Predicting the laser weldability of dissimilar polymers

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ABSTRACT

The determination of whether two dissimilar polymers are weldable is still largely trial-and-error. Here we suggest a correlation between laser weld strength and the ratio of the equilibrium interpenetration depth (w_∞) of two immiscible polymers to the maximum entanglement tube diameter of the two polymers (a_{max}). Taking the weld strength from a published chart of qualitative weld strength, we find a correlation coefficient between this and the ratio w_∞/a_{max} of 0.67, where w_∞ is based on the simple Hildebrand solubility parameter.

This encouraged a quantitative study of the laser weld strength. This was done using a through transmission laser welding process with 0.4% carbon black as an absorber. The laser welds were tensile tested and relative weld strengths obtained. This strength was compared to the w_∞/a_{max} ratio, where w_∞ is based on experimentally determined Hansen solubility parameters. The results suggest weldability between dissimilar polymers is obtainable when w_∞/a_{max} is greater than 0.15.

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1. Introduction

Polymer weld interfaces are ubiquitous. Welded polymer joints appear in plastic products, ranging in size from microns to meters [1,2]. Besides conventional welded joints, polymer weld interfaces are also present in paints [3], injection molding weld lines [4,5], self-healing materials [6,7], and polymer blends [8,9].

Weld strength development at dissimilar polymer interfaces has also been investigated [10–12], especially for hot tool processes [13,14]. In an earlier publication from our group, high-strength welds between high density polyethylene (HDPE) and poly-propylene (PP) have been achieved in a through transmission laser welding (TTLW) process [15].

Polymer laser welding has drawn a lot of attention over the last decade due to its relatively low cost, high quality, and flexibility towards mass customization [16]. The specific process of interest is the TTLW process, where the basic idea is that a laser transparent (not optical transparent) polymer is placed on top of a laser absorbing polymer. The incident laser beam heats up and melts the absorbing polymer, which wets, heats up, and melts the trans-parent polymer. When both polymers are molten and wetting has occurred, the polymers will inter-diffuse and entangle. The latter requires that the polymers are weld compatible [3,16,17].

To determine whether two polymers are weldable, i.e., weld compatible, weldability charts are often used [18]. An extract of an industrial weld chart is presented for 6 selected materials in Table 1. The numerical values are translations of the weld quality from the chart, where a "good welded joint" = 1, "satisfactory welded joint" = 2/3, "poor welded joint" = 1/3, and "no welded joint" = 0. The quantification is suggested by the authors of this paper.

The TTLW process is complex and difficult to understand and predict, and so is the weldability between dissimilar materials. Thus, the chart from Table 1 is based on trial-and-error approaches and accumulated experience from industry. The chart reveals that all 6 polymers are self-weldable and that material combinations are almost symmetric. This means that the weld strength is nearly independent of which of the two polymers absorb the laser beam.

The problem with these charts is that they are not based on a specific scientific standard. Also the charts require a huge amount of work to produce. Therefore, the challenge within this field of

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Table 1
A part of the weldability matrix from LPKF Laser and Electronics AG. 'a' refers to absorbing, while 't' refers to transparent [18].

	tHDPE	tPP	tPMMA	tPS	tPBT	tPC
aHDPE	1	0	1/3	0	0	0
aPP	0	1	1/3	0	0	0
aPMMA	1/3	1/3	1	2/3	1/3	0
aPS	0	0	2/3	1	2/3	1/3
aPBT	0	0	0	0	1	1
aPC	0	0	1	1/3	1	1

research is to investigate if any pattern exists from polymer–polymer weldability and physical/chemical properties of the welded polymers.

1.1. Reptation and polymer inter-diffusion

Reptative motion leading to inter-diffusion and entanglements is often taken to be the main mechanism of strength development at polymer–polymer interfaces [3,13,17]. Also, when welding dissimilar polymers, inter-diffusion and entanglements are necessary for achieving full strength [19–21]. The reptation time is the average time it takes a polymer of a given molecular weight at a given temperature to diffuse one radius of gyration [22–24].

From earlier publications from our group, the reptation times have been estimated for HDPE, PP, and PS (same grades as in this paper) to be a few milliseconds at temperatures lower than the typical melt zone temperatures in the TTLW [15,25]. That welding time is irrelevant for laser welding is also evident from the fact that all linear polymers are weldable to themselves [18].

1.2. Polymer miscibility

Polymer miscibility is relevant when welding dissimilar polymers. Polymer–polymer miscibility can be predicted using Flory–Huggins (FH) theory for polymer–polymer mixtures [26]. According to FH theory, mixing occurs if the Gibbs free energy (ΔG) is lowered. According to FH, the Gibbs free energy of a two-component polymer–polymer system is [23,27]:

$$\Delta G = \chi_{12} \cdot \varphi \cdot (1 - \varphi) + \frac{\varphi}{N_1} \cdot \ln(\varphi) + \frac{1 - \varphi}{N_2} \cdot \ln(1 - \varphi), \quad (1)$$

where N_i is the degree of polymerization of component number i, φ is the volume fraction of the one component, while $(1 - \varphi)$ is the volume fraction of the other component, and χ_{12} is the Flory–Huggins interaction parameter also known as the chi-parameter. In equation (1) the first term is the enthalpic term, which is positive if χ_{12} is positive, which is usually the case [28]. The last two terms are the entropic contribution, which is always negative. Therefore, entropy favors mixing, while enthalpy disfavors mixing (if χ_{12} is positive). When dealing with commercial polymers for structural components, the degree of polymerization is often large (>1000); thus, the entropic gain approaches zero, resulting in an immiscible polymer blend. Consequently, only a few combinations of high molecular weight polymer pairs are known to be miscible; consistent with the theoretical prediction that two polymers with a high molecular weight ($M_W \sim 10^5$ g/mol) will be immiscible, even when their solubility parameters (δ) are only slightly different [27].

When welding, polymer miscibility is obviously preferred; thus, from equation (1), the χ-parameter should be as low as possible, and ideally below the critical χ-parameter (χ_c), which determines complete miscibility [23]. The χ-parameter is given in terms of the Hildebrand solubility parameters as [29]:

$$\chi = \frac{V_m \cdot (\delta_1 - \delta_2)^2}{RT}, \quad (2)$$

where RT is the gas constant multiplied with absolute temperature, δ is the Hildebrand solubility parameter [$(MPa)^{1/2}$], and V_m is the molar volume [cm^3/mol]. The molar volume of the two polymer system is given as the geometric mean ($\sqrt{V_{m1} \cdot V_{m2}}$) [30]. Note that χ is dimensionless. Another way to estimate χ is using Hansen solubility parameters (HSP) [30]:

$$\chi = \frac{V_m \cdot \left((\delta_{D1} - \delta_{D2})^2 + \frac{1}{4}(\delta_{P1} - \delta_{P2})^2 + \frac{1}{4}(\delta_{H1} - \delta_{H2})^2 \right)}{RT}, \quad (3)$$

where the subscript D refers to the dispersion component, P refers to the polar component, and H refers to the component involving hydrogen bonds. Yet another and more precise way to determine χ is using small angle X-ray scattering (SAXS) and small angle neutron scattering (SANS) [31–35].

1.3. Helfand's theory

Even though polymer pairs often are immiscible, i.e., form two phases when mixed, there might still be a small interfacial zone between the two phases where both polymers can be present. This zone has a length of w_∞ and is referred to as the equilibrium interpenetration depth. w_∞ can be estimated from Helfand's theory stating [20,36]:

$$w_\infty = 2 \cdot \sqrt{\frac{b_1^2 + b_2^2}{12 \cdot \chi}}, \quad (4)$$

where b_i is the statistical segment length of polymer i. The infinity symbol implies that the molecular weight is assumed to approach infinity; i.e., to be high enough that the width of the interpenetration zone no longer depends on chain length. This is a very good approximation for commercial polymers [20].

1.4. Polymer network dimensions

For strength to develop at the interface, not only is w_∞ of importance, but also is the entanglement mesh spacing of the polymer melt. If the mesh size is large, w_∞ also needs to be large to ensure entanglement. And, vice versa, if the mesh size is small, entanglements can easily occur. A measure of the entanglement mesh size is the tube diameter (a) [24].

1.5. Hypothesis

To achieve mechanical strength in a polymer interface, it is necessary to maximize the number of interfacial entanglements. This is done by selecting polymer pairs with a large equilibrium interpenetration depth (w_∞) and a small tube diameter (a_{max}), where a_{max} is taken as the largest of the pair of tube diameters for the two materials. Thus, maximum weld strength should be attained by maximizing the ratio w_∞/a_{max}. Moreover, the temperature of relevance in equations (2) and (3) is the crystallization temperature (T_c), when going from molten to solid state, since this is the temperature where the polymer melt 'locks'. For amorphous polymers the glass transition temperature (T_g) found from cooling is used, see Section 2.5.

To investigate if w_∞/a_{max} results in improved weldability, the materials from Table 1 are evaluated using equations (2) and (4) (data is provided in Table 3). If the weldability is quantified as the