Nd:Yag laser irradiation of single lap joints made by polyethylene and polyethylene doped by carbon nanomaterials

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Abstract. Thermoplastic polyethylene can be welded by the transmission laser welding technique (TTLW) that exhibits some process related benefits with respect other conventional joining methods. This justifies its large use in wide fields, from the automotive to medical or domestic appliances. In this research, we studied single lap joints made by polyethylene pure and filled with carbon nanomaterials (0.2% in weight) to make the polymer laser absorbent. The joints were irradiated by a Nd:YAG laser operating at 1064 nm (first harmonic) with an intensity of $10^7$ W/cm² and 1÷30Hz, a maximum pulse energy of 300mJ and a laser spot of ≈1 cm² (no focusing lens were employed). The joints were characterized by morphological analysis, mechanical shear tests and calorimetric analysis. The results suggested that the laser exposition time must be opportunely balanced in order to avoid a poor adhesion between the polymer sheets and to realized efficient joints. In particular the mechanical test showed that the laser exposition time of 40 seconds is the best conditions to obtain the highest shear strength of the joints of 140 N. After too prolonged laser exposure times, degrading phenomena starts.

1. Introduction

The laser welding of polymers exhibits some process-related benefits compared to conventional joining methods such as bonding, ultrasonic, vibration or hot plate welding. For example, gluing of plastics mostly requires surface pre-treatment and needs organic solvents. Hot plate or hot air welding is cost-efficient, but slow and subject to wear. Ultrasonic or vibration welding expose the work pieces to high mechanical load [1].

Laser polymer welding has great advantages since it means low thermal and mechanical load to the component. It is clean, accurate and it has a high automation degree, but it does not uses any solvents. Moreover, laser welding does not generate any micro particles and this is a significant advantage in particular for fluid reservoirs and medical components [2]. Also, there is no melt ejection and therefore no distortion with laser welding. Moreover, the components do not have to be preprocessed before welding; this fact also contributes to a constant welding quality.

The main applications of laser welding of polymeric materials are in automotive engineering, electronics, communications, medical device technology, human care and domestic appliances (such as food and beverage packaging) [3,4].
During the laser welding process, the laser beam passes through the upper transparent polymeric layer until it hits the welding zone, where it is absorbed by the lower welding partner. The pressure of the tooling clamps enhances thermal conduction into the transparent joining partner, which also causes a local plasticization of the polymer to create a secure adhesive bond [5]. So this technique is suitable for applications where very high weld seam quality is specified [6].

In particular, in this work we applied the laser welding technique to a wide use thermoplastic polymer, ultra-high molecular weight polyethylene (UHMWPE). The special interest for this specific kind of polyethylene derives from its peculiar chemical and physical properties and the mechanical resistance. The polymer is a white, semitransparent and semicrystalline and it has a low absorption coefficient for visible light. In order to increase its absorption the UHMWPE was filled with pigments, like carbon nanostructures. We have employed a Nd:Yag laser at the wave length of 1064 nm and checked the mechanical strength of the joints welded at different times of laser exposure. The results showed that a good sealing action can be obtained at low exposure time. The highest shear load was of $\approx 140$ N, and it was exhibited in the joint realized with a laser exposition for 40 seconds, due to a deep and intimate interpenetration of the melted polymer at the sheet interfaces. We highlighted that higher exposition times induce severe damages of the polymeric structure.

2. Materials and methods

Ultra High Molecular Weight Poly Ethylene (UHMWPE-GUR 1020 $\rho = 0.930 \text{ g/cm}^3$, $M_w \approx 3 \times 10^6 \text{ g/mol}$) was supplied by Ticona and referred as “UH”. Powder of carbon nano materials (supplied by Good Fellow) with particle size of 50-100 nm order, was employed as filler and referred as “NC”. Nanocomposites were made by mixing the powder of UHMWPE and 0.2% weight percentages of NC with pure ethanol (Fluka), as dispersing medium; it was referred as “UH-NC”.

The mixing was kept in ultrasound bath at 25°C/2h. Than the solvent was separated and both the pure UH and the UH-NC nanocomposites were moulded in a hot press at 200°C/20 minutes ($P=20 \text{MPa}$), obtaining sheets 60mm$\times$60mm and 1mm of thickness by using releasing teflon films (123 micron) supplied by P.A.T.I. s.p.a.. The UH sheets had an appearance semitransparent while the UH-NC ones, black.

The welded joint was obtained by coupling two rectangular polymer sheets (30x20 mm, 1 mm thick), partially overlapped for a length of 15 mm and pressed between each other with pressure of about 70 kPa. The UH/UH-NC joints were then laser exposed for different times, within the range 30-90 seconds at 25°C/2 minutes by a 9 ns Nd:Yag laser operating at 1064 nm (first harmonic) in single pulse or at 1÷30 Hz repetition rate, with an intensity of about $10^7 \text{ W/cm}^2$, a maximum pulse energy of about 300 mJ and a laser spot of $\sim 1 \text{ cm}^2$ (no focusing lens were employed). The laser pulse energy had a Gaussian shape with a diametric FWHM of about 5 mm. The incident angle of the laser beam was 0°. They were referred as “UH-NC30, UH-NC40, UH-NC60, UH-NC75, UH-NC90” where the last number indicates the laser radiation time.

The Absorption Coefficient ($\mu$) of the laser light transmission in polyethylene was measured through the transmitted energy in thin films (90 $\mu$m thickness) with 1064 nm wavelength and 10 mJ pulse energy.

The shear test was carried out on the joints at 25°C by a LLOYD LR 10K universal testing machine with a crosshead speed of 5 mm/min. The specimens had a rectangular geometry, 20mm $\times$30 mm and 1 mm of thickness. For each irradiation dose 10 specimens were tested in order to give the average value.

Differential Scanning Calorimeter, DSC mod. SDT Q600 (TA Instruments) was performed from 30°C up to +180°C (heating rate of 5°C/min) under nitrogen flow (100 ml/min). The measurements gave an endothermic peak (melting peak) whose minimum value gave the melting temperature ($T_m$). Lamellar thickness ($L_c$) was calculated according to the Thomson-Gibbs equation [5].

The morphological observation of the laser spot was performed at 40x magnification by means of an optical microscope Mod. ZEISS Stemi 2000 C.
3. Results and discussion

The welding mechanism occurs at the interface where the laser highly light pass through the laser-transparent UH sheet placed on the top (irradiated face) and is absorbed by the dark UH-NC sheet laser-absorbent placed on the bottom (un-irradiated face; see figure 1a). The presence of NC powder in the polymeric sheet is necessary in order to make it laser-adsorbent [7]. The absorption coefficient values ($\mu$) of the pure UH and the nanocomposites film containing the NC are 25 cm$^{-1}$ and 81.1 cm$^{-1}$, respectively. This suggests that the carbon nano materials at $\lambda_0$=1064 nm gives an absorption power to the UHMWPE more than double its starting value.

![Figure 1. Scheme of a single lap joint irradiation by a laser source (a). Image of the UH/UH-NC30 (b), UH/UH-NC75 (c), UH/UH-NC90 joint (c). Optical microscope image of the of the laser spot in the UH-NC30 (e) and UH-NC90 joint (f).](image)

In order to verify if the absorption power of the doped polyethylene sheets can be enough to produce a good weld, several joints were prepared changing the laser radiation exposure time and then they were checked by means of shear tests. Three examples of joints with the dimensions of the sheets geometry and of the overlapped area are shown in figure 1 b-d. In particular the image of figure 1b,c shows the UH/UH-CN30 and UH/UH-CN75 joints where are present the circular spots in the black sheets. Their observation suggests that the laser light pass through both the two overlapped polymeric sheets. The laser spot has a regular circular shape, yellow-orange colored; its diameter is of about 11 mm. Its difference in color suggest that the NC structures are ejected from the polymer matrix by the laser energy. Besides, its morphology is homogeneous suggesting that the change of the material induced by the laser energy absorption is perfectly distributed in all the area. The heat can be regularly distributed in all the laser spot area so that no any visible degrading phenomena occurs. In figure 1d is presented the image of the UH/UH-CN90 joint; here the spot appears irregular and damaged, suggesting the starting of degrading phenomena after protracted exposition times such as 90 seconds. The laser spot diameter is of 10.1 mm, shorter compared to that of the UH/UH-CN30 joint above described. The optical images of the inner laser spot area of the UH-NC30 joint (fig.1e) and of UH-NC90 one (fig.1f) detached after the shear stress are shown in figure 1e,f. The images at 40x magnification highlights the presence of some little swollen areas in the spot of the UH-NC30 joint that became highly rough in the UH-NC90 joint with well visible damages. All the above described images suggest that the laser exposure time of 30-75 seconds is enough to produce a well sealed area among the two polymeric sheets while the time of 90 could be excessive.

The average load/displacement curves of the joints: UH /UH-NC30-60-75-90 are shown in figure 2. The curves indicates that the shear resistance of the joint improves with the laser exposure time in
the order: UH/UH-NC30 < UH/UH-NC60 < UH/UH-NC75, since the maximum load grows from 61 N to 153 N, enhancing of about 167%. Then it decreases again in the UH/UH-NC90 joint, since it results of 138N.

![Figure 2. Shear load-strain curves of the UH/UH-NC30-60-75-90 joints.](image)

The laser spot area of joints laser exposed for different times, within the range 30-90 seconds, progressively decreases from 105.3 mm$^2$ to 80.5 mm$^2$ and the shear stress values in the same range of radiation time increases from 1.58 MPa to 1.78 MPa.

In particular, the shear load is initially very low suggesting that the adsorbed energy is enough to give an appreciable weld. So, in these conditions, the welded joints exhibit a "poor" mechanical strength. After longer irradiation times, higher than 40 seconds, the shear stress value reaches a maximum that retains high up to 60 seconds of exposure. So, the sealing action induced by low irradiation exposure times (within the range 10-20 seconds) is not strong; the shear stresses are low, around 0.6 MPa. But when the exposure time overcomes a threshold value, such as 40 seconds, the adsorption power of the sheet becomes very high so that the heat developed at the interface is enough to engage a more intimate interconnection between the two polymeric sheets and the joint effectiveness grows reaching the maximum value. At very high exposure times, such as 90 seconds the shear load little decreases, due to the starting of degrading phenomena. So in the exposure range 40-75 seconds the sealing action is maximum. Anyway, since the shear strength values are very close, the time of 40 seconds can be considered enough to achieve a good joint. Therefore, it is considered as the optimal exposure time.

The difference in the welding area among the samples suggests that a low exposition time involves the more external layers of the polymeric sheet where the heat developed can be better distributed on a wider area. Instead a higher exposure time, such as 90 seconds, produces a great amount of heat during the irradiation; this generates a smaller welding area but deeper compared to the UH-NC30 one where the resulting shear strength are higher. More investigation tests will be necessary in order to demonstrate the correlation among the strength and the contact deepness of the layers involved in the lacerated joints. We have other experimental evidences already observed in iron oxide doped polyethylene sheets, red in appearance, studied in a previous paper [8].

Finally, the calorimetric DSC analysis was performed inside the laser spot of the colored sheets with the aim to observe the laser effect on the polymer with increasing the laser exposure time (figure 3). The measured heat flux versus the temperature gave the endothermic peaks whose minimum value indicates the melting temperature ($T_m$). The calorimetric parameters (melting temperature and lamellar thickness) were measured as a function of the radiation time of 0-90 seconds. The $T_m$ decreased from 134.6°C to 132.5°C (figure 3a) while the lamellar thickness improved from 23.9 nm to about 20.9 nm with increasing the laser exposition time (figure 3b). These changes highlighted a modify in the structural organization of the polymer after the laser irradiation due to the high temperatures reached during the irradiation. In particular the polymer organize itself in a different way since the crystals seems to be bigger in wideness but few in amount so that the melting temperature generally decreases.
4. Conclusions
In this paper were prepared joints based on UHMWPE and carbon nanostructures (0.2 weight%), welded by a Nd:Yag laser operating at $\lambda = 1064 \text{ nm}$ at different laser exposition times. Carbon nano-materials can be well employed as colored pigment to produce single lap polymeric joints. A good sealing action in polymeric joint can be obtained after a laser exposition of 40 seconds. The resulting joint showed a 150 N shear strength and a regular welded area. Structural polymeric modifications are induced during the laser irradiation in the polymeric material that lowers its melting temperature and enhance the lamellar wideness. A higher laser exposure times worsen the mechanical strength of the joint since damages are produced in the laser spot. Works are in progress in order to deep investigate about the carbon nanostructure amount affect in the laser welding process of the polyethylene.

References
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