Weldability of pipe grade polyethylenes as realized from thermal and mechanical properties assessments

Abstract: Since polyethylene (PE) has been widely accepted for the production of high-pressure fluid conveying pipelines, studies devoted to weldability of PE connections were always of major importance. In this study, two industrial PE grades designed for pipe production, namely PE80 and PE100, were injection molded, cut, and then welded as PE100-PE100, PE100-PE80, and PE80-PE80. The heat-welded joints were assessed by differential scanning calorimetry and tensile measurements. The results obtained from thermal and mechanical analyses were compared with equivalents for aged samples. Thermal analysis revealed that the melting point of the PE100-PE100 sample is obviously larger than the one for the PE80-PE80 joint, for the PE80 chains deteriorate the crystallization of PE100. Further, the PE80-PE80 sample showed the lowest lamellar thickness and crystalline molecular weight among the studied joints. The aging process was found to increase lamellar thickness and molecular weight, though in the PE100-PE100 sample such quantities very limitedly increased. The yield stress of aged joints was higher than that for just-prepared samples, while an inverse trend was seen for strain at break.

1 Introduction

Progress in the knowledge and engineering applications of polymeric materials has resulted in a great change in human life and led to an industrial revolution through which conventional materials have been replaced by new synthetic polymers. This uprising demand has brought about a new class of materials at lower cost and provided lighter goods having relatively reasonable mechanical properties [1–3]. Nevertheless, there have been at all times some difficulties in the way polymers are commercialized and introduced to the market, mostly arising from processing and/or in-service problems.

From the beginning of the polymer industry, a major proportion of the products were designed to be used in fluid conveying systems. With engineers lies prime responsibility to connect pipeline counterparts without sacrificing mechanical properties, specifically when they are subjected to dynamic multi-axial stresses. The invention of welding techniques has indeed provoked a drastic change in the product joints, but at the same time engineers in recent years have faced some new challenges. As a whole, welding is a process that involves the elimination of intermolecular distances at the interface of two pieces and the formation of a strong molecular-scale interface between them [4–6].

The proposed welding methods can be classified into two categories, namely hot and cold welding. In the hot welding process, the edges of single parts are fused, mixed with molten electrode or gap-filled metal and a joint is formed after cooling, while the intermolecular distance at the interface is detached by mechanical pressure in cold welding without melting the parts [7]. Electronic and laser
welding processes are among melt welding and ultrasound and friction welding are included in cold welding [8–10]. For polymeric pieces, however, mechanical welding methods are rarely used because of the high molecular weight of polymers as well as their low modulus, strength, molecular diffusion coefficient and thermal stability compared to metals [11–15]. On the other hand, the hot welding process is an adequate and cost-effective method for polymer welding due to an enhanced molecular motion at higher temperatures and a lower melting temperature with respect to metals. Molecular self-diffusion can be highlighted as one key in polymer welding that depends upon the temperature, crystalline phase and chain stiffness of the polymer counterparts [16–22].

A well-established application for polymer welding refers to water supply systems and oil and gas conveying pipelines. For carrying non-polar fluids, nylon pipes are applied, while for polar fluids such as water, polyethylene (PE) pipes are the most functional ones. This polymer has been noticed for manufacturing a wide variety of pipes; hence, it plays a vital role in conveying gas and water, especially in agricultural landscapes. Bearing in mind the drinking water shortage all around the world, the welds of PE pipes should gain more importance because a failed welding process leads to wastage of a huge amount of water.

To the best of the authors’ knowledge, the majority of publications in the field of polymer welding are patents in which parameters governing the efficiency of welding are not systematically or obviously monitored. Moreover, the existing reports suffer from being unable to track changes in the molecular scale, e.g. fluctuations in molecular weight and crystallinity. It is believed that such molecular criteria remote the resistance of weld interface against an external stress. In the present work, we intended to look at the PE pipe heat welds from a closer angle to understand the role of molecular-scale transitions on the weldability of PE counterparts. Two commercially available PE grades designed for high-pressure pipe manufacture, i.e. PE80 and PE100, were injection molded, cut, and welded. The joints were prepared via the butt welding process, and the crystallinity and mechanical behavior of just-prepared and aged welds were investigated. An attempt was made to uncover the interrelationship between microstructure and welding performance quantifying the molecular-scale parameters of the welded specimens.

2 Materials and methods

PE granules were purchased from Bandar Imam local Petrochemical Co. (BIPC), Mahshahr, Iran. The as-received properties of both PE grades (PE100 and PE80) are listed in Table 1.

Samples with dumbbell shapes and standard dimensions (150 mm length, 25 mm width, 3 mm thickness) were prepared using an injection molding machine (Krauss Maffei, Aktiengesellschaft 350-4300 CX, Munich, Germany). Mold fittings were in accordance with the standard procedure of ASTM DIN16963. The welding process was carried out at 200°C and 2 MPa pressure following the standard DVS 2201 using a butt-fusion welding machine (Rothenberger, Roweld P315 B, Johannesburg, South Africa). This test was run in such a manner as to avoid air bubble formation in the samples. The welded samples were named as P100-P100, P100-P80 and P80-P80. Figure 1 illustrates some of the welded samples. The length of the welded area was 2 cm×1 cm×0.5 cm.

The mechanical properties of the welded samples were measured by a Gotech tensile test machine (GT AI500, Taichung, Thailand) at 5 mm/min strain rate and ambient temperature in accordance with ISO 6259-3. The weld line was placed exactly in the middle of the dumbbell samples as can be seen in Figure 1. For the sake of accuracy, each sample was tested five times and the average

<table>
<thead>
<tr>
<th>Property</th>
<th>Standard</th>
<th>Unit</th>
<th>PE80</th>
<th>PE100</th>
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<tbody>
<tr>
<td>MFR (190°C/5.0 kg)</td>
<td>ISO 1133</td>
<td>g/10 min</td>
<td>0.45</td>
<td>0.25</td>
</tr>
<tr>
<td>MFRR (190, 21.6/190, 2.16)</td>
<td>ISO 1133</td>
<td>–</td>
<td>144.8</td>
<td>128.6</td>
</tr>
<tr>
<td>Density</td>
<td>ISO 1183</td>
<td>g/cm³</td>
<td>0.945</td>
<td>0.960</td>
</tr>
<tr>
<td>Minimum required strength</td>
<td>ISO 9080</td>
<td>MPa</td>
<td>8</td>
<td>10</td>
</tr>
<tr>
<td>Yield strength</td>
<td>ISO 527-2</td>
<td>MPa</td>
<td>22</td>
<td>25</td>
</tr>
<tr>
<td>Elongation at break</td>
<td>ISO 527-2</td>
<td>%</td>
<td>–</td>
<td>&gt;600</td>
</tr>
<tr>
<td>Tensile modulus</td>
<td>ISO 527-2</td>
<td>MPa</td>
<td>850</td>
<td>–</td>
</tr>
<tr>
<td>Charpy notch impact</td>
<td>ISO 179</td>
<td>kJ/m²</td>
<td>12</td>
<td>16</td>
</tr>
<tr>
<td>Hardness</td>
<td>ISO 868</td>
<td>Shore D</td>
<td>60</td>
<td>60</td>
</tr>
</tbody>
</table>
The degree of crystallinity of the samples was estimated using Eq. (1), as follows:

\[ X_c = \frac{\Delta H_m}{\Delta H_c} \times 100\% \]  

(1)

where \( X_c \) is the degree of crystallinity (%), \( \Delta H_m \) is the measured melting enthalpy (J/g) and \( \Delta H_c \) is the melting enthalpy of PE at 100% crystallinity, the value of which is reported to be 293 J/g [23].

3 Results and discussion

The DSC analysis results of the welded samples and their melting points and crystallization degrees are plotted in Figure 2. The PE100-PE100 weld in Figure 2B represents relatively a very high melting point and crystallinity percent, while the PE80-PE80 sample shows the lowest melting point among the studied specimens. Noticeably, the melting point decreases by the attachment of PE80 to PE100, but the PE100-PE80 sample comparatively shows the lowest degree of crystallinity.

It can be explained that PE100 macromolecules in the PE100-PE100 sample create crystals that are fairly more ordered with an acceptable lamellar thickness, when compared to PE80-PE80 connection. This was liable for higher degree of crystallinity and melting temperature in case of PE100-PE100 connection. On the other hand, blending of PE80 with PE100 forms chains with lower possibility of crystallization in the interfacial welding zone. Thus, the presence of PE100 chains poses the diluting effect on the crystallization process of the PE80 chains and somehow hinders them from being able to form ordered spherulites. In other words, PE80 chains with lower molecular weight, larger entropy, and more rapid configurational changes at elevated temperatures harden the contribution of PE100 chains to the spherulite formation, thereby reducing the melting temperature and degree of crystallinity of the resulting binary sample. In opposition to this weld, the PE80-PE80 sample behaves like a homo-polymer with lower molecular weight compared to PE100-PE100 and permits constructing crystals having smaller thickness. In such systems, more spherulites are formed in the absence of a hindrance such as PE100 molecules. As a result, the PE80-PE80 specimen shows a larger crystallinity degree and a smaller melting temperature. In order to evaluate the crystallization behavior of welded samples more precisely, we can apply some mathematical models, through which molecular criteria are considered. The amount of molecular weight

![Figure 1: Polyethylene welded samples: (A) PE100-PE100 and (B) PE100-PE80.](image-url)
involved in the crystalline phase \((M_w)\) and lamella thickness \((L)\) are determined according to Eqs. (2) and (3), respectively [24]:

\[
M_w = \frac{2RT_{\text{max}}}{\Delta H(T_{\text{max}} - T)} \tag{2}
\]

\[
L = \frac{2\sigma T_{\text{max}}}{\Delta H(T_{\text{max}} - T)} \tag{3}
\]

where \(T_{\text{max}}\) (K) is the maximum melting temperature corresponding to a perfectly crystallized polymer, \(R\) is the molar gas constant and \(\Delta H\) is the specific heat of fusion for a 100% crystalline polymer. Also, \(\sigma\) is the lamella surface energy \((\text{J/cm}^2)\) and \(T\) (K) stands for the melting temperature of the polymer [25]. Diffusion of macromolecules in the molten state and recrystallization behavior in the course of cooling of welded surfaces is affected by the molecular weight distribution that can be estimated by the melt flow rate ratio (MFRR) index (see Table 1) [26]. Molecular diffusion controls the performance of the welding process. Using melting DSC curves, the crystalline phase molecular weight \((M_w)\) and lamella thickness were calculated (Table 2). The crystal thickness and molecular weight for the PE100-PE100 sample were higher than those for the PE80-PE80 welded sample, where the one for PE100-PE80 sample situated between two welded homo-polymers.

The crystallization behavior assessment suggests that the physical properties of PE100-PE100 are better than those of the PE80-PE80 sample, which can be explained in view of the higher molecular weight of the former. Accordingly, a similar trend in mechanical properties could be expected. Figure 3A illustrates the stress-strain curves obtained for the welded samples. Accordingly, a necking phenomenon was observed for all the samples; thereby the samples experienced fracture as they were elongated from the welding zone. Moreover, all specimens showed yield stress as a result of tension. Notably, the stress-strain behaviors of the PE80-PE80 and PE100-PE100 specimens were comparable to each other, whereas the heterogeneous sample, i.e. PE100-PE80, showed a different trend and failed in bearing high elongations until it broke. Bearing in mind the crystallization behavior of the PE100-PE80 sample discussed earlier, it can be interpreted that the PE80 part in this sample was solely stretched and experienced necking, while the PE100 part did not contribute to necking. Despite this fact, all PE100-PE80 welds failed from their interface. Another notable difference in the mechanical behavior of this joint with regard to the other two was its lower elongation at break, 160% on average, which is in vivid contradiction to the PE80-PE80 and PE100-PE100 samples broken at strains above 400%. Since PE100 has larger crystalline phase domains than PE80, much more polymer segments gradually migrate from its crystalline phase to the amorphous regions during stretching experiments; hence the plastic phase domains increase and a higher elongation to break is gained. From the perspective of dissimilarity, two PE grades in the PE100-PE80 weld end at a much weaker interface than

Figure 2: (A) Non-isothermal DSC thermograms showing the melting behavior of samples and (B) the alteration of melting point and degree of crystallinity for welds with different compositions.

<table>
<thead>
<tr>
<th>Sample code</th>
<th>(L) (nm)</th>
<th>(M_w) (g/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PE100-PE100</td>
<td>33.25</td>
<td>1025.68</td>
</tr>
<tr>
<td>PE100-PE80</td>
<td>24.68</td>
<td>857.78</td>
</tr>
<tr>
<td>PE80-PE80</td>
<td>20.49</td>
<td>708.15</td>
</tr>
</tbody>
</table>
other ones and this constitutes a major impediment to the crystallization process of both phases. Our observations during the tensile test revealed that necking takes place at the weld line. Regarding the PE100-PE80 sample, one can hypothesize that the jump in the curve between 100% and 300% strains is a signature of slow crack formation in the vicinity of interface of the two samples. Figure 3B schematically shows the proposed mechanism for the stretched samples. An increased micro-cavity contribution to the weld zone of the PE100-PE80 joint is expected when the sample experiences stretching. As a result, some micron-sized cavities are formed at the interface of two PE grades.

Figure 4 confirms the generation of micro-cavities or micro-voids at the weld interface of the PE100-PE80 sample during the tensile test, which stems from the reconfiguration of the polymeric chain segments and crystalline lamella. The shape and the number of cavities depend on the original molecular arrangement. Since the formation and population of cavities in a stretched sample are responsible for crack initiation, larger micro-cavities are believed to reason smaller elongation to beak. Such cavities are resulted mostly from different crystallization rates of phases at the welding region. The difference in the crystallization rate is induced by dissimilar macromolecular structures of PE100 and PE80 polymers, despite the fact that some are more and less from a unique family. It is to be noted that the long-term properties of polymeric parts and especially welded zones, e.g. crystallization behavior, might experience severe fluctuations during harsh service circumstances.

All the welded samples were aged to investigate the effects of time and temperature on their physical and mechanical properties. The thermal analysis results of the aged samples are illustrated in Figure 5A. A comparison of the results in Figure 2A demonstrates that the melting peaks are shifted to higher temperatures and occupied larger areas after aging, suggesting a significant change in the crystallization behavior. In a similar fashion, the molecular weight and thickness of the crystals for aged samples are calculated by using Eqs. (1) and (2), and the results are presented in Figure 5B. For the sake of comparison, the ones for originally tested samples are also plotted in this figure.

It can be observed in Figure 5B that the aged samples have relatively higher molecular weight in the crystalline regions and reveal a larger crystal thickness with respect to the originally tested ones. Through the aging period, which is long enough at the same level of chain diffusion time, an opportunity is provided to the macromolecules to

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**Figure 3:** (A) Engineering stress-strain curves of the prepared welds and (B) the proposed mechanism for the PE80-PE100 welded sample.

**Figure 4:** Weld fracture cross section of the PE100-PE80 sample.
migrate from the amorphous domains to spherulitic ones making them more ordered. As a direct result of this phenomenon, either the crystalline phase molecular weight or lamella thickness is enhanced; thereby melting temperature and crystallinity degree are augmented.

In parallel with alterations observed in case of crystallization behavior, the mechanical properties of aged samples were changed. The stress-strain curves of the aged samples are shown in Figure 6A. By comparing these results with the ones illustrated in Figure 3A, it is proved that a considerable change in the mechanical behavior has been introduced due to aging. To evaluate such changes more accurately, the yield stress and strain at break of aged and normal samples are measured and featured graphically in Figure 6B and C. It can be realized from these figures that aged joints comparatively have much smaller elongations at break and demonstrate larger yield stress. The plausible explanation relates to crystallinity alteration due to aging, where a rise in the crystallinity percent brings about some obstacles to the chain configurational state [19]. In other words, the crystalline regions, as physical crosslinks, worsen the polymer plasticity, diminish the intermolecular micro-cavities and improve the material stiffness and modulus. Consequently, the yield stress increases by aging, in vivid contradiction to the strain at break [16]. This explanation is consistent with observations irrespective of welding type. In contrast to the PE100-PE100 sample, which shows the maximum yield stress among the studied samples, the PE100-PE80 joint has the lowest amount of yield stress due to the heterogeneous nature of interface and lower phase compatibility. As a result of higher Gibbs free energy of the system, the crystallinity degree of this sample takes the smallest value and does not exceed as much as others because of aging. Another striking feature of Figure 6 is the highest strain at break of the PE80-PE80 sample among the aged samples, which can be ascribed to the very low degree of crystallization of this sample. The chains of PE80 have lower molecular weight and larger intermolecular micro-cavities; hence the larger freedom of molecular movements impedes the segmental rearrangements to enter the crystalline cells during aging period.

4 Conclusions

In this work, PE, as one of the well-known materials for producing high-pressure polymeric pipes, was chosen and the properties of different welded joints prepared from two commercial pipe grades, i.e. PE100 and PE80, were discussed. Three different types of welds, namely PE100-PE100, PE100-PE80 and PE80-PE80, were prepared using the heat welding process. Thermal and tensile analyses were carried out to correlate welding performance with molecular-level situation. The results of non-isothermal DSC tests demonstrate that the PE100-PE100 sample has comparatively the highest crystallinity weight fraction and melting point among the studied cases, whereas the PE100-PE80 and PE80-PE80 specimens show the lowest crystallinity degree and melting point. The findings verify that PE80 chains only play the role of a diluent agent in the PE100 crystallization process and consequently decrease the resultant crystallinity weight fraction of the PE100-PE80 sample. In addition, the prepared samples were aged and different
crystallization parameters including lamella thickness and crystalline average molecular weight were determined. The obtained data provide a proof that the aging process enhances the crystal thickness and molecular weight on account of macromolecule diffusion in crystalline cells during aging period. However, PE80-PE80 has shown the lowest lamella thickness and crystalline molecular weight. Since aging promotes the crystallization process, the aged specimens show larger yield stress and considerably lower elongation at break. It can be concluded that the welding of similar polymers results in the strongest joint and among the studied samples here, the PE100-PE100 specimen offers the most effective weld.

References


Figure 6: (A) Engineering stress-strain curves of aged samples. A comparison of (B) yield stress and (C) strain at break of aged and just prepared samples.


