# SELECTIVE HEATING APPLICATIONS FOR THE PROCESSING OF POLYMER-POLYMER MATERIALS

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## Abstract

*Polymer-Polymer composites, or self-reinforced materials, imply the combination of a reinforcing thermoplastic fibre and a supporting thermoplastic matrix from the same polymer family. The involved project, named ESPRIT, try to develop methodologies to selectively heat the polymeric matrix whilst not heating the reinforcement. The target is to achieve a state of mouldability as fast as possible with minimum impact on the morphology of the fibre. For this reason the application of electromagnetic energy, absorbed by susceptors distributed only in the polymeric matrix, can heat quickly the matrix minimizing reinforcing fibre damage. This work focuses its efforts in the study of the effects of susceptors dispersed in polymeric matrices, under microwave and induction radiation. The efficiency of the heating methods has been addressed and possible applications explored.*

## 1 Introduction.

Microwave technology provides an alternative to conventional heating methodologies suitable for some widely home applications, cooking, chemical synthesis, treatment of gas effluents, curing of thermoset and rubber materials, adhesives [1], ovens for ceramic manufacture. Basically provides an alternative methodology to conventional heating, with several advantages like: penetrating radiation, controllable electric field distribution, rapid heating selective heating of materials, and self-limiting reactions. Two major effects are responsible for the heating which results from this interaction: dipolar polarization and conduction [2], [3]. Microwave heating present important disadvantages, mainly related to the lack of uniform and selective heating over a large volume and the transparency of most of the materials to microwaves.

The microwaves do not interact with the majority of polymeric materials due the lack of dipolar moment. For this reason, additives as heating susceptors can be used to prepare materials able to absorb microwaves. These additives are conductive, or have dielectric properties significantly different from the polymeric matrix. The electric field will modify the polymer environment and heating profiles will be different from the base polymer. Depending on the nature of the susceptor the material answer to the microwaves can be due to one specific mechanism or combination of several of them [4].

The literature has tested the effectiveness of different microwave susceptor taking into account the size, shape, concentration, electrical resistivity and the distribution and dispersion in the polymeric matrix. The most widely used and effective susceptors are carbon structures. Other suitable particles as silicium carbide, titanium dioxide, metal flakes, zinc oxide or talc have been tried [5].

The electromagnetic induction is based on Lenz's law and the principles found by Michael Faraday [6]. The heating phenomena of ferromagnetic and conductive materials, which appear when such materials are exposed to an alternating electromagnetic field, are based on eddy currents and magnetic polarization effects (hysteresis).

Induction heating of polymeric materials are commonly used for welding and heating applications. Induction welding of reinforced polymers has been developed as a fast and local joining technique for thermoplastic materials. Usually carbon fibers or additional metallic grids are used as susceptors for the electromagnetic field. In the case of carbon fibers, the so called heating or welding promoters have not only heating functions but often serve as reinforcement for the material. If the joining partners do not contain any conductive material, a metallic grid is used as a susceptor, which is placed within the fusion zone [7]. The AC magnetic field heats the susceptor, from which the heat is consequently transferred to the surrounding region, which begins to melt. Most applications do not take the use of particulate heating promoters intoconsideration. Suwanwatana et al. have begun to develop a material system which contains magnetic particles for the bonding of thermoplastic components by inductive means [8, 9]. They achieved a very good bonding strength by using a polysulfone film doped with nickel particles. The particle loading was chosen between 10 and 20 vol.-% with varying particle sizes in the nano and micro range. The best heating results using an AC magnetic field with 2.25 MHz were obtained from smaller particles (79 nm) [8].

## 2 Materials and testing methods.

Both heating methodologies, microwave and induction, were applied to study the susceptor effect of different additives in different polymeric materials. The final objective is to prepare self-reinforced polymeric matrices able to be selectively heated, minimizing the fibre damage (see figure 1).

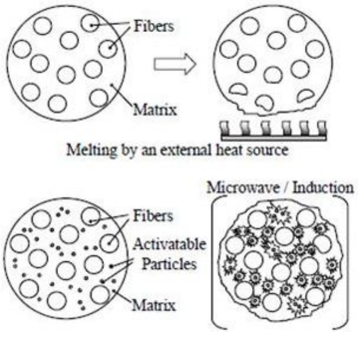

**Figure 1.** Selectively heating methodology of self-reinforced polymers based on electromagnetic susceptors.

For this purpose, different equipments for microwave and induction heating techniques were selected. In detail, for microwave studies it was selected a closed oven with the following dimensions: (320x340x215) mm. The magnetron had a maximum power of 2000W and worked at 2.45GHz of frequency. The oven was adapted with an infrared camera ThermaCAM P640 to measure the temperature into the chamber. For induction trials it was used a source the Trumpf Hüttinger Truheat MF and Trumpf Hüttinger IG 10/2000 devices (500-2500 kHz) and a spiral coil with 100 mm of diameter and internal cooling system.

For the preparation of composite materials based on polymeric matrices and microwave and induction susceptors, co-rotative twin screw extruder W&P model ZSK25 has been used.

The materials selected as polymeric matrices have been a high density polyethylene (HDPE) INEOS Rigidex HD6070EA and polyamide 6 Dynalon B3S25. The table 1 shows the different susceptors studied for microwave and induction heating processes.

| Name | Brand Name | Manufacturer |
|---|---|---|
| Carbon black (CB) | Raven L Ultra | Columbian Chemicals Company |
| Cast iron powder | FG 0000/0300 | Gotthart Maier Metallpulver GmbH |
| Magnetite powder ($Fe_3O_4$) | FO 0000/0090/HA | Gotthart Maier Metallpulver GmbH |
| Carbon nano tubes (CNT) | NC 7000 | Nanocyl S.A. |
| Lead zirconium titanate (PZT) | PCM55 | Noliac |
| Nickel | Nickel 99 % | TLS-Technik GmbH & Co Spezialpulver KG |
| Ilmenite | 90W AU | Mineralmühle Leun |
| SiC | No.357391 | Sigma & Aldrich |
| $TiO_2$ | Tioxide TR-92 | Hunstsman |

**Table 1.** Susceptor additives used for microwave and induction heating processes.

## 3 Results and discussion.

### *3.1. Microwave process.*

Based on multimode oven technology it has been studied the susceptor efficency of the different polymeric systems with promoter additive. Microwave power and irradiation time were fixed to calculate the slope between temperature rise and susceptor loading (%).This parameter is related with the susceptor efficency in microwave heating process [10]. The effectiveness between carbonous susceptor and other susceptors showed strong differences. Suitable heating conditions for carbon nanotubes and carbon black additives were fixed at 400W of power and 20 seconds of exposure time. Those experimental conditions were too low for non carbonous particles due the lack of microwave absorbance. It was necessary to increase the power up to 1000W and 120s to measurable heating process. Table 2 shows the heating efficiency calculated for 1000W of power and 120s of irradiation.

| **Matrix System** | **Heating Efficiency (°C)/%) (2 min)** |
|---|---|
| HDPE/CB | 10.9 |
| HDPE/$Fe_3O_4$ | 0.85 |
| HDPE/PZT | 0.29 |
| HDPE/SiC | 0.77 |
| PA6/CB | 0.67 |
| PA6/$Fe_3O_4$ | 3.23 |
| PA6/PZT | 0.04 |
| PA6/SiC | 2.01 |

**Table 2.** Heating Efficiency calculated for compounded matrix systems.

As it was expected, the non-carbon based susceptors showed low heating efficiencies, whereas carbon black was the responsible of a fast heating process. In fact, as it has been previously discussed, the used 1000W power was too high for the HDPE/CB compounds, which was revealed by material burning and generation of hot spots. In the case of polyamide 6 we have observed an extreme low heating efficiency for carbon black, compared with other polymeric matrices. In detail, the microwave susceptor effect of carbon black can be seen in the figure 2.

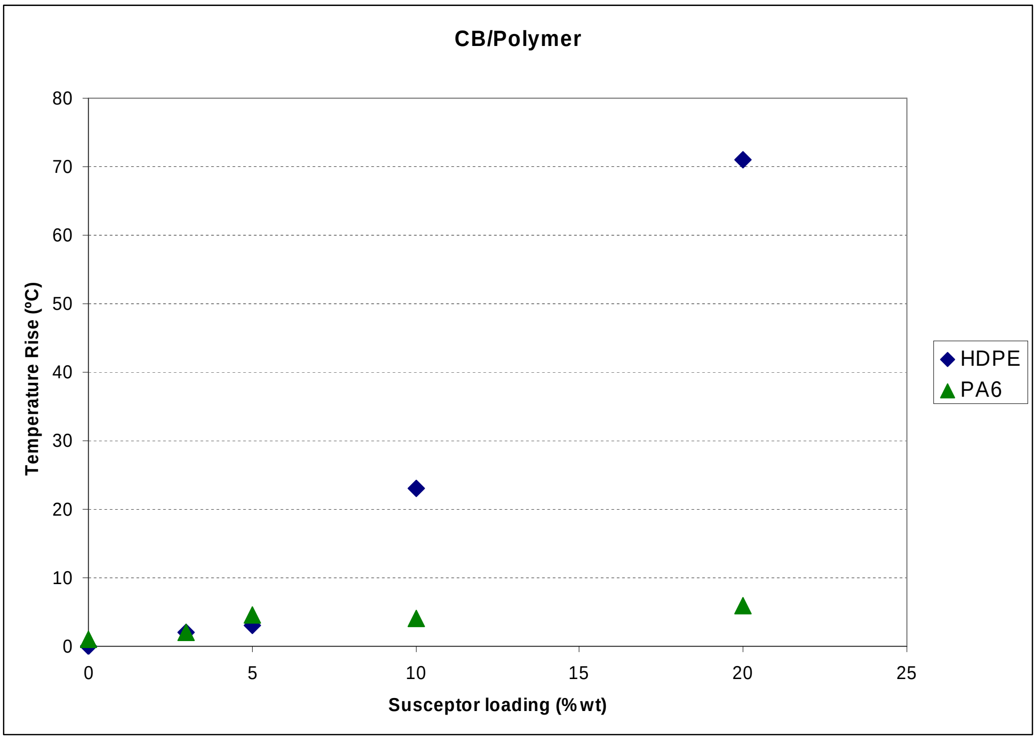

**Figure 2.** Temperature Rise of CB/polymer systems at 400W of power and 20s.

Other carbonous particles can be studied as heating promoters, attending the dependence of heating efficiency with active surface (particle size). In this way, important improvements have been found.

### *3.2. Induction process.*

The comparison of filler degree influence on the heating rate shows an identical behavior for different particle materials. All obtained curves follow an exponential relation (Fig. 2). The highest heating rates were observed with the ferromagnetic material, from the highest, cast iron powder and magnetite, to the lowest, which was nickel in the experimental series. The electrically conductive carbon black reveiled an insufficient heating rate for the use as an induction heating promoter. This behavior was also observed with 1 wt-% carbon nano tube doped material. The domination of ferromagnetic properties on the particulate induction heating behavior, which has been previously stated by other research groups, was hereby confirmed [8,11]. Due to this effect, the iron particles, which possess higher magnetic properties, showed a significant higher linear heating rate than materials with lower magnetic properties. Additionally, the incorporation of more particles into the matrix consequently led to higher heating rates. Nevertheless, the amount of high susceptor fractions was counterproductive to be used with a light-weight material since the density of the compound was increased by the high-density ferromagnetic susceptors. Due to this, a susceptor fraction of 5 to 10 wt.-% was set as maximum economically feasible limit.

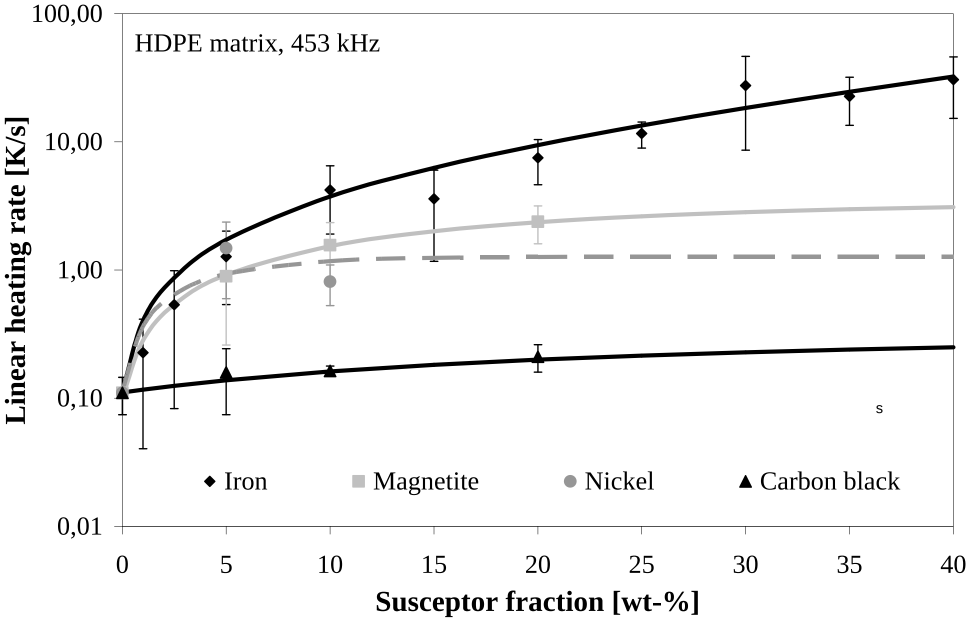

**Figure 3.** Linear heating rate of inductively heated HDPE in relation to filler degree and susceptor material

By regarding the influence of the magnetic field strength and in this case the related current provided by the generator, a quadratic influence on the heating rate was expected [12]. This could be verified for all tested cast iron sample series with high accordance (Fig. 3). At currents higher than 20 A, the heating rate of 20 wt-% iron doped HDPE lies approximately four times higher than the one of low doped materials. By reducing the generator current to 10 A and lower, the linear heat rate drops for all cases close to zero. Consequently, the effect of the filler fraction is negligible in this case. Thus, the transferred energy at low currents is so small that it is hardly enough to compensate the heat loss to the surrounding environment.

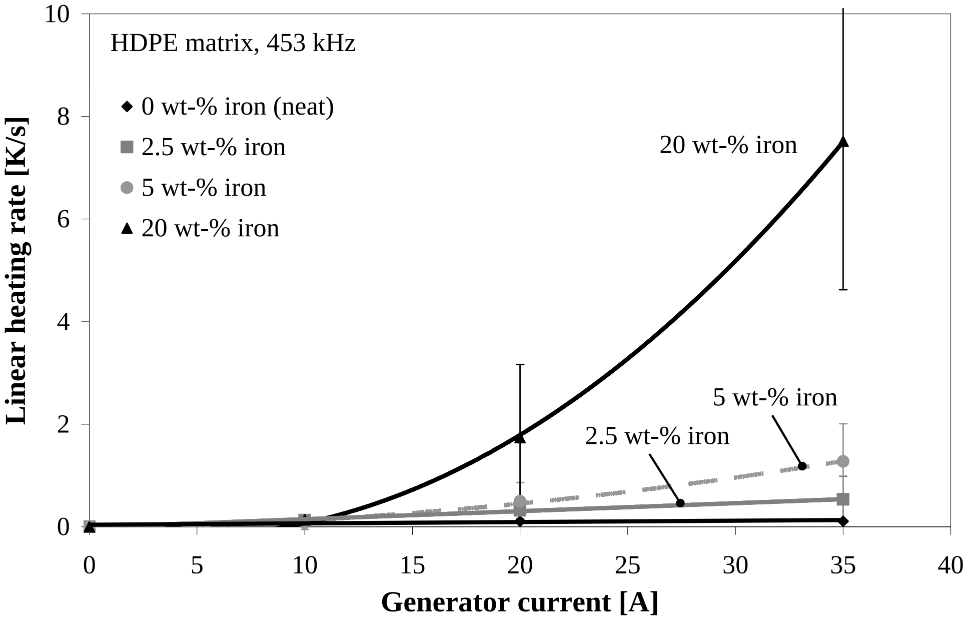

**Figure 4.** Linear heating rate of inductively heated HDPE in relation to generating current of electromagnetic field

An expected exponential relationship between linear heating rate and coupling distance, which describes the distance between sample and coil, was also confirmed (Fig. 4). It was proven that at increasing distance, the difference between different fillers decreases. At 12 mm coupling distance, the filler fraction of magnetite in the HDPE sample (5 wt-% vs. 10 wt-%) had only a subordinate influence on the measured heating rate.

For an industrial application, the effect of coupling distance should be precisely regarded according to the magnetic field distribution. For this study, the minimum coupling distance was 2 mm and mainly limited due to lack of space when mounting the samples on the coil.

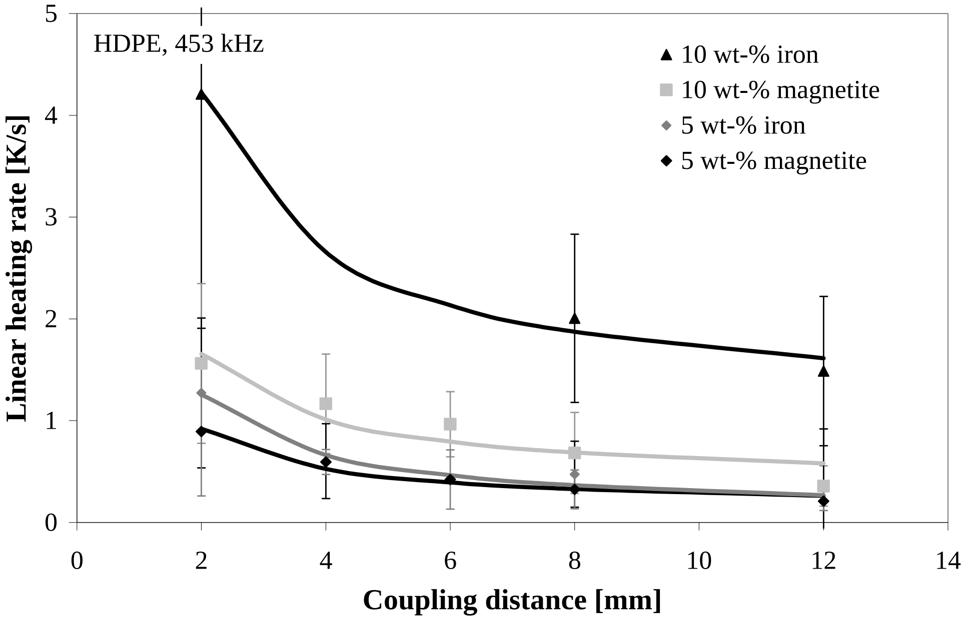

**Figure 5.** Reciprocal decrease of linear heating rate due to increased distance between coil and sample

As a result from the preliminary test series, the maximum generator current at 450 kHz was used for the study with ferromagnetic particle doped composite material. The coupling distance remained at its minimum of 2 mm.

As polymer-polymer composite, a polypropylene (PP) material with polyester (PET) fibers was used, which provided a high melting temperature gap between matrix and reinforcement. Consequently, the heating process was easily controllable. As heating promoter, cast iron powder, which showed the highest heating rates, was applied. Two filler fractions were tested: 5 wt-% in the matrix, which is regarded as the minimum content for an industrial process, and 20 wt-%, which provided a linear heating rate close to 10 K/s.

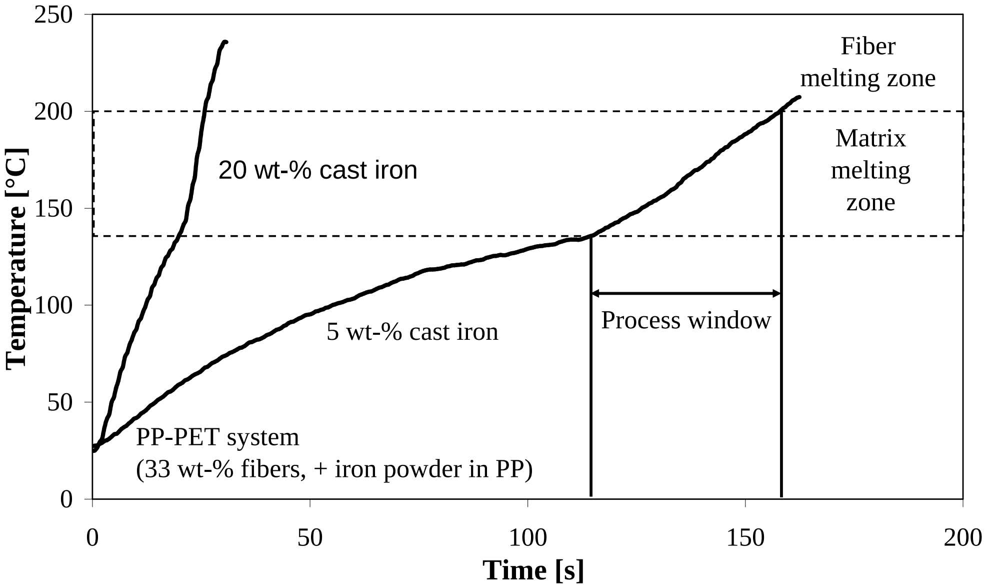

**Figure 6.** Application of particulate induction heating for the heating of polymer-polymer material

The obtained results from the heating with a polymer-polymer composite demonstrated a very quick heating of the highly doped material, which exceeded the melting temperature of the reinforcement already after only 25 seconds (Fig. 6). Although the melting temperature gap was approx. 80 °C, the quick heating led to a complete exceeding of the process window, which is characterized by a melted matrix and an intact reinforcement. Consequently, the reinforcement was melted in this case. Generally, the process window is dependent on the polymer-polymer system and shrinks with decreasing melting temperature of reinforcement fibers. For 20 wt-% iron in a PP-PET compound, it was exceeded within five seconds. The lower doped compound reached the process window later, but needed longer to cross it, which provided more safety time before reaching the melting temperature of the reinforcement. High fiber integrity of the reinforcing fibers was detected, when the process was stopped within the process window. By passing the process window, the fibers were damaged, which led in the extreme case to a burning-in of a hole at the sample exposed to the highest magnetic field density.

In order to optimize the heating process and reduce the danger of melting reinforcement, the use of an external pyrometer, which controls the generator power, was examined. This led to a stable temperature of the compound in the process window even at higher filler degrees. Alternatively, high filler degrees could be activated by a lower magnetic field strength as shown before, but this solution was regarded as counterproductive for economic and technical reasons.

## 4 Summary

It has been observed that using specific additives as susceptors can change the heating behavior of polymeric materials. The carbonous substances are the most effective for microwave heating, due to the high electrical conductivity of the particles. For induction heating purposes the system has been optimized to achieve the maximum heating effect and the minimum damage to the material, mainly using iron particles. Selective heating materials can be useful for different applications. The initial purpose of this work has been the modification of compression and injection moulding processes, adding microwave or/and induction devices and tools. Currently we are working on the preparation of self-reinforced polymers, selectively melting the polymeric matrix, and minimizing the damage of the reinforcing fibres. Another possible application of this selective heating is the development of new and more effective routes to polymer welding.

## 5 Acknowledgement

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