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Correlation between the spatial weld seam morphology and the spatial-temporal temperature profile in laser transmission welding of polypropylene

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Abstract

Laser welding of thermoplastic polymers is a well-known joining technology that is particularly efficient for joining thermoplastic polymers. Although the process is already in industrial use, the basic process-structure-property relationships are not fully understood and still subject of current research. The key to understand the correlations between process parameters and final weld properties are the mechanisms of origin of crystallinity, crystal phase, and spherulite size. Understanding is made difficult by the fact that the laser welding process is a highly dynamic thermomechanical process and therefore very sensitive to experimental circumstances and parameters. In addition, the spatial and temporal distribution of the temperature leads to different melting, consolidation and crystallization conditions within the weld seam, so that it is to be expected that this will also influence the spatial distribution of the microstructural features.
In this study, the spatial distribution of microstructural features inside the weld seam is investigated. For this purpose, the size of spherulites in the weld seam is examined by microscopy as well as differently occurring phases of polypropylene (α- and β-phase). Moreover, differential scanning calorimetry was performed in order to measure the crystallinity. The results are correlated with the spatial-temporal temperature profile inside the weld seam which is derived by a thermal simulation model applied with COMSOL.

Keywords: Laser transmission welding; weld seam morphology; temperature field simulation; polypropylene

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1. Introduction

Laser transmission welding enables the production of high-quality weld seams with high weld seam strength, while low thermal and mechanical component load is applied during the welding process [1]. During this joining process, both joining partners are positioned in overlap geometry and brought in contact with a clamping device. The laser initially transmits the upper joining partner and is then absorbed in the lower joining partner. The absorbed energy is thereby converted into heat and the lower joining partner is melted. By heat conduction from the lower to the upper joining partner, the latter is also melted and a weld seam is formed. For high quality welds, sufficient clamping pressures as well as proper process parameters are necessary [2].

The determination of suitable process parameters is usually done by parameter screening with successive mechanical testing. The reason is that laser welding is a highly dynamic thermomechanical process which is affected not only by material and laser beam parameters but also by the geometry of the joining parts and the applied clamping forces. All these parameters influence the spatial-temporal temperature profile and thus melting and crystallization process inside the weld seam area. Finally, the resulting weld seam performance depends on the microstructural properties achieved.

Looking at recent studies on laser transmission welding, the focus has mainly been on the investigation of the weld seam strength as a function of the line energy, the role of optical properties of the joining partners, the investigation of gap-bridging capability, the measurement of the temperature for process control, or the welding of new material combinations [3–5]. Thorough studies of the resulting microstructures within the weld and their impact on mechanical weld properties are quite rare, but of utmost importance in understanding the correlation between process parameters and the final mechanical properties of the parts.

The final mechanical part properties are mainly dependent on the molecular weight, the crystallinity, the nature of the spherulite phase, and the spherulite size [6]. Spherulites are spherical aggregates of crystallites that arise from the melt when it cools [7]. Studies of effects of spherulite size on the general mechanical properties of injection molded parts go back a long way. Several authors stated that impact strength and spherulite size are inversely related, meaning that increasing the spherulite diameter leads to a decrease in impact strength [8–10]. Similar correlations are known for tensile strength [11]. However, it was also found that the tensile strength passes through a maximum with decreasing spherulite diameter and decreases again with further decreasing spherulite diameter [12]. Though, it is generally agreed that the formation and size of spherulites is mainly dependent on nucleating agents and cooling rates during crystallization [13].

It must be mentioned, however, that the correlations described only apply to injection-molded base material. During the welding process, the process conditions differ fundamentally from those of injection molding, since on the one hand only local melting takes place and on the other hand the heating and cooling rates are much higher [14]. So far there are only a few studies on the influence of morphology on the mechanical properties of thermoplastic weld [5, 15]. Klein found, for example, that laser-welded samples made of polyoxymethylene fail at the transition from the coarse-spherulitic structure of the absorbent part to the fine-spherulitic structure of the transparent part [15, 15]. However, a detailed analysis of spatial formation of spherulitic structures as a function of the spatial-temporal temperature profile in the weld seam does not take place.

This paper attempts to understand the origin of the spatial distribution of microstructure features within the weld of laser fused polypropylene samples. For this purpose, the size of spherulites in the weld seam is examined by microscopy as well as differently occurring phases of polypropylene (α- and β-phase). Moreover, differential scanning calorimetry was performed in order to measure the crystallinity. The results are compared with the spatial-temporal temperature profile inside the weld seam which is derived by a thermal simulation model applied with COMSOL.

2. Experimentals

2.1. Experimental setup and characterization methods

For welding tests, a diode laser (TruDiode 301, Trumpf Laser) with a wavelength of 980 nm was used. The setup is shown in Figure 1 (left). The maximum output power is 300 W (cw) with a beam quality of 8 mm · mrad. A galvanometer scanner (PS1-10, Cambridge Technologies) and an F-theta lens (S4LFT3162, Sill Optics) were used for deflection and focusing of the laser beam. A pneumatic clamping device was used to position and fix the samples in overlap joint geometry. The applied joining pressure is 3 bar for an overlap area of 20 × 40 mm^2. For the experimental tests, contour welding was performed with a laser power of P_L = 15 W, a beam diameter of 2 mm and a feed rate of 20 mm/s, which proved to be appropriate during preliminary tests.

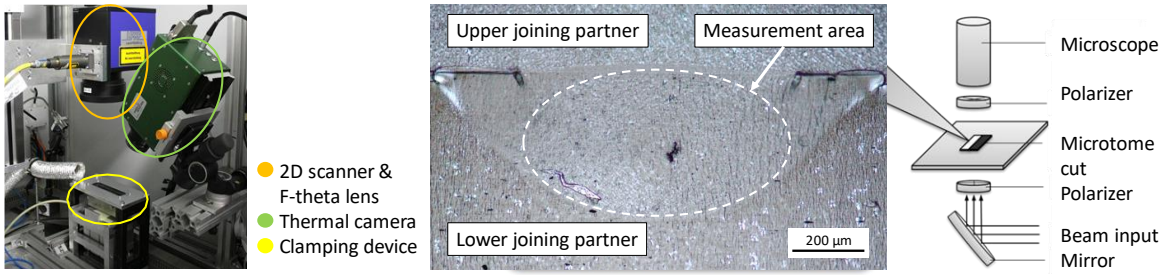


Fig. 1. Experimental setup (left) and schematic of polarization microscopy with an exemplary thin section of the weld seam and the measurement area (right)

In order to analyze the weld morphology, 10 µm thin sections were made across the weld using a microtome and examined by means of polarization microscopy, as demonstrated in Figure 1 (right). Using polarization microscopy, locally varying crystalline structures in the weld seam and in the surrounding edge areas can be detected and measured. In addition, the thin sections can provide information about the characteristics of the heat-affected zone in the upper and lower joining partners.

2.2. Thermoplastic materials

All tests were carried out with injection-moulded samples of natural and carbon black filled isotactic polypropylene (Moplen HP501L), since this material is widely used in industry. The absorbent joining partner contains 0.1 wt.-% carbon black without further additives compared to the natural polypropylene. The examination of optical properties of the PP samples is carried out by spectrometer measurements with wavelengths in the near infrared range (940 nm to 1,070 nm) of the diode laser using a UV-VIS-NIR spectrophotometer (Shimadzu UV-3600). For laser radiation with wavelengths in NIR region, the laser-transparent PP shows a transmittance of 63 ± 3 %. Both joining partners have the geometry of a half-shoulder test rod with a material thickness of 2 mm.

For polypropylene, it is known that above all the spherulite diameter and the phase fractions (α-, β- and γ-phase) decisively influence the mechanical properties [16]. For example, the tensile strength increases with decreasing spherulite diameter, while the impact strength, tensile strength and elongation at break increase and the modulus of elasticity decreases with increasing β-phase fraction [17]. In addition to the occurrence and size of the spherulites, the degree of crystallization is another essential parameter of morphology. With increasing crystallinity, the stiffness, the density and the yield stress increase, whereas the impact strength and the tendency to creep decrease.

3. Thermal simulation of laser welding process

In order to determine certain parameters such as maximum temperatures or heating and cooling rates during the welding process, a thermal finite element model based on the work of Devrient et al. is developed further with COMSOL [1]. The simulation model is built up to determine the temperature field and the cooling rates in the process necessary for the morphological studies and improve process understanding. The model is based on Fourier's general differential equation of heat conduction:

$$\rho \cdot c_p \cdot \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + Q \tag{1}$$

The specific heat capacity c_p and the density ρ were modelled as temperature-dependent. The temperature-dependent specific heat capacity c_p was determined from the DSC measurements, the density ρ and the thermal conductivity k were taken from literature [18]. In the volume heat source Q, both the moving laser beam with a Gaussian intensity distribution and the laser beam-material interaction are modelled according to Beer-Lambert law. The volume heat source Q in turn can be defined as a function of laser power P_l, beam radius r_l, feed rate v_l, absorption coefficient α, coordinates x, y, z and time t [4]. The model thus calculates the local and temporal temperature field during the welding process as a function of the process and material parameters.

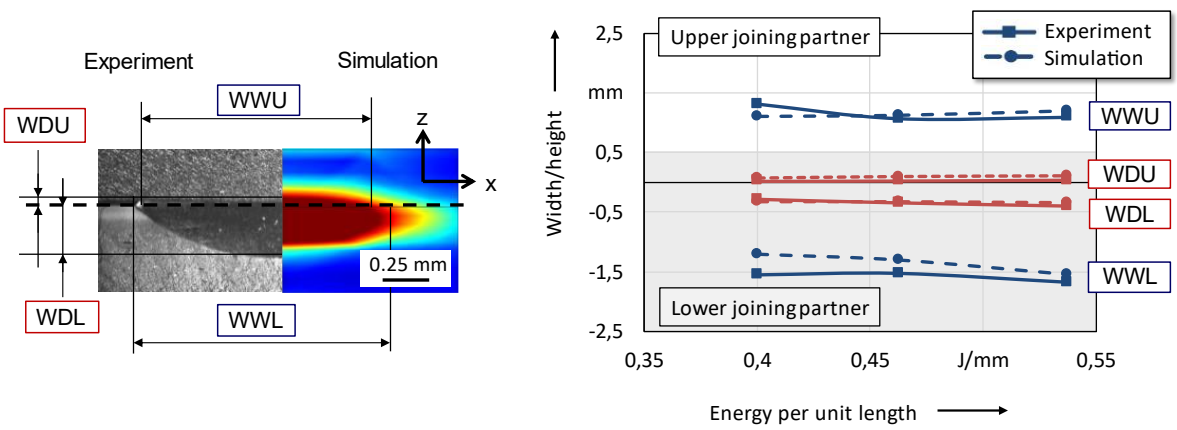


Fig. 2. Cross-sectional view of simulated temperature field at time t = 0.2 s and weld seam dimensions for contour welding of PP in overlap joint (left) and comparison of the weld seam width and depth between simulation and experiment (right)

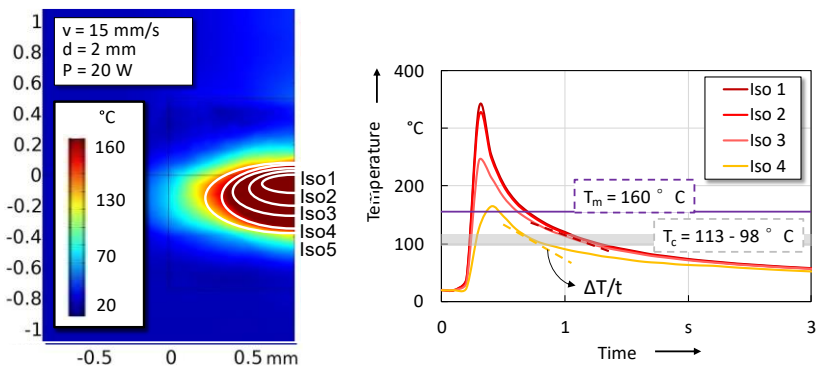
To verify the thermal finite element model, the weld seam was compared by simulation and experiment. A distinction was made between weld width of the upper joining partner (WWU), weld depth of the upper joining partner (WDU), weld width of the lower joining partner (WWL) and weld depth of the lower joining partner (WDL). The results are shown in Figure 2, where the abovementioned weld seam width and depth of the upper and lower joining partner are plotted as a function of the laser line energy. It can be stated that a high agreement between the experimental and the simulative values of the weld seam characteristics is achieved. In conclusion, the model is able to represent the formation of the temperature field during contour welding of polypropylene with adequate accuracy and draw conclusions about the duration in which the weld seam is within the relevant crystallization temperature range (T_C) during cooling.

4. Results and discussion

4.1. Effect of spatial temperature distribution on microstructure

In the following, the effects of the temperature field on the weld seam morphology during laser transmission welding are investigated. When the laser radiation impinges the absorbing joining partner, a spatial and temporal temperature distribution is formed. For the temperature curves shown in Figure 3, it is evident that the temperature in the welding center reaches its maximum value. This can be explained by the increasing amount of energy input in the welding center caused by the Gaussian intensity distribution of the laser beam. With increasing distance from the welding center, heat conduction within the thermoplastic material leads to a point-symmetric temperature decrease, forming isotherms around the welding center, see Figure 3 (left).

In order to spatially analyze the crystallization behaviour, the cooling rates were determined as a slope of the temperature curve in the crystallization range of polypropylene (113 - 98 °C). Here it can be seen that the cooling rates in this range increase with decreasing maximum temperature. The reason for this is that the melt pool volume in the center of the weld takes longer to cool down to room temperature than in the peripheral area. Due to the exponential cooling, the crystallization temperature in the center of the weld is only reached when the temperature curve has already flattened out. Thus, higher maximum temperatures cause lower cooling rates in the crystallization range of polypropylene, see Figure 3 (right).



Test point	Distance to interface	Cooling rate ΔT/t
Iso 1	100 µm	71.2 K/s
Iso 2	200 µm	74.0 K/s
Iso 3	300 µm	92.2 K/s
Iso 4	400 µm	114.0 K/s
Iso 5	500 µm	T_{max} too low

Fig. 2. Simulated temperature fields and weld seam dimensions for contour welding of PP (left) and segmentation of the weld in different isothermal regions with corresponding cooling rates in the crystallization area (right).

In our thermal simulation, cooling rates in the range of 71 – 114 K/s could be determined within the melt pool. Compared to e. g. injection moulding, the cooling rates that occur during laser transmission welding within the melt pool vary in a relatively small range, which also means that the diameters of the spherulites occurring in the weld seam show only minor differences [7].

The investigation of the weld seam morphology is performed using polarization microscopy. The predominant crystal modification is the α- and β-phase, whose determination of the diameter is shown exemplarily for some β-spherulites, see Figure 4 (left). The samples are generated with a laser power of P_L = 15 W at a constant feed rate of v_s = 20 mm/s and a beam diameter of d_D = 2.0 mm in contour welding. The weld morphology of the joined polypropylene samples is analyzed by polarization microscopy and correlated to the temperature profile of the thermal finite element model. The dependence of the spherulite diameter and distribution of the β-phase on the spatially resolved cooling rates in the crystallization range of polypropylene is shown in Figure 4. In the edge area of the weld seam, the total amount of spherulites (> 5 µm)

per mm² is higher compared to the amount in the center of the weld. Simultaneously, the size of the spherulites decreases from the center of the weld seam (Figure 4; Iso1, Iso2) towards the edge region (Figure 4; Iso3, Iso4). The crystal growth can be attributed to the cooling behaviour in the crystallization temperature range (T_C) of polypropylene. The slow heat dissipation during cooling in the welding center compared to the edge area leads to a lower cooling rate in the crystallization range of polypropylene. Due to the prevailing lower cooling rate, the region remains in the crystallization range for a longer period of time and therefore promotes crystal growth. At the same time, the amount of spherulites tends to increase from the welding center towards the edge area, which can be attributed to the increasing lateral temperature gradient in the weld seam, as shown in previous investigations [3].

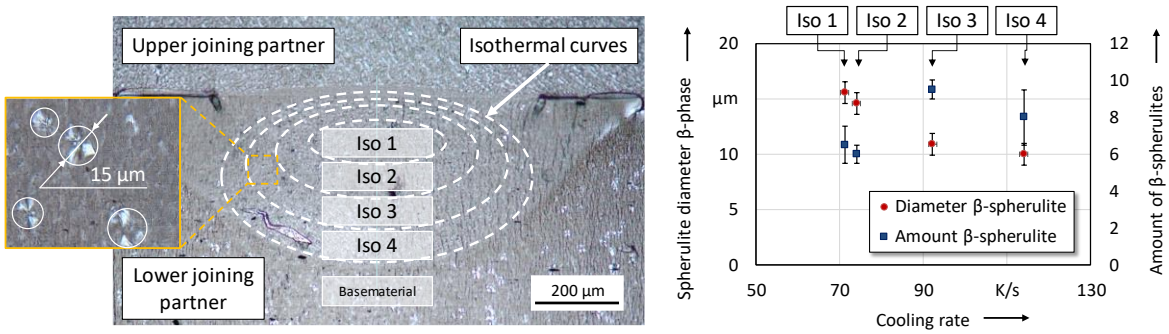


Fig. 3. Cross-sectional view of PP weld seam generated with a laser power of P_L = 15 W, a feed rate of v_s = 20 mm/s and a beam diameter of d_D = 2.0 mm with corresponding isothermal curves (left) and occurring and size of spherulites within the isothermal regions (right)

As known from literature, the spherulite diameter mainly depends on nucleating agents and the duration in which the material is within the relevant crystallization temperature range (T_C) during cooling[19]. Therefore, it cannot be excluded in the welding tests that the soot particles in the lower absorbing joining partner act as nucleating agents and thus have an influence on the spherulite size, which prevents the formation of larger spherulites. However, any influence can at least be considered constant, as all samples contain an identical filling content of 0.1 wt.-% of homogeneously distributed carbon black. Consequently, only a different cooling behaviour during the welding process can be the cause for the occurring and size of the spherulites.

4.2. Effect of spatial temperature distribution on degree of crystallization

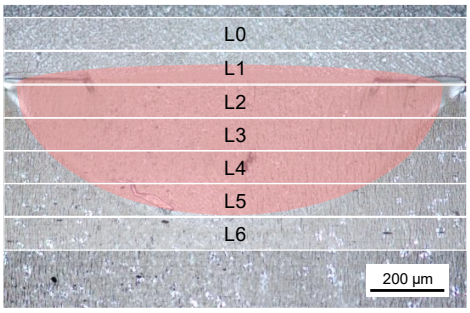
As already observed in the analysis of the microstructure, the occurrence and size of the spherulitic structures depend mainly on the cooling rate during laser welding and vary within the location of the weld. In addition, the melting and recrystallization of the material leads to a change in the degree of crystallization (proportion of the crystalline, ordered phase compared to the amorphous, disordered phase) and is dependent on the process parameters. The degree of crystallization is determined by means of DSC measurements and calculations of the melting enthalpy. In order to spatially resolve the degree of crystallization in the weld seam, microtomy is used to extract material from the joined polypropylene samples, see Figure 4. For each region, the melting enthalpy $\Delta H(T_{melt})$ is measured and set in relation to the literature value $\Delta H^0(T_{melt})$ = 207 J/g for completely crystalline polypropylene material:

$$C = \Delta H(T_{melt}) / \Delta H^0(T_{melt}) * 100\% \qquad (2)$$

Table 1 shows the subdivision of the samples layer by layer and the resulting degree of crystallization for an investigated set of parameters. The samples are generated with a laser power of P_L = 15 W at a constant feed rate of v_s = 20 mm/s and a beam diameter of d_D = 2.0 mm in contour welding. The results of the measurements show a lower degree of crystallization in the weld seam relative to the base material. The lower degree of crystallization can be explained by the cooling rates during laser welding in the crystallization range of polypropylene (113 – 98 °C). According to the calculations of the thermal finite element model, the cooling rates in the laser welding process are in the range of approx. 70 – 115 K/s, while cooling rates of several 100 K/s occur in injection moulding [7]. The shorter time of the material melted by laser radiation in the range of the crystallization temperature thus leads to a lower degree of crystallization compared to the base material.

Table 1. Subdivision of weld seam into layers with 100 µm height and 2.000 µm width and resulting degree of crystallization for each area

Position in weld	Degree of crystallization (%)	Std. dev.
L0 (base material)	38.5	0.3
L1	38.4	0.9
L2	39.8	1.3
L3	40.6	1.2
L4	40.5	0.8
L5	41.1	0.9
L6 (base material)	41.5	0.4



It is known that the degree of crystallization significantly influences the mechanical properties of thermoplastic components. With increasing degree of crystallization, e. g. the stiffness or the modulus of elasticity increases, whereas the creep tendency and the impact strength decrease [20]. Processing by means of laser radiation enables the adjustment of the spatial and temporal temperature distribution and thus the degree of crystallization within the weld seam.

In general, however, the results from the measurement of the degree of crystallization show only small differences between weld seam and base material. The reason for this is that the material extracted from the samples exceeds the weld width and represents an averaged value over several isothermal regions. In turn, previous investigations show that an increase in the degree of crystallization of polypropylene by 10 % can double the elastic modulus and thus generate a large leverage effect on the mechanical properties of the welded joint [21]. For a clear correlation between the degree of crystallization and the spatial-temporal temperature profile, the characterization of the degree of crystallization must be carried out on even smaller, isothermal regions within the weld seam and therefore requires a more extensive sample preparation.

5. Conclusion

This paper attempts to understand the origin of the spatial distribution of microstructure features within the weld seam of laser fused polypropylene samples. For this purpose, the size of spherulites in the weld is examined by microscopy as well as a relevant occurring phase of polypropylene. Based on the development of a thermal finite element model, the experimental results could be verified and correlated with the spatial-temporal temperature profile inside the weld seam. It is shown that the occurrence and size of spherulites in the weld seam of PP is mainly dependent on the spatially different cooling rate in the weld seam. From the center of the weld seam towards the edge area, the amount of spherulites per mm^2 increases, whereas the

spherulite diameter decreases, meaning that spherulite occurrence and size are inversely related to the spatial-temporal cooling rate. In addition, it is shown that the degree of crystallization of the weld seam is lower in comparison to the injection moulded base material, which is related to the reduced cooling rates in the relevant crystallization temperature range of polypropylene. The investigated correlations between spatial-temporal temperature profile and morphology of the weld seam can now be used as a basis for the influence of the mechanical properties of the weld.

Further studies will include a characterization of the degree of crystallization with higher spatial resolution to develop a correlation between the microstructure and the mechanical properties of welded components. This way, together with investigations including a variation of laser process parameters, the results of this paper can be used for targeted adjustments of the mechanical properties of the weld seam.

Acknowledgements

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References

[1] M. Devrient, M. Kern, P. Jaeschke, U. Stute, H. Haferkamp, and M. Schmidt, *Experimental investigation of laser transmission welding of thermoplastics with part-adapted temperature fields*. Hannover: Gottfried Wilhelm Leibniz Universität Hannover; Technische Informationsbibliothek (TIB), 2013.

[2] U. A. Russek, *Prozesstechnische Aspekte des Laserdurchstrahlschweißens von Thermoplasten*. Zugl.: Aachen, Techn. Hochsch., Diss., 2006. Aachen: Shaker, 2006.

[3] B. Geißler, T. Laumer, A. Wübbeke, T. Frick, V. Schöppner, and M. Schmidt, “Analysis of the Weld Seam Morphology of Polypropylene in Laser Transmission Welding,” *J. Manuf. Sci. Eng*, vol. 140, no. 11, 2018, doi: 10.1115/1.4040876.

[4] B. Geißler *et al.,* “Analysis of the interaction between the temperature field and the weld seam morphology in laser transmission welding by using two different discrete laser wavelengths,” *Journal of Laser Applications*, vol. 30, no. 3, p. 32408, 2018, doi: 10.2351/1.5040617.

[5] M.-L. Röhricht, T. Stichel, S. Roth, P. Bräuer, M. Schmidt, and S. Will, “Correlation between weld seam morphology and mechanical properties in laser transmission welding of polypropylene,” *Procedia CIRP*, vol. 94, pp. 691–696, 2020, doi: 10.1016/j.procir.2020.09.119.

[6] P. Tordjeman, C. Robert, G. Marin, and P. Gerard, “The effect of α, β crystalline structure on the mechanical properties of polypropylene,” (in En;en), *Eur. Phys. J. E*, vol. 4, no. 4, pp. 459–465, 2001, doi: 10.1007/s101890170101.

[7] G. Ehrenstein, *Polymer-Werkstoffe: Struktur ; Eigenschaften ; Anwendung,* 3rd ed. s.l.: Carl Hanser Fachbuchverlag, 2011.

[8] S. M. Ohlberg, J. Roth, and R. A. V. Raff, “Relationship between impact strength and spherulite growth in linear polyethylene,” *J. Appl. Polym. Sci.*, vol. 1, no. 1, pp. 114–120, 1959, doi: 10.1002/app.1959.070010118.

[9] D. Wright, R. Dunk, D. Bouvart, and M. Autran, “The effect of crystallinity on the properties of injection moulded polypropylene and polyacetal,” *Polymer*, vol. 29, no. 5, pp. 793–796, 1988, doi: 10.1016/0032-3861(88)90134-6.

[10] W. G. Perkins, “Polymer toughness and impact resistance,” *Polym. Eng. Sci.*, vol. 39, no. 12, pp. 2445–2460, 1999, doi: 10.1002/pen.11632.

[11] L. S. Remaly and J. M. Schultz, “Time-dependent effect of spherulite size on the tensile behavior of polypropylene,” *Journal of Applied Polymer Science*, vol. 14, no. 7, pp. 1871–1877, 1970, doi: 10.1002/app.1970.070140720.

[12] J. L. Way, J. R. Atkinson, and J. Nutting, “The effect of spherulite size on the fracture morphology of polypropylene,” (in En;en), *J Mater Sci*, vol. 9, no. 2, pp. 293–299, 1974, doi: 10.1007/BF00550954.

[13] S. Koltzenburg, M. Maskos, and O. Nuyken, *Polymere: Synthese, Eigenschaften und Anwendungen*. Berlin, Heidelberg: Springer Spektrum, 2014. [Online]. Available: https://www.researchgate.net/publication/316792098_Polymere_Synthese_Eigenschaften_und_Anwendungen

[14] V. Wippo *et al.,* “Evaluation of a Pyrometric-based Temperature Measuring Process for the Laser Transmission Welding,” *Physics*

Procedia, vol. 39, pp. 128–136, 2012, doi: 10.1016/j.phpro.2012.10.022.

[15] H. M. Klein and E. Haberstroh, “Laserschweißen von Kunststoffen in der Mikrotechnik: Fakultät für Maschinenwesen,” [Mainz]; Zugl.: Aachen, Techn. Hochsch., Diss., 2001. [Online]. Available: http://publications.rwth-aachen.de/record/59988

[16] L. S. Remaly and J. M. Schultz, “Time-dependent effect of spherulite size on the tensile behavior of polypropylene,” *Journal of Applied Polymer Science*, vol. 14, no. 7, pp. 1871–1877, 1970, doi: 10.1002/app.1970.070140720.

[17] J. Varga, “β-MODIFICATION OF ISOTACTIC POLYPROPYLENE: PREPARATION, STRUCTURE, PROCESSING, PROPERTIES, AND APPLICATION,” *Journal of Macromolecular Science, Part B*, vol. 41, 4-6, pp. 1121–1171, 2002, doi: 10.1081/MB-120013089.

[18] T. Frick, *Untersuchung der prozessbestimmenden Strahl-Stoff-Wechselwirkungen beim Laserstrahlschweißen von Kunststoffen*. Zugl.: Erlangen-Nürnberg, Univ., Diss., 2007. Bamberg: Meisenbach, 2007.

[19] S. Koltzenburg, M. Maskos, and O. Nuyken, *Polymere: Synthese, Eigenschaften und Anwendungen*. Berlin, Heidelberg: Springer Spektrum, 2014.

[20] *Technische Thermoplaste*. München, Wien: Hanser, 1998.

[21] A. Menyhard *et al.,* “Direct correlation between modulus and the crystalline structure in isotactic polypropylene,” *Express Polym. Lett.*, vol. 9, no. 3, pp. 308–320, 2015, doi: 10.3144/expresspolymlett.2015.28.