Laser transmission welding of absorber-free semi-crystalline polypropylene by using a quasi-simultaneous irradiation strategy

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Abstract

Unlike other joining techniques, laser transmission welding offers unique advantages such as selective and contactless energy deposition. This enables the fabrication of flexible seam geometries at low mechanical and thermal stresses. However, the use of absorbing additives for the lower joining partner such as carbon black is crucial as most polymers are transparent in the spectral range of typical beam sources (800–1100 nm). A novel approach is the application of beam sources emitting radiation within the polymeric intrinsic absorption bands between 1500 and 2000 nm. This enables absorber-free laser welding of transparent polymers for medical or microfluidic applications such as Lab-on-a-Chip devices. The main drawback on the other hand is the large heat affected zone (HAZ) due to the volume absorption which is extending over the entire cross section. A possible way to overcome this disadvantage is a quasi-simultaneous irradiation strategy. It could be proved in the past that the HAZ of polycarbonate (PC) can be reduced in the vertical direction by up to 30% compared with contour welding. Since the effects of light scattering on the absorber-free quasi-simultaneous irradiation strategy are still unknown, the beam propagation was simulated in polypropylene (PP). Based on the results, a thermal simulation of the welding process was carried out using the finite element method (FEM). The simulation model was then evaluated by comparing the results with experimental trials.

Keywords Polymer welding · Laser · Semi-crystalline polymers

1 Introduction

Since its first industrial introduction in the 1990s, laser transmission welding of plastics has been successfully established in various application areas. Unlike other joining techniques,

this method offers the advantage of a non-contact and highly precise energy deposition. This enables the fabrication of complex and narrow seam geometries which is especially favourable for the encapsulation of sensitive components in the automotive, medical and electronic industry. As most

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Fig. 1 Absorption curves of PP, PPSU and PC (left) and vertical intensity progression in classic and absorber-free laser transmission welding (right) [3]

polymers are transparent in the wavelength range of traditional laser sources at $\lambda = 800\text{--}1100\text{ nm}$ (Fig. 1, left), a modification of the optical properties is crucial in order to melt the joining partners [1, 2].

In general, the joining partners are welded as an overlap joint. A clamping device provides an exact positioning of the joining partners as well as a thermal contact. The lower joining partner is doped with an absorber such as carbon black which leads to a black colouring. The upper joining partner on the other hand is transparent to the laser radiation. During the welding process, the laser beam is being transmitted by the upper transparent joining partner and then absorbed at the surface of the lower absorbent joining partner. The electromagnetic energy is converted into heat which then fuses both parts in the interface area due to heat conduction. The welding seam is created as soon as the melt solidifies [2]. Apart from carbon black, it is also possible to use near-infrared absorbers which do not or slightly affect the colour of the component [4]. However, the disadvantage is the significantly higher price compared with carbon black. This has a strong impact especially on the production of high volumetric components. In some areas such as the biotechnology or medical sector, the use of absorbers is inadmissible as the functionality or biocompatibility of the product may be affected.

2 Absorber-free laser transmission welding

2.1 Fundamentals

In absorber-free laser transmission welding, fibre or diode lasers are used which emit radiation in the wavelength area of the polymeric absorption bands ($\lambda = 1500\text{--}2000\text{ nm}$) [5]. The absorption bands are caused by the first harmonic of various bonds such as methyl and methylene groups (Fig. 1, left) [6].

Similar to classic laser transmission welding, about 4–5% of the incident laser radiation is reflected depending on the angle of incidence. The major difference is, however, that the laser radiation is also being absorbed by the upper joining partner. In case of a collimated laser beam, the vertical intensity progression inside the material follows the formula of Lambert-Beer [3]:

$$I(z) = (1-R) \cdot I_0 \cdot e^{-\alpha \cdot z}$$

where $I(z)$ is the intensity progression in z -direction, α is the absorption coefficient, R is the reflectance and I_0 is the incident laser intensity. The reciprocal value of the absorption coefficient ($\delta = 1/\alpha$) is defined by the optical penetration depth δ . It is defined as the distance from the surface at which

Fig. 2 Simulated temperature profile (left) and HAZ in PC in quasi-simultaneous welding (right)

Table 1 Optical properties of PP at 1940 nm (thickness = 2 mm)

Optical properties		Calculated values	
Transmission T [%]	68.89	Scattering coefficient C_s [mm^{-1}]	0.39
Reflection R [%]	6.71	Absorption coefficient C_a [mm^{-1}]	0.24
Collimated Transmission T_c [%]	48.93	Anisotropy factor g [–]	0.63

the incident laser intensity decreases to $1/e$ (about 37%) of its original value. The different intensity progressions in classic and absorber-free laser transmission welding can be seen in Fig. 1.

In classic laser transmission welding, the absorbent joining partner has a high absorption coefficient which implies a low optical penetration depth, whilst in absorber-free laser transmission welding, the absorption coefficient is identical in both joining partners. As no absorber material is needed to adjust the optical properties, this new method offers the possibility of joining transparent or coloured polymers. However, the volume absorption of the laser radiation leads to a HAZ which extends over the entire cross section. Especially in thin plastic components with high seam densities such as Lab-on-a-Chip devices, the increased thermal stress due to the higher melt generation can lead to cracks and distortions. Therefore, the reduction of the HAZ in the vertical direction has been the subject of research for many years. A promising approach is a quasi-simultaneous irradiation strategy of absorber-free polymers which was first introduced by Mamuschkin et al. [3].

2.2 Quasi-simultaneous welding of absorber-free thermoplastics

In quasi-simultaneous welding, a galvanometric scanning system is used. The laser beam is moved along the welding contour multiple times at high feed rates of up to several m/s which leads to a nearly simultaneous heating of the entire welding contour [7]. The advantages compared to contour welding are the higher gap bridging ability as well as the short process time which leads to an increasing interest especially for mass production [8, 9]. Compared with contour welding, the heating process inside the polymer is carried out more gradually with the interaction phase t_i followed by the cooling phase t_c (see Fig. 2 left side). In a study which was presented by Mamuschkin et al., a thulium fibre laser with a wavelength of 1940 nm was used [3]. It could be shown for PC that during

the cooling phases, heat is dissipated from the upper and lower surface of the joining partners. On the other hand, heat is accumulated in the interface due to the low thermal conductivity of polymers (e.g. $0.2 \text{ W/m}^2\text{K}$ for PC). With increasing number of passes and feed rate, the increase in temperature becomes lower which leads to a smoother heating process. In case of PC, this technique contributes to a more favourable and precise energy deposition as the vertical extent of the HAZ is reduced by up to 30% compared with contour welding. It was assumed that the lower melt generation leads to a lower thermal stress during the welding process. In contrast to contour welding where the HAZ has a cylindrical shape, the smooth heating process in quasi-simultaneous welding leads to a more elliptical shape (see Fig. 2, right) [3].

Due to the high process dynamics and therefore the higher demands on the equipment, the total investment costs for a quasi-simultaneous welding system are higher which is one of the main drawbacks of this method. Another aspect is the limitation of the maximum seam length which depends on the scanning field of the optic as well as the maximum laser power output [3, 8]. Whilst the advantages of quasi-simultaneous laser welding of absorber-free amorphous polymers such as PC or TPE-A were shown in [3, 10], there is currently no work available on the influences on the HAZ of scattering materials such as semi-crystalline polymers.

The presented work is divided into a theoretical part, where the scattering behaviour of polypropylene (PP) is calculated using the four-flux model [11]. Further information about the material can be found in Section 4.1. For this purpose, the optical properties of PP were measured using a UV-VIS-NIR spectrometer Lambda 1050 (PerkinElmer Inc., Waltham, Massachusetts). Based on the scattering behaviour, the laser propagation inside the material was calculated using the ray tracing software ZEMAX®. The resulting heat source is then used to simulate the temperature distribution during the quasi-simultaneous welding process. The presented approach is described in detail by Aden [12–14]. In the experimental part,

Table 2 Components of the experimental setup

Laser system		Beam properties	
Beam source	IPG TLR-120-1940	Focal diameter	90 μm
Scanner	SCANLAB IntelliSCAN III 14	Rayleigh length	1.43 mm
Focal length	100 mm

Fig. 3 Scattered light propagation inside the material (left) and absorbed flux on the detector (right)

quasi-simultaneous irradiation trials were carried out using PP. The HAZ dimensions were investigated at different process parameters and compared with the simulated results.

3 Modelling the scattering behaviour in semi-crystalline polypropylene

3.1 Optical characterization

The laser beam propagation inside a scattering material can be described by the scattering coefficient C_{sc} , the absorption coefficient C_{ab} and the anisotropy factor g . The anisotropy factor has a value between -1 and 1 and is a factor for the average scattering direction. For instance, the material is a forward scattering medium for $g > 0$ and a backward scattering medium for $g < 0$. For $g = 0$, the medium is an isotropic scattering material. These values are calculated using the four-flux model with the help of the transmittance T , collimated transmittance T_c and reflectance R of the sample using an UV-VIS-NIR spectrometer. In contrast to T , the scattered fraction of the transmitted light is neglected in T_c [12]. The refractive index of PP was taken from the literature. The measured and calculated optical properties of PP with a thickness of 1 mm are summarized in Table 1.

The calculations of the presented values by using the four-flux model are specified in [12].

After the determination of the absorption and scattering properties, the laser beam propagation was calculated using ZEMAX®. Here, the mean free path of a ray is C_s^{-1} . The light scattering direction inside the material is described by the Henyey-Greenstein phase function [15]. The calculation was carried out for a flat PP sample with a thickness of 2 mm which is the overall thickness of both the upper and lower joining partner in the experimental part. The beam is a Gaussian beam. Due to the simulation of a large number of rays ($> 10^6$), deviations in the ray propagation are compensated. The transmitted rays are detected inside the sample by the detector which is a plain area. As a result, the heat source Q_a on the detector is calculated and normalized to 1 W (see Fig. 3). Q_a describes the absorbed laser intensity in a volume element. It can be seen that due to the scattering of the material, the highest absorbed flux is located in the upper joining partner.

3.2 Modelling the temperature distribution of the welding process

The temperature distribution during the welding process was simulated using COMSOL Multiphysics® (COMSOL Inc.,

Fig. 4 Experimental setup

Fig. 5 Analysis of the HAZ dimensions

Stockholm, Sweden). The heat conduction equation was solved using a finite element method:

$$\frac{\partial(\rho \cdot c_p \cdot T)}{\partial t} + \nabla \cdot (K \cdot \nabla T) = Q_a$$

where ρ is the density, c_p the specific heat capacity, K the heat conduction and Q_a the heat source. Apart from Q_a , these values depend on the temperature which makes the equation non-linear. The specific heat capacity was obtained by DSC measurements, and the mass density was taken from PVT diagrams. The heat conduction is taken from the literature [16].

The joining partners were modelled as one element as a perfect thermal contact was assumed. The overall thickness of both joining partners was 2 mm. The temperature field in a cross section perpendicular to the welding direction was calculated in order to investigate the HAZ. Furthermore, the temperature progression during the welding process was simulated as well. The isotherm at 160 °C is an approximation of the HAZ as it is the melting temperature of PP. The calculated HAZ dimensions are compared to the experimental results in Section 4.3.

4 Experimental work

4.1 Experimental setup

The experimental work was carried out using the setup in Fig. 4 with PP (SIMONA AG, Kirn, Germany). Instead of welding two PP plates with a thickness of $d = 1$ mm each, one bulk sample with a thickness of $d = 2$ mm was irradiated. The reason is that both joining partners have the same properties in absorber-free laser transmission welding. Furthermore, this facilitates the subsequent sample preparation for microtome cuts. As beam source, a thulium fibre laser with a wavelength of 1940 nm (TLR-120-WC, IPG Laser GmbH, Burbach, Germany) was used with a maximum laser power of 120 W. A summary of the components is given below.

The specimens were positioned by using a clamping device. The sample holder is made of aluminium which has a high heat conductivity ($K = 237 \text{ W/(m} \cdot \text{K)}$). Furthermore, the high reflectance couples the reflected laser beam back into the specimen. At last, the focal level was set 5 mm below the interface as impurities on the surface can lead to burnings due to the high peak intensity of the Gaussian beam. The welding seam length was 16 mm.

After irradiation, microtome cuts were made perpendicular to the beam motion using a rotary microtome RM2255 (Leica Biosystems Nussloch GmbH, Nussloch, Germany) and investigated using a polarization microscope Axio Imager A2.m of the company Carl Zeiss Microscopy GmbH, Jena, Germany, in order to measure the HAZ dimensions. Both the height and width of the HAZ were measured (see Fig. 5).

Contour welding trials were carried out as a preliminary test in order to determine appropriate parameters for the quasi-simultaneous welding trials. Here, line contours were irradiated with different line energies E . The line energy E is defined as follows:

Fig. 6 HAZ in contour welding at $E = 1.5 \text{ J/mm}$ (left) and $E = 2.5 \text{ J/mm}$ (right) using polarization microscopy

Fig. 7 HAZ in quasi-simultaneous welding at $E = 2.1$ J/mm (left) and $E = 2.5$ J/mm (right) using polarization microscopy

$$E = \frac{P}{v} * n$$

where P is the laser power, v is the feed rate and n is the number of passes.

4.2 Preliminary contour welding trials

In the contour welding trials ($n = 1$), the line energy was kept constant at 1.5 J/mm and 2.5 J/mm, whilst P and v were varied. The corresponding HAZ dimensions were measured. The following figure (Fig. 6) shows the microscopic analysis of the HAZ for $E = 1.5$ J/mm and 2.5 J/mm at different process parameters.

At $E = 1.5$ J/mm, an offset of the HAZ can be identified towards the upper half of the sample compared to contour welding of amorphous polymers as shown in Fig. 2. Due to the offset, no melting of the lower half could be observed. The reason for this observation is that the semi-crystalline structures lead to light scattering and therefore to a broadening of the intensity distribution. Furthermore, the optical path in the upper joining partner is longer compared with the light propagation in an amorphous polymer. Because of that, the absorption is shifted towards the upper joining partner as well.

At a line energy of 2.5 J/mm, the plastic is melted along the entire cross section. The reason for this is the higher absorbed laser power. The HAZ has a cylindrical shape. The smallest width is observed on the top side of the sample. The highest width on the other hand is located on the bottom side for all investigated parameters. This is in contradiction to the results in 1.5 J/mm where only the top side was melted. A possible reason is that the incident laser radiation is coupled back into the polymer due to reflection on the aluminium plate. Furthermore, it can be seen that the area near the aluminium plate was not melted. Due to the high thermal conductivity of aluminium, no significant heating occurs here. On the upper side, however, no melting of the material can be observed for 1.5 J/mm. At 2.5 J/mm on the other hand, the top surface is not melted at a feed rate of 4.5 mm/s and 9 mm/s. The results show that it was not possible to precisely deposit the radiation energy into the interface by using contour welding.

4.3 Quasi-simultaneous welding trials

Based on the contour welding trials, quasi-simultaneous irradiation trials were carried out for $E = 2.1$ J/mm and $E = 2.5$ J/mm. Compared with contour welding, the HAZ has an elliptic form due to the cooling phases as it was previously reported

Fig. 8 HAZ dimensions for $E = 2.1$ J/mm and $E = 2.5$ J/mm in dependence of the number of passes

Fig. 9 Temperature progression during contour and quasi-simultaneous welding at different positions (left = interface, right = upper and lower surface)

for PC in [3]. Generally, the offset of the HAZ is lower compared with contour welding. One reason could be that as the material is being molten after each irradiation pass, more laser radiation can penetrate into the material volume as less radiation is scattered in the polymer melt. On top of that, the optical path of the laser beam is reduced. Another reason is that due to heat accumulation, the highest temperature is shifted towards the middle of the sample. It can be seen that at 2.1 J/mm, the HAZ dimensions are significantly lower than at 2.5 J/mm. However, higher deviations can be observed at 2.1 J/mm. It is assumed that the process boundary for a generation of the HAZ is reached at that point. This leads to an unstable welding process with high deviations.

The following figure (see Fig. 8) shows the HAZ dimensions depending on the number of passes. At $E = 2.1$ J/mm, a reduction of 64% can be observed from $n = 1$ to $n = 150$ for the HAZ height. At $E = 2.5$ J/mm, the HAZ height is reduced by 37%. Additionally, it can be seen that the highest reduction is reached from $n = 1$ to $n = 10$ for both line energies. For higher number of passes, no significant change in the HAZ height can be observed. At $E = 2.1$ J/mm, the HAZ increases slightly with higher number passes. As the process boundary for the generation of a HAZ is reached at this line energy, the welding process becomes unstable which explains the

deviations in the HAZ dimensions. The increasing number of passes also leads to a reduction of the HAZ width. From $n = 1$ to $n = 150$, the width is reduced by 58% at $E = 2.1$ J/mm, whilst a reduction of 27% is achieved at $E = 2.5$ J/mm. The results show that similar to quasi-simultaneous welding of amorphous polymers, a reduction of the HAZ dimensions can be achieved. In case of contour welding, light scattering causes an offset of the HAZ towards the upper joining partner. If insufficient line energies are applied, only the upper joining partner is melted. In quasi-simultaneous welding on the other hand, heat accumulation compensates the offset.

The reduced thermal stress can also be detected by the simulation of the temperature progression during the welding process. Figure 9 shows the temperature progression during quasi-simultaneous welding at different number of passes and contour welding at $E = 2.5$ J/mm.

In the interface (Fig. 9, left), it can be seen that in contour welding, the melting temperature is exceeded as soon as the laser beam passes the reference point. On the other hand, quasi-simultaneous welding leads to a smoother temperature increase which leads to a delayed melting process. Furthermore, the temperature jumps become smaller with increasing number of passes. As the line energy was kept constant at $E = 2.5$ J/mm, the feed rate was adapted to the number

Fig. 10 The simulated and experimental HAZ

Fig. 11 Comparison between the HAZ of the welding trials and the simulation

of passes. The temperature progression at the upper surface has a similar development as in the interface. The difference is, however, that the melting temperature was not exceeded for both contour and quasi-simultaneous welding as the heat is dissipated through the glass plate. At the lower surface, no temperature increase could be detected. The reason is that the lower surface is in contact to the aluminium sample holder which has a high heat conductivity.

Figure 10 shows a comparison between the simulated HAZ and the corresponding real HAZ for three exemplary parameter sets. The simulated HAZ also shows an elliptical shape for $n = 50$ and $n = 150$ and a cylindrical shape for $n = 1$. It can also be seen that the simulation predicts a bigger HAZ. The highest temperature is located in the upper joining partner. Due to the reduction of the HAZ height, the temperature maximum is moved towards the interface at $n = 50$ and $n = 150$.

The calculated HAZ dimensions were compared to the experimental results which are shown in Fig. 11. For all investigated parameters, the simulation model predicts higher HAZ dimensions. Furthermore, a reduction of the HAZ dimensions can be detected with increasing number of passes. There is a high deviation in case of the HAZ width with an average deviation of 52.5%. Here, the deviation increases with increasing number of passes. The HAZ height is described more precisely as the average deviation is 16.6%. The deviation is almost constant for all investigated parameters with an exception for $n = 1$ where the simulated and experimental results have nearly the same value.

There are several reasons for the deviations between the simulation model and the real experiments. In the theoretical model, homogenous material properties (e.g. crystallinity) were assumed, whereas under real conditions, the distribution and size of the crystalline structures depend on the production parameters. Furthermore, the optical and scattering properties were considered temperature-independent. Because of that, the dissolution of the crystalline structures during the melting process was neglected.

5 Conclusion

In the presented work, the scattering behaviour of PP was modelled using the four-flux model. Here, the optical properties of PP were measured using a UV/VIS/NIR spectrometer. Based on the obtained results, the laser beam propagation as well as the temperature distribution and progression inside the material during the welding process were simulated. It could be shown that the highest laser beam absorption was located in the upper joining partner due to the scattering properties of PP. In the experimental section, the quasi-simultaneous irradiation trials showed that the offset of the HAZ can be reduced which enables a more precise energy deposition compared with contour welding. The reason is that the highest temperature is shifted towards the middle of the sample due to heat accumulation. It could also be shown that the vertical extent of the HAZ could be reduced which leads to lower thermal stress. In future works, different strong scattering materials need to be investigated to confirm the identified advantages compared to contour welding. Furthermore, the precise energy deposition inside the interface should be further investigated. This could be done by varying the optical parameters such as the focal length. Additionally, the seam strength should be investigated using tensile tests. At last the effects of the seam length on the HAZ should be investigated as a longer seam length affects the cooling phases after each irradiation pass.

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