RALF LACH[1) *)], PAVEL HUTAŘ[2)], PETR VESELÝ[1)], EVA NEZBEDOVÁ[3)], ZDENEK KNÉSL[2)], THOMAS KOCH[4)], CHRISTIAN BIERÖGEL[1)], WOLFGANG GRELLMANN[1)]

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Assessment with indentation techniques of the local mechanical behavior of joints in polymer parts

Summary — The paper summarizes our investigations performed with micro- and nanoindentation techniques and results presented in literature, concerning local mechanical behavior of welded joints [polyamide 6, polypropylene, polyethylene (high density and crosslinked polyethylene), poly(vinyl chloride)] and weld lines (polypropylene, polycarbonate, polystyrene) of amorphous and semicrystalline polymers. For amorphous polymers, minimum of hardness and indentation modulus in the joint are found or no change in the properties were stated, whereas for semicrystalline polymers, hardness and indentation modulus mostly pass through a maximum in the weld line, the welded joint or the heat-affected zones.
Keywords: welded joints, weld lines, microindentation techniques, nanoindentation techniques, hardness, indentation modulus.

OCENA ZA POMOCĄ METODY WCISKANIA WGŁĘBNIKA LOKALNYCH WŁAŚCIWOŚCI MECHANICZNYCH ZŁĄCZY CZĘŚCI WYKONANYCH Z MATERIAŁÓW POLIMEROWYCH

Streszczenie — W pracy podsumowano wyniki badań wykonanych metodą wciskania wgłębnika w zakresie mikro i nano oraz wyniki przedstawione w literaturze, dotyczące lokalnych właściwości złączy spawanych [poliamidu 6, polipropylenu, polietylenu (dużej gęstości i usieciowanego), poli(chlorku winylu)] i linii spoiny polimerów amorficznych i semikrystalicznych (polipropylenu, poliwęglanu, polistyrenu). W przypadku polimerów amorficznych wykazano w złączu minimum twardości i modułu sprężystości przy wciskaniu wgłębnika lub stwierdzono brak zmian tych właściwości. W przypadku polimerów semikrystalicznych twardość i moduł sprężystości osiągają maksimum w linii spoiny, złączu spawanym lub w strefach wpływu ciepła.
Słowa kluczowe: złącze spawane, linia spoiny, technika mikroindentacji, technika nanoindentacji, twardość, moduł sprężystości przy wciskaniu wgłębnika.

INTRODUCTION

Joints in polymer parts such as welded joints, adhesive bonds, weld lines and others have a great importance especially for more complex assemblies and larger components used, for example, in structural constructions, used in piping, production of chemical equipment and large machine parts [1]. Due to the widespread application of jointed parts, the formation of joints is also the subject of intensive scientific research. To evaluate the quality of joints, there are some suitable experimental testing and evaluation methods. In comparison with the non-destructive testing methods widely used, there are several mechanical testing techniques available which also allow an experimentally supported quantitative evaluation of the joint quality. For welded joints, for example, the tensile test gives the welding coefficient as a quantitative parameter being the quotient of the tensile strength of the weld and that of the basic material [2—4]. In each case, the welding coefficient allows a quantitative evaluation and ensures certain comparability; it does not, however, permit evaluation under a practice-oriented view, because it is an integral characteristics of the whole specimen, which does not take the locality of the weld into account. In contrast, extensive scanning of the weld influence zones by means of recording microhardness testing permits a relatively detailed description of the hardness and elastic properties, depending on the location of testing on the one hand. On the other hand, for the evaluation of the local and temporal load and damage course, hybrid methods such as laser extensometry are necessary, by which not only the local resolution of the

1) Martin-Luther University Halle-Wittenberg, Centre of Engineering, 06099 Halle/Saale, Germany.
2) Academy of Sciences of the Czech Republic, Institute of Physics of Materials, Ziskova 22, 616 62 Brno, Czech Republic.
3) Polymer Institute Brno, Tkalcovska 36/2, 656 49 Brno, Czech Republic.
4) Vienna University of Technology, Institute of Materials Science and Technology, Favoritenstraße 9, 1040 Vienna, Austria.
*) Corresponding author; e-mail: ralf.lach@psm.uni-halle.de

mechanical properties, but also parameters reflecting the local mechanical behavior can be determined [4, 5].

Compared to metals, joints in polymer parts have been investigated using indentation techniques very rarely [5—11]. Aside from the group of Prof. Grellmann [5, 9] and Dr Koch [10, 11], only the Balta Calleja team [6—8] have been interested in this problem in recent years. First results of microhardness measurements performed on amorphous polymers such as polycarbonate (PC) and polystyrene (PS), passing through the weld line, were presented by Garcia Gutierrez *et al*. [8] in 1999 (see also references [6, 7]). Furthermore, results preliminary presented by Veselý *et al*. and Lach *et al*. for polyethylene (PE-HD) [5, 9] and by Koch for polypropylene (PP) [10, 11], as well as unpublished data on polyamide 6 (PA 6), polypropylene (PP), poly(vinyl chloride) (PVC) and crosslinked polyethylene (PE-X) are introduced into the following discussion.

The aim of this study is to gather indentation data, taken from different sources, concerning welded polymer parts to evaluate the influence of the morphology of the polymer material (amorphous or semicrystalline). Our own results either unpublished or published only in conference proceedings [5, 9] or book chapters [10] will be presented together with those described in the literature.

EXPERIMENTAL

Materials

High density polyethylene (PE-HD) material was used in the form of commercial single-wall polyethylene plastic pipes manufactured by Pipelife Czech s.r.o. (see www.pipelife.cz). These pipes (outer diameter 110 mm and wall thickness 6.3 mm, *i.e.* with standard dimension ratio *SDR* = 17.6) made by extrusion from PE 100 (a PE-HD grade) are typically used for transportation of gases (CSN EN 1555) [12].

Injection molded slabs (60×138×4 mm) made from a commercial grade polyamide (PA 6) were supplied by Leuna Miramid GmbH. Furthermore, slabs of the same dimensions cut of extruded plates of a commercial grade poly(vinyl chloride) (PVC) provided by Simona AG were used.

Compression molded, 4 mm thick plates of a commercial polypropylene (PP) grade supplied by Polymer Institute Brno (α-nucleated and non-nucleated, *i.e.*, with and without nucleating agent) were applied to study.

Specimens cut from electron-beam crosslinked polyethylene (PE-X) pipes, provided by Das Kunststoff-Zentrum (SKZ) Halle/Saale were investigated.

Welding of investigated materials

PE-HD. The conditions (pressure p and temperature T) of heated tool butt welding performed at the Polymer Institute Brno were varied (optimum conditions, optimum pressure and lower and higher temperature, optimum temperature and lower and higher pressure). Rectangular specimens (80×10×6.3 mm) across the welded region were cut from the pipes and analyzed using different techniques. For further details see reference [9].

PA 6 and PVC. Both, PA 6 and PVC slabs, were cut into two halves and subsequently re-jointed by heated tool butt welding performed at the Kunststoff-Zentrum (KuZ) gGmbH Leipzig. Dumbbell shaped specimens cut from the slabs were also used for the tensile tests using laser extensometry (for more information see reference [4]). Furthermore, slabs cut from extruded plates of commercial grade PVC were also welded at the KuZ using the same technique.

PP. In the case of PP, two sets of specimens were used. On the one hand, compression molded, 4 mm thick plates of a commercial grade PP were welded using a plate--welding installation (T = 190—200 °C, p = 0.1 N/mm^2). For further information see references [10] and [11]. On the other hand, dumbbell shaped injection molded specimens also made from a commercial PP were applied. Due to the two-point gating process (injection molding machine 320 S 500-150/150 from Arburg), weld lines were formed in the middle of these specimens.

PE-X. Specimens cut from PE-X pipes were welded using an advanced heated tool welding technique, by Das Kunststoff-Zentrum (SKZ) Halle/Saale (for general information see references [13—15, 16]).

Methods of testing

Microstructure analysis

The microscopic investigations of the morphology close to the welding connections were carried out by cutting ultrathin sections using a microtome. These cuts several micrometers thick were analyzed by transmission optical microscopy (Leica RM-DX in case of PE 100 and Zeiss Axiolab in case of PP, respectively) in polarized light. In order to determine the crystallinity degree of PE 100, differential scanning calorimetry (DSC 820, Mettler--Toledo) was applied.

Micro- and nanoindentation tests

The hardness and elastic properties of most samples (welded samples of PE-HD, PA 6, PVC and PE-X as well as injection molded PP specimens) were determined at the room temperature using a depth-sensing microhardness tester Fischerscope H 100C XYp equipped with a Vickers diamond indenter. Samples were prepared from the specimens and polished with differently fine-grade sandpapers. The indentation results are average values from the minimum of three measurements across the welding connection as it is shown in Figure 1. From load-indentation depth diagrams measured on the sam-

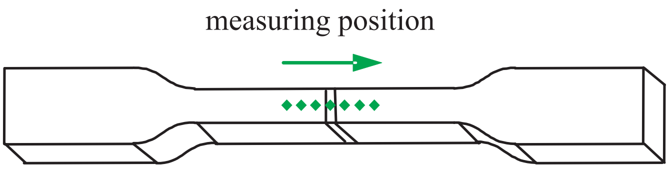

Fig. 1. Schematic representation of the measuring positions at the sample during indentation tests

ples up to the maximum load of 1 N with a constant small loading rate similar to the unloading rate (loading and unloading within 20 s), the Martens hardness *HM* and indentation modulus (*E*) or value of expression $E/(1-\nu^2)$ (ν is the Poisson's ratio) were estimated as defined in ISO 14577-1:2002.

For the welded samples of compression molded PP a minimum of eight load–indentation depth diagrams were determined using a recording nanohardness tester Nanoindenter XP (for more information see reference [10]) equipped with a Berkovich diamond indenter, to calculate indentation hardness (H_{IT}) and modulus according to the method described by Oliver and Pharr [17]. Similarly to the micro range loading and unloading occur within 20 s to keep a constant small loading rate similar to the unloading rate, however, including a dwell time of 30 s between loading and unloading. From the welded PP plates sections close to welded regions were cut and imbedded into cold-curing epoxy resin. These samples were polished with differently fine-grade sandpapers, and additionally with fine-grained diamond and magnesium oxide powder.

RESULTS AND DISCUSSION

Local hardness and modulus close to joints in polymer parts

Welded joints

Due to the fact that the injection molding or extrusion processes are related to cooling rates typically in the order of up to 1000 deg/min (*i.e.* the whole crystallization process is mainly completed after a few seconds), the degree of crystallization of semicrystalline injection molded or extruded samples is much smaller than in the case of isothermal crystallization. This also means that a processes running in the timescale of minutes, such as welding, are combined with increasing degree of crystallization in the welded joints compared to the basic material. For PE 100 it was found that the degree of crystallization in the welded joints is 56 %, whereas that in the basic material is only 50 % [5, 9]. Changes in the structure due to welding are also clearly visible in the image of the welded region using polarized light presented in Figure 2. Based on the well-known observation that the degree of crystallization strongly influences the mechanical properties, maximum values of the indentation modulus, for example, are measured in the welded joint (see Figure 3a for PA 6 and Figure 4 for PE). This behavior was also found for PE-X (Figure 3a) [16].

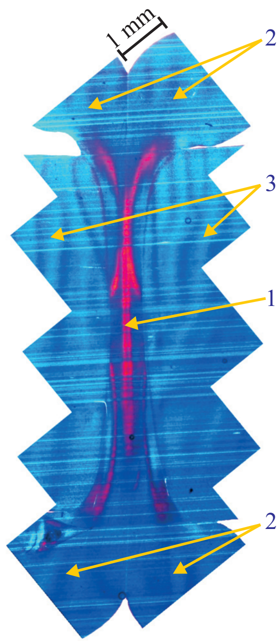

Fig. 2. Polarized-light microscopic image of the welded region of PE 100 (1 — welded joint, 2 — weld beads, 3 — basic material)

The modulus profile of pipe grade PE-HD (PE-100) is shown in Figure 4 for different welding parameters (pressure or temperature) in comparison with optimal conditions [only the modulus $E/(1-\nu^2)$ was plotted in this study, because both *HM* and $E/(1-\nu^2)$ show the same trend]. Both increase in welding pressure and decrease in welding temperature lead to decreasing values of *HM* and *E* moduli in the welded region compared to the optimal conditions. Sharpening of the hardness profile is evident in the material welded with higher pressure (Figure 4a). In the other case, when the temperature was varied, an inverse trend of the mechanical properties between lower and higher temperature was obtained compared to changes in properties induced by variation in pressure (Figure 4b). Whereas the material welded at higher temperature shows enhanced hardness in the welding area, this is not apparent in the material welded at lower temperature, which can be correlated to changes of the microstructure. Especially the influence of temperature on the crystallization process in the welding zone is an important factor for final properties.

The findings obtained for compression molded PP (see Figure 5) differ in principle from the results found for PE and PA 6, which is clearly due to the preparation process of the samples (compression molding for PP instead of extrusion or injection molding for PE or PA 6). For PP, increasing modulus *E* and hardness do not occur in the welded joint but in the heat-affected zones (Figure 5). Beside refinement of the spherulithic morphology, the enhanced nucleation of PP results in an increasing degree of

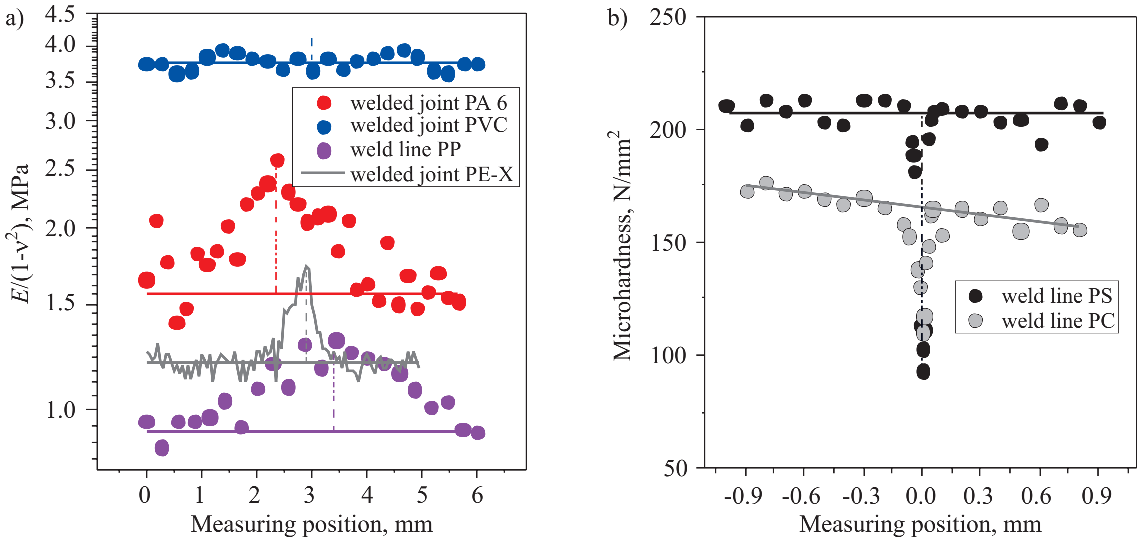

Fig. 3. Indentation modulus (E) expression close to welded joints of PA 6, PVC and PE-X, and weld line of PP (a), microhardness close to weld lines of PS (melting temperature 230 °C) and PC (melt temperature: 270 °C) at the distance of 5 mm from the sample edge (data from [6]) (b)

crystallization in the basic material (which is related to higher values of E and H_{IT}), so that a welding-induced increase of stiffness and hardness in the heat-affected zones is less apparent (Figure 5b).

For amorphous polymer materials such as PVC, no changes of the modulus caused by the welding process were detected (Figure 3a).

Weld lines

As a consequence of increased time for crystallization due to reduction of flow velocity of the polymer melt during injection molding, the degree of crystallization is increased close to the weld line in semicrystalline polymers such as PP. Because both modulus and hardness increase with increasing degree of crystallization for semicrystalline polymers, the modulus, for example, passes through a maximum within the range of the weld line (Figure 3a).

The behavior of amorphous polymers such as PS and PC is very different from that of semicrystalline polymers (see Figure 3b). Microindentation studies based on the optical measurement of the residual impression have shown that the weld line in PS and PC is correlated with a sharp minimum in microhardness [6]. Moreover, the microhardness profile across the weld line depends on the conditions of the injection molding process [melting temperature: 270—300 °C (PC), 230—270 °C (PS)] and the distance from the sample edge (2—5 mm) or the obstacle (Figure 3b) [6—8]. Under specific conditions, therefore, the weld lines for PC and PS — similarly to the welded

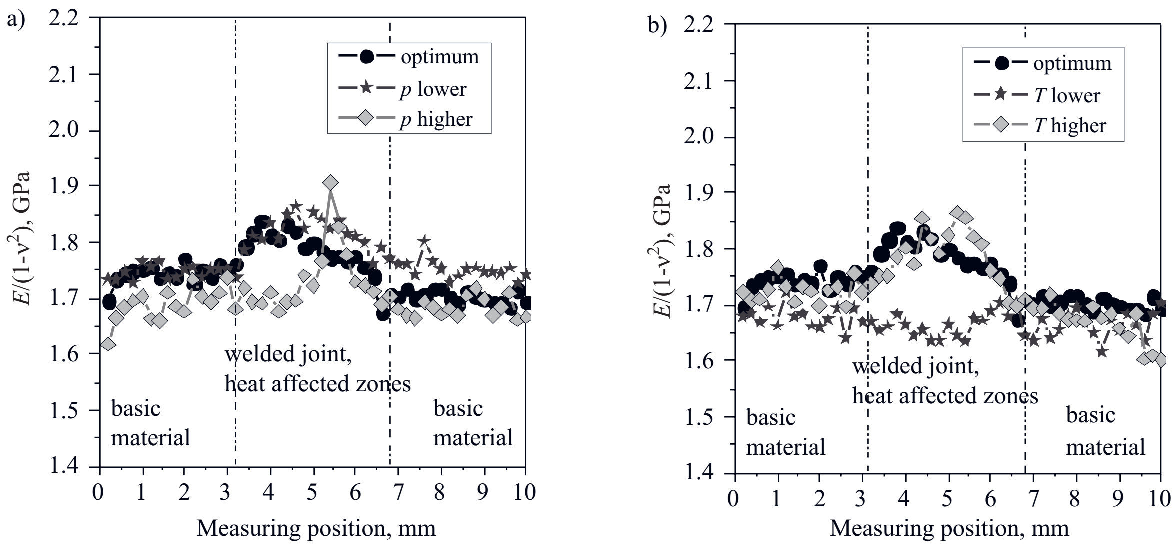

Fig. 4. Indentation modulus (E) expression across the welded region in PE 100 (data from [5, 9]) as a function of welding pressure p (a) and temperature T (b)

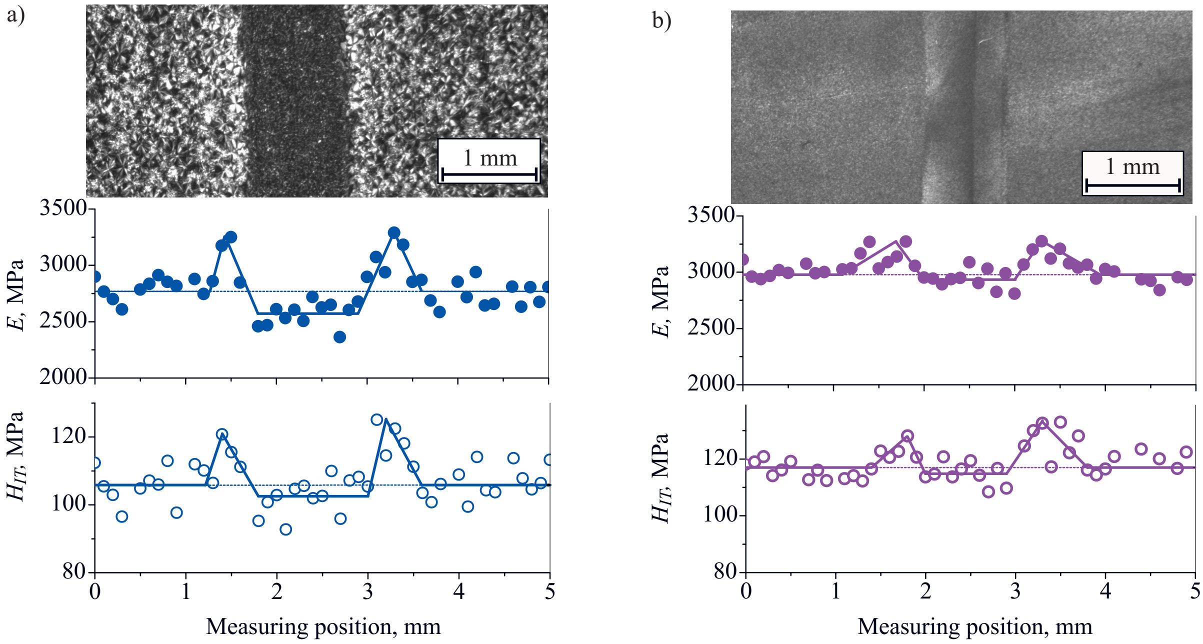

Fig. 5. Microstructure close to welded joints of non-nucleated (a) and nucleated PP (b) and its influence on indentation modulus and indentation hardness (data from [10, 11])

joint for PVC (compare Figure 3a) — cannot be detected using indentation techniques [6, 7].

Application of indentation modulus measurements to assessment of joint stability

To assess the stability of a weld joint from the fracture mechanics point of view, knowledge of the local material properties close to the weld joint is essential. Therefore, a finite element model has been recently developed to simulate crack propagation in a region with inhomogenously distributed mechanical properties [18]. A welded pipe three-dimensional model with a semi-elliptical axial crack located in the centre of the weld, propagating through the pipe wall thickness, as it is shown in Figure 6, was used.

The material inhomogeneity can be given by the ratio between the maximum (E_{max}) and the minimum (E_{min}) value of the elastic modulus in the weld region. This ratio depends on the specific polymer material and welding conditions. In cases of PE/PE and PP/PP, E_{max}/E_{min} are equal to 1.07 and 1.32, respectively (see Figure 3a and 4). In numerical simulations, the change in mechanical properties caused by the welding process is usually ignored. However, for some specific welding conditions the ratio of E_{max}/E_{min} can reach relatively high values and then the stress intensity factor is incorrectly estimated. Ševčík *et al.* [18] proposed a simple engineering equation for the estimation of the stress intensity factor K_I in the welded region:

$$K_I = K_I^{homogenous\ pipe} \times (E_{max}/E_{min}) \quad (1)$$

where: $K_I^{homogenous\ pipe}$ — the stress intensity factor estimated for crack in a homogenous pipe (see *e.g.* reference [19]).

Eq. (1) yields values of the stress intensity factor within 5 % of that determined from numerical simulation when $E_{max}/E_{min} < 1$. In the case of $E_{max}/E_{min} = 2$, the maximum error stretches to 10 %. Nevertheless, the relationship presented in eq. (1) is suitable for a conservative calculation of the stress intensity factor in a welded pipe and takes into account that this factor is always higher in the welded region due to the material inhomogeneity with respect to the homogenous case. Neglecting the local material inhomogeneities can lead to a dangerous underestimation of the pipe lifetime.

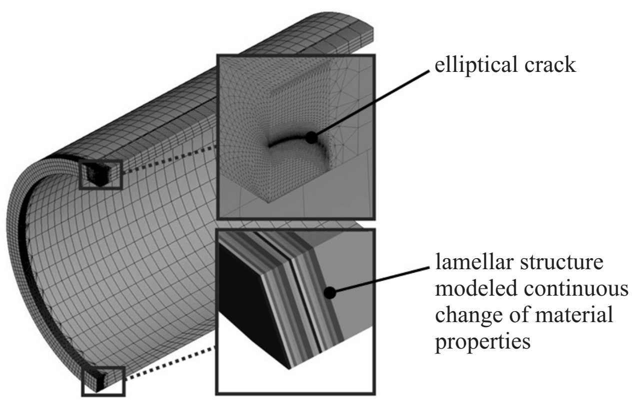

Fig. 6. Typical finite element mesh with a crack and lamellar structure of the modelling polymer weld (according to [18])

CONCLUSIONS

— For amorphous polymers (PC, PS, PVC) either microhardness (PC, PS) or both microhardness and modulus (PVC) present a minimum (PC, PS) or no change (PC, PS, PVC) in the region adjacent to the joint compared to the material outside the joint.

— For semicrystalline polymers (PA 6, PP, PE-HD, PE-X), hardness and indentation modulus mostly pass through a maximum in the weld line (injection molded PP), the welded joint (PA 6, PE-HD, PE-X) or the heat-affected zones (compression molded PP).

— A three-dimensional analysis of a polymer welded pipe with a crack located in the center of the weld predicted an increase in the stress intensity factor due to an increase of the ratio between the maximum and the minimum of Young's modulus values within the welded region, E_{max}/E_{min} (see reference [18] for details). The effect of the material inhomogeneity in the weld region can be significant for ratios $E_{max}/E_{min} > 1.2$.

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