methane oxidative coupling, respectively, all observing favourable outcomes under microwave conditions. Chen et al. [51, 52] and Bond et al. [9] investigated methane's oxidative coupling in conventional and microwave settings. According to the findings of both study groups, the process under microwave heating is conducted at temperatures between $200\,\mathrm{K}$ and $400\,\mathrm{K}$ lower than those of traditional heating while maintaining the same level of product selectivity.

Koch et al. explored hydrogen cyanide (HCN) synthesizing and discovered higher selectivity compared to the conventional process [53]. A thermal gradient could explain this because the catalyst's surface was at a higher temperature than the gas phase. Additional research, such as those conducted by Sinev, clarifies the complex impacts of microwave radiation on gas–solid catalytic processes. Sinev's study on ethane dehydrogenating revealed varying impacts depending on catalyst composition[54]. Menendez et al. investigated microwave–assisted dry reforming of methane with activated carbon as a catalyst and microwave receptor, finding up to a 40% higher conversion of both CO2 and CH4 compared to electric furnace heating under similar conditions [55].

Furthermore, the utilization of microwaves in methane dehydroaromatization (MDA) presents a promising avenue for methane valorization into higher–value hydrocarbons [56]. Advanced reactor designs, such as structured reactors combined with microwave heating, offer the potential for inhibiting coke formation and extending catalytic stability in MDA processes [17].

Integrating microwaves into catalytic reactions, particularly in gas–solid systems, holds immense promise for enhancing reaction selectivity, which means that the solid catalyst (microwave absorbing media) can be selectively heated. As a consequence, the temperature of the solid catalyst surface, where chemical reactions typically occur, is higher than that of

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the gas phase. Unlike conventional heating, this selective microwave heating phenomenon may result in significant energy efficiency improvements [28, 57, 58]. Moreover, efforts to improve selectivity and overcome challenges in non–selective gas–phase reactions, such as in the CO2–driven isobutane dehydrogenation process, have demonstrated promising results under microwave heating conditions, offering high yields essential for commercial implementation[59].

In addition to this, microwave–assisted catalytic processes have shown promise in addressing environmental concerns and rapid start–up/shut–down. The reduction in reaction temperatures and shorter reaction times associated with microwave heating can contribute to lower energy consumption and reduced greenhouse gas emissions.

Several high–potential processes studied in microwave–assisted systems are listed in the Table 1.2.

Table 1.2: Examples of improved promising processes under microwave-assisted systems.

Process	**Features**	**Ref.**
Ammonia for hydrogen production	Ammonia conversion improvement by factor of 2	[60]
Methane dry reforming	Microwave-assisted system resulted at higher conversion than electrical heating	[61]
Methanol steam reforming for hydrogen production	Higher conversion of methanol for MW-assisted-system with similar average bed temperature	[29]
Oxidative (non-oxidative) coupling of methane	Higher methane conversion at lower operating temperature with the absence of oxygen	[12]
Acetylene production	Increase in methane conversion over activated carbon using microwave heating	[38]

Despite the substantial advantages demonstrated by microwave–assisted catalytic reactions, there remains a gap in understanding the precise mechanisms underlying these enhancements beyond mere heat generation. Various hypotheses, including non-thermal effects and selective heating, have been proposed to explain the observed improvements in reaction conversion and selectivity [12]. However, challenges in obtaining reliable temperature measurements under microwave heating hinder the validation of these hypotheses [62]. According to Kappe's instructional review, precisely measuring the temperature in a system that is heated by microwave is a challenging endeavor [63].

Coke formation is generally unavoidable in heterogeneous hydrocarbon catalysis, and it reduces catalyst activity. Several studies found that coke formation was less pronounced in microwave–assisted systems [64, 65]. In conventional systems, the deactivation of the catalyst is compensated by increasing the temperature to maintain a certain conversion. The microwave interaction with the catalytic bed, on the other hand, may improve coke formation due to its excellent carbon dielectric properties. Non–homogeneous coke formation in the catalytic bed, on the other hand, may introduce greater temperature non-homogeneity.

Another critical challenge in the microwave–catalyst system is to identify a suitable material for both functions: microwave energy conversion and the desired chemistry. Hence, the best catalyst and chemistry for conventional heating and microwave heating are frequently different. Numerous researchers have looked into microwave–assisted heterogeneous catalytic processes– see for instance [8, 20, 38, 60, 66–69]. In addition to this, scaling up microwave systems presents distinct challenges compared to conventional processes, such as the need to adjust the operational frequency from 2.45 GHz to 915 MHz to increase reactor size, requiring thorough studies to account for changes in catalyst penetration depth with frequency variation before designing the scaled–up reactor [70].

Overall, microwave technology offers a viable path forward for chemical process advancement, with potential applications ranging from petrochemicals to environmental remediation to renewable energy generation. It will take ongoing research efforts to clarify the underlying processes and optimize process conditions for microwave–assisted catalysis to reach its full potential for efficient and sustainable chemical transformations.

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1.4. Research Questions and Outline of Thesis

This study aims to investigate the feasibility of process scale–up using microwave energy and to apply an appropriate solution to create a well-controlled, highly uniform heating profile along the catalyst bed. In this manner, a new type of equipment design is required that would introduce the microwave field uniformly along the catalyst bed structure. In order to determine the possibility of achieving these goals, the specific research questions are formulated as follows:

1. What are the main limitations of mono–mode and multi–mode cavities in scalling–up the microwave applicators?
2. What are the specific challenges with the design of a traveling microwave wave-guide in accordance with process intensification principles?
3. What requirements need to be met in the design of traveling microwave equipment in order to obtain a fairly uniform microwave field distribution along the reactor?
4. What is the role of impedance matching in the design of traveling microwave reactor? Aside from empty reactor design, which physical limitations and properties have to be taken into account to acquire a uniform heating profile along the catalyst bed while the reactor is loaded with a microwave–susceptible catalytic fixed–bed?
5. What is the role of the operational mode of a traveling microwave catalytic reactor? In particular, can we increase the uniformity of the temperature distribution inside the catalyst bed by periodically switching the direction of the microwave flow?

The thesis consists of six chapters, including a general introduction in this chapter, Figure 1.2. The content of the remaining five chapters is outlined as follows:

Chapter 2

This chapter investigates the physics of standing–wave and traveling–wave concepts. The basic types of microwave reactor systems are introduced, along with the traveling microwave reactor principle. It also discusses the current status and future perspectives of using microwaves as a selective and locally controlled heating method in heterogeneous catalytic flow reactors. Various factors related to the microwave–catalyst interaction and the design of microwave–assisted catalytic reactor systems are analyzed. The analysis clearly shows the superiority of the traveling–wave systems over the mono–mode and multi–mode cavity–based systems when it comes to

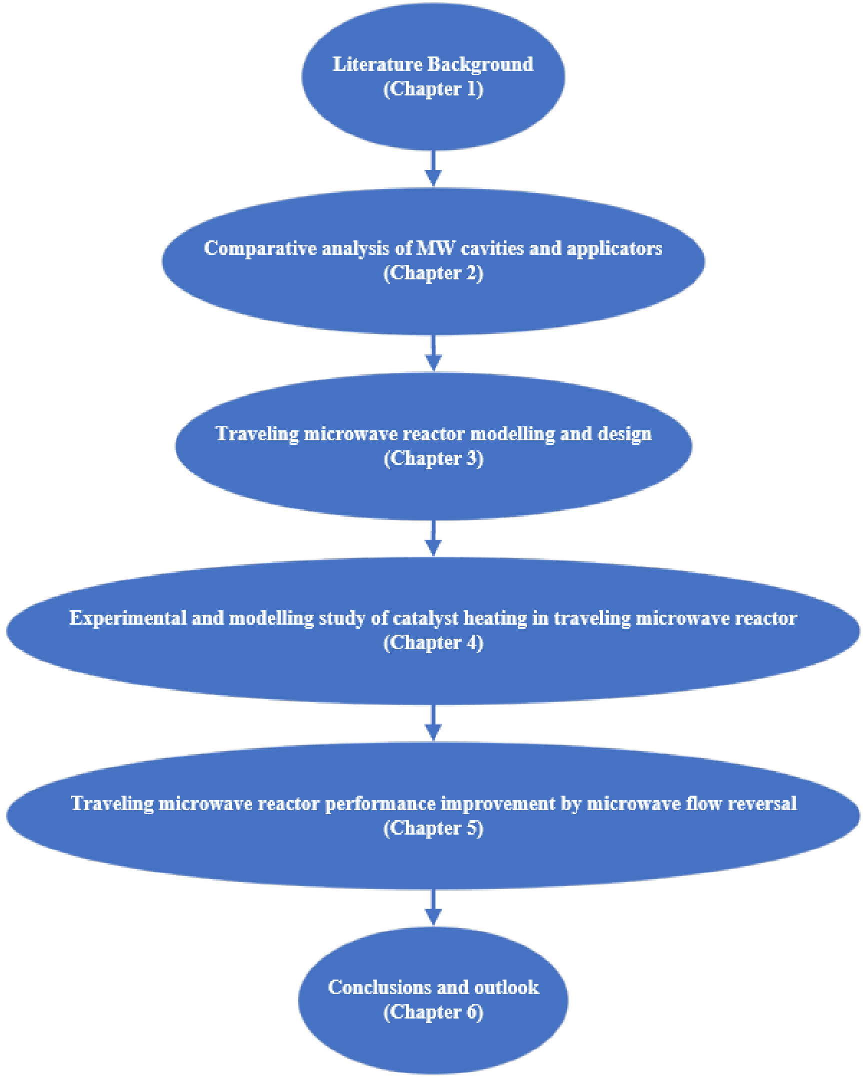

Figure 1.2: Outline of the Thesis

the design and application of microwave flow reactors at relevant production scale–up.

Chapter 3

This chapter addresses the superiority of the traveling microwave reactor over the typical microwave applicators in process scale–up. The applicability of typical multi–mode and mono–mode cavities is limited because both systems, due to generating standing microwaves, either suffer from non–uniform heating or size limitations. Contrary to common cavity–based microwave applicators, the traveling microwave reactor has the potential to enable process scale–up, a better coupling of microwave energy with microwave–susceptible catalysts, and consequently highly uniform microwave heating. In this chapter, the engineering challenges entailed with the design of a traveling microwave waveguide are explained, and appropriate solutions are developed. The new traveling microwave reactor with a coaxial waveguide structure is presented. Furthermore, in order to keep the impedance matching and minimize the microwave reflections while the reactor is loaded with catalyst samples, the new reactor's loading patterns are introduced. The uniformity of the catalyst's heating profile for the proposed method is demonstrated via the simulation results.

Chapter 4

The traveling microwave reactor experimental setup is demonstrated in this chapter. The results of the heating test are shown for different types of catalysts. The experimental results prove the validity of the simulations.

Chapter 5

In this chapter, a novel heterogeneous catalytic reactor concept is introduced, based on the coaxial wave–guide structure, to improve the performance of microwave heating operations. The Reverse Traveling Microwave Reactor (RTMR) 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. The study provides new insights into the design and scale-up of microwave-assisted catalytic flow processes.

Chapter 6

Chapter 6 summarizes the results and findings of this research work. It also proposes recommendations for further development of the research.

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2

Microwave Reactor Systems

This chapter investigates the physics of standing wave and traveling wave concepts. Basic alternative types of microwave reactor systems are discussed. Various factors related to the microwave–-catalyst interaction and the design of microwave–-assisted catalytic reactor systems are analyzed. The analysis clearly shows the superiority of the traveling–wave systems over the mono–mode and multi-–mode cavity-–based systems when it comes to the design and application of microwave flow reactors at an industrially relevant production scale.

Parts of this chapter have been published in The Chemical Record Journal **19** (2019) [1].

2.1. Introduction

Despite the encouraging results of various laboratory-scale studies, commercial implementations of microwaves in heterogeneous catalysis are nonexistent. This is primarily due to the complexity of the interactions between the microwaves and solid catalysts, as well as to several important design factors influencing the performance of a continuously operated microwave–assisted flow reactor.

2.2. Standing Wave and Traveling Wave Concepts

A wave can be described as a disturbance that travels through a medium, transporting energy from one location (its source) to another location without transporting matter.

As a wave is observed traveling through a medium, a crest is seen moving along from particle to particle. The crest is followed by a trough that, in turn (in the form of a sine wave), traveling through the medium. This sine wave pattern continues to move in uninterrupted fashion until it encounters another wave along the medium or until it encounters a boundary with another medium. This type of wave pattern seen traveling through a medium is referred to as a traveling wave, (see Figure 2.1). Traveling waves are observed when a wave is not confined to a given space along the medium. The most commonly observed traveling wave is an ocean wave.

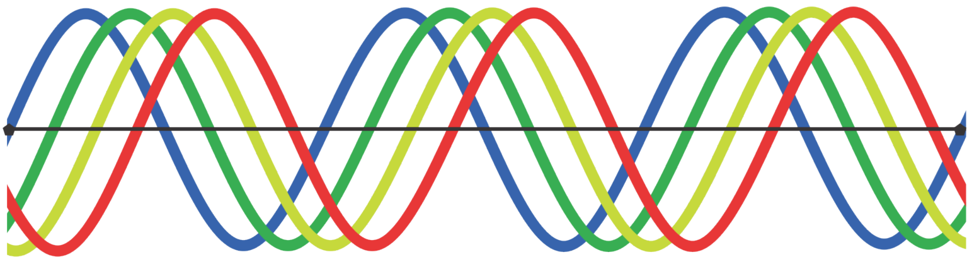

Figure 2.1: Traveling wave pattern. Different colours show different snapshots of the wave, taken a small time interval apart, while the wave is traveling.

The standing wave, also called the stationary wave, is a combination of two waves moving in opposite directions, each having the same amplitude and frequency. The phenomenon is the result of interference; that is, when waves are superimposed, their energies are either added together or cancelled out. In the case of waves moving in the same direction, interference produces a travelling wave; for oppositely moving waves, interference produces an oscillating wave fixed in space. A vibrating rope tied at one end will produce a standing wave, as shown in Figure 2.2. At all times, there are positions (N) along the rope, called nodes, at which there is no movement at all; there, the two wave trains are always in opposition. On either side of a node is a vibrating antinode (A). The antinodes alternate in the direction of displacement.

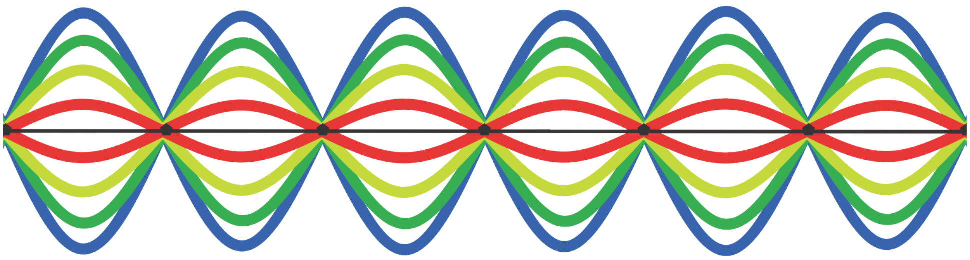

Figure 2.2: Standing wave pattern. Different colours show different snapshots of the wave, taken a small time interval apart, while the positions of the nodes (the points that never move) are fixed along the wave.

2.3. Basic Types of Microwave Reactor Systems

2.3.1. Reactors Based on Multi–Mode and Mono–Mode Cavities

Microwave heating takes place in the so-called ”microwave applicators” or ”cavities” in our kitchens and in the industry. These applicators are classified into three types: multi-mode, mono-mode, and traveling wave.

Multi–mode applicators, such as microwave ovens in households, can be represented simply as cuboidal metal cavities (Figure 2.3). Here, microwaves are reflected from the cavity walls and (partially) from the heated sample (load). Because of these reflections, multi–mode cavities exhibit a chaotic interaction with the sample and a non–homogeneous microwave field, resulting in a non–uniform heating profile and the formation of hot spots. To that end, in those cavities, a rotating disk and a mode stirrer (rotating reflector, see Figure 2.3) are used to make the microwave field distribution as homogeneous as possible. Despite the heating uniformity issue, multi–mode cavities are commonly used in industry due to their low cost, ease of construction, capacity to heat up large loads, and versatility. A common issue in multi–mode applicators is a low field density compared to the high generated microwave power (1000 W to 1400 W), resulting in poor performance in the case of small–volume samples [2].

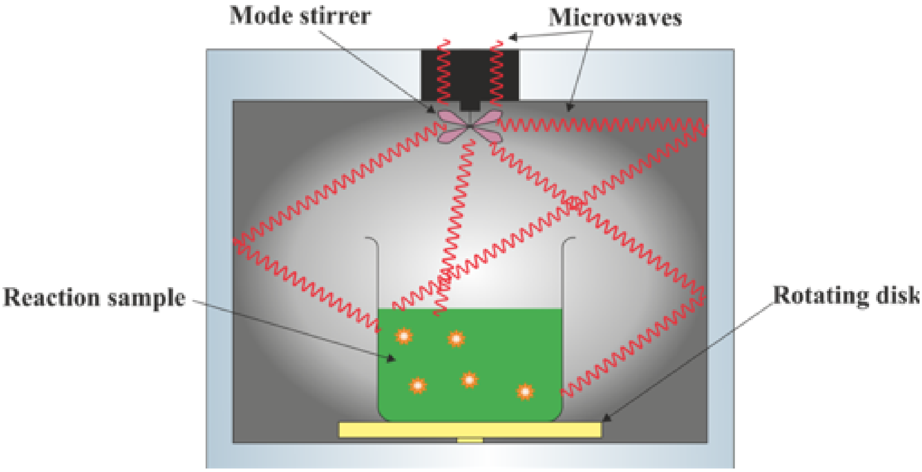

Figure 2.3: Multi–mode microwave cavity, adapted from reference [3]

In mono–mode cavities, however, only one mode exists and generates a standing wave inside the cavity. The irradiated material is placed where the electromagnetic field is at its strongest (Figure 2.4) [3].

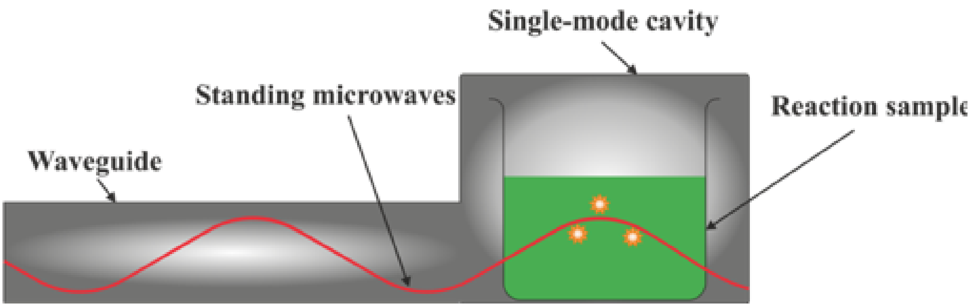

Figure 2.4: Mono–mode microwave cavity, adapted from reference [3]

Because of this feature, the size of mono–mode cavities is strictly limited to less than half of the wavelength, in order to provide a more uniform heating profile. As a consequence, knowing that the typical wavelength of microwave is of the of the order of $10\,\mathrm{cm}$, mono–mode applicators are typically used to process volumes of up to $200\,\mathrm{mL}$, which is their primary limitation [3]. However, when compared to multi–mode cavities, mono–mode cavities can provide higher field strength while consuming less energy. Although frequency changes and the position and dielectric properties of the heated sample can have a strong influence on the microwave field pattern inside a mono–mode cavity, Patil et al. have demonstrated the effect of the shape and dimensions of a milli–reactor setup on controlled and efficient microwave heating in a mono–mode cavity [4].

As shown above, the applicability of typical multi–mode and mono–mode cavities is limited because both systems suffer from non–uniform heating and are highly dependent on the frequency and dielectric properties of the sample. These negative characteristics may not be critical in the case of drying or thawing, but they are critical in the case of controlled chemical reactions.

2.3.2. Reactors Based on Traveling Wave Principle

The traveling wave reactor, in contrast to the previously described cavities, has the potential to provide highly uniform microwave heating by avoiding reflections and resonant conditions [5]. The microwave field inside the reactor in a properly designed traveling wave system travels in only one direction to avoid non–uniform electromagnetic interference patterns and, as a result, non–uniform heating along the reactor's axial direction. Traveling wave reactors can be thought of in a variety of configurations. In recent years, the most popular TWR concept, with coaxial cable configuration, has been studied [6, 7]. Figure 2.5 shows a cross–sectional longitudinal view of a coaxial traveling wave reactor, where microwave energy delivered at the inlet is absorbed by the catalyst and dissipated at a high heating rate.

The stainless–steel inner and outer conductors form a coaxial wave–guide structure and their diameters are adjusted so that the wave–guide's characteristic impedance remains constant with the feed transmission system's characteristic impedance ($50\,\Omega$). Stainless–steel is a common material for MW wave–guides as it is easy to manufacture, can withstand high temperatures, provides resistance to eddy currents, and has high thermal conductivity. This reactor is cylindrically symmetric along the inner conductor's central axis, and catalysts of various shapes, such as particles, foams, or monoliths, can be placed in the annular space between the conductors, with process fluids or reactants flowing in the axial direction through the catalyst load.

2.4. Design Challenges in a Microwave Reactor System

As stated in the preceding section, the microwave field pattern within a mono–mode cavity may be highly influenced by frequency changes as well as the location and dielectric properties of the sample. This may cause a thermal runaway, hot spots in the material, and, in some cases, a loss of interaction between the microwave energy and the material.

There are several mechanisms that should be considered in this framework for effective energy transfer to the substrate, compensating for varying dielectric properties and frequencies, and maintaining the coupling between the material and the applicator throughout the process. Currently, the best approach to this issue is to use an inline monitoring and feedback–control mechanism for temperature actuation and microwave energy input, as well as auto–tuning of the microwave field by tuning elements (e.g., stub–tuner and plunger; i.e., movable walls). The direction of the tuner influences both standing wave patterns and the amplitude of the electric field along the waveguide, i.e., cavity. As a result, the tuner alters the actual length of the cavity in the direction of microwave propagation. Subsequently, tuners will adapt and optimize the interaction between various materials (with various dielectric properties) and the microwave. Figure 2.6 depicts various electric field patterns at 2.45 GHz in a mono–mode rectangular microwave cavity.

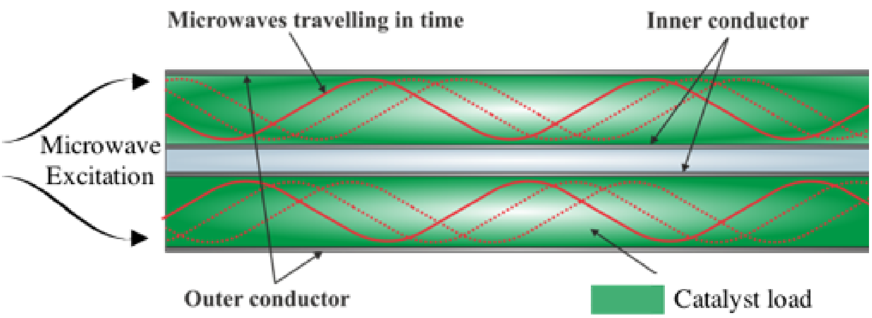

Figure 2.5: Coaxial traveling microwave reactor configuration.

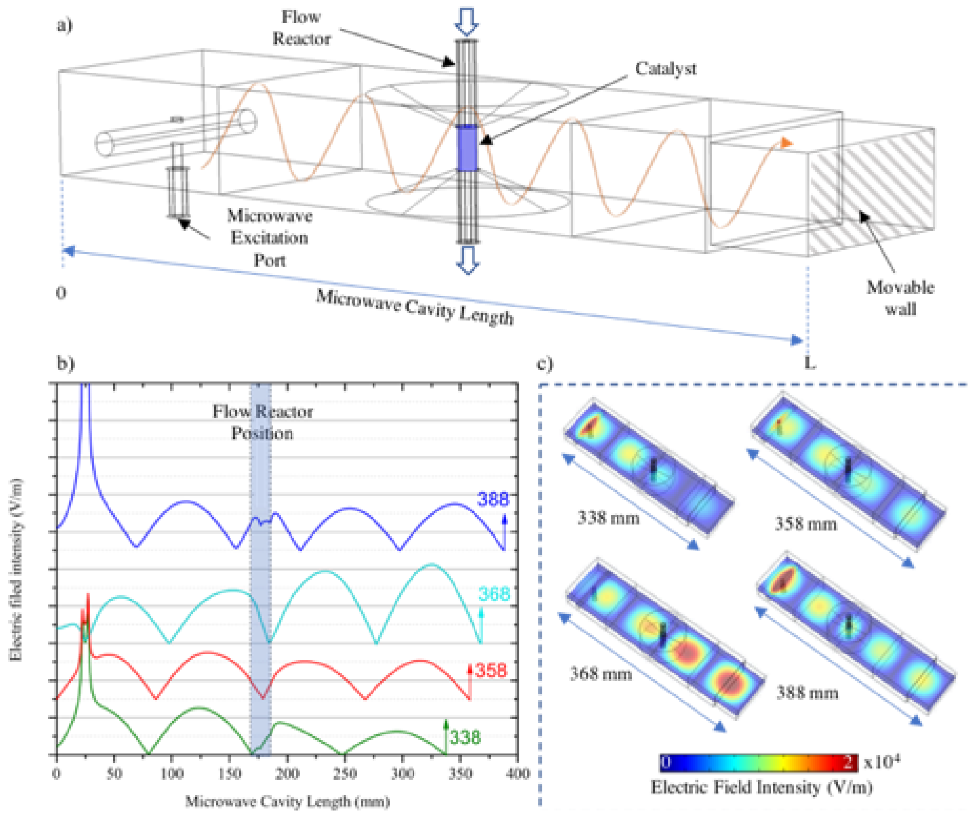

Figure 2.6: a) Perspective view of mono–mode rectangular microwave cavity with an adjustable tuner (i.e., movable walls), and b-c) simulated normalized electric field distributions, at the centre along the cavity, in different microwave cavity lengths. The simulations were performed in COMSOL Multiphysics (v. 5.3a) simulation environment using a Radio Frequency module at $2.45\,\mathrm{GHz}$ with an input power of $100\,\mathrm{W}$.

It is clear that changing the cavity length affects the standing wave patterns within the cavity, as seen in the Figure 2.6b-c. In this layout, the optimal cavity length for the flow reactor location is $388\,\mathrm{mm}$, considering the flow reactor position (Figure 2.6b). At this length, the maximum amplitude of the standing wave corresponds to the direction of the flow reactor. This match causes the substance (catalyst) present in the reactor to heat up. It is important to emphasize that this simulation is performed at room temperature. Dielectric properties, in general, change with temperature and material composition. This could result in a variety of standing wave patterns. To sustain adequate operating conditions and maximize the interaction between the catalyst and the microwave field in a mono-mode cavity-based reactor, tuners must be continuously adjusted. Otherwise, the object would no longer be able to be heated. Furthermore, the wave frequency is also a factor that can influence the electric field propagation within the microwave cavity.

Contrary to the mono–mode cavity, there is no need to deal with frequency variations throughout the operation in the traveling wave reactor. The only elements that should be considered in TWR design are the reactor size and microwave reflections along the whole system, in order to ensure that no standing wave is produced throughout the structure.

The reactor measurements are chosen based on electromagnetic wave propagation constraints, and as a result, the diameter of the outer conductor, i.e., the outer wall of the entire coaxial structure, cannot exceed half of the electromagnetic wavelength. Therefore, the upper limit of the diameter is roughly $61\,\mathrm{mm}$ for the typical frequency of $2.45\,\mathrm{GHz}$ [3].

Microwave reflection is critical to the efficiency of a traveling-wave reactor. A smaller microwave reflection leads to faster heating and fewer energy losses since more microwave energy penetrates the sample. To reduce electromagnetic reflections in the coaxial structure TWR, the characteristic impedance of the waveguide should be set and retained at the feed transmission system's characteristic impedance. It is also important to maintain impedance matching when adding the dielectric load (catalyst) within the waveguide. To do this, the reactor must be progressively loaded in order to prevent abrupt impedance changes. Otherwise, it can reflect a large portion of the incident waveback. This will result in the formation of a standing wave within the waveguide and, consequently, a non–uniform heating profile and the formation of hot spots.

Figure 2.7 depicts a simulated heating profile in a traveling microwave reactor for two different loading patterns under identical conditions. As seen in Figure 2.7a, the catalyst cannot communicate with the electromagnetic wave effectively due to the large microwave reflections (more than 12%). Even with an increased energy supply, the catalyst cannot be heated, and hot spots form within the reaction region. On the other hand, in Figure 2.7b, where the microwave load is gradually introduced to the system, the temperature profile is distributed more smoothly without forming any hot spots. In the latter configuration, the white space between the conical catalyst load and the reactor outer conductor is assumed to be filled with a microwave–transparent material, e.g., air or quartz wool. The catalyst material, used for this simulation study, is SiC with constant dielectric properties of $\epsilon_r = 17 - i0.70$. More detailed information about the simulated configurations in Figure 2.7 can be found in Table 2.1.

Table 2.1: Simulated Configurations Details

Reactor Dimension		
Inner Diameter	**Outer Diameter**	**Length**
15.7 [mm]	54.8 [mm]	30 [cm]
Catalyst Bed Configuration	**Figure 2.7(a)**	**Figure 2.7(b)**
	Cylindrically filled with solid SiC	Conically filled with solid SiC

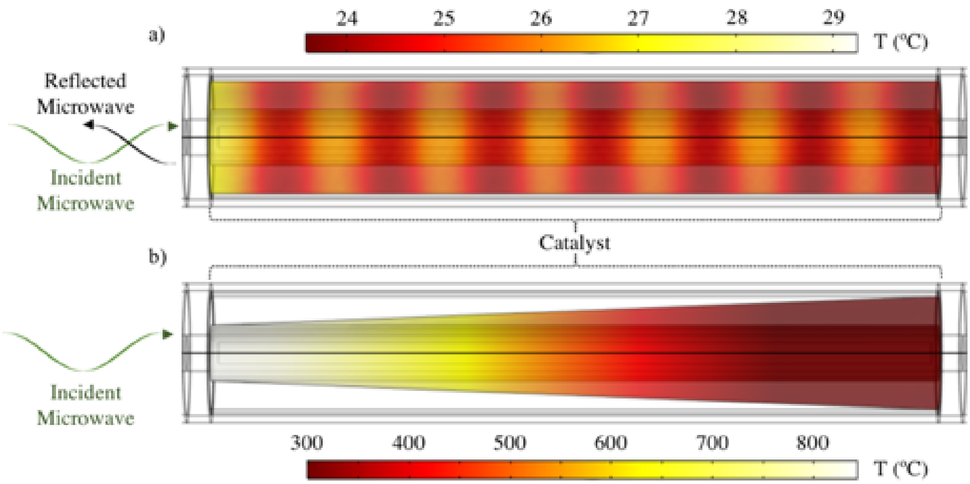

Figure 2.7: Simulated catalyst heating profile in coaxial traveling wave reactor in different catalyst loading configurations: a) Fully b) Partially filled by the catalyst.

2

2.5. Conclusions

The most important conclusion drawn from this chapter is that microwaves must flow in order to achieve a practical and scalable microwave-assisted heterogeneous catalytic process in a flow system. On larger scales, cavity–based structures, whether standing–wave or multi–mode, cannot ensure a uniform, well–controlled temperature distribution in the catalyst bed. In traveling-wave systems, on the other hand, the temperature profile control can be achieved by other means like controlling the distribution of dielectric properties in the catalyst bed, the waveguide geometry, and the frequency of the microwave. A multidisciplinary approach and collaboration involving catalysis, material science, electrical and chemical engineering are required for success here. In the following chapters we will show our first steps into this multidisciplinary field.

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3

Coaxial Traveling Microwave Reactors: Design Challenges and Solutions

Microwave chemistry applications have been investigated for more than three decades. The applicability of typical multi–mode and mono–mode cavities is limited because both systems, due to generating standing microwaves, either suffer from non–uniform heating or size limitations. Contrary to common cavity–based microwave applicators, the traveling microwave reactor has the potential to enable process scale–up, better coupling of microwave energy with microwave-susceptible catalysts, and consequently highly uniform microwave heating. In this work, the engineering challenges entailed with the design of a traveling microwave waveguide are explained, and appropriate solutions are developed. A new traveling microwave reactor with a coaxial waveguide structure is presented. Simulation results show that there is no standing wave generated along the structure. Furthermore, in order to keep the impedance matching and minimize the microwave

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reflections while the reactor is loaded with catalyst samples, new reactor's loading patterns are introduced. Simulation results show that for the proposed method, a microwave–susceptible catalytic fixed–bed could interact more efficiently with microwave energy and produce a uniform heating profile.

3.1. Introduction

Microwave–assisted processes offer several advantages over the traditional ones in terms of reaction acceleration, volumetric heating, improving product yields and selectivity, and reducing undesired side reactions [2–5]. Lower bulk gas temperatures further enhance reaction selectivity, particularly with microwave-absorbing catalysts [6, 7]. However, a major challenge in industrial applications is scaling up microwave reactors reliably [8].

Three main types of microwave applicators are currently used: multi–mode, single–mode (mono–mode), and traveling microwave reactors (TMR) [9–11]. Multi–mode and mono-mode cavities have limitations due to non–uniform heating and sensitivity to sample properties. In contrast, traveling microwave reactors offer highly uniform microwave heating by avoiding resonant conditions, enabling potential process scale–up beyond cavity system limitations [10, 11]. TMR design focuses on reactor size and managing microwave reflections to prevent standing wave patterns in the system.

Herein, we present a new traveling microwave reactor design, explain the engineering challenges entailed with this design, and provide opportunities to develop solutions required for this type of reactor. In this context, we present the critical factors for the design procedure of a new coaxial traveling microwave reactor and describe its advantages through numerical simulations. Moreover, the effects of the catalyst geometry and loading pattern on its heating profile are studied and optimized to have a more efficient and uniform heating profile along the reactor.

3.2. Traveling Microwave Reactor Design

3.2.1. The Characteristic of the Waveguide

The first step in the design of a microwave reactor is the selection of the most appropriate type of waveguide for the given application. TMRs can be implemented in different waveguide types and configurations. Common types of waveguides, which can be considered for the required scale, are rectangular, circular and coaxial, where each has some advantages. The concept of TMR with a coaxial cable configuration has been studied in the last few years [10–16]. This configuration has several advantages over the other types of waveguides in terms of having no cut–off frequency for the transverse electromagnetic (TEM) mode of propagation [17, 18]. In the TEM mode, electric field lines run radially while magnetic field lines run in circles around the inner conductor, see Figure 3.1-b. Consequently, the available operating frequency range of this waveguide is much broader than of other types.

Designing a microwave excitation section in coaxial waveguides is much simpler due to the coaxial nature of the most common microwave generators. Furthermore, the entire inner space of the waveguide can be used for process functionalities because the electromagnetic field is uniform over the circumferences of conductors in coaxial waveguides, while in the case of rectangular and circular ones, this space is limited to only a portion of their sides. However, it is also important to design the cross-section of the reactor in such a way that the electromagnetic field effectively couples with the catalytic bed inside.

Axial and radial field distributions in a traveling microwave reactor, in which the inner and outer conductors form the coaxial waveguide structure, are depicted in Figure 3.1. Microwave energy delivered at the inlet is absorbed by the dielectric material, e.g., a catalyst, and dissipated into heat.

The reactor dimensions are selected based on the restrictions imposed by the electromagnetic wave propagation. In that sense, the first requirement is to adjust the impedance matching of different parts to minimize the unwanted microwave field reflections. The second is to make sure that the device works in a single TEM mode by adjusting the cut–off frequency well above the working frequency. The design process, considering these two requirements, is explained below.

The Dimensions of the Waveguide

Microwave reflection presents an important aspect of reactor performance. A smaller microwave reflection leads to faster heating and less energy loss since more microwave energy penetrates into the dielectric sample (e.g., catalyst). In order to minimize the electromagnetic wave reflections in the coaxial structure reactor, the characteristic impedance of the waveguide should be adjusted to be maintained at the characteristic impedance of the feed transmission system. The most common industry standard for the characteristic impedance is $50\,\Omega$ since it presents a great compromise between power handling and low loss [19, 20]. All the equipment, e.g., microwave generators, used in this study are assumed to have an impedance of $50\,\Omega$.

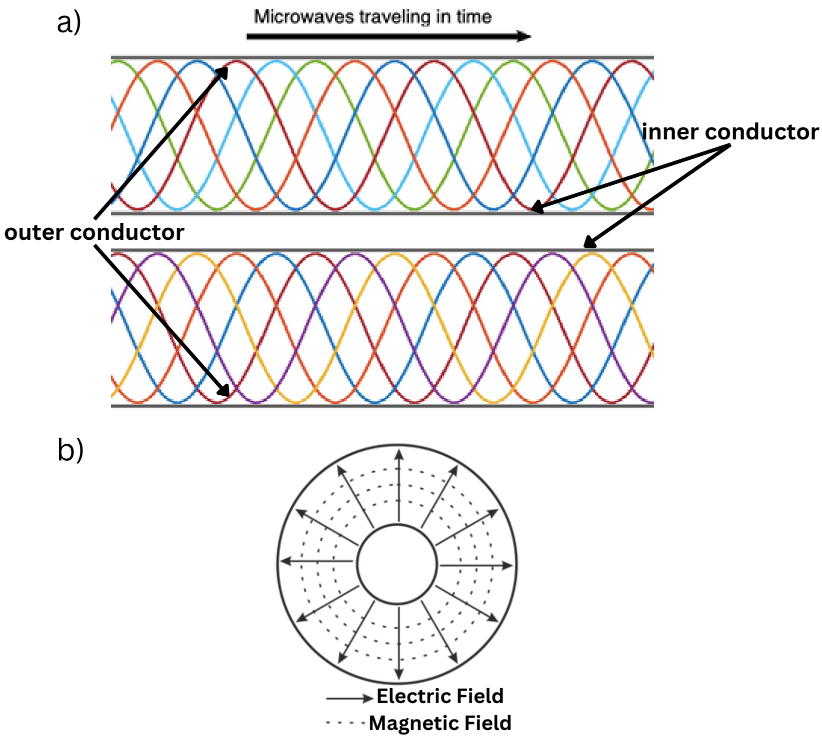

Figure 3.1: a) Axial and b) radial electric field distribution in the coaxial traveling microwave reactor.

Characteristic impedance is the ratio of the voltage of the transmission line to the current, which is described for the TEM mode in a coaxial waveguide as [19]:

$$Z_0 = \sqrt{\frac{L}{C}} = \frac{\sqrt{\frac{\mu}{\epsilon}}}{2\pi} \ln \frac{r_o}{r_i} = \frac{60}{\sqrt{\epsilon_r}} \ln \frac{r_o}{r_i} \tag{3.1}$$

where L stands for the total self-inductance of the two conductors, C is the capacitance of two conductors, r_o and r_i are the radii of the outer and inner conductors, respectively, and ϵ_r is the relative permittivity of the dielectric material placed between the conductors (insulator).

For the case discussed herein, the dielectric material of the waveguide was assumed to be air $(\epsilon_r \approx 1)$. Therefore, considering Equation 3.1, in order to keep the characteristic impedance of the customized waveguide equal to $50\,\Omega$, the necessary ratio of the radii of the outer and inner conductors was found to be 2.3.

3

Cut–off Frequency of the Waveguide

The cut–off frequency of a coaxial transmission line is the frequency at which the excitation of the next unwanted transverse electric, e.g., TE11 [1], mode starts to propagate. This mode has a different propagation velocity and microwave field distribution than the desired TEM mode. This causes perturbations in the electromagnetic behaviour of the device and the heating pattern of the sample. The cut–off frequency in a coaxial transmission line can be calculated using the following formula [19]:

$$f_c = \frac{c}{(r_o + r_i)\pi\sqrt{\epsilon_r}} \tag{3.2}$$

where c is the speed of light in vacuum. Considering this equation and assuming that air is the filling medium, to adjust the dimensions to have the cut-off frequency above $2.45\,\mathrm{GHz}$, the sum of the radii should be lower than $38.94\,\mathrm{mm}$. Otherwise, microwave reflections are noticeable which lead to a standing wave pattern instead of a traveling one.

Combining the results of the constraints achieved from equations 3.1 and 3.2, the radii were selected to result in $Z_0 = 50\,\Omega$ and $f_c = 2.6\,\mathrm{GHz}$. It means that only the desirable TEM mode can propagate at frequencies below the cut-off frequency of $2.6\,\mathrm{GHz}$.

3.2.2. The Microwave Transition Part and the Absorber

In the previous section, the optimized dimensions of the custom-design coaxial microwave reactor were obtained. The next step is deciding how the microwave field will be introduced into the coaxial microwave reactor. In this sense, designing a microwave transition part is essential to excite the reactor through a standard $50\,\Omega$ type–N coaxial connector with minor reflections.

[1]TE11 mode describes waves in a cylindrical tube where electric field lines sway side to side while making one loop around the tube and oscillate once along its length.

Different methods can be employed in designing the transition section, but the most practical one with the least manufacturing complexity is the tapered coaxial transition. The taper curve can be exponential, logarithmic or even linear, each delivering a compromise between operating bandwidth and transition length [21]. In the current study, we are not bound by strict dimensional or bandwidth requirements, thus we can adopt a simple linearly tapered curve in the design of the transition part. This will lead to a conical-shaped coaxial microwave transition, which is placed between the type–N coaxial connector and reactor intersection, see Figure 3.2.

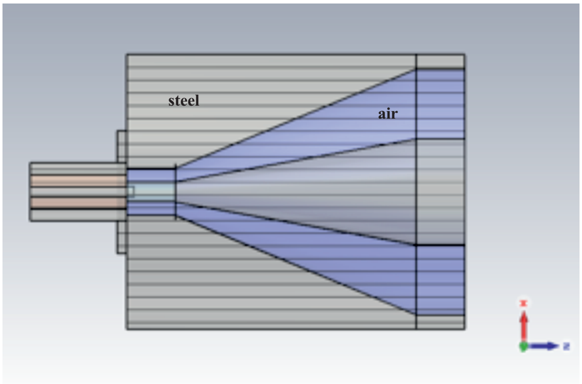

Figure 3.2: Coaxial transition part cross–section.

Since the dimensions, i.e., the radii of the outer and inner conductors, of the coaxial microwave reactor and type–N connector are already known, the only optimization parameter would be the length of the conical shape microwave transition part. Microwave theory textbooks advise that when two different impedances are expected at each end of a waveguide transition, the length should be extended about one half–wavelength along the impedance change direction. In the present design, the input and output impedances are the same. In fact, the transition is needed to only adapt two physically un–identical $50\,\Omega$ coaxial waveguides. In this case, the length of the transition is expected to have less impact on the overall reflection performance. To investigate the effect of transition length on the reflection factor, a full–wave electromagnetic field analysis was performed in the simulation environment, CST Studio Suite [22]. A time–domain method was employed for solving Maxwell's equations and obtaining a fast estimation of electromagnetic performance over the desired bandwidth ($2.43\,\mathrm{GHz}$–$2.47\,\mathrm{GHz}$).

The obtained simulation results showed that varying the transition length over a wide range of values has an insignificant effect on the impedance performance of the device, see Figure 3.3. Therefore, for the purpose of reducing the length and weight of the overall structure, we have chosen $50\,\mathrm{mm}$ as the length of the transition for future analyses, which has been achieved with acceptable reflection at the input port (less than 0.3% of the input MW power).

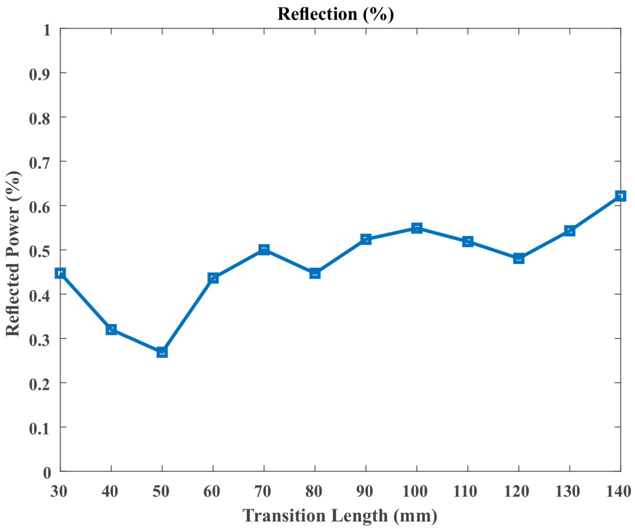

Figure 3.3: The simulated results of a coaxial microwave transition part. [2]

As the microwave energy travels along the coaxial microwave reactor, part of this energy is going to be absorbed by microwave–susceptible media, i.e., catalysts or catalyst supports, and dissipated into heat in the reaction zone, and the rest is going to be leaked to the environment. To prevent the unwanted microwave leakage, designing a microwave absorber to be placed at the end of the reactor is crucial to absorb the remaining microwave energy safely.

In this manner, a commercial microwave absorber material was selected due to its good permittivity, permeability, and attenuation properties and the easiness of machining. The empty space inside the absorber section was considered air–insulated to have matching waveguide size in the reactor and the absorber intersection. The absorbing part could be designed in a conical shape, see Figure 3.4, to have a smooth transition from $50\,\Omega$ waveguide to the absorber.

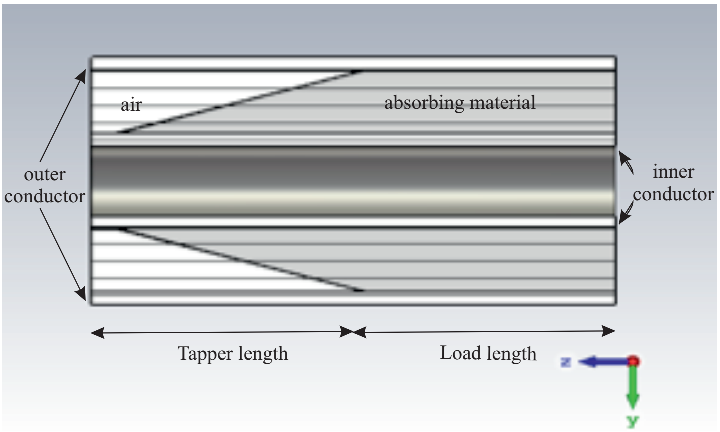

Figure 3.4: Coaxial microwave absorber cross–section.

The simulated results for the optimization of the length for each of these two sections are presented in Figure 3.5. As it can be inferred from the results, increasing the taper length, more than $50\,\mathrm{mm}$, produces smaller input reflections and due to that, there would be insignificant improvements in the performance. However, changing the length of the load part has no meaningful effect on the reflection but can decrease the level of radiation significantly. Considering the optimization results, the taper length and load length are both selected to be $50\,\mathrm{mm}$.

3.3. Performance Characteristics of the Integrated System

In the analysis so far, the coaxial microwave waveguide, its connection to the microwave generator system, i.e., the microwave transition part, and the microwave absorber section have been investigated and optimized to produce negligible reflections. In this section, all the above designed parts have been integrated to constitute a coaxial traveling microwave reactor. The proposed microwave reactor should enable a highly uniform heating

[2]Simulated length changes are made stepwise, resulting in fluctuations or discontinuities in curves as the system reacts to each discrete change.

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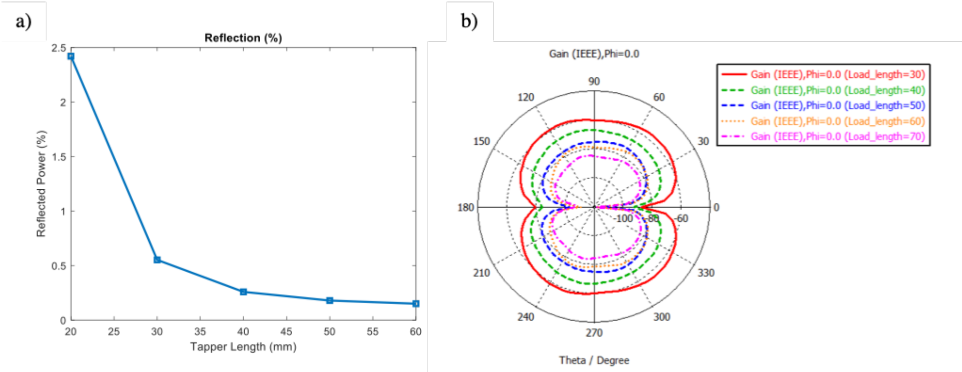

Figure 3.5: The simulated results of a coaxial microwave absorber a) Effect of changing Taper Length in the Absorbing Section on the input reflection (Load Length=$50\,\mathrm{mm}$) and b) Effect of varying Load Length of Absorbing Section on the external radiation (Taper Length=$50\,\mathrm{mm}$)

profile [23].

The cross–sectional view of the proposed microwave reactor along the axial direction is presented in Figure 3.6. Here, the stainless–steel inner and outer conductors form the coaxial waveguide structure. The reaction zone, is the annular space between the two concentric quartz tubes ($\varnothing_{in}$: $27\,\mathrm{mm}$, $\varnothing_{out}$: $46\,\mathrm{mm}$, $Length$: $300\,\mathrm{mm}$), and the accessible reactor volume is $0.327\,\mathrm{L}$.

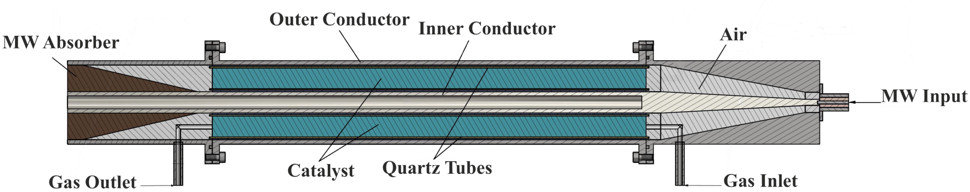

Figure 3.6: A cross–sectional schematic of the proposed traveling microwave reactor.

The quartz tubes form a sealed toroidal reactor inside the waveguide and play the role of a dielectric insulator (between the catalyst and the conductors) in the coaxial waveguide. This provides a way to have a relatively high–temperature reactor inside a cooler waveguide structure while maintaining fluid containment. Different forms of catalysts and/or catalyst supports can be placed in the space between the quartz tubes, and the process gases flow in the axial direction along the catalyst section. The conical–shape coaxial transition part, filled with air, is placed between the microwave excitation port and the reactor intersection in order to adapt the two devices. The tapered coaxial absorber is provided at the end of the waveguide to prevent any microwave leakage to the environment.

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The electromagnetic field simulation software CST Studio Suite was used for the analysis of the reactor characteristics. The simulated results of the normalized microwave field distribution inside the reactor without a catalyst bed inside are presented in Figure 3.7. The results reveal that in the optimized design, the microwave field is perfectly traveling along the reactor, and the remaining energy is dissipated in the absorber section.

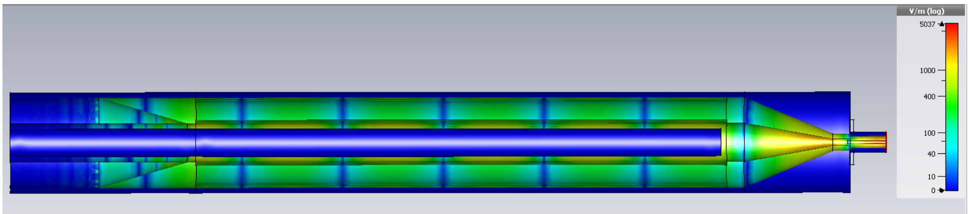

Figure 3.7: Normalized electromagnetic field distribution of the optimized reactor.

Figure 3.8 depicts the radiation pattern of the proposed design in three-dimensional cylindrical coordinates. Assuming the reactor to be extended along the z–axis, it is customary to evaluate the radiation pattern in two orthogonal planes, i.e., $phi = 0$ (XZ–plane) and $phi = 90$ (YZ–plane), while sweeping the $theta$ value. The red curves in the figures show that the EM field at $1\,\mathrm{m}$ distance from the reactor is less than -70 db, which according to the reference is safe. This can offer an acceptable approximation for the three-dimensional radiation pattern of any device. The presented results show that the leaked waves from different parts of the reactor are at least $-65\,\mathrm{dB}$ $(3 \times 10^6$ times) weaker than the source-generated power. This is well below the FDA and European directive approved range of safe radiation for microwave heating devices [24, 25].

Obtaining proper traveling waves inside the reactor requires good impedance matching between the parts of the structure. Simulation results show that

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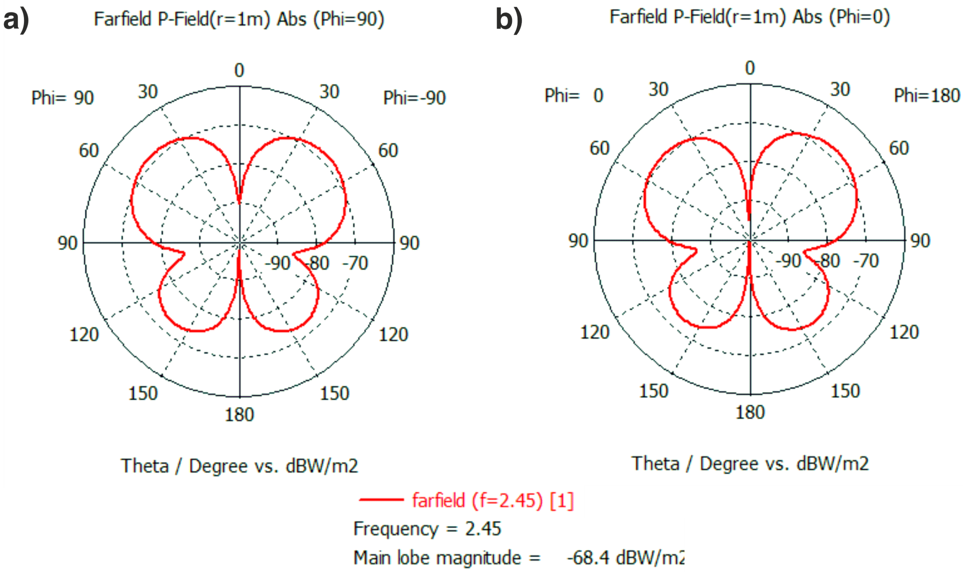

Figure 3.8: Radiation pattern in the environment: a) $phi = 90$ and b) $phi = 0$.

the reflection coefficient of the overall structure in the working frequency range is approximately $-20\,\mathrm{dB}$ ($\approx\%\,1$) at the working frequency of $2.45\,\mathrm{GHz}$. This is a proof that there is no standing wave generated along the structure.

3.4. Controlling the Catalyst Heating Profile

The simulation results discussed so far concerned an empty traveling microwave reactor system, optimized for impedance matching. This way, the microwave reflections could be minimized. In a reactor loaded with a catalyst as a microwave absorbing media, the impedance matching should obviously be retained. To this end, the catalyst volume/mass should increase gradually over the reactor length, avoiding any sudden impedance changes. Otherwise, it would behave as a mirror at the intersection in the way of the microwave and reflect a considerable 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, in the case where the catalyst is evenly and longitudinally filled along the reactor, the microwave field intensity and consequently the heating rate would attenuate exponentially.

To illustrate the effect of the impedance matching in the reactor loaded with catalyst, different loading schemes were studied with the length of the catalyst cone ranging from 50 mm to 300 mm with 5 mm intervals, see Figure 3.9.

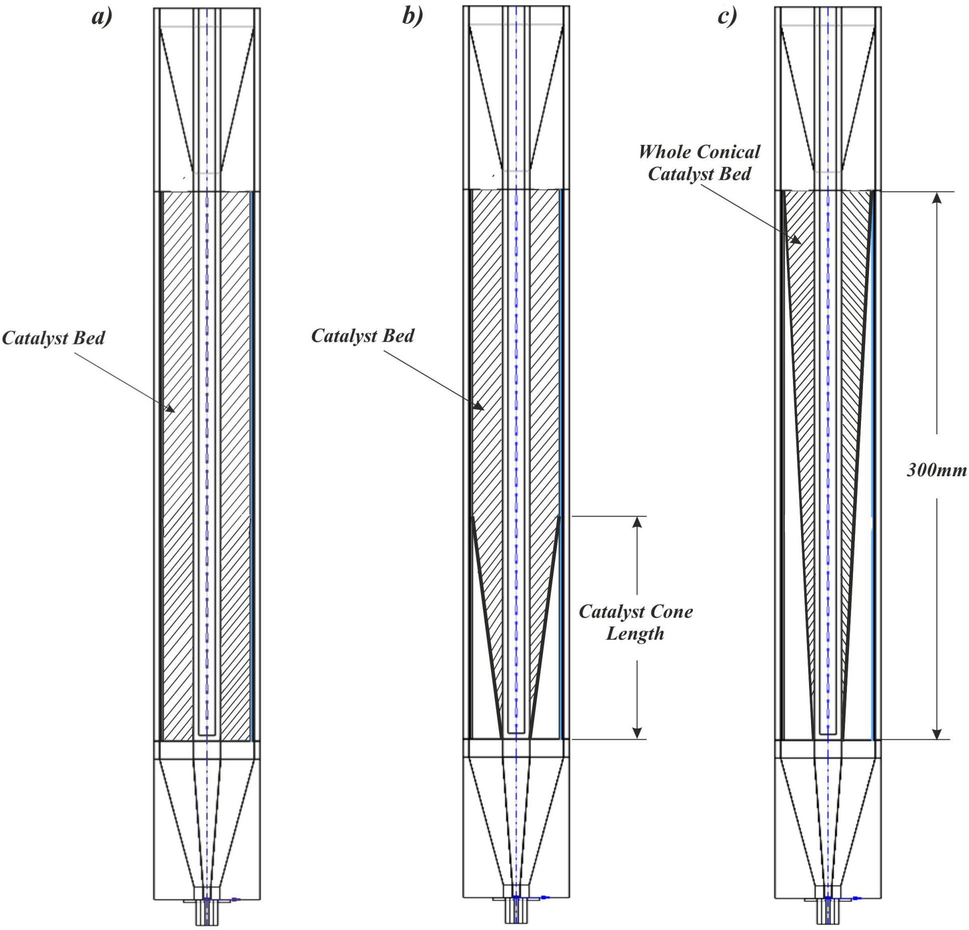

Figure 3.9: Schematic diagram of different catalyst loading patterns: a) Fully loaded; b) Gradually increasing the catalyst concentration cone length from 50 mm to 300 mm.

The heating profiles were simulated for all cone lengths, and the minimum and maximum temperatures of the catalyst bed are extracted and compared in Figure 3.10. In all cases, the reactor is assumed to be loaded with platinum on carbon, Pt/C catalyst. The temperature–dependent dielectric of that catalyst was provided by L.S. Gangurde et al. [26]. The heat–transfer and electromagnetic field equations were coupled using COMSOL Multiphysics 5.3 simulation environment [27]. The assumed MW power input is 400 W.

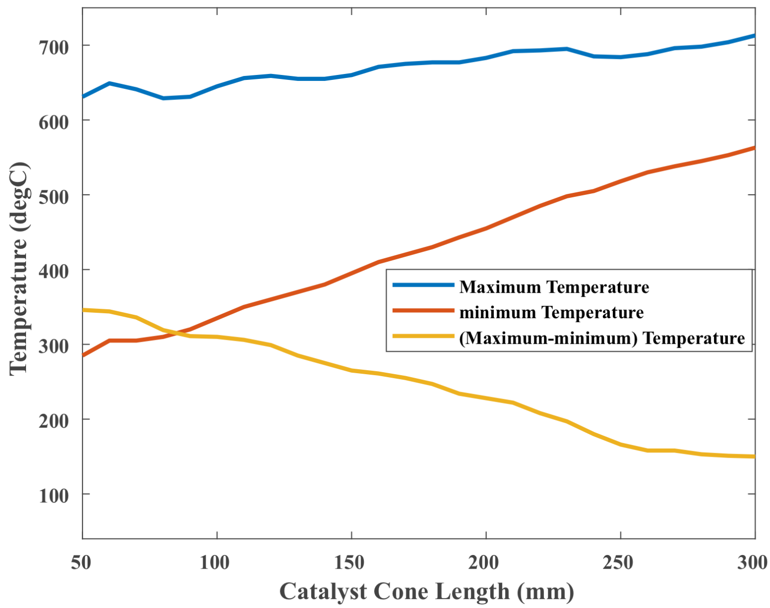

Figure 3.10: Effect of catalyst cone length on the heating profile of the catalyst bed and its homogeneity.

As can be seen in Figure 3.10, the difference between the maximum and minimum temperatures decreases with the increasing length of the conical part of the catalyst bed reaching the minimum for the case when the cone extends over the entire reactor length (300 mm, case (c) in Figure 3.9). This is mainly due to the good impedance matching of the catalytic bed with the MW system.

Figure 3.11, compares the heating profiles of the normal (uniform cylindrical) and conical (gradually increasing) catalyst loading pattern. As illustrated in Figure 3.11(a), due to significant microwave reflections, the uniformly loaded catalyst cannot interact with the electromagnetic waves efficiently and even with the same energy input the catalyst cannot be heated properly ($570°C$ at most). On the other hand, in the case presented in Figure 3.11(b), where the catalyst load is gradually increased (with the intention of keeping the impedance matching), the maximum temperature achieved is much higher (above $700°C$) and the temperature profile is smoother, without forming hot spots. Also, the uniform heating zone is much larger.

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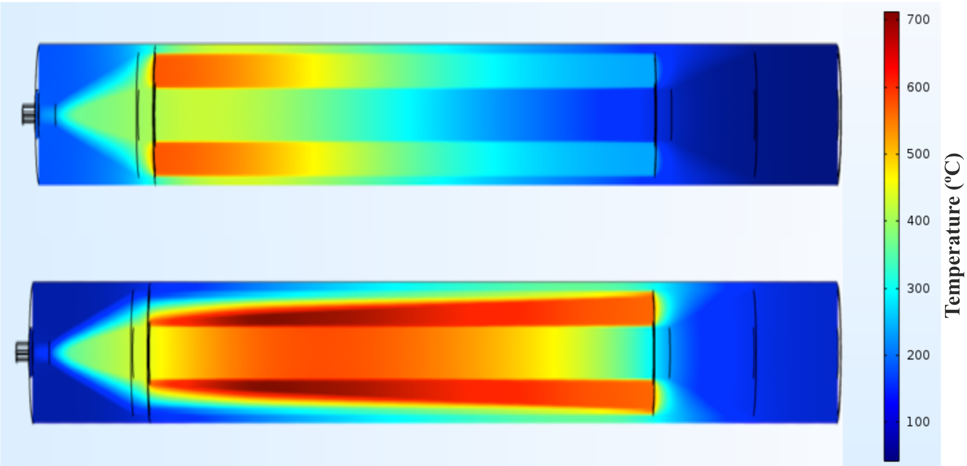

Figure 3.11: Catalyst heating profile of different loading pattern: a) Fully b) Gradually loaded by the catalyst.

To examine the heating uniformity in the radial direction, the temperature at three different axial positions (i.e. $z = 73\,\mathrm{mm}$, $150\,\mathrm{mm}$, and $300\,\mathrm{mm}$) is plotted in Figure 3.12. It can be seen from the figure that even in the radial direction, the catalyst zone is heated up quite evenly.

It is notable that the noticeable difference between Figure 3.11 and Figure 2.7 is due to the different materials of the load in the reactor, PtC and SiC, respectively.

In fact, the two latest figures show that in the current design, the temperature distribution in both axial and radial directions is fairly uniform, and no macroscopic hot spots occur along the catalyst bed. However, from the simulation results and the heating profile depicted in Figure 3.11, the temperature in the absorber section is considerably lower than the reactor section. It is due to the fact that the amount of power which reaches the

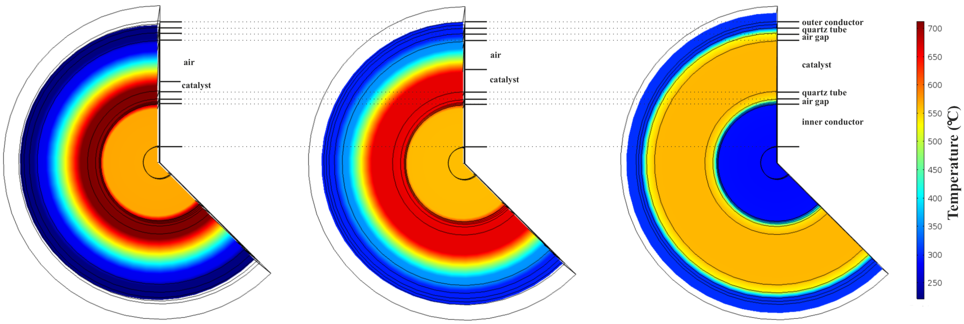

Figure 3.12: Catalyst heating profile of gradually loading pattern at different axial locations: a) $z = 73\,\mathrm{mm}$ b) $z = 150\,\mathrm{mm}$, and c) $z = 300\,\mathrm{mm}$ (at the end of the bed).

absorber is practically negligible, and this proves the efficiency of the design.

3.5. Conclusion

This chapter addresses the critical aspects of designing a traveling microwave reactor. A coaxial traveling microwave reactor is proposed as a potential solution to the process scale-up and catalyst heating profile issues. Simulated results demonstrate that the reflection coefficient of the overall structure is $-20\ dB$ ($\approx\%\ 1$) at the standard working frequency of $2.45\,\mathrm{GHz}$. This proves that there is no standing wave generated along the structure. Additionally, a catalyst loading pattern is proposed in order to keep the impedance matching and minimize the microwave reflections. Simulation results show that the proposed gradual loading pattern enables more efficient coupling between the microwave energy and the catalyst, which leads to having a more uniform temperature distribution inside the bed.

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4

Catalyst Heating in Traveling Microwave Reactor

In this chapter, our primary objective is to experimentally validate the catalytic heating performance of the designed traveling microwave reactor (TMR). Leveraging silicon carbide (SiC) samples as microwave absorption media and catalyst support in a packed-bed configuration, we meticulously explore the TMR system's capability to achieve uniform temperature distribution. Through rigorous simulation and experimentation, our findings underscore the robustness of the TMR model in accurately predicting temperature profiles and reaction dynamics within the reactor. The validation improves our understanding of microwave-assisted catalytic processes and emphasizes the importance of model validation for maintaining reactor efficiency and reliability.

4.1. Introduction

The primary focus of this chapter is to experimentally verify the effectiveness of the proposed traveling microwave reactor. We use silicon carbide (SiC) samples as both microwave absorbers and catalyst supports in a packed–bed setup to investigate the TMR's ability to achieve uniform temperature distribution.

First, we explain the details of the traveling microwave reactor setup designed and developed to evaluate the performance of the microwave coupling and heating uniformity. A brief overview of the different components of the experimental setup, including the traveling microwave assembly, solid–state microwave generator, and temperature measurement technique, is presented. Additionally, we conduct extensive simulations to model the behavior of the TMR and validate its predictions against experimental data. The results of the experimental validation of the catalyst heating tests are discussed, along with the findings from the simulation modeling and validation process.

The successful validation of the TMR model provides a solid foundation for future studies, offering researchers a reliable framework for exploring and optimizing microwave-assisted catalysis processes on a larger scale. It also underscores the importance of employing computer modeling techniques to address the limitations associated with temperature measurements in microwave reactors.

4.2. Experimental Setup

A scheme of the experimental setup is presented in Figure 4.1. Detailed information about the design challenges of the reactor and the solutions to these challenges can be found in Chapter 3. The essential parts of the setup are: the traveling microwave reactor assembly, the microwave generator, and the temperature measurement and control system. The P&ID of the setup is depicted in Figure 4.2.

4.2.1. Traveling Microwave Reactor Assembly

The assembly consists of a custom–made coaxial traveling microwave reactor with a microwave absorber at the end to avoid any leakage of microwave radiation into the surroundings. The absorber is made of carbon, an excel-

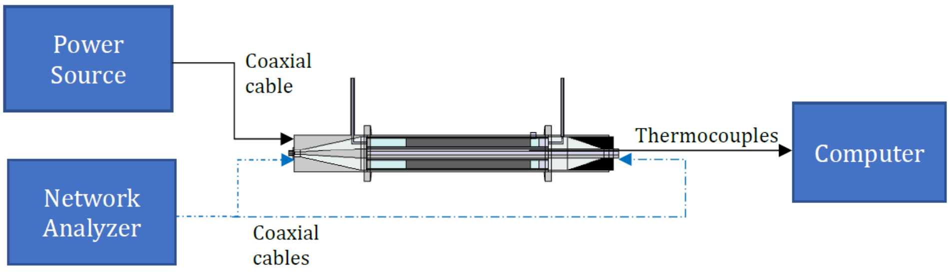

Figure 4.1: Schematic representation of the experimental setup.

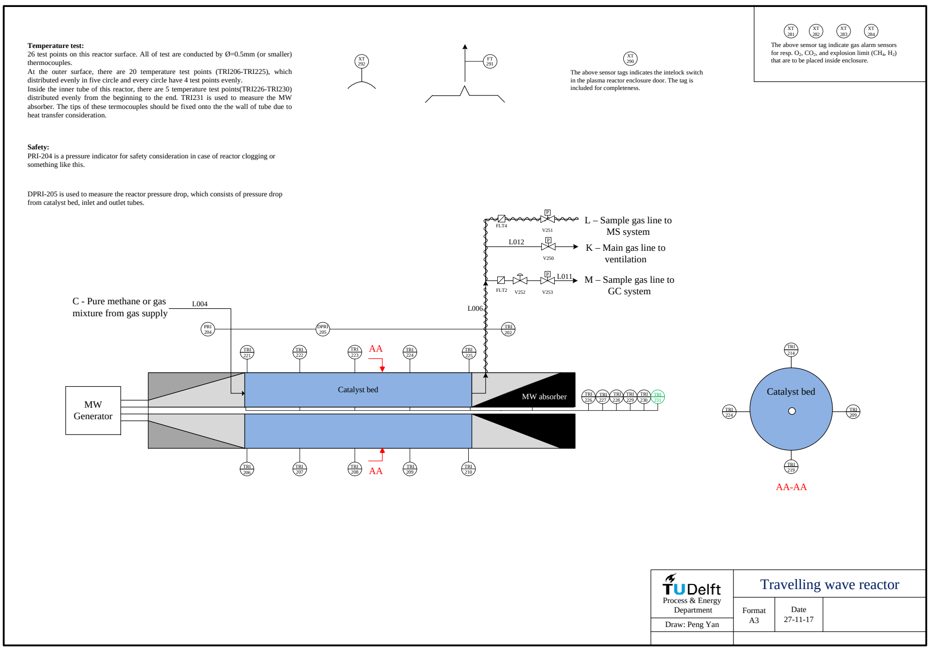

Figure 4.2: P & ID of the traveling microwave reactor setup.

lent microwave absorber, in a PTFE matrix. A schematic drawing of the reactor can be seen in Figure 4.3.

The outer and inner conductors of the reactor are made of stainless steel 310S, the inner diameter of the outer conductor and the outer diameter of the inner conductor are 54.8 mm and 15.7 mm, respectively. These diameters are of special relevance for microwave engineering since they will deter-

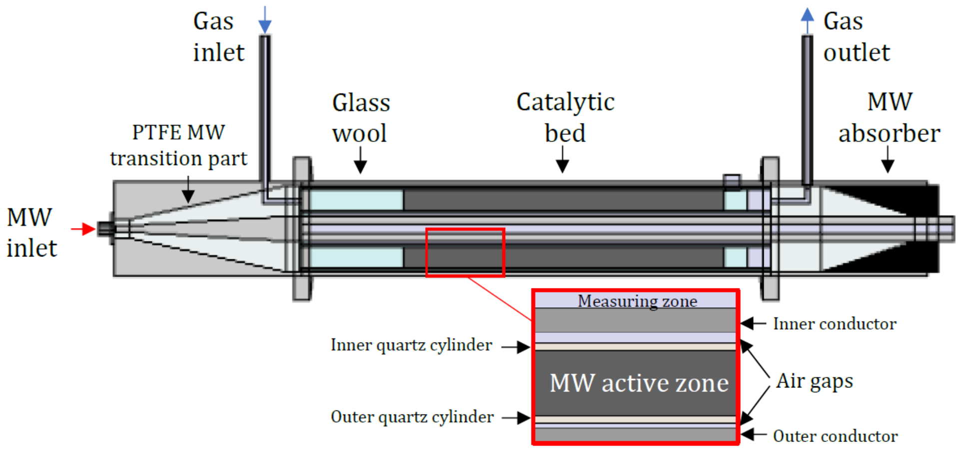

Figure 4.3: Representation of the axial section of the traveling microwave reactor.(This figure is adapted from [1])

mine the characteristic impedance of the medium and the cut–off frequency. It is on the surface of these two conductors that an oscillating current will occur, creating an electric field in the radial direction and a magnetic field in the angular direction.

Both at the inlet and outlet, two conical shapes, the so–called MW transition parts, are provided because a progressive variance of the impedance reduces the reflection of the electromagnetic wave in the system. Both cones are filled with PTFE. This material was selected due to its low dielectric losses. The catalytic bed is originally confined in between two quartz cylinders of $50\,\mathrm{mm}$ and $23\,\mathrm{mm}$ diameter, which prevent direct contact between the catalyst and both conductors. The space before and behind the catalyst bed is filled with quartz wool. The length of the catalytic zone is $300\,\mathrm{mm}$. The inner conductor has an aperture through which a thermocouple can be introduced to measure the temperature. This hole reaches the end of the catalytic zone; the diameter of the orifice is $6\,\mathrm{mm}$.

4.2.2. Microwave Generator

The microwave generator used in the study is the solid–state MiniFlow 200SS generator made by SAIREM, demonstrated in Figure 4.4. The device is specifically designed for laboratory use [2]. It is connected to the traveling microwave reactor by a coaxial cable to transfer the microwave energy.

The generator has adjustable power from 0 to 200 W with 1 W power increment, and controlled variable frequency from 2430 MHz to 2470 MHz with 0.1 MHz increment [2].

The advantages of using a solid–state microwave generator are as follows [2]:

1. Compact size and light weight.
2. Stable operation starting from microwave power levels as low as 0.5 W and power adjustable in 1 W steps.
3. No magnetron, resulting in longer lifetime and no high voltage.
4. Very good frequency spectrum even at low power; the frequency spectrum of magnetron–based generators has poor stability below 100 W to 150 W.
5. Built–in internal protection against mismatching and reflected power interlock.
6. Built–in isolator with automatic power reduction or switch off.
7. True RMS detector with linear measurement of reflected and forward power.
8. Very low ripple (0.2% RMS).
9. Possibility to adjust microwave frequency ±20 MHz, i.e., from 2430 MHz to 2470 MHz.

4.2.3. Network Analyzer
A network analyzer is an instrument that measures the network parameters of electrical networks (scattering parameters, or s–parameters), which show the electromagnetic wave reflection and transmission coefficients at different ports of an electrical network. We used the Agilent Technologies E5071C, Santa Clara, CA, USA network analyzer, which can cover the frequency range of 300 kHz to 14 GHz, see Figure 4.5

4.2.4. Temperature Measurements
In microwave heating, temperature measurements present significant challenges due to the lack of appropriate tools [3]. Two options are typically considered:

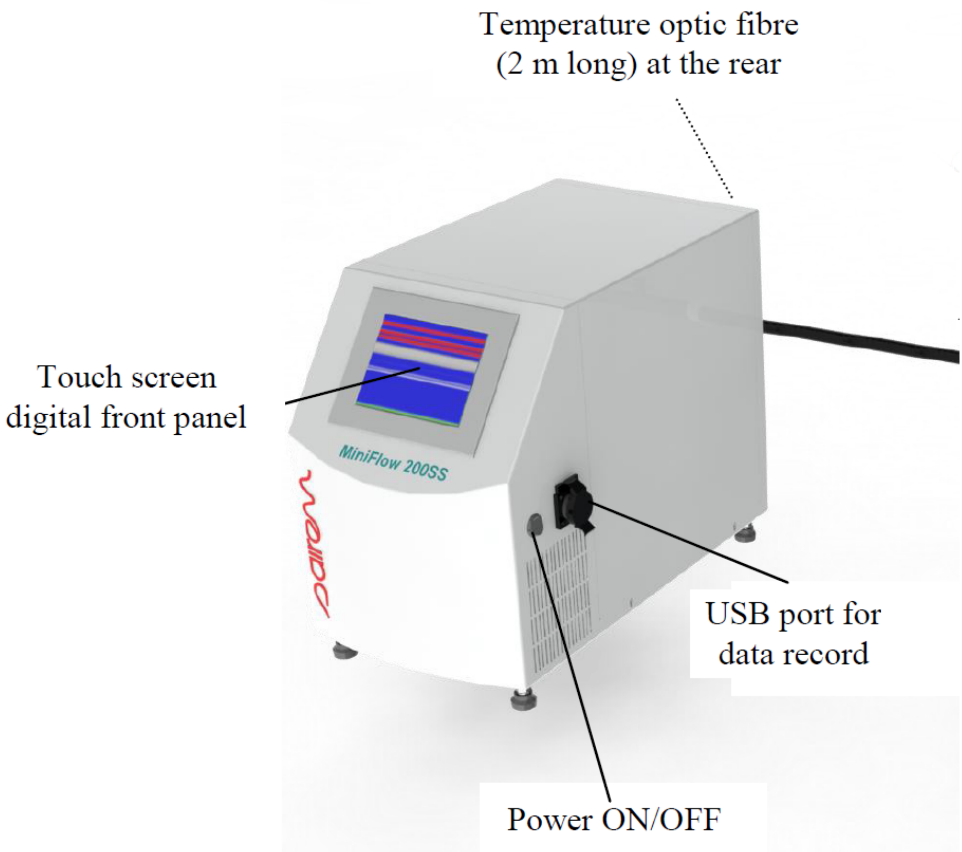

Figure 4.4: Solid–state MiniFlow microwave generator.

1. Fiber–optic sensors.
2. Pyrometers or infrared thermometers.

The first option requires the probes to be inserted in the reactor, without compromising the fluid containment. Moreover, commercial fiber optic sensors are not suitable for high-temperature processes as they can operate in temperatures not exceeding 250 °C. Moreover, typical infrared sensors are not able to look through the quartz wall of the reactor.

An alternative solution could be an optical spectrum pyrometer. However, the use of an infrared camera to inspect the temperature of the catalytic bed is limited. Since the infrared waves cannot penetrate through metals, only the surface temperature of the outer conductor could be measured.

Figure 4.5: Network analyzer: Agilent Technologies E5071C.

Another way to measure the temperature is to use thermocouples. Unfortunately, thermocouples cannot be introduced directly inside the microwave cavities because they can disturb the electric field distribution and may provoke sparking. To overcome this problem, the hole of $6\,\mathrm{mm}$ diameter inside the inner conductor could be considered for introducing the thermocouples. There, the electric field is $0\,\mathrm{V/m}$, so such a technique is safe.

In the current study, due to the discussed limitations of optical fiber sensors and infrared cameras, the fixed–bed temperatures were measured using four thermocouples (type K), with an operational temperature of 1300 °C. These thermocouples are introduced inside the hollow inner conductor of the TMR (where no electromagnetic wave is presented) and connected to a computer, where the output is displayed. Positions of the thermocouples are shown in Figure 4.6.

4.3. Catalyst Heating Tests

In the following section, the results of the catalyst heating tests in the traveling microwave reactor are presented. The ultimate objective is to use the experimental measurements to validate the simulated model and to verify whether the reactor can reach high temperatures ($700\,°\mathrm{C}$ or higher),

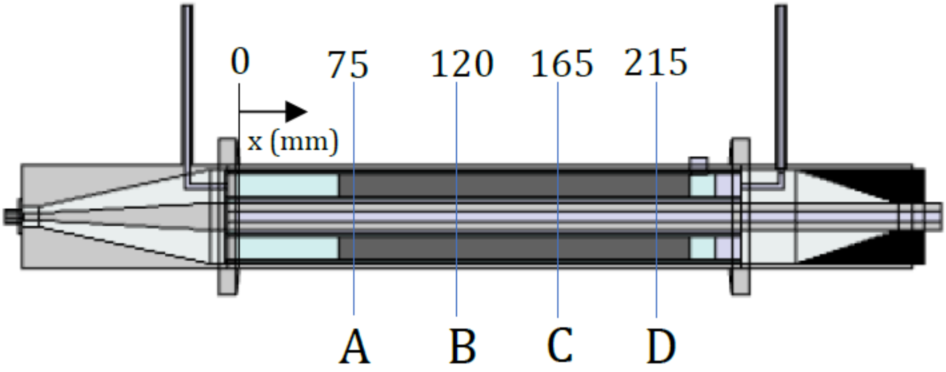

Figure 4.6: Relative position of the thermocouples inside the inner conductor.

using the microwave radiation as a sole heating source. All tests were carried out under non–reactive conditions.

4.3.1. Activated Carbon Experiments

The microwave susceptive packed–bed has a total mass of 302.7 g. The packed–bed is divided into five sections, each having a 10, 20, 30, 40, and 50 wt.% of activated carbon, the rest consisting of quartz sand. The total mass of presumably microwave–active material would be 90 g. The mixing between the sand quartz and the carbon particles is done mechanically, and different batches with different percentages are introduced in the reactor to have a progressive loading. This means that the amount of activated carbon closer to the microwave inlet is small and increasing till the end of the annular bed, where most of the material would be activated carbon particles (see Figure 4.7). According to the results of Chapter 3, progressive loading generates a more homogeneous temperature distribution. The results of the heating test with 60 W microwave power inlet are shown in Figure 4.8.

Activated carbon is known for being a good microwave absorber. Its high electrical conductivity and its large specific surface area make this material highly polarizable, and the energy is dissipated more easily due to the numerous surface defects expected in the particles. This is seen in the experimental results shown in Figure 4.8. Point D, which is situated at the end of the bed, is less heated since most of the energy is consumed in the earlier part of the packing. We also observe that the temperature of point A reaches circa 150 °C, nearly 25 °C less than the temperature of points B

Figure 4.7: Photograph of the packed-bed, which consists of a mixture of progressively loaded activated carbon and quartz sand.

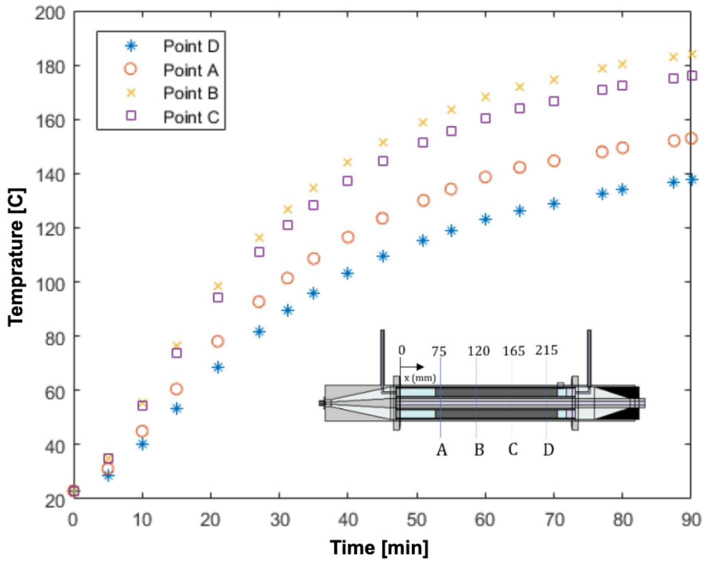

Figure 4.8: Experimentally measured temperature profile of a progressively loaded activated carbon in traveling microwave reactor irradiated with 60 W microwave power.

and C. This is due to the fact that the load at the front consists of only 10 wt.% activated carbon, which is not enough to raise the temperature in that section of the reactor.

Figure 4.9 presents the dependency of the maximum temperature difference in the bed on the microwave power used. The difference increases with the inlet power. Such behaviour is expected since the absorbed power depends on the square of the electric field intensity, while other transport mechanisms of energy present in the system depend linearly on the temperature. Increasing the power inlet would decrease the dissipation of energy characteristic time, while it would not affect the conduction or convection mechanisms. This may be seen as a drawback, but it presents a common aspect of microwave heating, even at small scales. Despite this, very high and stable conversions have been obtained for reactions like dry reforming [4, 5].

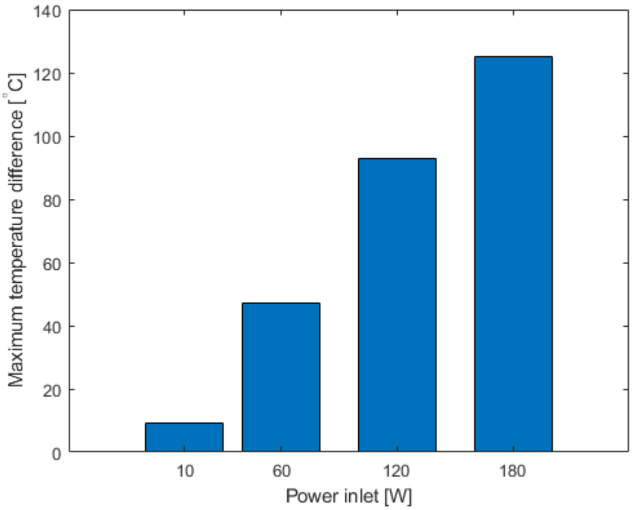

Figure 4.9: Maximum axial temperature difference depends on MW power input. Results for a packed–bed consisting of a mixture of activated carbon and quartz sand.

4.3.2. Silicon Carbide Foams Experiments

Following the activated carbon study, it is decided to switch to another material that can be studied in the air atmosphere. Silicon carbide is known for being an excellent microwave absorber. The material passivates at room temperature, and a thin layer of SiO_2 formed should protect the structure from further oxidation [6].

Next to the above, silicon carbide has outstanding thermal, mechanical, and electrical properties. It is already being used in electric furnaces as resistance heating elements [7]. It presents an excellent option when high–loss materials in high–temperature conditions are required. With regard to the scope of the present project, SiC overcomes some of the major drawbacks of alumina, silica, and carbon as the most commonly used supports for heterogeneous catalysis. Its higher thermal conductivity compared to alumina and silica makes it easier to heat. It is also more stable than carbon-based supports under oxidative conditions thanks to its superficial passivation [8]. Therefore, it is a material that can support the active phase while at the same time dissipating the electric energy of the microwaves to heat the system, as shown for the microwave—-assisted methane dry reforming and toluene cracking [9, 10]. In the present study, two different forms of silicon carbide packing, a crushed foam α–SiC and β–SiC extrudates have been investigated.

Crushed Foam SiC

The size of the crushed foam particles is between $3\,\mathrm{mm}$ to $5\,\mathrm{mm}$ of lump size. The length of the packed–bed is set to $210\,\mathrm{mm}$. This is equal to $109.25\,\mathrm{g}$ of silicon carbide foam; see Figure 4.10. The packed–bed porosity is a relevant parameter in microwave engineering. It will not only determine the effective properties of the bed, e.g., thermal conductivity and specific heat, but will also have an effect on the characteristic impedance of the medium, its electric conductivity, and thus the electric field intensity in the reactor.

The total porosity of the α–SiC crushed packing (being a sum of the voidage of the foam pores and the interparticle voidage) is calculated as follows:

Figure 4.10: Crushed $\alpha - SiC$ foam packed bed.

$$\theta = \frac{V_{air}}{V_T} = 1 - \frac{V_{solid}}{V_T} = 1 - \frac{m_{solid}}{V_T.\rho_{solid}} \tag{4.1}$$

where θ is the total bed porosity, V_i is the volume of a specific phase, V_T is the total volume, m_i is the mass, and ρ stands for the density. Considering the density of the bulk silicon carbide equal to $3.21\,\mathrm{g\,cm^{-3}}$ [11], the calculated total porosity of the bed is $\theta = 0.9$.

This bed is exposed to a MW inlet power of $60\,\mathrm{W}$. From the heating results shown in Figure 4.11, it is clear that the heating of the crushed $\alpha - SiC$ foam, contrary to expectations, is disappointing when compared to a mixture of activated carbon and quartz sands in Section 4.3.1.

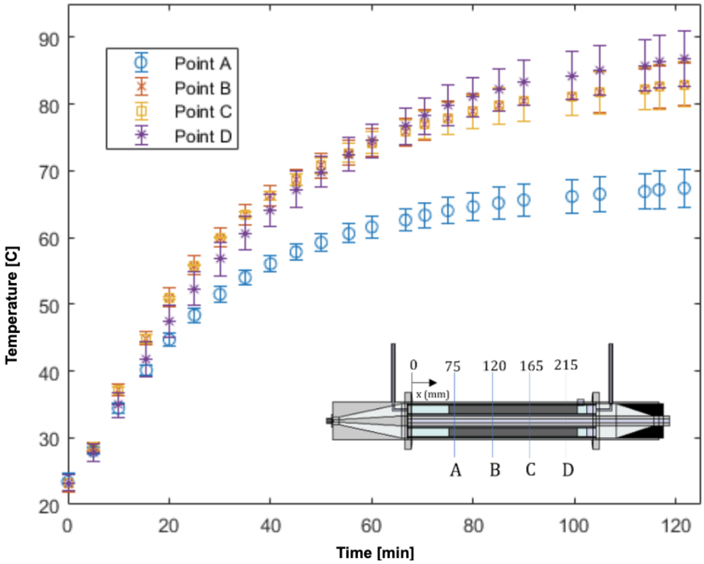

Figure 4.11: Experimentally measured temperature profile of the crushed–foam $\alpha - SiC$ in traveling microwave reactor irritated with $60\,\mathrm{W}$ microwave power.

It can also be seen that the hottest thermocouple is the closest to the absorber, which shows that the crushed SiC foam bed does not absorb MW energy efficiently. For that reason, the majority of the MW energy is received by the MW absorber, where it is dissipated as heat.

β–SiC Extrudates

Cylindrical extrudates of $5\,\mathrm{mm}$ of lumped size made of SiC, were used in the next series of heating experiments. The bed porosity of this shape of extrudates is between 0.3 to 0.4 [12]. Compared to the previous experiments, a denser bed is obtained because we now deal with solid extrudates instead of foam; see Figure 4.12. Therefore, a better dissipation of heat can be seen; see Figure 4.13.

Figure 4.12: Microwave active bed of $\beta - SiC$ extrudates of 5 mm lumped size

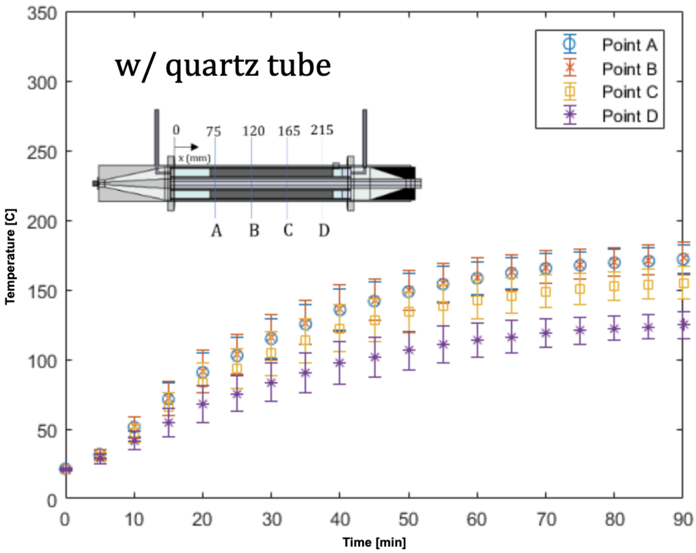

Figure 4.13: Experimentally measured temperature profile of $SiC{-}\beta$ in extrudates form in traveling microwave reactor irritated with 60 W microwave power.

As observed, the hottest thermocouple is no longer the closest to the absorber. This indicates that most of the energy is absorbed before reaching the end of the bed. In comparison with the previous results, this type of packing has a much better capacity to dissipate heat. The heating of the bed can be further improved by removing the inner quartz tube. In such cases, a further increase of more than 100 °C is obtained; see Figure 4.14.

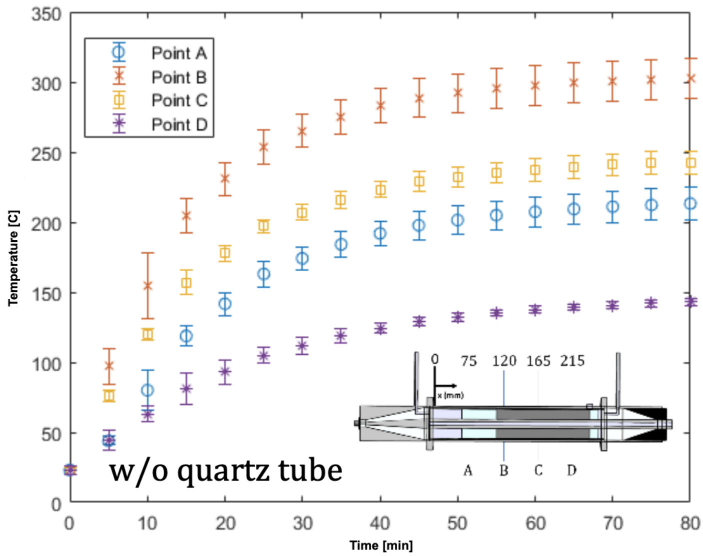

Figure 4.14: Experimentally measured temperature profile of $\beta{-}SiC$ in extrudes form in traveling microwave reactor irritated with 60 W microwave power–Without the inner quartz tube.

Finally, the last experiment has been repeated with an increasing microwave power input. At a power of 200 W, the maximum temperature reached at point B was around 600 °C, see Figure 4.15.

As a result of the high temperatures observed, oxidation occurred in the inner conductor. The temperature reached can be identified by the colour of the metal rod; see Figure 4.16. Considering the relation proposed by British Stainless–Steel Association [13], the observed colours would match the registered temperature and distribution.

Based on the experiments described here, it is evident that $SiC{-}\alpha$ and $SiC{-}\beta$ have significantly different microwave heating profiles. The reason for this divergence is that they have distinct crystal structures and properties, which influence their response to microwave energy. There are differences in microwave dissipation properties among these polytypes, as well as other characteristics that are particularly relevant to microwave heating applica-

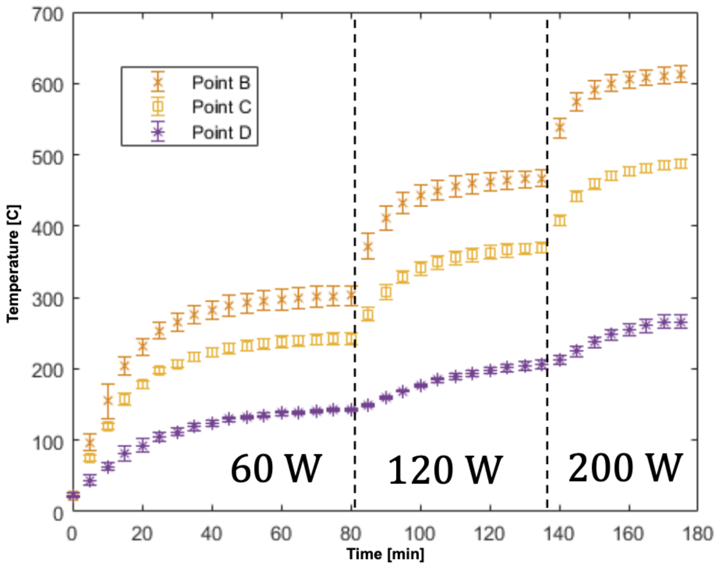

Figure 4.15: Experimentally measured temperature profile of SiC–β in extrudes form in traveling microwave reactor irritated with 200 W microwave power–without the inner quartz tube.

Figure 4.16: Tinted inner conductor due to oxidation after reaching temperatures around 600 °C.

tions [14].

In conclusion, the heating experiments have shown that the temperature of the packed–bed can be efficiently raised to high temperatures. The goal is then shifted to achieving more uniform temperatures in the catalyst bed. As a result, a model of the reactor is created to conduct a more in–depth investigation.

4

4.4. Reactor Model and Its Validation

The electromagnetic waves and heat transfer physics are fully coupled in the reactor's 3D model using the COMSOL MULTIPHYSICS simulation environment. The numerical problem is solved using the finite element method (FEM), and the governing equations used for each physics module are described as follows.

Electromagnetic waves

To determine the electric field distribution in the coaxial waveguide, the wave equation derived from Maxwell's equations is solved [15, 16] in the domains between the waveguide, i.e., the air domain, catalyst and quartz tubes:

$$\nabla^2 \mathbf{E} + \omega^2 \epsilon \mu \mathbf{E} = 0 \tag{4.2}$$

where **E** is the electric field vector (V/m), μ and ϵ stand 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 has been determined, the power dissipated in the material can be calculated using the following equation [17]:

$$Q_{MW} = \pi f \epsilon_0 \epsilon_r'' \mathbf{E}.\mathbf{E}^* \tag{4.3}$$

where Q_{MW} represents the dissipated power density in the material (W/m^3), f is the incident microwave frequency (Hz), ϵ_0 is the permittivity of free space, ϵ_r'' is the dielectric loss of the material, and $\mathbf{E}^*$ is the complex conjugate of **E**.

Heat transfer in solids

This module is applied to all the solid domains (catalyst, quartz tubes, inner and outer conductors) of the reactor. In this physic interface, the following transient equation is solved [18]:

$$\rho C_p \frac{\partial T}{\partial t} + \nabla.(-k\nabla T) = Q_{MW} \tag{4.4}$$

where ρ is the material density ($\mathrm{kg/m^3}$), C_p is the specific heat capacity ($\mathrm{J/(kgK)}$), T is the temperature (K), k stands for the thermal conductivity ($\mathrm{W/(mK)}$). It is notable that in the equation above, no fluid flow has been simulated, hence the convective term will be zero. As the reactor is exposed to the air, external convection cooling was taken into account by adding a heat flux boundary for the outer walls of the waveguide. The heat transfer coefficient between the wall and the air was set to $10\,\mathrm{W/(m^2K)}$, according to experience [1]. The initial temperature for the overall system was set to $20\,°\mathrm{C}$.

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 Equation 4.4, 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.

4

Next, the electromagnetic model is validated by comparing the mathematical model's results to the TMR's scattering parameters. The results of the heating experiments with catalytic active beds made of crushed foam of $\alpha - SiC$ or regular extrudates of $\beta - SiC$, are then used to validate the entire model.

4.4.1. Electromagnetic Model Validation

The reflected microwave power, or the first scattering parameter of the TMR structure, is measured with the Agilent network analyzer, described in 4.2.3. The validation procedure was carried out in the lab using various materials that could be measured and compared to the simulation results. The following available materials were tested:

1. The empty TMR structure;
2. TMR uniformly filled with crushed $\alpha - SiC$ foam;
3. TMR uniformly filled with crushed $\beta - SiC$ foam;

The experimental and simulated scattering parameter values are in good agreement, as shown in Table 4.1. This match demonstrates that the elec-

Table 4.1: Comparison of the results of the mathematical model with the scattering parameters of the TMR.

Model	Experimental S_{11}		Simulated S_{11}	
	dB	Power Reflection	dB	Power Reflection
1	-18.46	1.43 %	-19.03	1.25 %
2	-5.47	28.38 %	-5.97	25.29 %
3	-4.38	36.48 %	-4.96	31.92 %

tromagnetic model's behaviour is consistent with the physical setup, indicating that the model is electromagnetically valid.

In addition to this, the simulation results of the complete model, including Maxwell's equations and heat generations, are compared to the practical values obtained by exciting the two different active materials (which have been tested in the previous section) with a solid-state microwave generator. It is notable that these experiments were performed by Alberto Gonzalez, a master student of the Intensified Reaction & Separation Systems (IRSS) group at TU Delft, and the results are presented in the Catalysis journal [1]. The presented research demonstrates the feasibility of creating a dependable model for predicting the heating characteristics of a microwave-absorbing solid material within the Traveling Microwave Reactor over time.

4.5. Conclusion

We empirically demonstrated that the configuration of the Traveling Microwave Reactor can efficiently heat a solid microwave-absorbing catalyst. Furthermore, the heating characteristics of a microwave-absorbing solid material inside this microwave reactor can be accurately modelled, and this model can serve as a valuable tool for guiding the development and design of such reactors. Starting from this model, adjustments in reactor dimensions and materials can be fine-tuned to achieve not only elevated temperatures within the reactor bed but also a more uniform temperature distribution. The findings of this investigation also highlight the potential of employing computer modeling techniques to address the limitations associated with temperature measurements in Traveling Microwave Reactors.

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4

5

Reverse Traveling Microwave Reactor – Modelling and Design Considerations

Everything should be made as simple as possible,
but not simpler.

Albert Einstein

Microwave heating presents a potentially green alternative for energy supply to chemical and catalytic reactors as it can be based on electricity from renewable sources. The Reverse Traveling Microwave Reactor (RTMR) is a new 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 chapter, 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

Parts of this chapter have been published in *Chemical Engineering Science* (2021) [1] *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.*

5.1. Introduction

In Chapter 3 [2], 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 have 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, Dp, is defined as the distance from the material's surface at which microwave power drops to $1/e(=37\%)$ from the strength at the surface. This value in a microwave-susceptible material is a good indicator of the material to convert microwave energy to heat [3] and gives an idea of the heat distribution within the material. When the magnetic susceptibility is high (materials with higher loss factor ε_r''), the material could absorb the microwave energy faster while its penetration depth is low [4]. 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 the microwave, absorbs more energy and accordingly dissipates more heat in that region.

In this chapter, we further study the uniformity of the temperature distribution inside a microwave–susceptible material load in a packed–bed configuration. This catalyst 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 new 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 to avoid temperature differences along the load.

5.2. Reverse Traveling Microwave Reactor Design

The operational principles of a simple "one–way" traveling–wave reactor have been discussed in Chapter2 and [2]. For the fully loaded reactor with a catalyst sample, which is microwave absorbent, large temperature gradients along the axial direction of the TMR are 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 [5–11]. Coaxial waveguide could potentially provide several advantages in terms of having no cut–off frequency for TEM propagation mode [12, 13]. 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 $2.45\,\mathrm{GHz}$, the upper limit of the diameter is approximately $61\,\mathrm{mm}$ [11]. 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. 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 ($50\,\Omega$).

To this end, the first consideration is to adjust the parameters to achieve 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 diameters of the outer and inner conductors are determined as $51\,\mathrm{mm}$ and $22\,\mathrm{mm}$, respectively.

Additionally, 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 [14]. According to the previous discussions, the length of the transition part does not have a significant effect on the microwave power reflections, so $50\,\mathrm{mm}$ is chosen for the length of the transition part, which contains air $(\varepsilon_r = 1)$.

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 Figure 5.1. The dimensions of the coaxial waveguide are determined as described in the previous section, and the length of the catalyst zone is $300\,\mathrm{mm}$. Microwave radiation can be introduced from both ends of the reactor, while one port is ON, the other port needs to be switched OFF.

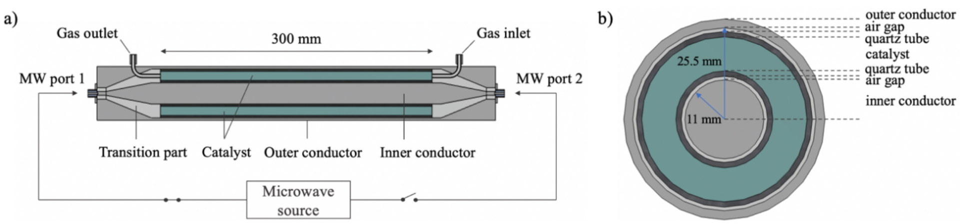

Figure 5.1: Cross-sectional schematic of reverse traveling–wave microwave reactor, a) axial, b) radial (at the middle of the bed)

5.2.1. Simulated 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 [15].

During the model development, several assumptions have been made:

- The materials are isotropic media.
- The permeability of all the materials used is assigned as free space ($\mu_r = 1$), since there is no magnetic material.
- 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 [16].
- The temperature-dependent dielectric properties of catalyst material (PtC) were obtained from our previous study [17], and a constant value is used for the temperatures above $850°C$.
- No fluid dynamics.
- A continuity condition was used at the interface of two different domains 1 and 2: $-\mathbf{n} \cdot (-k_1 \nabla T_1) - \mathbf{n} \cdot (-k_2 \nabla T_2) = 0$ [16].

The simulated result of the normalized electric field distribution inside the reactor without a catalyst bed is presented in Figure 5.2. The simulation results reveal that the reflected power of the overall structure is less than 1% at the working frequency of $2.45\,\mathrm{GHz}$, which proves that no standing–wave is generated along the structure and the microwave field is ideally traveling along the reactor.

5.3. Heating Profile Control

All the optimizations, have been done so far, concerned with the empty reactor. Once we load the reactor with a catalyst as a microwave–absorbing material, microwave energy would be absorbed differently depending on the material dielectric properties. This may occur due to many reasons, e.g., not having the impedance matching along the reactor, which

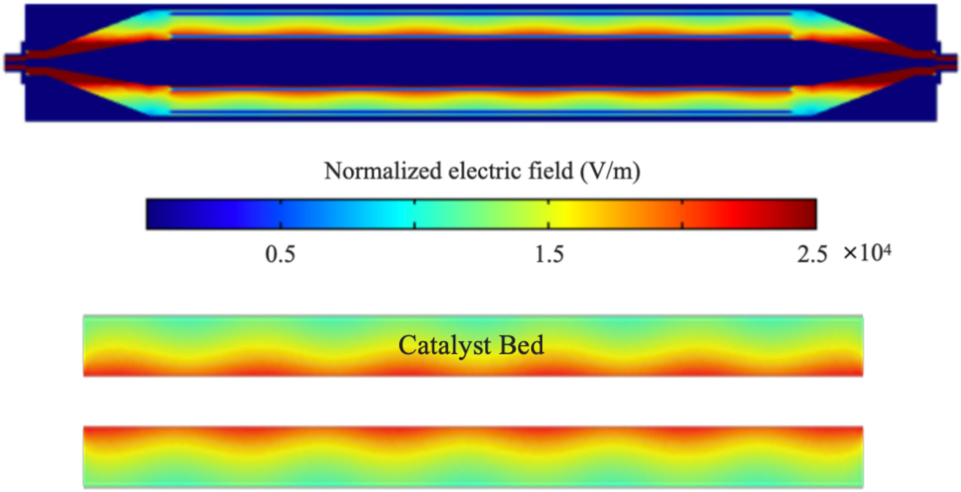

Figure 5.2: Normalized electric field distribution inside the reactor (microwave energy introduced from the left port): a) the whole configuration, b) catalyst section.

results in hot zone formation and un–uniformed correlated heating profile. However, in the reverse traveling microwave reactor, loaded with a catalyst, we could improve the heating homogeneity by different factors. The most effective parameters which could significantly influence the heating distribution, are investigated below.

5.3.1. Microwave power switching time

One of the items, which enables us to control the heating pattern, is the microwave power switching time. By being able to introduce 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 could 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 time in the loaded reactor, the maximum and minimum temperatures within the packed bed with different switching times were extracted and compared. In this study, platinum on carbon (Pt/C) catalyst is used due to its excellent microwave absorbing

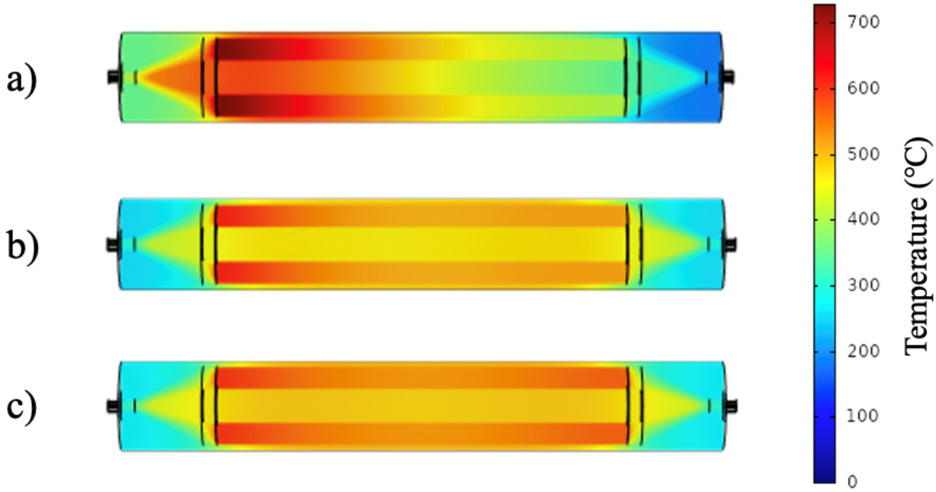

Figure 5.3: The catalyst heating profile for different switching times: (a) No switching (b) 5 minutes (c) 30 seconds.

ability. The temperature–dependent dielectric properties of the catalyst are provided by Gangurde et al. [17]. 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 Figure 5.3(a), if the microwave is not reversed during the process (i.e., previous design [2] TMR), the maximum–minimum temperature difference along the bed is 324 °C, after 60 minutes heating with 400 W MW power. On the other hand, as illustrated in Figure 5.3(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 time is defined as the time elapsed between adjacent switches. In this case, the heating of the catalyst bed is symmetric, with the minimum temperature in the middle. The temperature difference along the bed is 156 °C and 75 °C for the switching times of 5 minutes and 30 seconds, respectively. The maximum and minimum temperatures of the packed bed with the corresponding switching times are presented in Figure 5.4. The results show that a lower switching time 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 time of less than 30 seconds, 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 times approaching zero, which generates less heat before each switch.

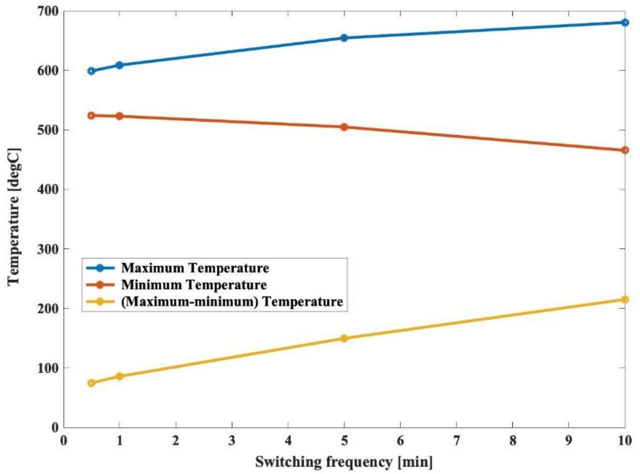

Figure 5.4: Effect of microwave switching times on the maximum and minimum temperature of the packed bed.

Changing the maximum and minimum temperature of the catalyst over time for the switching time of 30 seconds is plotted in Figure 5.5. The temperature keeps increasing over time, but the temperature difference along the packed-bed is stabled around 75 °C after 45 minutes 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 times, such as 30 seconds, 1 minute, and 5 minutes. The material properties determine the heating pattern, but the value of the temperature difference varies depending on the switching time.

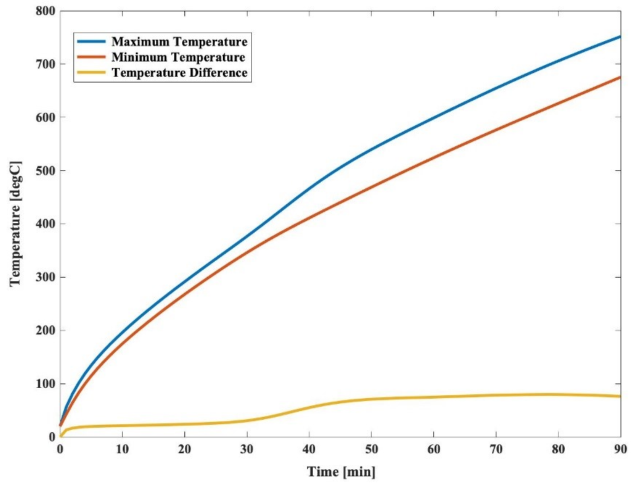

Figure 5.5: The maximum and minimum temperature for half-minute switching time over time.

5.3.2. Effects of the catalyst geometry (impedance matching)

According to the simulation results, when the microwave power is switched between the two ports, the centre 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 the 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 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 spot formation. Moreover, for the case where catalyst is 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 Figure 5.6. 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 side).

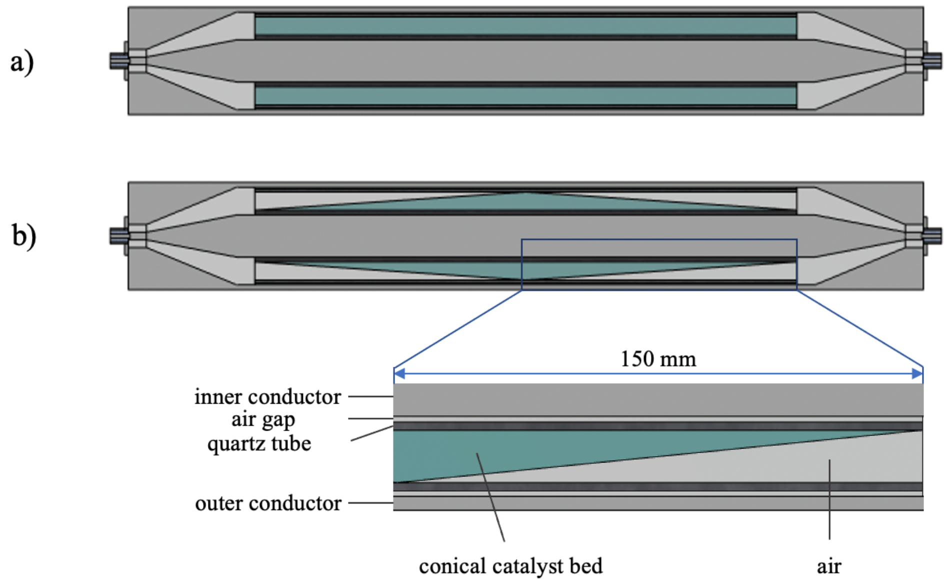

Figure 5.6: Schematic diagram of different catalyst loading patterns: (a) Fully loading, (b) Conical–shaped loading (Gradually increasing the catalyst mass from both sides of the reactor.)

The temperature distribution of normally and gradually loaded RTMR after 60 minutes of heating can be found in Figure 5.7. In both cases, the microwave switching time between the ports is considered to 30 seconds 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

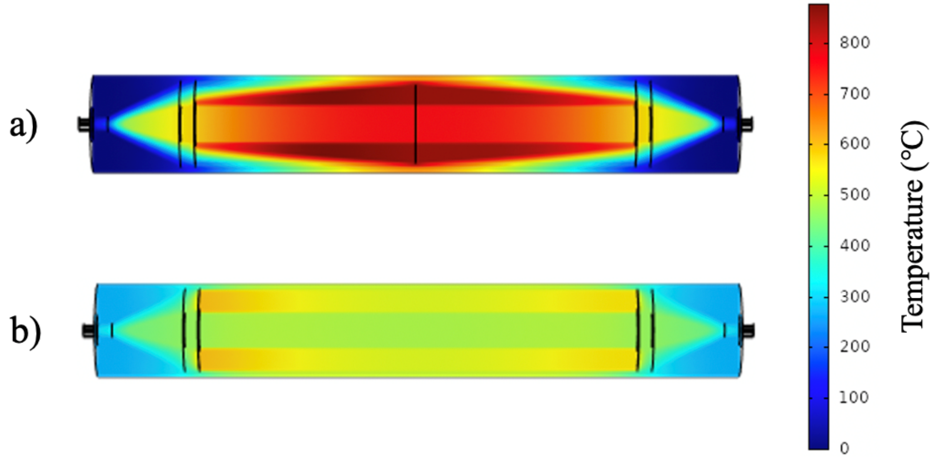

Figure 5.7: Heating profile of the reactor for different loading patterns: (a) conical shaped bed, 30 seconds and (b) fully loaded bed with 30 seconds switching time.

loading patterns along the heating time is extracted and compared in Figure 5.8. According to the results, the temperature difference of both types of bed is stabilized after 50 minutes of heating. 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 are 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 is 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.

5.4. 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

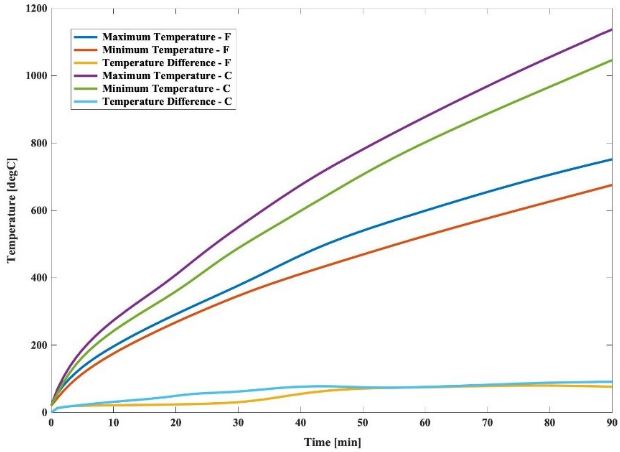

Figure 5.8: Maximum and minimum temperature for 30-second switching time over time: F = fully loaded bed, C = conical shaped bed.

effectivity of the microwave switching time 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.

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6

Conclusion and Outlook

Go to the edge of the cliff and jump off,
build your wings on the way down.

R. Bradbury

6.1. Overall Conclusion

This thesis explores the potential applications of microwave radiation for heterogeneous catalytic processes and addresses the challenges encountered in scaling up these processes.

In Chapter 2, the physics of the standing wave and the travelling wave concepts were investigated. Basic types of microwave reactor systems were introduced along with the travelling microwave reactor principle. The most essential finding is that microwaves must flow in order to establish a feasible and scalable microwave–assisted heterogeneous catalytic process in a flow system. Various aspects of the microwave-catalyst interaction, as well as the design of microwave-assisted catalytic reactor systems, were investigated. When it comes to the design and deployment of microwave flow reactors at appropriate industrial scale-up, the study clearly revealed the advantage of travelling-wave systems over mono-mode and multi-mode cavity-based systems.

A traveling microwave reactor with a coaxial construction was suggested in Chapter 3 as a possible remedy for problems with catalyst heating profile and process scale-up. At the standard operating frequency of 2.45 GHz, the simulation results demonstrated that the suggested structure had a reflection coefficient of -20 dB (≈% 1), indicating that no standing wave was generated along the structure. In order to maintain impedance matching and reduce microwave reflections, a catalyst loading pattern was also suggested. This allowed for a more effective coupling of the microwave energy with the catalyst and resulted in a more even temperature distribution inside the bed.

Chapter 4 demonstrated that the heating characteristics of a microwave-absorbing solid material inside a traveling microwave reactor can be accurately modeled. The model can be used to optimize reactor dimensions and materials, resulting in higher temperatures inside the bed and a more uniform temperature distribution. The results of this study also showed that computer modeling techniques can be used to overcome temperature measurement limitations in microwave reactors. Furthermore, experiments showed that a microwave-absorbing solid catalyst can be heated efficiently using the TMR configuration.

The introduction of the Reverse Traveling Microwave Reactor (RTMR) in Chapter 5 is a breakthrough in addressing operational challenges observed in one-way traveling microwave systems. It offers a potential solution for the scale-up of microwave-driven catalytic processes. A comprehensive simulation-based study in this chapter evaluates the effectiveness of microwave switching frequency in achieving temperature uniformity within the catalyst bed. The novel catalyst loading pattern introduced in the RTMR configuration facilitates the penetration of microwave energy deeper into the catalyst sample, improving impedance matching and subsequently enhancing the uniformity of catalyst bed heating. Results demonstrate a more efficient coupling between the catalyst sample and microwave energy, especially with the proposed conical loading pattern.

6.2. Addressing Research Questions

In this section, we delve into each of the research questions posed at the beginning of this thesis, providing concise answers based on the insights gained from our research:

1. What are the main limitations of mono–mode and multi–mode cavities in scalling–up the microwave applicators?
   Mono-mode and multi-mode cavities present challenges in scaling up microwave applicators due to size limitations and non-uniform heating. However, the TMR, with its traveling wave approach, overcomes these limitations by enabling better coupling of microwave energy with catalysts, leading to highly uniform heating.

2. What are the specific challenges with the design of a traveling microwave wave-guide in accordance with process intensification principles?
   Designing a TMR requires considerations for minimizing microwave reflections while loaded with catalyst samples. The proposed coaxial TMR structure and optimized loading patterns address these challenges effectively, resulting in a more even temperature distribution.

3. What requirements need to be met in the design of traveling microwave equipment in order to obtain a fairly uniform microwave field distribution along the reactor?
   The TMR design emphasizes optimizing microwave-catalyst interactions, ensuring a fairly uniform microwave field distribution along the reactor, which is crucial for consistent catalyst heating.

4. What is the role of impedance matching in the design of traveling microwave reactor? Aside from empty reactor design, which physical limitations and properties have to be taken into account to acquire a uniform heating profile along the catalyst bed while the reactor is loaded with a microwave–susceptible catalytic fixed–bed?
   Impedance matching plays a crucial role in TMR design to avoid microwave reflections and ensure efficient energy transfer to the catalyst. Physical properties of catalyst materials and reactor dimensions are carefully considered to achieve a uniform heating profile along the loaded catalyst bed. On the other hand, To maintain impedance matching and reduce microwave reflections while the reactor is loaded with a microwave–susceptible catalytic material, an innovative approach to loading the catalyst material is proposed. This allows for a more effective coupling of the microwave energy with the catalyst and results

in a more even temperature distribution inside the bed.

5. What is the role of the operational mode of a traveling microwave catalytic reactor? In particular, can we increase the uniformity of the temperature distribution inside the catalyst bed by periodically switching the direction of the microwave flow?
   The Reverse Traveling Microwave Reactor (RTMR) introduces a novel operational mode by periodically switching the direction of microwave flow. This innovative approach improves temperature uniformity within the catalyst bed, overcoming temperature gradients typically observed in one–way traveling microwave systems.

6.3. Comparison with Other Types of Microwave Reactors

The research presented in this thesis highlights the development and optimization of a Traveling Microwave Reactor (TMR) for heterogeneous catalytic processes, emphasizing its advantages over traditional mono–mode and multi–mode cavity–based systems. This section compares the TMR with other types of microwave reactors, specifically focusing on the Rotary Monolithic Microwave Reactor (RMMR) [1] and the Fluidized Bed Microwave Reactor (FBMR) [2].

The Rotary Monolithic Microwave Reactor (RMMR), illustrated in Figure 6.1, uses stationary cavity resonators to provide consistent microwave exposure while heating a monolithic catalyst structure. This design reduces hot spots and temperature gradients, resulting in a steady temperature distribution and excellent reaction efficiency. Its rotational mechanism and fixed cavity offer a simple and reliable way to maintain consistent heating, making it suitable for small to medium–sized catalytic operations [1].

A Fluidized Bed Microwave Reactor (FBMR), illustrated in Figure 6.2, combines the principles of fluidization with microwave heating. In a fluidized bed, solid particles (typically catalysts) are suspended in an upward–flowing gas or liquid, creating a fluid–like state. By incorporating microwave heating, the reactor ensures efficient and uniform heating of the fluidized particles, enhancing reaction rates and improving process efficiency. The fluidized nature of the bed allows for excellent heat and mass transfer, but the design and operational complexity, particularly for large-scale applica-

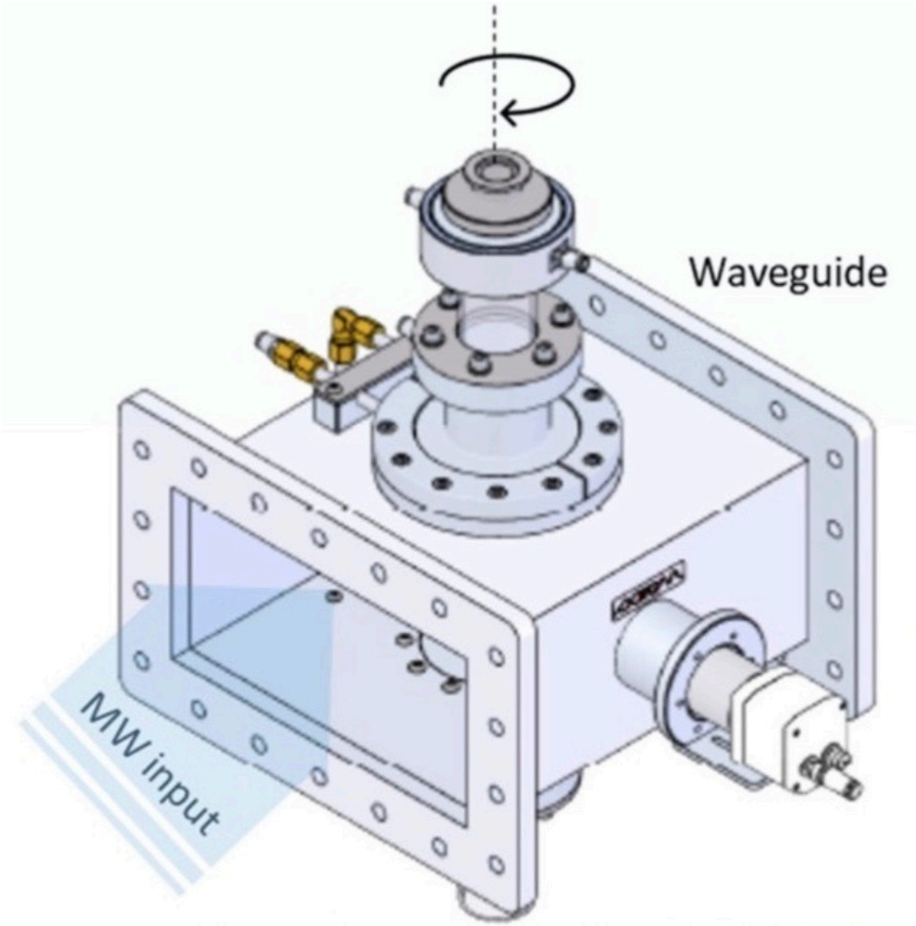

Figure 6.1: Scheme of the rotary monolithic reactor inside the rectangular cavity (adopted from [1])

tions, can present challenges [2].

The FBMR is superior in terms of scalability due to its ability to manage small and large production sizes. The TMR and RMMR, on the other hand, face challenges in scalability. The TMR's bed porosity distribution and maximum feasible bed length restrict its scalability, whereas the RMMR's larger cavity resonators and more complicated rotational mechanisms make scaling difficult. As a result, the FBMR is most suitable for scaling, while the TMR and RMMR are equally challenging to scale up.

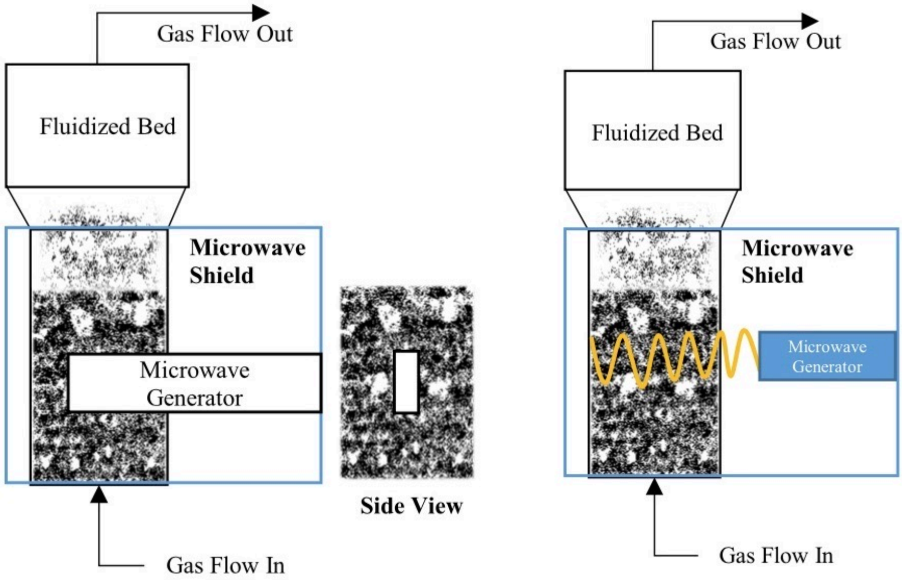

Figure 6.2: Scheme of the fluidized–bed reactor heated by a) Internal MW and b) External MW (adopted from [2])

In conclusion, each reactor type offers specific advantages for microwave–-assisted catalytic processes. The rotary monolithic microwave reactor is ideal for straightforward and reliable heating of smaller catalyst volumes, while the fluidized bed microwave reactor excels in providing excellent heat and mass transfer characteristics for small to large–scale operations. The traveling microwave reactor, however, stands out in achieving uniform microwave heating and is suitable for small to medium–scale catalytic processes.

To provide a clearer comparison, Table 6.1 summarizes the key differences and advantages of the traveling microwave reactor (TMR), rotary monolithic microwave reactor (RMMR), and fludized bed microwave reactor (FBMR):

This comparative summary provides a foundational understanding of the technologies and their implications. While the TMR stands out for its advanced capabilities in scalability and energy efficiency, the RMMR and FBMR also present valuable advantages for specific applications, highlighting the diverse potential of microwave–assisted catalytic processes.

Table 6.1: Comparison of Traveling Microwave Reactor (TMR), Rotary Monolithic Microwave Reactor (RMMR), and Fluidized Bed Microwave Reactor (FBMR).

Aspect	Traveling Microwave Reactor (TMR)	Rotary Monolithic Microwave Reactor (RMMR)	Fluidized Bed Microwave Reactor (FBMR)
Heating Method and Efficiency	Continuous irradiation with traveling wave, uniform and efficient heating	Stationary cavity with rotating monolith, ensuring even exposure to microwaves and reducing hot spots	Fluidization with microwave radiation, ensuring uniform heating through mixing and rapid heat transfer
Scalability and Industrial Application	Challenging scale-up due to bed porosity and length limitations, small to medium scales	Challenging scale-up due to complex rotational mechanisms, small to medium scales	Good scalability, continuous mixing, suitable for small to large scales
Advantages	High energy efficiency, versatile, continuous flow	Uniform heating, high reaction rates, energy efficient	Excellent heat and mass transfer, high catalytic activity, rapid heating
Drawbacks/ Challenges	High initial setup and operational costs, precise control needed	High initial costs, scalability challenges, complex rotational mechanism	Complex design and operation, precise control needed, potential particle issues

6

6.4. Suggestions for Future Studies

The research presented in this thesis provides a foundation for further studies in the field of microwave-assisted heterogeneous catalysis. Several possible areas of future research include:

- **Experimental Validation:** Future studies could focus on the experimental validation of the models and simulations presented in this thesis to verify their accuracy and reliability in further applications.

- **Comparative Analysis:** Researchers could explore the performance of the Reverse Traveling Microwave Reactor (RTMR) configuration in various catalytic processes and compare its performance to the recent microwave reactor configuration, such as the Zaragoza's monolith rotary reactor.

- **Catalyst Material Variation:** Investigations into the effects of different catalyst materials and shapes on heating and uniformity in the catalyst bed could be fruitful for optimizing microwave-assisted catalytic reactions.

- **Advanced Modeling Techniques:** Future research could involve the development of advanced models and simulation techniques to further optimize the design of traveling microwave reactors, with a focus on improving heating efficiency and uniformity.

- **Diversified Applications:** Investigating the potential for using microwaves in other types of catalytic processes, such as biocatalytic processes, and evaluating their performance.

The potential of microwave radiation as a practical heating technique for heterogeneous catalytic processes is highlighted throughout this thesis, along with a detailed examination of the design and optimization of traveling microwave reactors for catalyst heating. The findings provide valuable insights into how various reactor designs affect temperature distribution in the catalyst bed and open opportunities for further research in microwave–assisted heterogeneous catalysis.

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A special thank you to Dr. Johan Padding, who stepped in during the closing phases to assist me pull everything together. Johan, your mentorship and support throughout these final, critical stages made all the difference. Thank you for stepping in and seeing me through to the finish; I'm lucky to have had you by my side.

I'm also incredibly grateful to TU Delft's staff, whose expertise, kindness, and willingness to help make this journey easier and more pleasurable. And to my friends and colleagues at the Process and Energy faculty—thank you for the inspiration, the laughs, and just being there to make this experience so memorable.

I want to take a moment to share my heartfelt gratitude to my wonderful partner, Maysam, and my amazing son, Radin. Maysam, you've been my rock through all of this. Your endless patience and understanding, especially during the most demanding phases of this research, have been my anchor. And Radin, your bright smile and boundless energy are the perfect reminders of what truly matters, always lifting me up when I need it the most. I love you both beyond words, and you know it better than anyone.

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Curriculum Vitæ

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List of Publications

3. **F. Eghbal Sarabi**, Jiawen Liu, Andrzej Stankiewicz, Hakan Nigar, *Reverse Traveling Microwave Reactor – Modelling and Design Considerations*, Chemical Engineering Science **246**, p. 116862 (2021).

2. **F. Eghbal Sarabi**, Andrzej Stankiewicz, Meisam Ghorbani, Hakan Nigar, *Coaxial Traveling-Wave Microwave Reactors: Design Challenges and Solutions*, Chemical Engineering Research and Design **153**, pp. 677–683 (2020).

1. Andrzej Stankiewicz, **F. Eghbal Sarabi**, Abdul Baubaid, Peng Yan, and Hakan Nigar, *Perspectives of Microwaves–Enhanced Heterogeneous Catalytic Gas-Phase Processes in Flow Systems*, The Chemical Record **19**, pp. 40–50 (2019).

Propositions

accompanying the dissertation

Traveling Microwave Reactor: Design Challenges and Solutions

by

Farnaz Eghbal Sarabi

1. By applying flip thinking, this research aims to transform common challenges in microwave heating into unique opportunities for innovation and advancement. (this thesis)
2. The shape of catalyst particles within a reactor significantly impact the efficiency of microwave heating, affecting both the distribution of electromagnetic waves and the resulting thermal profiles. (this thesis)
3. Accurate modeling of dielectric properties is essential for predicting and controlling the heating behaviour and ensuring the stability and efficiency of the reaction process. (this thesis)
4. In certain contexts, silence is a more powerful and strategic tool than actively proving a point.
5. Prejudgment, rooted in our biases and preconceived notions, often distorts our perception of others and hinders genuine understanding and connection.
6. We should evaluate our lives, considering our circumstances and requirements, but not based on the beautiful appearance of others' lives.
7. While the fear of missed opportunities often drives bold action, the consequences of regretting impulsive decisions can outweigh the remorse of inaction.
8. When you think you have reached the end, it is just the beginning.
9. If you want to succeed, do everything hard; work hard, sport hard, laugh hard, ...

These propositions are regarded as opposable and defendable and have been approved as such by the promotors prof. dr. ir. A.I. Stankiewicz and prof. dr. ir. J.T. Padding.