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Effect of Pigments on Laser Beam Transmission in Diode Laser Transmission Welding of Poly(propylene)

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Abstract: Welding technologies are state of the art for joining polymer composites, with one of the two joining parts considered laser transmissive (LT) and the other laser absorptive (LA). Pigments are often added to LT to enhance the crystallinity of the polymer matrix. However, pigments lead to internal scattering of the laser beam and the rate of transmission or the laser energy density decreases. Depending upon the type and amount of pigments added in the formulation of LT, the percentage of the laser beam transmitted, absorbed, or scattered differs. Laser welding performance depends on the laser energy available for welding after considering the losses. In the present study, optical transmission of injection molded isotactic polypropylene (iPP) samples were analyzed with a varying dosage of organic pigment (neat PP, 2%, 3%, 4%, 5%, 6%, 8%, and 10%) using a LPKF TMG 3 transmission tester. The device uses a wavelength of 980 nm and simulates the optical radiation conditions of diode laser transmission welding (LTW). The percentage transmission varied with the sample thickness and the composition percentage of pigment. The modified Bouguer-Lambert law described the transmission energy and apparent extinction coefficient. The model was validated with the experimental value of transmittances of the samples with varying sample thicknesses of iPP. There was a decrease in the percentage of laser transmission with an increase in the pigment content of the samples. It was found that the apparent extinction coefficient is a function of the pigment levels.

Keywords: Polymer Composites, Pigments, Optical Transmission, Polypropylene, Extinction Coefficient

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Introduction

Laser transmission welding (LTW) is a technique that is used to join two thermoplastic (TP) materials: laser-transmissive (LT) and laser-absorptive (LA). An incident beam of radiation containing nucleating agents (NA) or pigments passes through LT [1]. The amount of radiation transmitted depends on the size, type, dispersion, distribution, etc., of the pigments. After transmission, the radiation gets absorbed by LA depending on the laser-absorbing pigment (mostly carbon black: CB) in LA. As a result, thermal energy is transferred from LA to LT, leading to the inter-diffusion of the polymer and ultimately forming a weld joint upon cooling. The optical properties of LT and LA determine the amount of laser beam energy transmitted and distributed over the interface. Optical characteristics of polymers are related to the welding performance, considering polymer thickness, wavelength, and various additives for joining two surfaces. For a strong joint, sufficient thermal energy must be available at the weld interface to melt the polymer and at the same time not degrade the polymer due to excessive energy [2]. Various models were developed to understand the effect of some of the factors on the melting and re-solidification phenomenon [3].

Theory

Most of the polymeric materials are transparent to the near-infrared (NIR) laser beam (800~ 1064nm) [4]. Depending on the type and amount of pigments added in the formulation, the amount of laser beam transmitted, scattered or reflected differs. The rate of transmission for organic pigments is higher compared to inorganic pigments [5]. For semi-crystalline (SC) polymers, the phenomenon depends upon the spherulite structure size. Finer spherulites have higher transmittance than coarse spherulites [6]. Due to reflections and refractions, the light gets scattered randomly [7]. The actual optical path of the laser beam is prolonged in SC polymers as the rays get refracted and scattered on the crystalline superstructures. As a result, the absorption increases [8]. In LTW, preferential energy deposition occurs with subsequent TP material melting at the interfacial zone, as shown in Figure 1. The interaction between the laser radiation and TP depends on three crucial parameters: total transmittance (TT), total absorbance (AT), and total reflectance (RT). They are related, as shown in Eq. (1)

$$1 = T_T + A_T + R_T \qquad 1)$$

Depending on the polymer morphology, sample thickness, and compounding ingredients, the percentage of T_T, $A_{T,}$ and R_T differs.

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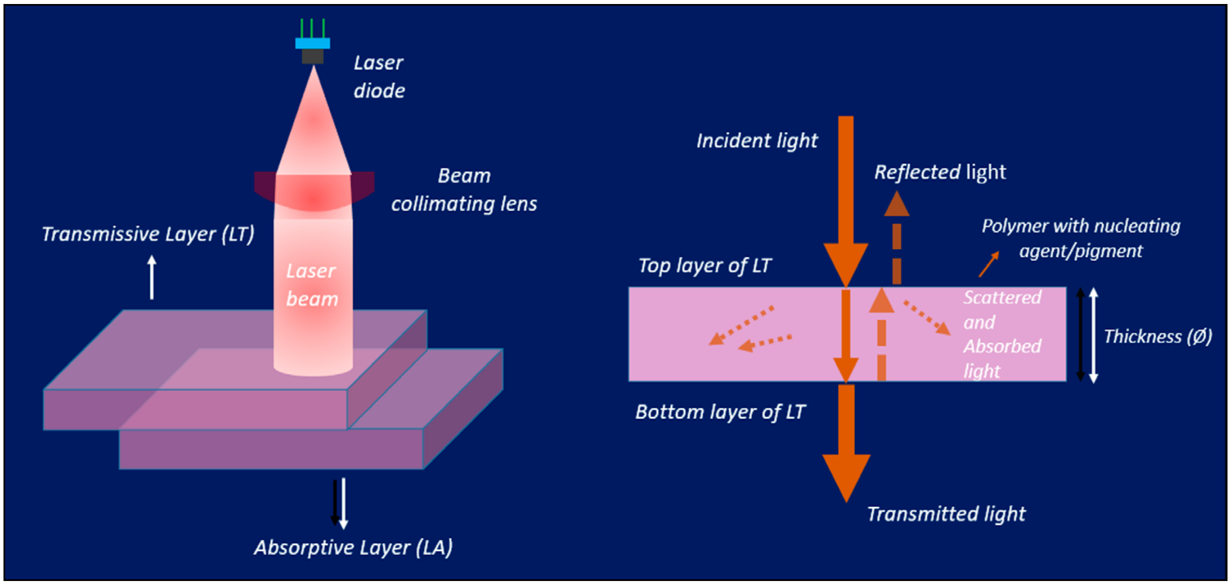

Figure 1. Schematic of transmission laser welding of lap geometry of materials (left) and light transmission through a transmissive layer with reflected, scattered, and absorbed light phenomena (right) [9]. Reproduced with permission from [9]. Copyright (2021)CC BY 4.0, Frontiers in Materials.

For LT, it is difficult to separate absorption and scattering [8,9]. Some studies measure the laser light scattering, such as x-y scanning pinhole [8], charged coupled device, small light sensor [10], etc. The presence of pigments increases the internal scattering of the laser beam, which increases the effective beam diameter. With an increase in the pigment level, the percentage of scattering may vary [11]. When laser radiation falls on the laser transmissive surface, the intensity of the radiation decreases as it propagates, which can be elaborated by integrating Eq. (2) over a definite path length F [12]:

$$\int_{I_\lambda=I_\lambda(0)}^{I_\lambda(F)} \frac{dI_\lambda}{I_\lambda} = -\int_{F^*=0}^{F} \beta_\lambda (F^*)dF^* \qquad 2)$$

where $I\lambda$ (0) is the incident laser radiation from the laser head on a given volume at F = 0 (F is a dummy variable of integration), and $\beta\lambda$ is the extinction coefficient. The integration yields the radiation intensity at location F in the given direction ω:

$$I_\lambda (F) = I_\lambda(0)\, e^{\left[-\int_0^F \beta_\lambda(F^*)dF^*\right]} \qquad 3)$$

Eq. 3 is known as the Beer's, Bouguer's or Lambert-Bourguer law, which shows that as the laser radiation passes through an absorbing-scattering medium, the intensity is attenuated exponentially. The local emission or scattering into the direction ω is not taken into consideration in this law. The extinction coefficient (βλ) is a summation of absorption coefficient (κλ) and scattering coefficient (ϕλ) [12].

$$\beta_\lambda = \kappa_\lambda + \varphi_\lambda \tag{4}$$

Considering that the scattering of the radiation in the laser transmissive part is negligible, Eq.(4) reduces to

$$\beta_\lambda = \kappa_\lambda \tag{5}$$

The interaction between the laser radiation and the crystalline structure of the polymer occurs if the size of the spherulites is in the range of the visible or near-infrared (NIR) light (400-1060 nm) which determines the amount of scattering. Pigment distribution, size, and nature of scattering centers influence the scattering behavior. Beam broadening occurs in polymers with higher crystallinity [13]. Many simulations have been carried out using the Bouguer-Lambert law [14,16]. However, an assumption is made that the laser radiation travels in a straight line through LT and LA, but there will be specular reflective losses (ηi) apart from the absorptive losses, which can be calculated by Eq. (7)

$$\eta_i = \left(\frac{n_1 - n_0}{n_1 + n_0}\right)^2 \tag{6}$$

Model Development

In the case of polymers that are non-scattering, single-scattering or multi-scattering, the total energy transmission can be described by the Bouguer-Lambert law [7]. M. Chen et al. [16] developed and modified a model for measuring the absorption coefficient for LA material filled with CB using the measured transmittances with varying thicknesses. The total laser radiation intensity (Iλ) after passing through the polymer of a given thickness can be given by [7]:

$$I_\lambda = T_T\, I_\lambda(0) = I_\lambda(0)(1 - R_T)(1 - \eta)e^{-\beta_\lambda t} \tag{7}$$

where,

RT: total reflectance of the polymer from the top incident surface as well as the polymer bulk
η: surface reflectance at the polymer bottom surface
t: total thickness of the laser transmissive part or laser absorptive part

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Eq. (7) will be referred to as the modified Bouguer-Lambert law derived from Beer-Lambert law. For simplification, it is assumed that the total reflectance (RT) contribution from the polymer bulk takes place in a local volume close to the top of the incident surface.

$$\ln T_T \approx \ln(1 - R_T - \eta) - \kappa_\lambda t \qquad 8)$$

Eq. (8) will be used in the experimental analysis for validating the modified Bouguer-Lambert law. The samples of 2 mm thickness were stamped by cutting the polypropylene (PP) coupons (Figure 2) with varying pigment content with neat PP, 2%, 3%, 4%, 5%, 6%, 8%, and 10%. Thin cross-sections were also taken as 120 μm, 240 μm, and 360 μm using a microtome.

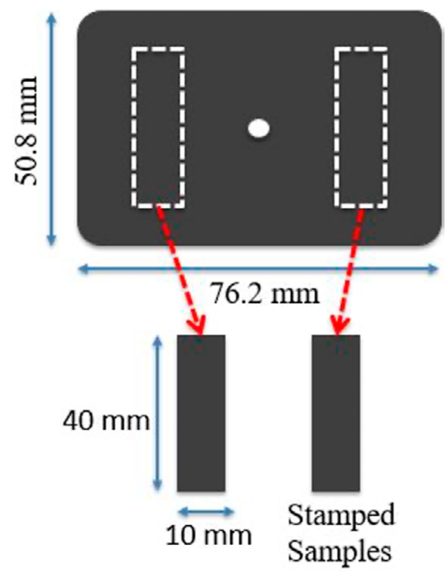

Figure 2. PP sample stamped for analysis.

The LPKF TMG 3 transmission tester (class 1 according to DIN EN 60825-1) was utilized to determine the optical transmission of thermoplastics. The maximum radiation intensity is on the aperture plate of the device. The specifications of the device are mentioned in Table 1. The device is designed to simulate the optical radiation conditions of laser transmission welding of thermoplastic as realistically as possible. The laser beam is emitted from a diode of the tester and penetrates the thermoplastic sample to be measured. The intensity of the radiation after the transmission is calculated based on the photoelectric effect, representing the value for the optical permeability of the thermoplastic at a wavelength of 980 nm. The scattered rays are blocked with an aperture of 3 mm, and the remaining photons are measured on the detector. The data obtained was utilized to validate the modified Bouguer-Lambert law.

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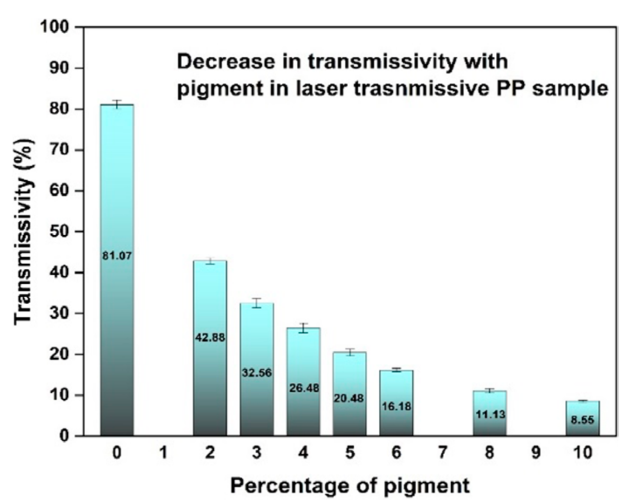
Table 1. Specification of LPKF TMG 3.

Data	Value	Unit
Laser class	1	
Wavelength	980	Nm
Max. power	<1	mW
Diameter of the measuring aperture	3	mm
Focus diameter on the measuring aperture	~1.5	mm

Three measurements on the samples were taken, and an average was considered. The data was utilized to validate the modified Bouguer-Lambert law. The measured transmission values vary according to the wall thickness and the visual properties of a thermoplastic component. The diameter of the measuring beam is approximately 1.5 mm.

Results and Discussion

The addition of NA/pigments changes the crystallinity of the semi-crystalline polymers (PP). Due to changes in the morphology and thickness of the samples, the amount of laser radiation (optical properties) passing through the laser transmissive sample differs. It can be observed in the graph, Figure 3, there is a decrease in the percentage of laser transmission with an increase in the pigment content. Figure 3(b) shows that the fraction of the laser beam energy transmitted depends on the thickness of the laser transmissive layer. It decreases with an increase in thickness [17].



(a)

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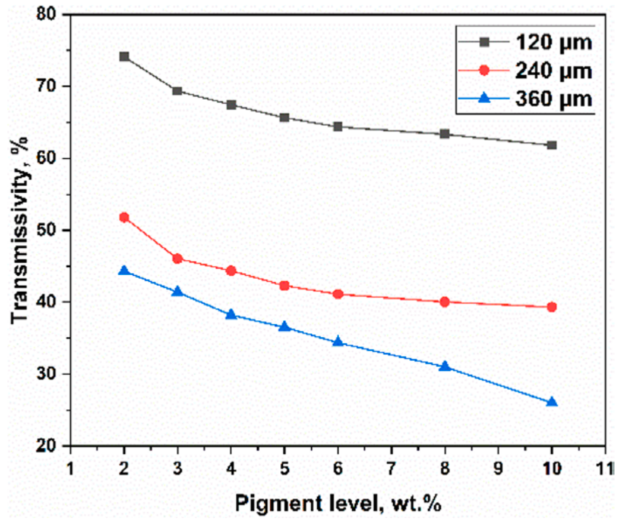

(b)

Figure 3. Decrease in transmissivity with (a) pigment content (b) thickness variation.

A linear relationship between lnTT and thickness t validates the modified Bouguer-Lambert law (Figure 4). The apparent κλ for every pigment content was determined as shown in Table 2 with the R-square value.

Table 2. Apparent κλ for samples with varying pigment content.

Sl. No.	Pigment level (wt.%)	Apparent κλ (1/mm)	$R_T + \eta = 2\eta$	R-Square
1	2	0.85	0.05	80.64
2	3	0.86	0.13	62.65
3	4	1.08	0.12	77.54
4	5	1.16	0.14	76.78
5	6	1.33	0.14	83.82
6	8	1.69	0.10	94.12
7	10	2.32	0.03	99.99

The values of the apparent κλ are plotted with the pigment level, as shown in Figure 4. A linear relationship between the apparent κλ and pigment level with R2: 95.51% was observed experimentally. Equation 9 obtained from the graph can be used to determine apparent κλ as a function of pigment for PP samples. Apparent κλ increased from 0.85 1/mm for 2 wt.% pigment to 2.32 1/mm for 10 wt.% pigment. Higher κλ at higher pigment content can affect the thermal profile, which may facilitate the welding process.

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$$y = 0.204x + 0.194 \tag{9}$$

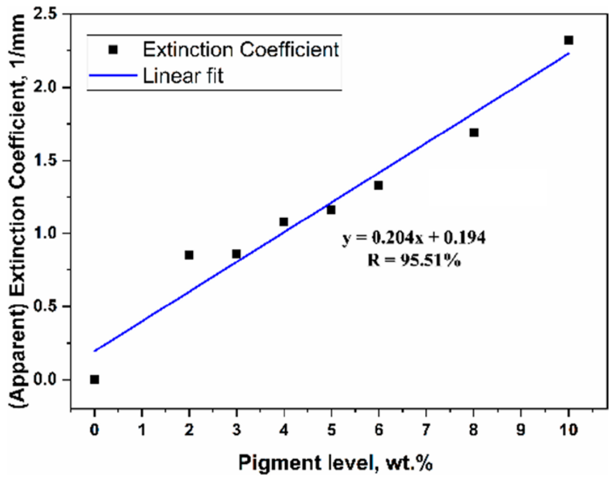

Figure 4. κλ Vs pigment level for PP.

Conclusion

The Bouguer–Lambert law was utilized to study the effect of pigment content and sample thickness on the laser beam transmission. A linear relationship between the apparent **κλ** and the pigment content was observed. At higher pigment content, the percentage of transmission decreases. Hence, there is less energy available for the laser absorptive layer to absorb the laser beam. The amount of melting may not be sufficient for the interdiffusion for welding. This could decrease the weld strength. However, higher **κλ** at higher pigment content can affect the thermal profile, which may help the welding process. Hence, an optimum pigment percentage needs to be quantified in future work. Also, the effect of pigment size and its distribution on **κλ** and polymer crystallinity will be carried out.

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