IEEE Access
Multidisciplinary ⋮ Rapid Review ⋮ Open Access Journal

Received 21 March 2023, accepted 16 April 2023, date of publication 19 April 2023, date of current version 25 April 2023.

Digital Object Identifier 10.1109/ACCESS.2023.3268538

RESEARCH ARTICLE

A Novel Combined Design of Vessel and Resonant Cavity for Microwave Multi-Frequency Heating Chemical Reactor Using Antennas as Applicators

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This work has received funding in part by the European Union's Horizon 2020 research and innovation programme under grant agreement No 820665-polynSPIRE project, and in part by the European Union's Horizon Europe research and innovation programme under grant agreement No 101058540-PLASTICE project. The work of María Campo-Valera was supported by the Postdoctoral Program Margarita Salas—Spanish Ministry of Universities (financed by European Union—NextGenerationEU). The work of Ignacio Rodríguez-Rodríguez was supported by Plan Andaluz de Investigación, Desarrollo e Innovación (PAIDI), Junta de Andalucía, Spain.

ABSTRACT In this work a new design concept in the field of microwave heating assisted chemical reactors is proposed focusing on two main innovations. The first one consists in combining the resonant cavity and the vessel in the same volume, improving the durability of the system by using a metallic material for the vessel. The second consists of integrating antennas as applicator ports for energy transmission from the magnetron into the cavity, instead of waveguides, allowing greater design flexibility in terms of cavity and system dimensions. In this work, a microwave reactor with four electromagnetic energy emitting antennas is optimized. This innovation is evaluated by means of a simulation model that solves the finite element method (FEM) using the commercial software COMSOL Multiphysics coupling radio frequency and heat transfer physics. Within this work, simulations have demonstrated the microwave heating process in the configured chemical reactor where the implementation of standard waveguides would not be possible due to the incompatible size required in the selected diameter dimension for 106 litres. Therefore, the results achieved lay the foundation for the construction of a microwave reactor intended to drive the industrial process of chemical recycling of polymers on a large industrial scale.

INDEX TERMS Microwave heating technology, electromagnetic antennas, resonant cavity, FEM simulation, multiphysics engineering application.

I. INTRODUCTION

Plastic have caused a revolutionary change in the materials market, decreasing the price of final products. It is a light, cheap, durable and easy to adapt to the needs material so all this can explain why its use is so widespread throughout the world. Classic recycling causes degradation of polymers as a consequence of heat and mechanic cycles. Chemical recycling involves the modification of the molecular structure of polymeric waste in order to create new molecules that can be utilized to produce a new polymer with similar properties to virgin polymer, or alternatively, to manufacture other products.

There is a trend in which more and more plastic is produced and, in most cases, it is not managed properly. Since 1950 till 2015, it is estimated that about 6,3 billion tons of plastic waste was generated, of which around 9% was recycled, 12% was incinerated and the rest (79%) was

The associate editor coordinating the review of this manuscript and approving it for publication was Tutku Karacolak.

stored in landfills or was released directly into the environment. [1] This tendency, added to the slow degradation of polymers in nature and all the implications their presence in the environment has, make necessary to gestionate them.

As mentioned previously, chemical recycling presents several advantages over conventional recycling, and that is why there is a great interest in developing more efficient techniques. Chemical reactors are the main point in the depolymerization process, and the heat source is the requirement more important int terms to make feasible this process from the energetical point of view [2], [3], [4]. Microwaves play a key role in this development since compared to traditional heating techniques, they are more energy efficient, temperature homogeneity is higher, milder reaction conditions can be reached, shorter heating time is required and higher yields can be achieved [4], [5]. In addition, microwave heating technology applied to chemical compounds as acids or amines is highly effective due to its ability to absorb microwave radiation. This work aims to validate through simulations, an innovative design capable of being developed and brought to reality of a type of microwave reactor whose design will be highly flexible by integrating antennas instead of waveguides as energy transmission elements towards the cavity.

Microwave instruments can be classified into two types: monomode and multimode reactor [6]. In monomode reactors, only one reduced size reactor vessel can be irradiated, being provided a highly homogenous energy field of high-power intensity, resulting in fast heating rates oriented toward the reaction vessel [6], creating a standing wave. Monomode reactors have the advantage of that microwaves can be applied using a coaxial dipole antenna as applicator inside the reacting medium [7], [8], [9], which make them highly efficient in heating (since all the energy is absorbed by the medium), flexible and easily controllable, enabling its easy industrial scale-up [5]. In contrast multimode reactors provide better reproducibility [10] and they are implemented for larger cavities and volumes that may accommodate one or several vessels for heating at once, where the electromagnetic waves are reflected from the walls in a chaotic way and oscillating its mode by stirrer. In multimode devices, to optimize the homogeneity of the electromagnetic field distribution in the material, commercial cavities use a mode stirrer or turntable reaction places [6]. In the presented application focused on an industrial development where high process volumes are required, the selection of the type of cavity can only be multimode due to the indicated characteristics.

It is also interesting to present a metal-based vessel and reactor combined in order to increase the irradiance power over the reaction medium, increasing the effective volume of microwave energy transportation and reducing parts and construction materials at the same time that it is solved problems like material resistance against pressure operation or corrosion. Bulk metals tend to reflect the incident waves giving place to constructive and destructive interferences caused by the overlap of waves [11].

On the field of plastic chemical recycling, they are usually recycled by high pressure hydrolysis processes, where microwaves can be employed. In this process, microwaves help water to penetrate into polymer chemical structure, enhancing the rupture of their bonds and therefore, reaching high degradation rate compared with conventional heating [12], [13], [14].

With that proposal, dipole antennas can be used as microwave interstitial applicator, substituting conventional microwaves oven type reactors attending to its promising performances. They are an interesting alternative due to their simplicity, flexibility and very high efficiency in the transfer of energy from the microwave source (magnetron) directly to the medium. [8]. However, the implantation of this technology requires a deep numerical and experimental study to define both important experimental and security aspects for its implementation.

The authors in [15] proposed a study focused on microwave heating operation over chemical reactors with allocated vessels inside, using multifrequency energy applicators. This work presents a new study designing a reactor that combines the resonant cavity and the vessel, and implements antennas as applicators for heating processes during the depolymerization of plastics in the chemical recycling. This paper is organized as follows. Section II describes the methodology and framework of the presented innovation in a chemical reactor. In Section III, the authors presents the FEM simulation results analysis focused on electric field and temperature. In Section IV main achievements are presented. Finally, additional innovations and improvements over the designed model are indicated in Section V.

II. METHODOLOGY

The use of microwave heating as a method of heating and processing materials has gained increasing attention in recent years due to its unique advantages, including faster heating rates, more uniform heating, and the potential for energy savings [16], [17]. In this paper, we present this heating technology applied to chemical reactors where target temperature is estimated between 150°C and 200 °C, being a suitable method once checking that chemical products used in the mixing material as chlorhydric acid is highly susceptible to be heated by the proposed heating method.

Through simulations, it is demonstrated the advantages of using microwave heating over traditional methods, including improved processing times and reduced energy consumption. Furthermore, we believe that our model has broad implications for industry, as it provides a powerful tool for optimizing the use of microwave heating in a wide range of applications where the processed material would be compatible to the microwave heating process. Through a detailed discussion of the motivations and contributions of our research, we aim to provide readers with a comprehensive understanding of the significance of our model and its potential impact on the field.

Multimode cavities are the most widely used form of microwave applicators [7], [8]. In essence, any metal container a few times larger than the wavelength can serve as a basic multimode microwave applicator. Since multimode cavities are not limited in their dimensions, they can be used to process large or complex shaped products [18], [19]. The microwave energy, carried by a waveguide or a transmission line into the cavity, is deposited into a lossy material placed within.

The proposed microwave-assisted chemical reactor design combines resonant cavity and vessel, offering cost-effectiveness, flexibility in size, and improved performance, replacing traditional waveguides and their required elements by antennas used as ports that they are easier to be installed and their size allows be integrated where size restrictions are presented. The combination of multi-frequency heating and a stirrer, results in a more uniform temperature distribution avoiding hot points and reduced heating time. However, optimizing antenna distribution and their geometric design remains a challenge, and computational time may increase when considering dielectric properties variation at various temperature points. Despite these limitations, the presented reactor design has potential applications in chemical processes, sterilization, drying and dehydration, biofuels and biomass production, and nanopowder production in metals and ceramics.

In this paper, a multimode cavity reactor is defined. During reactor detailed design process, some factors have appeared as limitation for a feasible construction:

- Material availability
- Pressure compensation system
- Structural behaviour and security
- Electronic components availability

Thus, a new reactor design has been generated with a final volume of 106 L, unifying the reactor cavity and vessel as the same recipient and volume in a metallic structure able to work under pressure and corrosive conditions, and including antennas as microwave energy applicators. The described model as follow Fig. 1.

The main innovation presented by the authors is the use of antennas instead of waveguides for transmitting energy into the resonant cavity of a microwave-assisted chemical reactor. This approach allows for greater flexibility in the design and size of the reactor, as the resonant cavity and vessel can be integrated in the same volume and smaller reactor diameters can be used. If traditional waveguides were used for energy transmission, higher cavity dimensions would be required to accommodate the 915 MHz standard waveguide (WR975) and introduce this energy inside the cavity due to 915 MHz wave size. Therefore, the use of antennas provides a more efficient and adaptable approach to microwave-assisted chemical reactions.

A preliminary study was conducted wherein the configuration of the model was chosen by positioning antennas at an arbitrary height where the maximum electromagnetic field was detected for energy transmission. While this model validates the conceptual design, future efforts will be directed towards optimizing the position and number of antennas to enhance the uniformity of the electric field.

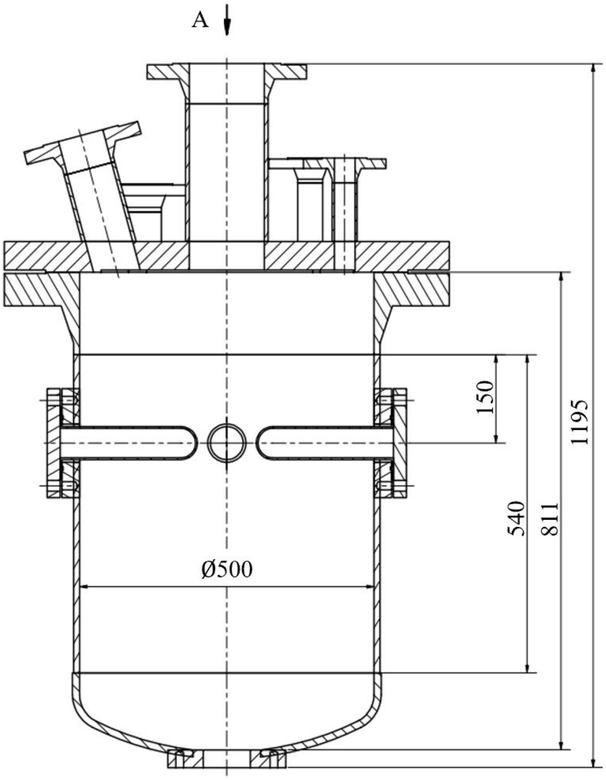

FIGURE 1. Resonant cavity dimensions of the model in mm, cross section.

A reactor configuration using the same working volume for reactor cavity and vessel will improve the equipment in terms of security by using metallic material as resonant cavity and vessel that is working under pressure conditions, but their manufacturing requires specific alloy material resistant against acid corrosion, being selected Hastelloy C22 or Hastelloy C276 for project requirements. A multifrequency operation system has been deployed to precisely regulate the position and power output of every microwave applicator port. The system has a total of four generators. Three of these generators are rated at 6 kW and operate at a frequency of 2.45 GHz while the fourth generator is rated at 5 kW and operates at a frequency of 915 MHz. It is decided for each generator to implement an antenna as microwave energy applicator inside the cavity, distributing each one around the reactor cavity, as shown in Table 1. All these assumptions are introduced in the model as boundary conditions where is defined:

- Main volume as a domain defined by material mixing to be heated and air.
- Electromagnetic ports as coaxial inputs configurated each one according to their work frequency, power and wave phase.

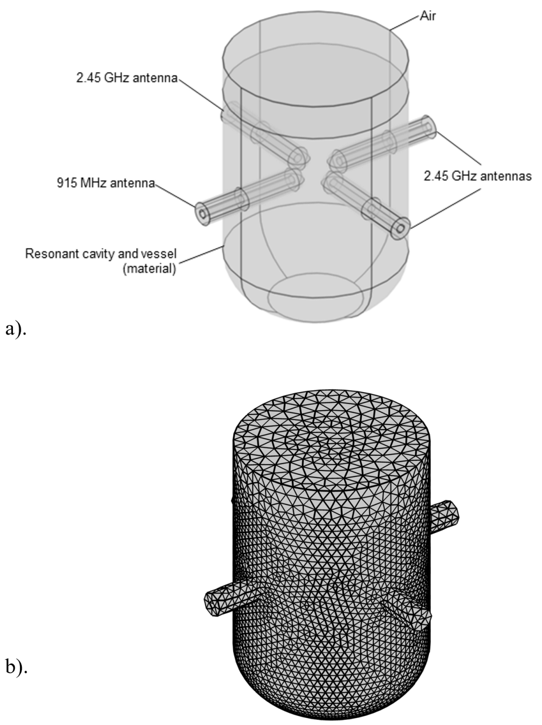

FIGURE 2. **(a) Configuration used in the detailed design resonant cavity, 106 L; (b) FEM simulation meshing in COMSOL Multiphysics.**

TABLE 1. **Magnetron frequency, power, and waveguides reference of the microwave system.**

Magnetron frequency	Power	Quantity	Waveguide reference
2.45 GHz	6 kW	3	WR340 (antenna)
915 MHz	5 kW	1	WR975 (antenna)

- External boundaries are selected as an Impedance boundary condition that will create the condition of a material that reflects electromagnetic waves.

In addition, the simulation study is modelled with the following assumptions:

1) Mixing material to be heated is modelled as liquid.
2) Work pressure inside the volume is constant fixed in 12 bar.
3) No phase change is considered.
4) Fluid rotational movement is modelled applying a described velocity that generates an axial movement.

A. NUMERICAL METHOD: MICROWAVE HEATING

Microwave heating is a multiphysics phenomenon involving electromagnetic waves and heat transfer; therefore, any material that is exposed to electromagnetic radiation will heat up. Rapidly varying electric and magnetic fields lead to four sources of heating. Any electric field applied to a conducting material will cause current to flow. In addition, a time-varying electric field will cause dipolar molecules, such as water, to oscillate back and forth.

This section introduces the Finite Element Method using the commercial software COMSOL Multiphysics [20]. The preprocessing, processing and postprocessing used in the simulation of the vessel was implemented as follows:

- Preprocessing: The geometry used for the simulation is a vessel of cylindrical material with a cavity of 106 L. Regarding the frequencies used for the simulations, we have 915 MHz and 2.45 GHz. The meshing has been carried out with $\lambda/6$ with a maximum frequency of 2.45 GHz as depicted in Fig. 2.
- Processing: The creation of the proposed numerical model using the Finite Element Method (FEM) involves utilizing the radio frequency (RF) module is utilized to simulate the process of electromagnetic heating. The model is based on the assumption of a time-harmonic electromagnetic field and is derived by implementing Maxwell's equations [21], as described in the accompanying equation (1).

$$\nabla \times \left(\mu^{-1} \nabla \times \vec{E}\right) - (\omega^2 \varepsilon - i\omega\sigma)\vec{E} = \vec{0} \qquad (1)$$

where ∇ is nabla gradient operator which calculates the gradient of a scalar field; i is the imaginary symbol; E is the complex-valued electric field; μ is permeability; ω is the angular frequency; ε is the permitivity; and σ is the electrical conductivity.

In this model, the two modes of heat transfer, namely convection and diffusion, are taken into consideration. Regarding heat transfer in a fluid, three important contributions to the heat equation are considered. The first, consists of the transport of energy, which is represented as a convective term in the heat equation. The second, has to do with the viscous effects produced by heating in the fluid, which are noticeable in fast flow in viscous fluids. The last one, is the fluid density which depends on the temperature. Taking into account these contributions, we obtain the expression (2) which represents the transient heat equation governing the temperature field within a fluid.

$$C_p\rho \frac{dT}{dt} + \rho c_p u \cdot \nabla T = \alpha_p T \left(\frac{\delta p}{\delta t} + u \cdot \nabla p\right) + \\ = \tau : S + \nabla \cdot (k \nabla T) + q \qquad (2)$$

where $C_p\rho$ is the effective heat capacity; C_p is fluid heat capacity; ρ is the fluid density; u is the fluid velocity field; T is Temperature; α_p is the coefficient of linear thermal expansion; k is the effective thermal conductivity; q is heat source.

Building upon the earlier mentioned development of the numerical model using FEM, equations (1) and (2) are incorporated into the multi-physics model to analyze the microwave assisted heating process.

- Postprocessing: Regarding the post-processing, the results are studied in the frequency and time domain. It is obtained the electrical values necessary for converting transmitted power into heat in the frequency domain.

In the time domain, for each time studied (1 min, 20 min and 40 min) the heat transfer conditions are applied to the material domain by the model. Finally, the analysis of time-dependent electromagnetic heating involves the coupling of the two aforementioned physics. At each studied time, the solver assesses the amount of energy transferred to each element of the mesh by evaluating the heat transfer boundary conditions.

III. RESULTS

Under the aim of understanding how the electric and electromagnetic fields are distributed throughout the reactor cavity, a series of simulations were carried out. A study about the distribution of the electric field considering the frequencies of 915 MHz and 2.45 GHz individually, as well as their coupled effect and its correlation with the formation of hot and cold spots.

It also was simulated the electromagnetic field density distribution to evaluate the energy conversion in the vessel volume which was completed with a further study of the effect of stirring on temperature distribution.

A. ELECTRIC FIELD

Fig. 3 displays graphical representations of the electric field coupling. The results obtained from simulating the electric field for the frequencies of 915 MHz, 2.45 GHz and coupling frequencies are presented below.

1) 915 MHz

Having just one applicator antenna inside the cavity, a specific area affected by this frequency is located close to the port, being introduced until intermediate depth. The results of the simulation indicate that the average electric field within the vessel volume is 2137 V/m, with a standard deviation of 967 V/m based on the configuration used. The standard deviation represents in this evaluation the homogeneity measurement of the electric field, being more homogeneous fields whose showing lower standard deviations.

2) 2.45 GHz

Due to three different 2.45 GHz antenna applicators are distributed along the resonant cavity volume, the simulation results indicate a more uniform distribution of the electric field. It is observed that the areas of high electric field intensity are concentrated in small zones near the ports along the cavity. Regarding 2.45 GHz configuration, the electric field mean is higher to previous case, resulting in 4132 V/m because higher power is applied with 3 magnetrons, being the standard deviation 1743 V/m.

3) COUPLING FREQUENCIES

Combining the indicated frequencies (915 MHz and 2.45 GHz) results in an uniform distribution of the electric field throughout the volume of the vessel, both in the lower and upper regions. Moreover, the electric field values are higher when the frequencies are combined, compared to using each frequency individually. The average electric field value and its standard deviation were 4945 V/m and 2110 V/m, respectively.

TABLE 2. Simulation results according isolated frequency of magnetron.

Frequency	Electric field (V/m)	Standard deviation (V/m)
915 MHz	2137	967
2.45 GHz	4132	1743
Coupling	4945	2110

Table 2 compiles obtained results of electric field and their standard deviation for every operated frequency.

The numerical results show that the 915 MHz energy applicator produces the strongest field with an average of nearly 9500 V/m, but with low uniformity. On the other hand, the use of the 2.45 GHz antennas leads to a more uniform field, but the electric field strength only increases by 30% and the power output is increased by 300%. When the two frequencies are merged, the resultant field exhibits the highest degree of uniformity compared to the other fields where frequencies are operating in isolation, but differences between slices of the vessel can still be observed. These differences could be improved by optimally distributing the antenna ports vertically.

Regardless of establishing a configuration that maximizes the homogenization of the electric field throughout the entire volume, the simulation reveals the existence of both hot and cold spots. The cross-sectional view presented in Fig. 4 illustrates these findings, where hot spots appear with the same power level and distributed over the slice, at the same time that a cold area is keeped in the inner.

At 915 MHz simulations, a large hot point is found close to antenna with a low gradient level, whereas the vessel volume is absorbing a reduced level of energy from the MW source, as depicted in Fig. 3a. The simulation at 2.45 GHz indicates a shallower wave penetration depth compared to the 915 MHz simulation. However, the presence of hot spots is largely mitigated, except in the inner region, as illustrated in Fig. 3b. The simulation results show that combining both frequencies of 2.45 GHz and 915 MHz leads to a more consistent electric field intensity across each of the ten slices along the vessel height. Furthermore, if the applicator antennas are positioned vertically, the homogeneity of the volume can be further improved, as demonstrated in Fig. 3c.

B. ELECTROMAGNETIC POWER LOSS DENSITY

The electromagnetic power loss density (W/m^{-3}) is visualized in Fig. 5. It is showing a homogeneous energy conversion along the vessel volume. The most intense electromagnetic power loss density is located at plane where applicator antennas are operating.

C. TEMPERATURE AND STIRRER EFFECT

A study of time-dependent heat transfer was performed using FEM multiphysics simulation to investigate two different sce-

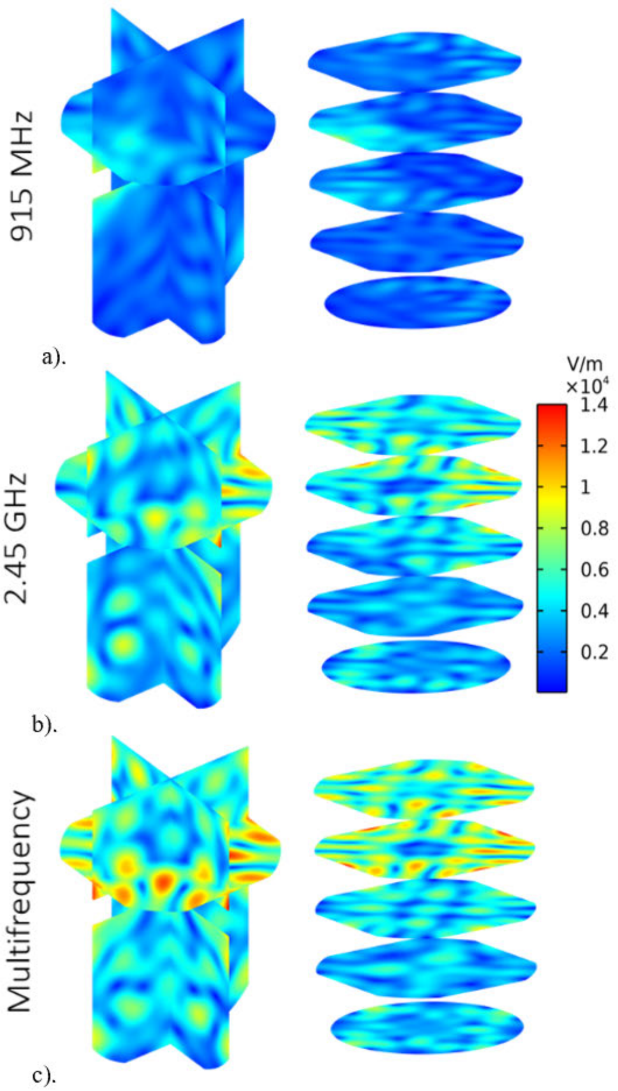

FIGURE 3. Normalized electric field (v/m) using three different frequency operating scenarios. The left panel shows the longitudinal section, while the right panel shows the cross-section along the Z-axis. The three frequency scenarios are as follows: (a) 915 MHz, (b) 2.45 GHz, and (c) coupled frequencies.

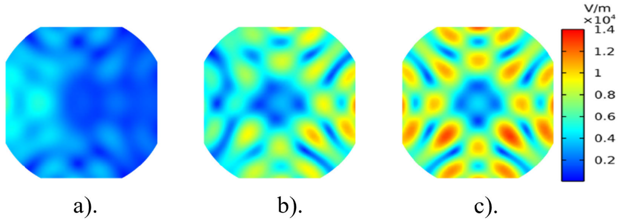

FIGURE 4. Normalized electric field (v/m) using three different frequency operating scenarios. Cross-section in Z-axis at 0.475 m from the reactor floor. (a) 915 MHz; (b) 2.45 GHz and (c) coupled frequencies.

narios. In the context of microwave heating, time-dependent heat transfer analysis is utilized to investigate the dynamic distribution of heat within a system as it evolves over time. This type of analysis is crucial for assessing and optimizing

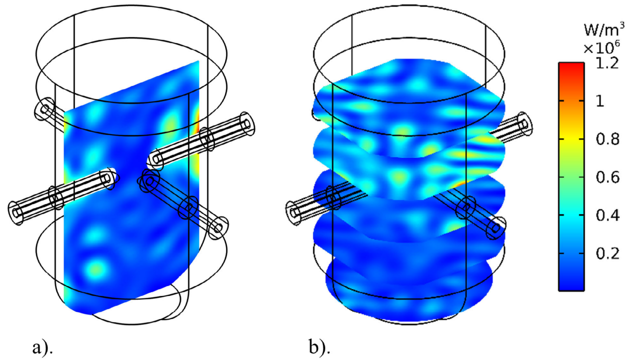

FIGURE 5. Electromagnetic power loss density (W/m^{-3}) with differents sections. (a) Longitudinal section and (b) Z-axis cross-section.

the performance of microwave heating systems, especially in applications where the uniformity of temperature distribution is critical. In the first scenario, a static material volume was considered, and the uneven distribution of electromagnetic losses along the vessel resulted in scattered hot spots and a highly non-uniform temperature distribution, as depicted in Fig. 6a. This non-uniform temperature distribution is primarily attributed to the electromagnetic heat loss, which serves as the primary source of heat in the microwave system and does not ensure consistent processing conditions throughout the batch. Similar electric field distributions can also be observed. In the second scenario, the dielectric properties were kept constant, but the material inside the vessel was modeled as a fluid in motion, resulting in a new temperature distribution where the temperature values were uniform throughout the vessel, as shown in Fig. 6b. These results demonstrate that the incorporation of a mixer significantly enhance the temperature distribution.

In addition, considering no stirrer influence, it is evaluated the average temperature for different cross-sections inside of reactor volume, at time intervals from 0 to 40 min. As represents Fig. 7.

It was checked and validated the influence of the stirrer system inside the chemical reactor with the aim to homogenize the temperature and avoid hot points. Inspecting the simulation model where a static material is processed hot points reaching until 650°C are observed, being a value five times higher that the mixing temperature achieved using a stirrer system, being represented in Fig. 8.

Considering the stirrer influence over the mixing material of the reactor achieving a total mixing of the material removing any hot spot, it is considering an evolution of the temperature of the material along time as it is shown in Fig. 9.

The results from the FEM simulation indicate that it is possible to reach the target temperatures set in the chemical experiment within a reasonable time frame. The combination of multi-frequency heating and the use of a stirrer leads to a homogeneous temperature distribution in the vessel, making this system suitable for the recycling of all polymers considered, as shown in Table 3.

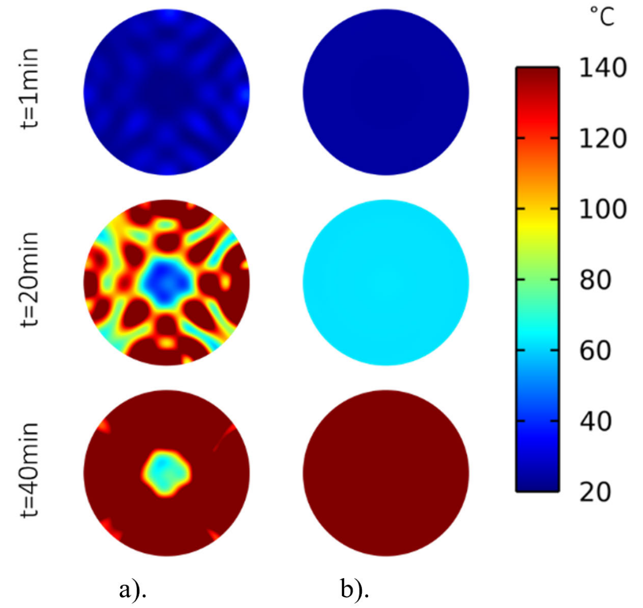

FIGURE 6. Comparison between different temperature distributions in a cross section 0.475 m from the bottom of the reactor. (a) Simulation considering a static material under heating and (b) fluid moving in the vessel.

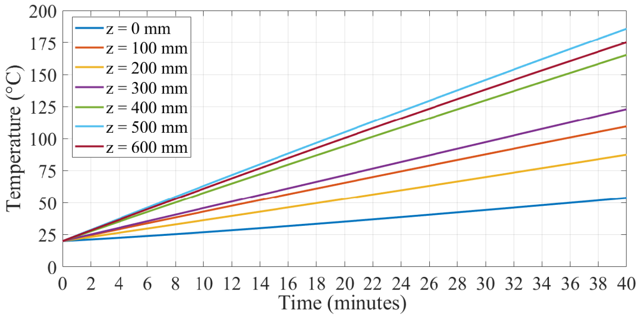

FIGURE 7. Mean temperatures at various cross-section within the reactor over time intervals spanning 0 to 40 min.

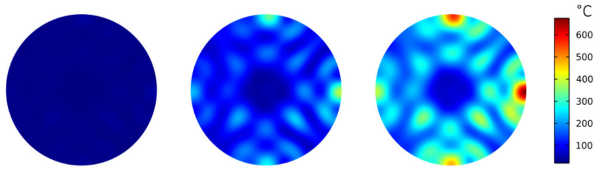

FIGURE 8. Found hot spots in a cross-section at a cross-section located 0.475 m from the bottom of the reactor at time intervals = 1 m, 20 min and 40 min.

TABLE 3. Experimental conditions extracted from laboratory tests.

Depolymerization material	Material temperature (°C)	Vessel pressure (bar)	Processing time (minutes)
PA66	170	4.6	30
PA66	190	10	10
PU	230	5.8	40

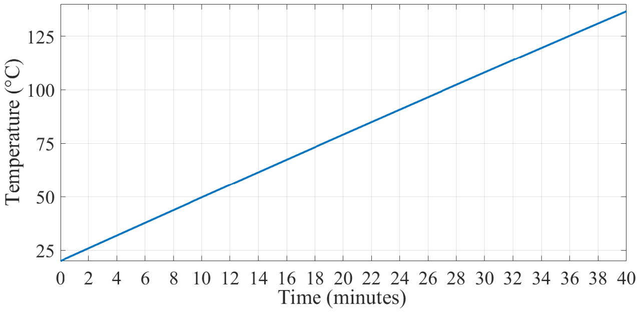

FIGURE 9. Temperature average in the reactor volume at time intervals from 0 to 40 min.

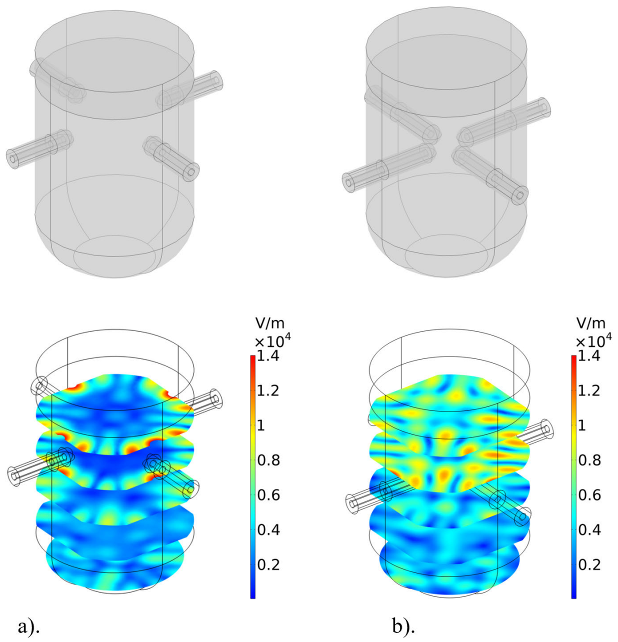

FIGURE 10. Inserted antennas. (a) Initial position and (b) Additional 120 mm inserted antennas.

In addition of above, one parameter that has high influence is the antenna design that is transmitting the microwave energy inside the cavity. It was checked by different simulation models that the inserted length of the antenna inside the cavity modifies the electric field and the final temperature achieved by the mixing material inside the cavity, being more than two times in the most optimized case found. As a first conclusion, as deeper is the antenna inserted in the cavity, higher electric field is generated inside the cavity, more energy is transmitted in an effective way and, thus, higher temperatures are achieved. It is an effect created both wave size and the interaction between each antenna and cavity. Graphically, it is explained by a comparison of electric fields of the same design by varying the inserted length of the antennas in Fig. 10. Further planned studies will optimize the distribution of antennas and their configuration of size, diameter and format.

IV. CONCLUSION

A design innovation in the field of chemical reactors assisted by microwave heating resonant cavities has been developed in this paper including two main specific characteristics: i) combination of resonant cavity and vessel where the chemical mixing material is processed using the benefits of metal material as container; ii) integrate antennas as applicator ports of microwave energy inside the cavity, obtaining a flexible model of the equipment in terms of selecting new formats or dimensions of the cavity independently of the frequency implemented.

Technically, during this work, the physical system utilizing microwave heating was transformed into two separate mathematical models, offering benefits for future design phases and system arrangements. These results establish a foundation for constructing a microwave reactor aimed at advancing the industrial process of polymer chemical recycling on a large industrial scale.

The modelled reactor was studied by adjusting the frequency, power, number, and placement of the microwave antennas, and the results of this study have shown that the frequency combination (915 MHz and 2.45 GHz) is a technical option that achieve a better homogenization of the electromagnetic field. In addition, the integration of a stirrer inside the resonant cavity reaches a more homogeneous temperature of the sample, minimizing the presence of thermal runaway in the processed material. Furthermore, compared to traditional heating technologies, the use of a microwave heating source enables a considerable decrease in heating time (at least 50% than using a convection heating technology), due to a direct heating increases the process's overall efficiency focusing directly on chemical material.

In addition to indicating the main advantages of implementing a chemical reactor where the heat source is provided by microwaves, by using antennas, they allow to design and create more flexible reactor formats and sizes, in this case unifying resonant cavity and vessel, and using lower diameters, because if microwave energy were transmitted by traditional waveguides, it would be required higher cavity dimensions for adapting 915 MHz standard waveguide (WR975).

V. FUTURE WORKS IN MICROWAVE CHEMISTRY

Future efforts would be focused in the antenna design and position, and their distribution along the resonant cavity, creating sensitive and a parametric studies about position and geometrical design of the antenna, based in a mathematical simulation.

This will provide more optimiced results independlty of the stirrer effect applied to the processed material. In additon to configurate the specific dielectric propeties of the material for each temperature point will provide more realistic results, although a higher computational time will be required.

Theoretical developed model can be implemented in different sectors where specific material conditions are required according the processed substances or lower sizes are better with the aim to optimize budget or improve the design of the resonant cavity. Current and future trends in the microwave reactors industry include the following:

- recycling (plastic and composite materials).
- pharmacy (sterilization).
- drying and dehydration.
- biofuels and biomass.
- nanopowder productions in metals and ceramics.

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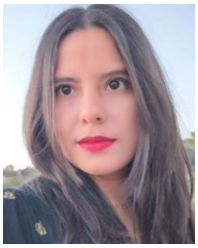

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