Finite element model for microwave heating used in chemical recycling of plastic waste material

A. Frisa-Rubio[1]*, C. González-Niño[1], N. García-Polanco[1], S. Fießer[2], T. García-Armingol[1,3]
1. Fundacion CIRCE, Zaragoza, SPAIN
2. Fricke und Mallah Microwave Technology GmbH, Peine, GERMANY
3. Instituto Universitario de Investigación Mixto CIRCE - (Fundacion CIRCE - Universidad de Zaragoza), Zaragoza, SPAIN
*Corresponding author: afrisa@fcirce.es, circe@fcirce.es

Abstract

Over the last years, there has been an increasing interest in plastic recycling as an alternative to reduce fossil fuel dependency. In this light, chemical recycling has been presented as a promising strategy to maximise the recycling rate, although this technology presents drawbacks such as its high energy consumption. In addition, in case of using conventional heating technologies to provide this energy, the potential environmental benefit of recycling can be reduced. As a possible alternative to minimize the deleterious effect on environmental impact, the substitution of conventional heating technologies by microwave (MW) heating can bring important benefits. From this perspective, the aim of the current work is to design and optimise a MW assisted reactor for polymer recycling using up to eight ports emitting electromagnetic waves at two different frequencies, simulating the physical process of material heating inside the vessel.
Several scientific publications are available in the literature concerning single-frequency simulation (S-F) for different geometries and applications, however, few research articles delve into multi-frequency (M-F) simulation. In this study *COMSOL Multiphysics®* was used to perform the modeling, the finite elements method (FEM) simulation of the microwave heating of a reacting mixture is achieved coupling the "Electromagnetic Waves, Frequency Domain" physics from the RF Module with the "heat transfer in fluids" physics from the Heat Transfer Module through the "electromagnetic heating" multiphysics. A prediction of the electric field and temperature distribution in the reactor over time was obtained using the proposed framework under different geometrical configurations. Special care was taken to avoid the induction of temperature hotspots in the domain of the chamber, ensuring uniformity of processing conditions. The main results consist in the design of an appropriate MW assisted reactor for depolymerization, capable of producing a homogeneous electric field and reducing the heating time to reach the target temperature (170-200 °C).

Keywords

Electromagnetic heating, design optimization, microwaves, plastic recycling, multi-frequency microwaves, polyamide, polyurethane.

1. Introduction

The present research is focused on polyamide (PA) and polyurethane (PU) depolymerization processes based on chemical reactions. Chemical recycling technology allows recovery of the original monomers without altering their properties [1]. The first step is to identify and describe the chemical reactions taking place, adding chemical agents to activate and speed them up. From this step, key parameters are obtained such as pressure conditions, process temperature or depolymerization time. The second step is to combine the heating and mixing processes to accelerate the chemical reaction. This work shows the introduction of microwave heating technology in the chemical recycling field, where microwave radiation allows a faster heating process [2]. The benefits of this approach are two-fold, since this technology also leads to an increase in the sustainability of the industrial process when compared to conventional heating methodologies that mainly rely on fossil fuels that produce polluting gases [3].
Our design of a MW-assisted reactor, as shown in Figure 1, consists of two cylinders that share a common axis. The exterior cylinder is fitted with two 915 MHz (2.5 kW each) and six 2.45 GHz (3 kW each) waveguides for a total power of 23 kW and it is designed as a metallic multimode cavity. The internal one is built in a material transparent to MW (PTFE) [4] and acts as the reservoir for the reacting mixture, where its low thermal conductivity [5] will benefit the heating process in the reservoir, minimizing heat losses to the environment.

2. Numerical Model

In this model, the finite elements simulation of the microwave heating of the reacting mixture is achieved by means of coupling the "Electromagnetic Waves, Frequency Domain" physics from the RF Module with the "Heat Transfer in Fluids" physics from the Heat Transfer Module through the "Electromagnetic Heating" Multiphysics. This complex problem also included temperature dependent permittivity values that were obtained experimentally for each frequency used in the reactor.
In order to simulate the entire reactor and the heating process to achieve the depolymerization, two different *COMSOL Multiphysics®* modules are used in this work: Radio Frequency (RF) and Heat Transfer (HT). In the RF module, the Maxwell equations and the Poynting theorem are used to simulate the electromagnetic heating process. In the HT module, the energy conservation law is used to simulate the heating process.

2.1 Electromagnetics and RF modeling

Assuming time-harmonic EM field, Maxwell equations reduce to a vector Helmholtz equation with complex valued components [6] - these are formulated in *COMSOL Multiphysics®*:

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

where μ is magnetic permeability, ω is angular velocity, σ is electrical conductivity and $\vec{E}$ is complex-valued electric field. Dissipated power density q due to MWs [7] is given by

$$q = -\nabla\cdot\vec{S} \quad (2)$$

Where $\vec{S}$ is the Poynting vector [6], which is computed as

$$\vec{S} = \frac{1}{2}Re(\vec{E}\times\vec{H}^*) \quad (3)$$

where $\vec{H}$ is magnetic field intensity.

2.2 Heat transfer modeling

Two heat transfer mechanisms are present in our model - convection and diffusion. For the Boussinesq approximation, the temperature equation is used

$$c_p\rho\frac{dT}{dt} = \lambda\Delta T + q \quad (4)$$

where c_p is the isobaric heat capacity, λ is thermal conductivity and q is the heat source due to MWs, which is given by equation 2.

2.3 Heat transfer in a fluid modeling

Due to the fluid motion, three contributions to the heat equation are included:

- The transport of fluid implies energy transport too, which appears in the heat equation as the convective contribution. Depending on the thermal properties on the fluid and on the flow regime, either the convective or the conductive heat transfer can dominate.
- The viscous effects of the fluid flow produce fluid heating. This term is often neglected, nevertheless, its contribution is noticeable for fast flow in viscous fluids.
- As soon as a fluid density is temperature-dependent, a pressure work term contributes to the heat equation.

Accounting for these contributions, in addition to conduction, results in the following transient heat equation for the temperature field in a fluid:

$$c_p\rho\frac{dT}{dt} + \rho c_p u\cdot\nabla T = \alpha_p T\left(\frac{\partial p}{\partial t} + u\cdot\nabla p\right) + \tau{:}S + \nabla\cdot(\mathrm{k}\nabla\mathrm{T}) + q \quad (5)$$

3. Simulation strategies with *COMSOL Multiphysics*®

3.1 Boundary Conditions

Impedance boundary conditions are imposed upon all sides of the cavity and the waveguides. The wave ports are located at the entrance of the waveguides (see Figure 1) and configured to propagate through them in the transverse electric 10 (TE_{10}) modes. Different wave equations were defined for the complete geometry and specifically for the cavity of the reactor (as shown in cross section of Figure 1, left), which will present time-dependent permittivities that were considered in the study. This will be explained in detail in the next section covering the simulation strategy employed.

3.2 Model setup

This simulation work was based around the idea of the multi-frequency heating of a reacting mixture. As is widely known, the permittivity of materials is a function of different parameters, including temperature and, more importantly in our case, frequency [8].

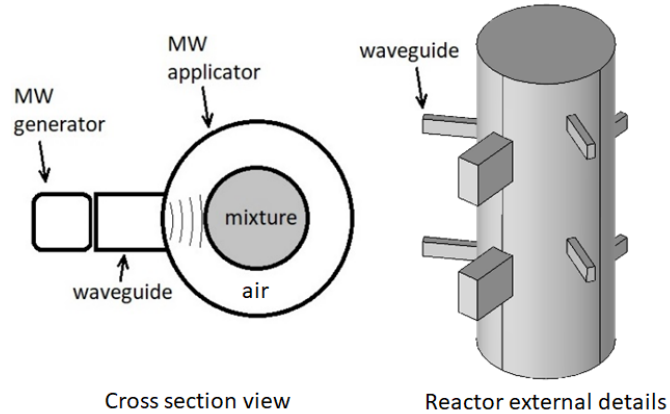

Figure 1. Schematic of the cross-section of the reactor (left). External details of the reactor (right). As shown, there are two types of waveguides, for the two different frequencies.

In the light of this fact, permittivity values were experimentally determined for the two frequencies of interest in the reactor (915 MHz and 2.45 GHz) and under different temperatures (25, 100 and 200 ºC). These values were introduced in the model using the local tables within the two interpolation nodes that were included under ‘global definitions.’ In this fashion, for every frequency two tables were introduced: one for the real part of the permittivity (dielectric constant) and the other for the imaginary part (dielectric loss factor).

Therefore, two different EM Waves physics were employed in the model builder, one per frequency. Under each physics node, two wave equations were defined. The first one, covering the whole component, is used to calculate the displacement field from the permittivity values of the materials assigned to the different domains of the component. The second one, encompassing only the domain of the reservoir, overrides the first one and makes use of user-specified permittivity values that reference the interpolation nodes to obtain the temperature-dependent permittivity values for each physics.

This strategy allows for the modelling of such a complex system, as the software is able to evaluate the time-dependent permittivity for each frequency present in the physics of the reactor.

Regarding the reservoir, the heating of a completely static substance was compared with a fluid moving in it. In order to achieve this while keeping computational costs to reasonable limits, a user-defined velocity field was employed, mimicking the behaviour that a mixer (stirrer effect) would impose on a fluid. Such field included the spinning of the fluid around the axis of the reservoir as well as vertical velocities. These promote longitudinal mixing, moving from the top to the bottom in the central part of the geometry and upwards close to the reservoir walls. In all cases, all velocities were kept null at the boundaries of the reservoir, imitating the behaviour of a no-slip boundary condition.

3.3 Mesh Independence Test

As introduced before, the geometry includes two different volumes composed of air and a lossy material, respectively, for which different permittivities are described as functions of temperature for each frequency. As a consequence of the different dielectric constants of the materials, the wavelength of the electromagnetic waves will be different in these media according to equation 6, where λ is the wavelength in the medium, c_0 is the speed of light in free space, f is the frequency of the EM wave and ε_r is the dielectric constant of said propagation medium [9].

$$\lambda = \frac{c_0}{f\sqrt{\varepsilon_r}} \tag{6}$$

Looking at equation 6 again, it is immediate to observe that the wavelength not only depends on the dielectric constant, but also on the frequency of the EM wave. In order to set an appropriate mesh, the most challenging values of the permittivity and frequency leading to the shortest wavelengths were used. In air, this expression yields a wavelength of 12.24 cm for the most restrictive case of 2.45 GHz; while for the dielectric material these corresponded to the same frequency at 200 °C, giving a wavelength of 5.75 cm. The former is resolved by using between 4 and 5 elements per wavelength without incurring further complications, while the latter poses a computational challenge due to the large size of the geometry and the high mesh density required in order to solve the problem effectively. Consequently, a mesh independence test was carried out for a varying number of elements in the domain correspondent to the lossy medium, ranging from the minimum of 2 elements per wavelength necessary to solve the problem and up to 3.75, corresponding to the maximum number of elements that our equipment can solve while avoiding memory errors. The CPU time to solve the electromagnetic waves was of 49 minutes on an Intel® Xeon® CPU ES-2630 v4 at 2.20 GHz with 64 GB of RAM, running on Windows® Server R2 2012, 64 bits. Figure 2 shows how the electric field norm values at a specific point of the geometry in the reservoir converge from the third solution (corresponding to 3 elements per wavelength at 2.45 GHz for a total of 762473 mesh elements in the model) on, as the relative error between this case and the finest mesh able to solve the problem without memory issues amounts to solely 1.25%.

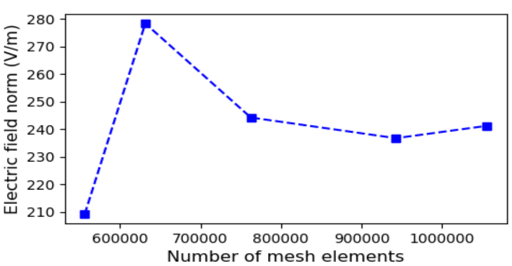

Figure 2. Grid independence plot.

4. Simulation Results

Regarding the RF simulations, different configurations in terms of frequency have been simulated in order to select the most convenient.
On the one hand, different phenomena can be observed when analysis was carried out for the longitudinal sections in Figure 3.

1- Simulations at 915 MHz: A higher penetration depth of the energy into the material is produced inside the vessel to activate and to heat the central area. In addition, generated electric field is localized just over the areas of influence close to MW generators.
2- Simulations at 2.45 GHz: A more homogeneous application of the electric field is achieved and therefore, the heat transfer is expected to be more efficient.
3- Combining both frequencies, the reactor achieves a more homogeneous distribution along the complete vessel, both inner areas and outer areas. In addition, the electric field values are increased.

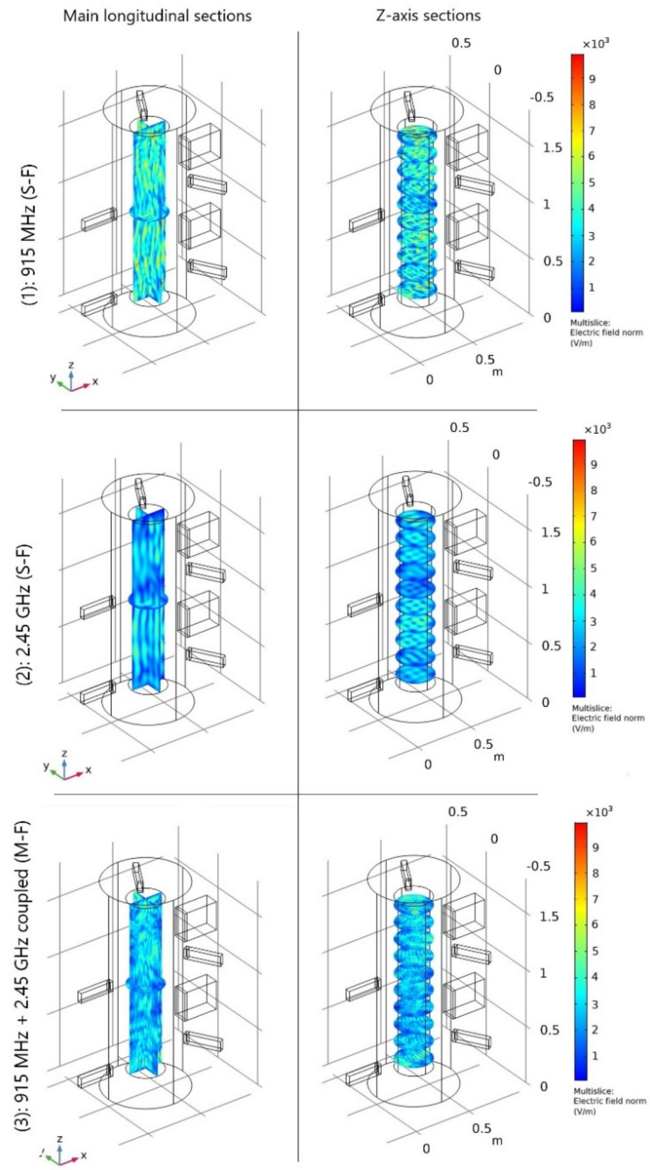

Figure 3. Electric field norm (V/m) comparative by using single frequencies (915MHz or 2.45GHz) or coupled (915MHz +2.45GHz). Longitudinal sections and Z-axis sections (model dimensions in m).

On the other hand, considering the generation of hot and cold points, different results (cases) can be observed in Figure 4.

1- 915 MHz simulations: Specific hot points are found whereas, the central part is not absorbing energy from the MW source.
2- 2.45 GHz simulations: Penetration depth of the wave is lower than the obtained for 915 MHz simulations, but hot points are not observed.

3- By combining frequencies, the slice shown (3) increases nominal values of electric field norm and the hot spots that can be observed have a lower gradient with respect to nearby areas.

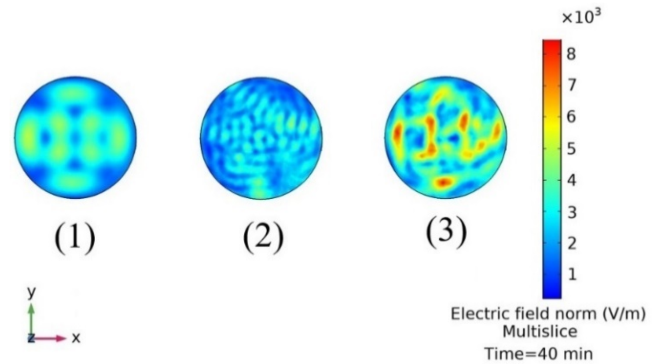

Figure 4. Comparison of electric field norm (V/m) at Z-axis z=0,5m, in the three cases: (1) 915MHz, (2) 2.45 GHz and combined cases (3) 915MHz + 2.45 GHz.

Following the electromagnetic waves study, a time dependent one was carried out for the heat transfer physics, contemplating two different scenarios. For the first one, a static material is considered, where the spatial distribution of the electromagnetic losses leads to a highly uneven temperature distribution with scattered hotspots that does not ensure uniformity of processing conditions in the batch (Figure 5, left). For the second scenario, the dielectric properties were kept the same, whereas the content of the reservoir is modeled as a fluid moving (see Section 3.1 of this paper), leading to an almost homogeneous temperature field in the vessel (Figure 5, right), highlighting the dramatic effect in temperature distribution that the inclusion of a mixer has in the MW heating of fluids (movement over the fluid is applied).

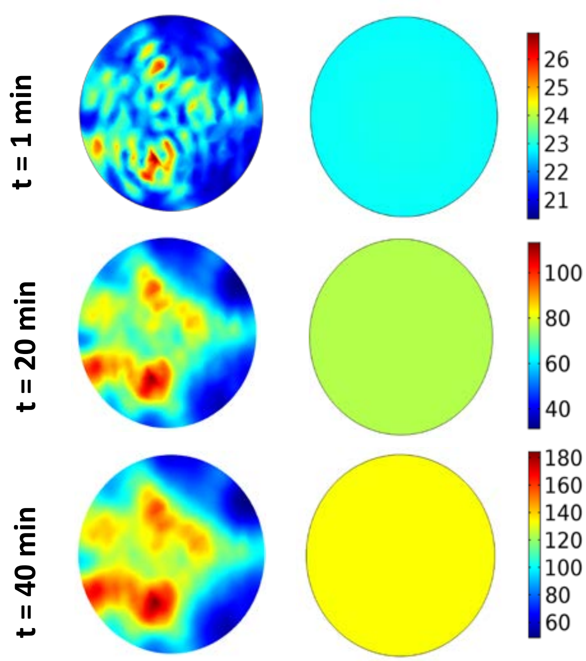

Figure 5. Comparison between the different temperature (ºC) distributions in a cross section at times = 1, 20 and 40 min for a model considering a static material subject to heating (left column) and a fluid moving in the reservoir (right column).

5. Conclusions

A complex physical system was reduced to two different mathematical models, taking advantage in design stage and process configuration by using *COMSOL Multiphysics®*. The result will allow the development of a new microwave system focused on a novel industrial process.

A complete simulation methodology for depolymerization of PA by means of microwave heating simulation has been presented. Two microwave frequencies and a coupled multifrequency models were used in order to obtain the simulation results in accordance with experimental measurements (performed studies under polynSPIRE project protected under confidential document).

Then, RF simulations have been performed to analyse the thermal behaviour of the mixing fluid under different MW configuration. The results presented have revealed that combining MW frequencies is expected to be the most promising strategy to improve the homogenization of the electromagnetic field distribution achieving a more homogeneous temperature, minimizing hotspots presence. This conclusion has been extracted after performing a comprehensive sensitivity study analysis varying the frequency, number of ports and position. In addition, MW also allows a significant reduction of heating time leading to an increase of the overall efficiency of the process.

Future efforts will address the total coupling between heating and chemical process with the aim of simulating the interaction of the MW heating with the chemical reactions.

The model generated was proven to be a useful tool for reactor design and optimisation, being able to capture temperature changes in the reacting mixture due to the electromagnetic heating induced

by the microwaves. Additionally, the framework developed for this model can be easily modified to predict the performance of other microwave-assisted reactions.

6. References

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