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Department of Mechanical and Manufacturing Engineering at Aalborg University, Fibigerstraede 16, 9220 Aalborg Ø, Denmark

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ABSTRACT

Properties of polystyrene/polystyrene (PS/PS) diode laser (808 nm) welded lap-joints were evaluated. First, the reptation time of the selected PS was determined using plate/plate oscillatory rheometry. The reptation time was critically dependent on temperature, but as long the temperature was above 200 °C, the reptation time was less than 8 ms. The reptation time is, therefore, smaller than the process time, permitting development of full weld strength.

Two methods were selected for mechanical testing; a *lap-joint shear test* to evaluate the weld strength $[kJ/m^2]$ and a *double cantilever beam* (DCB) method to evaluate the critical strain release energy (G_{Ic}) [J/m²]. The shear test suggested constant weld strength of 21 kJ/m² independent of laser heat input, while the DCB method resulted in varying G_{Ic} with an optimum at a laser power of 50 W and a laser speed of 153.8 mm/s. This difference was ascribed two different fracture mechanisms; *chain rupture* and *chain pull-out*.

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1. Introduction

Polymer joining, and in particular polymer welding, plays an ever increasing importance in today's industry [1,2]. Polymer welds are present in various applications including electronics, packaging, automotive, medical devices etc. Welding is considered one of the most critical steps in manufacturing [3], hence a better understanding is required. Also, from a scientific point of view, weld strength establishment is of importance because of, among others, novel research on smart or self-healing materials [4,5].

In modern manufacturing, laser welding of plastics has experienced increasing attention from industry since the 90s due to relatively low investment cost, flexibility towards mass customization, and high quality welds [6]. Since laser welding in plastics is a relatively new research area, only a sparse amount of research is available. However, research on modeling and process optimization has been reported [7,8]. For instance the molten time for polyamide-6 in a polymer laser weld is reported to be seconds with a peak temperature of 333 °C at a laser power of 60 W and a laser speed of 10 mm/s [9]. Another group reports the molten time of a PP/PP laser transmission weld to be approximately several seconds (>4 s) having $P_{\text{laser}} = 100$ W and $v_{\text{laser}} = 110$ mm/s [10]. On the other hand, mechanical testing of the established laser welds is not well investigated.

Reptation of molecules at the interface is responsible for most of the strength established in a welded joint. The reptation time reveals information on how long time it takes a polymer of a given molecular weight at a given temperature to diffuse one radius of gyration (R_g) [11,12]. In an earlier publication from this group, entitled "Investigation on High Strength Laser Welds of Polypropylene and High-Density Polyethylene" [13], it was concluded that laser welding of polypropylene and high density polyethylene was relatively easy, first of all because of a reptation time within milli seconds.

Polystyrene (PS) is often reported to possess poor weldability, which is explained by a reptation time of several minutes. Wool has reported the reptation time of PS with $M_w = 245,000$ g/mol at 118 °C to be 1860 min [11]. Thus, PS should not be weldable to even itself. However, in



^{*} Corresponding author. Tel.: +45 29293623.

E-mail addresses: tbjuhl@gmail.com, thjuhl@m-tech.aau.dk (T.B. Juhl), jc@m-tech.aau.dk (J.deC. Christiansen), i9eaj@m-tech.aau.dk (E.A. Jensen).

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practice this is not the case [14]. According to weld charts for laser welding, PS/PS is marked as an 'excellent weld' [15]. This might be explained from the reptation time being very temperature dependent, i.e., an increase in temperature should decrease the reptation time dramatically.

This paper focuses on laser welding in PS. The reptation time of a PS grade will be evaluated to see if interpenetration depth at the PS/PS interface is on the order of $R_{\rm g}$. Moreover, laser welded samples will be tested for mechanical properties, i.e., overall strength and critical energy release rate ($G_{\rm Ic}$). Additionally, the fracture mechanisms of the laser welded joints are discussed.

2. Theory

2.1. Polymer diffusion at interfaces

One way to quantify the diffusion of polymers at the interface is to use reptation models. The idea of reptation is to model the polymer melt entanglements as a chain constricted by a surrounding tube. The chain can then move back and forth within the tube and can only escape at the tube ends. The time it takes to escape the tube is defined as the reptation time and, therefore, during the reptation time a polymer chain has diffused one radius of gyration [11,16,17]. More information on determination of reptation time and polymer physics is given in [13].

The reptation model makes it possible to associate the macroscopic zero shear viscosity (η_0) with the microscopic reptation time (τ_{rep}) for an entangled linear polymer melt [17]:

$$\tau_{rep}(T) = \frac{20M_e}{\pi^2 R T_{\rho}} \eta_0(T), \tag{1}$$

where *R* is the gas constant, *T* is the temperature in Kelvin, M_e is the molecular weight between entanglements and ρ is the polymer melt density. Additionally, τ_{rep} is also provided as [17]:

$$\tau_{rep}(T) = \frac{\zeta(T) N_K{}^3 b_K{}^4}{\pi^2 k_B T a^2},$$
(2)

where $N_{\rm K}$ (= $M_{\rm W}/M_{\rm K}$) is the number of Kuhn segments, $M_{\rm W}$ is the molecular weight, $M_{\rm K}$ is the molecular weight of a Kuhn segment, $k_{\rm B}$ is Boltzmann's constant, $b_{\rm K}$ is the length of a Kuhn segment, a is the tube diameter and $\zeta(T)$ is the temperature dependent monomeric friction coefficient. ζ is a material constant, which only depends on temperature and follows the Volger-Fulcher equation [16]:

$$\ln\zeta(T) = A + \frac{B}{T - C},\tag{3}$$

where A, B, and C are fitting parameters.

2.2. Critical energy release rate (G_{Ic})

The most common parameter to describe the *fracture* toughness is the critical energy release rate (G_{Ic}). G_{Ic} [J/m²] is interpreted as the energy required to increase the area of a crack by 1 m². The "I" refers to the first mode of fracture,

also known as the opening mode [18]. G_{Ic} is a material constant which for polystyrene is reported as 1 kJ/m² [18,19]. This value only contains one significant digit, since G_{Ic} depends on temperature and crack propagation rate [20,21]. G_{Ic} is also reported as 0.3 to 0.8 kJ/m² [22].

Another point is that G_{lc} depends on molecular weight (M_w) . G_{lc} is constant above a certain M_w threshold M^* . As a rule of thumb $M^* = 8M_c$, where M_c is the critical molecular weight, which for PS is 32,000 g/mol [23,24]. G_{lc} below $8M_c$ can be calculated as [11]:

$$G_{Ic} \approx \left(0.3 \cdot 1000 \frac{J}{m^2} \cdot \frac{M_W}{M_C}\right) \left(1 - \left(\frac{M_C}{M_W}\right)^{\frac{V_2}{2}}\right)^2 \tag{4}$$

2.3. Fracture mechanisms

Basically, two different mechanisms of fracture in polymer interfaces exist; *chain pull-out* and *chain rupture* [5,25]. Chain pull-outs dominate when dealing with small stresses for long periods of time, e.g., when the crack propagation rate is very low. On the other hand, bond rupture is predominant when the stresses are large; for instance at large deformation rates [26].

3. Experimental

3.1. Materials

For laser welding in amorphous plastics of known molecular weight, a polystyrene PS MFCD00084450 from Sigma-Aldrich with $M_w = 192,000$ g/mol was selected. ENSACO 260 G carbon black from Timcal was utilized as laser absorber. Data relevant for determining reptation time and monomeric friction coefficient are presented in Table 1.

3.2. Rheometry

As indicated in section 2.1, the main purpose of using rheometry was to determine η_0 as a function of temperature, which was used to determine the reptation time as a function of temperature as presented in Equation 1.

The zero shear viscosity was determined with a Paar Physica MCR500 rheometer in a plate-plate configuration with a 25 mm disc and a gap height of 1 mm. In order to measure within the linear elastic regime, the oscillatory tests were performed with small strain amplitude, $\gamma = 0.05$. The angular frequency varied from 0.0628 to 628 rad/s. η_0

Table 1		
Polymer	physics	data.

Symbol	Description	PS
b _K [nm] ^a	Kuhn segment length	2.69
a [nm] ^a	Tube diameter	8.52
M _e [g/mol] ^a	Molecular weight between	13,280
	entanglements	
M ₀ [g/mol] ^a	Monomer molar mass	104.14
ρ _{melt} [kg/m ³] ^b	Melt density	969

^a Ref. [23].

^b Ref. [27].

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