# HEATING OF POLYMER-POLYMER COMPOSITES BY INDUCTIVE MEANS

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**Keywords**: *Inductive heating, ferromagnetic particle, HDPE, selective melting*

## 1 Introduction

Polymer-polymer composites consist of a polymer matrix and polymer reinforcement. When they are manufactured exclusively of components from the same polymer family, they are called self-reinforced polymers (SRP). Polymer-polymer composites offer the possibility of designing extremely light-weight components with excellent impact behavior and increased tensile properties [1]. Additionally, SRP offer increased sustainability potential, due to the application of similar molecular structures, which do not have to be disposed separately.

The processing of a SRP semi-finished material is challenging, since a too high process temperature may damage and possibly destroy the polymer reinforcement, even when matrix and fiber do not melt at the same temperature. Nevertheless, recent industrially realized approaches accept fiber melting during processing to a certain extent, which unintentionally weakens the nominal strength potential of the material.

A new approach for heating polymer-polymer composites by inductive means is presented in this paper. The induction based melting of non-polymer fiber reinforced thermoplastic composite materials has recently been enhanced in the field of welding applications [2,3]. The electrical properties of the reinforcement are mainly used in this case for matrix melting.

Since SRP consist exclusively of non-conductive polymers, which do not offer the possibility to be heated by an alternating magnetic field on their own, ferromagnetic heating promoters are added. The promoters have a particulate shape and are exclusively used in the matrix phase to limit the heating of the reinforcement and avoid its melting.

Particulate heating of polymers has been recently developed for polysulfone tape welding applications [4-6]. It was found, that nickel, as a promoter material, showed a dependency on particle size and oxidation state as well as on frequency, and magnetic field strength. The presented study proceeds with this approach and adapts it to SRP material, which demands accurate temperature control.

## 2 Experimental

For induction tests, particulate doped high density polyethylene (HDPE) sheets with the dimensions of 60 x 60 x 2 mm³ were exposed to a magnetic field in a distance of 2 mm to the coil with a medium frequency of 450 kHz. The field was provided by a pancake coil and driven by a Hüttinger TruHeat 5010 MF (Trumpf Hüttinger, Germany) generator. HDPE was chosen due to its low melting temperature and served as the matrix material for the later manufactured self-reinforced polyolefin SRP with polypropylene fibers.

Amongst the incorporated heating promoters, which have been added individually in various fractions, ferromagnetic (cast iron, magnetite, and nickel) and electrically conductive materials (carbon black, carbon nano tubes) were tested. A detailed description of the experimental work has been published elsewhere [7].

The obtained temperature profile was analyzed and compared according to the linear heating rate of each promoter material. The linear heating rate is characterized by its linear rise from the experimental start to the onset of crystallite melting. A typical temperature evolution of a particle doped sample is illustrated in Fig 1. The temperature rise decelerates in the region close to the melting temperature because the energy is consumed to melt the crystallites instead of further heating the sample. In the literature, this energy is referred to as latent heat

and is commonly described within a heat capacity graph, which is unique for each material. The temperature curve accelerates again when most crystallites have become molten.
Regarding the linear heating rate, the influence of particle material, generator power, and coupling distance has been examined with neat HDPE material in an experimental series for the later use with polymer-polymer material. The influence of other parameters, like frequency and semi-finished material shape, has been already investigated in a preliminary test series [8].
The matrix degradation, which is related to a shift of the melting temperature due to molecular chain changes, has been investigated by means of differential scanning calorimetry (DSC). Therefore, the melting temperature change of thermally damaged infrared radiated material was compared to induction heated material.

## 3 Results and Discussion

### 3.1 Induction with Neat Material

The comparison of filler degree influence on the heating rate shows an identical behavior for different particle materials. It was found that all curves follow an exponential relation (Fig. 2). The highest heating rates were observed in the ferromagnetic material, from the highest, cast iron powder and magnetite, to the lowest in this case, nickel. The electrically conductive carbon black shows an insufficient heating rate for the use as a SRP heating promoter. This behavior was also found for 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 [5,9]. Due to this effect, the iron particles, which have higher magnetic properties, show a significant higher linear heating rate than materials with lower magnetic properties. Additionally, the incorporation of more particles into the matrix consequently leads to higher heating rates. In this context, even with filler fractions as low as 5 wt-%, heating rates of 1 K/s can be reached.
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 [10]. This could be verified for all tested 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. The provided energy to the particles by the electromagnetic field at low currents is so small that it is hardly enough to compensate the heat loss to the surrounding environment.
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 could be shown 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-%) has only a subordinate influence on the 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.

### 3.2 Induction with Polymer-Polymer Material

As a result from the preliminary test series, the maximum generator current at 450 kHz was used for the study with particle doped SRP 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 provides a high melting temperature gap between matrix and reinforcement. Consequently, the heating process is easily controllable. As heating promoter, 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 provides a linear heating rate close to 10 K/s.
The obtained results show a very quick heating of the highly doped material, which exceeds the melting temperature of the reinforcement already after only 25 seconds (Fig. 5). Although the melting temperature gap is approx. 80 °C, the quick heating

leads to a crossing of the process window, which is characterized by a melted matrix and an intact reinforcement. 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 is crossed within five seconds. The lower doped compound reaches the process window later, but needs longer to cross it, which provides more safety time before reaching the melting temperature of the reinforcement. It was found that the reinforcing fibers survive the heat treatment, when the process is stopped within the process window. When passing the process window, the fibers are damaged, which lead 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 has been examined, which controls the generator power. This leads to a stable temperature of the compound even at high filler degrees. Alternatively, high filler degrees could be activated by a lower magnetic field strength as shown before, but this is counterproductive for evident reasons.

### 3.3 Degradation Study

The influence of the intrinsic particulate induction heating on the matrix performance and degradation is important for the evaluation of the hot spot difficulty. Since the particles can be regarded as single heat sources, the surrounding matrix is assumed to be subjected to extreme heat gradients. Consequently, the appearance of matrix degradation has to be evaluated.

Extrinsic heating methods, like infrared radiation on a surface, provide the highest power input directly to the surface-close region. Degradation artifacts can be observed easily by optical means, in best case in high degradation state with the naked eye. In comparison to that the use of optical means on the surface for intrinsic heating methods is useless, since the heat sources and the potential damages are distributed within the sample.

This study introduces an indirect approach for estimating the degradation state of an induction heated material. It is known from literature, that the degradation state is linked to morphological changes. Changes of the polymer chain length may also influence the melting temperature [11]. Consequently, the degradation analysis was performed by a comparative study of the melting temperature change of infrared radiated and induction heated material (Fig. 6). The infrared radiated samples, which were heated by a 1000 W infrared spot, were used as reference because the degradation state was directly observable on the surface by a color shift.

The melting temperature of infrared radiated samples decreases quickly with increasing radiation time. The used HDPE material is severely charred and optically damaged after 120 seconds. The melting temperature was reduced by approx. 8 °C in comparison to the initial state.

In contrast, the melting temperature of induction heated material increases up to 60 s. This is explained by the fact that the sample is only in a beginning melting state at a temperature of 100 °C, which is reached after 60 seconds. This leads to a further cross-linking of polymer chains and an increasing melting temperature [cp. 12,13]. With increasing induction heating time, the melting temperature decreases in a moderate way. It drops below its initial melting temperature when passing the 150 second mark and is further reduced with increasing radiation time. After five minutes the melting temperature has reduced by approx. 1.5 °C in comparison to its initial state.

Although the results convey the impression that induction heating causes degradation, it is regarded as low for the examined heating cycles. This implies that the occurrence of hot spots caused by induction heating is not problematic for the material.

## 4 Summary

The parameter influence of suitable heating promoters for the induction heating of polymer-polymer was examined. The heating ability of particulate heating promoters is dependent on various factors, including promoter material, magnetic field frequency, magnetic field strength, and coupling distance. The heating performance is independent from the matrix material. Matrix melting could be achieved within a time frame of 90 seconds and less. The presence of the reinforcement after inductive heating was verified. It was shown that a suitable process window exists, which

guarantees the processability of the material without reaching a reinforcement melting temperature and the associated fiber damage.
The detection of the degradation state of particulate induction heated materials has been conducted by an analysis of the melting temperature change during heating. The method revealed hardly any detectable degradation of inductively heated material in comparison to infrared heated samples within the same test duration.
In future studies, the heating method will be tested with other SRP material combinations, before a suitable process setup will be developed for an industrial application.


## Acknowledgement

The research leading to these results has received funding from the European Community's Seventh Framework Programme NMP-2007-2.4.1 under grant agreement 214355 (acronym: ESPRIT). The kind help of Dr. Miro Duhovic is gratefully acknowledged.


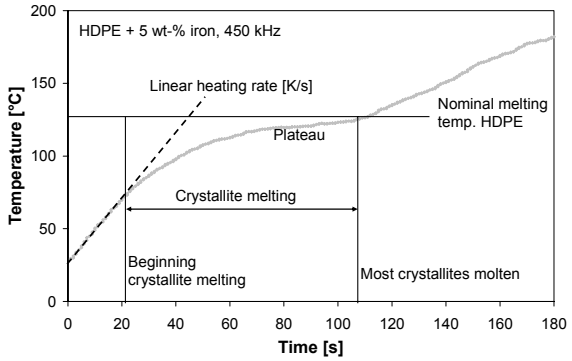


Fig. 1. Exemplary temperature evolution of a particle doped HDPE sample

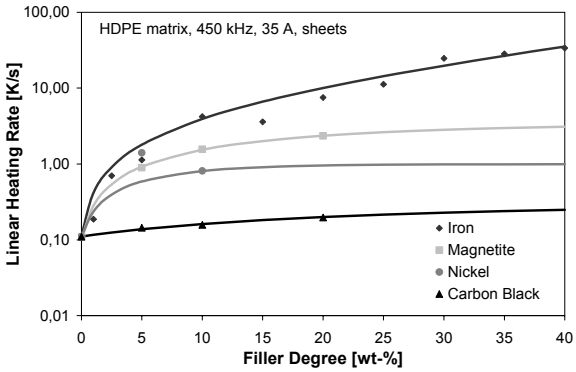


Fig. 2. Influence of materials and filler degrees on the linear heating rate of particle doped HDPE

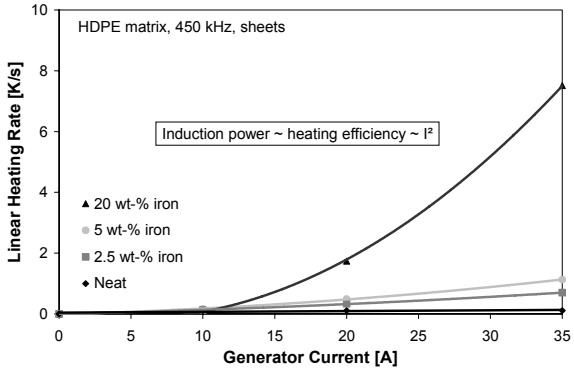


Fig. 3. Linear heating rate as function of generator power for iron particle doped HDPE samples

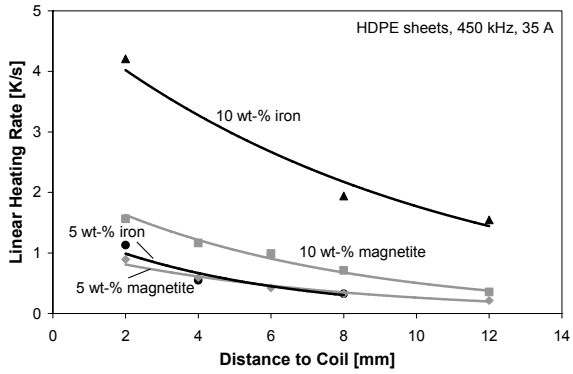


Fig. 4. Influence of coupling distance on linear heat rate for ferromagnetic promoters

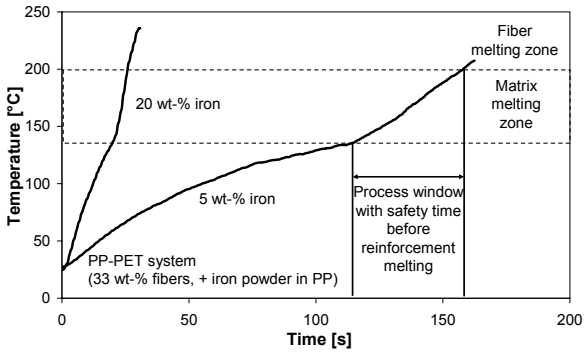


Fig. 5. Melting of polymer-polymer composite by particulate induction heating

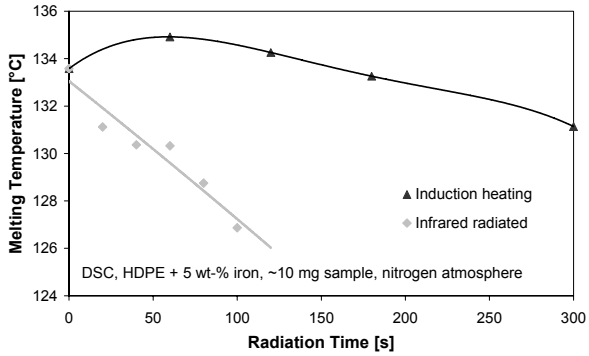


Fig. 6. Melting temperature reduction of HDPE during electromagnetic radiation

## References


[1] A. Kmetty, T. Bárány and J. Karger-Kocsis "Self-reinforced polymeric materials: A review". *Prog. Polym. Sci.*, Vol. 35, pp 1288-1310, 2010.

[2] L. Moser, P. Mitschang, A. K. Schlarb, "Induction welding of thermoplastic polymer composites using robotic techniques". *Sampe J.*, Vol. 44, No. 5, pp 43-49, 2008.

[3] B. K. Fink, S. H. McKnight, J. W. Gillespie Jr., S. Yarlagadda, "Ferromagnetic nano-particulate and conductive mesh susceptors for induction-based repair of composites". *Proceedings of Army Science Conference*, Norfolk, VA, USA, 1998.

[4] W. Suwanwatana, S. Yarlagadda, J. W. Gillespie Jr., An investigation of oxidation effects on hysteresis heating of nickel particles. *J. Mater. Sci.*, Vol. 38, pp 565–573, 2003.

[5] W. Suwanwatana, S. Yarlagadda, J. W. Gillespie Jr., "Influence of particle size on hysteresis heating of nickel particulate polymer films". *Compos. Sci. Technol.*, Vol. 66, pp 2825–2836, 2006.

[6] W. Suwanwatana, S. Yarlagadda, J. W. Gillespie Jr., "Hysteresis heating based induction bonding of thermoplastic composites." *Compos. Sci. Technol.*, Vol. 66, pp 1713−1723, 2006.

[7] T. Bayerl, A. B. Borrás, J. I. Andrés Gallego, B. Galindo Galiana, P. Mitschang, "Melting of polymer-polymer composites by particulate heating promoters and electromagnetic radiation", In: D. Bhattacharyya, S. Fakirov (Eds.), *Synthetic Polymer-Polymer Composites*, Hanser, in press, 2011.

[8] T. Bayerl, R. Schledjewski, P. Mitschang, "Induction heating of thermoplastic materials by particulate heating promoters", *Polym. Polym. Compos.,* submitted for publication , 2011.

[9] S. Dutz, R. Hergt, J. Mürbe, R. Müller, M. Zeisberger, W. Andrä, "Hysteresis losses of magnetic nanoparticle powders in the single domain size range". *J. Magn. Magn. Mater.*, Vol. 308, pp 305–312, 2007.

[10] V. Rudnev, D. Loveless, R. Cook, M. Black (eds.), *Handbook of Induction Heating*, Marcel Dekker, New York, 2003.

[11] C. M. Chan, S. Venkatraman, "Crosslinking of poly(arylene ether ketone)s. 1. Rheological behavior of the melt and mechanical properties of cured resin." *J. Appl. Polym. Sci.*, Vol. 32, pp 5933–5943, 1986.

[12] A. Holmström, E. M. Sörvik, "Thermooxidative degradation of polyethylene. I and II. Structural changes occurring in low-density polyethylene, high-density polyethylene, and tetrateracontane heated in air". *J. Polym Science, Polym. Chem.*, Vol. 16, pp 2555–2586, 1978.

[13] A. Holmström,: The course of thermooxidative degradation of LD- and HD-polyethylene under accelerated testing conditions. In: R. K. Eby, (Ed.):

*Durability of Macromolecular Materials*, American Chemical Society, Washington, pp 45–62, 1979.