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COMPATIBILITY OF MEDIUM DENSITY POLYETHYLENE (MDPE) FOR DISTRIBUTION OF GASEOUS HYDROGEN

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ABSTRACT

Numerous projects are looking into distributing blends of natural gas and different amounts of gaseous hydrogen through the existing natural gas distribution system, which is widely composed of medium density polyethylene (MDPE) line pipes. The mechanical behavior of MDPE with hydrogen is not well understood; therefore, the effect of gaseous H_2 on the mechanical properties of MDPE needs to be examined. In the current study, we investigate the effects of gaseous H_2 on fatigue life and fracture resistance of MDPE in the presence of 3.4 MPa gaseous H_2. Fatigue life tests were also conducted at a pressure of 21 MPa to investigate the effect of gas pressure on the fatigue behavior of MDPE. Results showed that the presence of gaseous H_2 did not degrade the fatigue life nor the fracture resistance of MDPE. Additionally, based on the value of fracture resistance calculated, a failure assessment diagram was constructed to determine the applicability of using MDPE pipeline for distribution of gaseous H_2. Even in the presence of a large internal crack, the failure assessment evaluation indicated that the MDPE pipes lie within the safe region under typical service conditions of natural gas distribution pipeline system.

Keywords: Fatigue; Fracture; Hydrogen; Polymers

1. INTRODUCTION

With an increased interest towards using renewable energy sources, hydrogen has gained attention due to its effective energy carrying and storing capability as an alternative to the use of carbon-based fuel [1, 2]. The use of existing natural gas distribution systems is being considered to distribute blends of gaseous hydrogen and natural gas, thus reducing the carbon footprint of this energy distribution pathway. It has been estimated that the cost for constructing new hydrogen infrastructure can be 10 to 20% higher compared to its natural gas counterpart [2], but more importantly replicating existing infrastructure for hydrogen is not economically feasible. Hence, the use of natural gas infrastructure for distribution of hydrogen could be an economical solution towards the development and penetration of hydrogen technology.

The natural gas pipelines are typically divided into transmission and distribution systems, the latter being operated at lower pressures. Due to the low operating pressure, newly-installed distribution systems are typically comprised of polymer pipes; additionally, polymer pipe is relatively easy to manufacture and install, low cost and resistant to corrosion and chemicals. Therefore, the effect of gaseous hydrogen on the material integrity of polymeric pipeline materials needs to be thoroughly understood prior to using natural gas pipeline system for transportation of gaseous hydrogen blended with natural gas and eventually as pure gaseous hydrogen.

Klopffer *et al.* [3] studied the effect of 3 MPa gaseous hydrogen on the static properties including tensile, creep, and ductile fracture of high density polyethylene (HDPE). Results showed that the variation observed in the mechanical properties for tests conducted in air and in hydrogen differ by 10%. Similarly, in a study conducted by Simmons *et al*. [4] on HDPE at higher pressure of 28 MPa hydrogen, the modulus was reported to drop by approximately 17%. However, they also

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report that after removing the hydrogen pressure, the modulus was recovered. Castagnet *et al.* [5] reported results from tests conducted on polyethylene thermoplastic pre-exposed to hydrogen at pressure of 10 MPa for 30 days. However, no effect on the tensile properties of polyethylene materials was observed.

Menon *et al.* [6] carried out a thorough investigation on the effect of high-pressure hydrogen on the deformation (swelling), density change, and storage modulus of elastomer (Buna N and Viton A) and semi-crystalline thermoplastic (Polytetrafluoroethylene (PTFE) and HDPE) materials. In general, the effect of high-pressure hydrogen was more prominent in elastomers compared to thermoplastics, which was attributed to the presence of crystalline structure in the thermoplastics and lack of crystalline phases in the case of elastomers. In addition, a set of tensile tests was also conducted on PTFE and HDPE thermoplastics [6]. Interestingly, tensile test results showed an increase in the stiffness of the material in the presence of high-pressure hydrogen.

Studies investigating the hydrogen embrittlement phenomenon on the static properties of polymeric properties are limited in literature [3-7], whereas, to our knowledge, effects of hydrogen on the fatigue and fracture behavior have not been reported in the open literature. In the current study, medium density polyethylene (MDPE), also known as yellow pipe, is the material of interest. One of the major differences between MDPE and HDPE is the crystallinity, which is higher for HDPE and lower in MDPE. The difference in the crystallinity, can eventually affect the hydrogen transport and consequently, their static as well as dynamic mechanical properties [6, 8]. The effect of gaseous hydrogen on the fatigue life and fracture resistance of MDPE are reported in this study. Based on the obtained results, a failure assessment diagram (FAD) was also constructed to inform the assessment of structural integrity of MDPE pipe for distribution of gaseous hydrogen.

2. MATERIAL AND EXPERIMENTAL SETUP

2.1 Material and specimens

As-manufactured medium density polyethylene (MDPE) pipes were provided by Southern Company Services for the current study (i.e., these had not been in service). The evaluated pipe was designated as IPS 6 DR 11.5: Nominal Pipe Size (IPS) of 6” with dimension ratio (DR) of 11.5, specifically outside diameter (OD) of 168 mm and wall thickness of 14.6 mm. Basic material properties for the MDPE thermoplastic based on ASTM D3350 [9] is shown below.

Table 1: MECHANICAL PROPERTIES OF MDPE BASED ON ASTM D3350 STANDARD [9].

Property	Specified Range
Density	0.926-0.940g/cm^3
Melt Index	<0.4 – 0.15
Flexural Modulus	552 - <758 MPa
Tensile Strength at yield	18 - <21 MPa
Color and UV stabilizer	Colored with UV stabilizer

Fatigue life testing was conducted using the circumferentially notched tensile (CNT) specimen; the detailed geometry is shown in Figure 1. Specimens were extracted in the longitudinal direction with a notch root radius of 0.152 mm and net diameter of 7.62 mm across the notch root. These specimens were threaded at the ends for gripping (½ inch-13 UNC 2A thread): thus, the notch root radius was designed carefully to avoid failure at the grip section during the fatigue loading. Based on the notch root radius and specimen geometry, the elastic stress concentration factor (K_t) was calculated to be approximately 5 at the circumferential notch, while the K_t at the threads was determined to be 4. As a result, none of the specimens failed at the grip section.

Compact tension (CT) specimens, with width (w) of 26.4 mm and thickness (B) of 12.7 mm were utilized to determine the fracture resistance of MDPE in air and in gaseous H_2. Additional dimensions of the specimen geometry are shown in Figure 2. CT specimens were also extracted from the IPS 6 pipe section in the CL orientation, i.e. the crack was aligned in the longitudinal (L) direction and loaded in circumferential (C) direction. Prior to testing, a sharp notch with an approximate depth (a) of 12.7 mm was introduced using a razor blade to minimize blunting at the crack tip, which the notch/crack depth is measured relative to the load line (i.e., center of the loading holes).

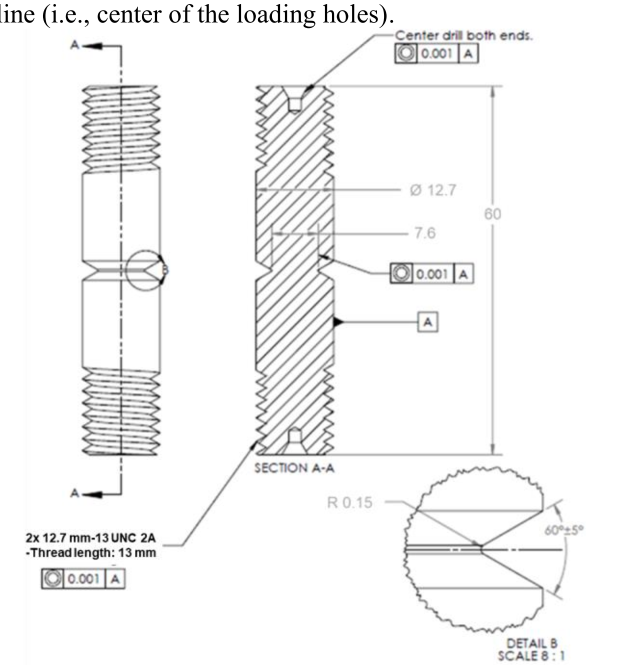

FIGURE 1: CIRCUMFERENTIAL NOTCHED TENSILE (CNT) SPECIMEN GEOMETRY USED FOR CONDUCTING FATIGUE LIFE TEST. ALL DIMENSIONS ARE IN MM.

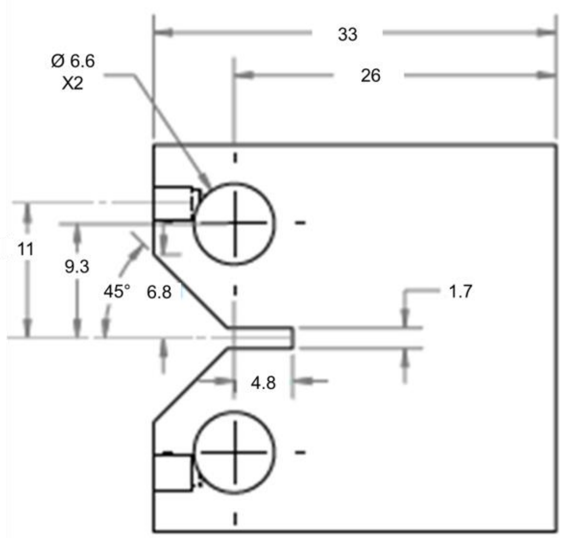

FIGURE 2: COMPACT TENSION (CT) SPECIMEN GEOMETRY USED FOR CONDUCTING FRACTURE RESISTANCE TESTS. ALL DIMENSIONS ARE IN MM.

2.2 Testing environment

Fatigue and fracture testing was carried out in air and in gaseous H_2 at pressure of 3.4 MPa. Additional fatigue life tests were conducted at higher H_2 pressure of 21 MPa to explore the effect of pressure. All of the tests were conducted using a custom-designed pressure vessel with the pressure rating of 138 MPa [10], which was integrated within a MTS servo hydraulic test frame to continuously expose the specimen to gaseous H_2 throughout the fatigue and fracture testing. Furthermore, to eliminate impurities such as moisture and oxygen, the testing volume was first purged to the test pressure with nitrogen three times and then with 99.9999% pure gaseous hydrogen an additional three times. A detailed description of the testing facility and capabilities at Sandia National Laboratories is presented in [10].

2.3 Fatigue life testing

Fatigue life tests in air were conducted on a MTS servo hydraulic test frame with 5 kN load cell, whereas the tests in gaseous H_2 utilized a custom-made load transducer (22 kN) located within the pressure boundary of the integrated test system (to avoid measurement of the frictional loads associated with the dynamic seals on the load system pull rods). Fatigue tests, both in air and in gaseous H_2 environment were conducted in load-control mode in tension-tension configuration with load ratio (R) of 0.1 and frequency of 1 Hz. Tests were conducted with maximum applied stress ranging from 20 to 27 MPa, based on the applied load and reduced (or net) cross-sectional area across the notch root. Conventional plots of (net section) stress versus cycles to failure, which we call fatigue life, were constructed to characterize the fatigue behavior of MDPE in both the high-cycle regime (up to about 200,000 cycles to failure) and the low-cycle regime (as low as about 2,000 cycles to failure). Due to the viscoelastic nature of polymeric materials, the specimen temperature is known to increase upon fatigue loading. Hence, for tests in air, the temperature at the specimen surface was monitored using a thermocouple and found to be unchanged (i.e., ~ 20° C) for these test conditions.

2.4 Fracture mechanics testing

Fracture resistance of MDPE was evaluated using CT specimens in air and in gaseous H_2. The same test systems as utilized for the fatigue testing were used for the fracture tests. The fracture tests were conducted under displacement control at the displacement rate of 0.51 mm/minute; specimens were monotonically loaded to fracture. The actuator displacement was used to control the test and to determine the load-displacement curves.

3. RESULTS AND DISCUSSIONS

3.1 Fatigue life

The maximum applied stress versus number of cycles to failure is plotted in Figure 3 for all tests (air and in gaseous H_2 at pressures of 3.4 MPa and 21 MPa). The value of maximum stress was obtained by dividing applied maximum load by cross-section area at the notch root. To place the tested fatigue stresses into perspective, the hoop stress was calculated for an internal pressure of 0.86 MPa (125 psi), OD of 168 mm, and pipe thickness of 14.6 mm, which is represented by the dashed line in Figure 3. The internal pressure of 0.86 MPa has been reported as maximum allowable operating pressure (MAOP) in some natural gas distribution pipeline systems [11]. It also needs to be noted that the value of hoop stress is not just a function of internal pressure but also pipe dimension. None the less, the range of maximum stresses used for conducting the fatigue life tests were significantly higher than the calculated hoop stress, and the fatigue lives for these high stresses were observed to be in the range of 2,000 to 200,000 cycles.

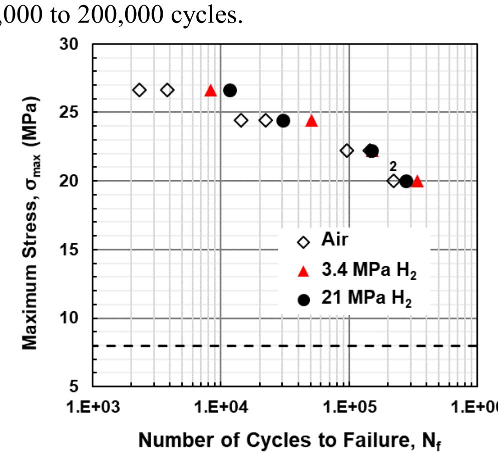

FIGURE 3: MAXIMUM STRESS VERSUS FATIGUE LIFE DATA OBTAINED FOR MDPE IN AIR AND IN GASEOUS H_2 ENVIRONMENT AT 3.4 MPA AND 21 MPA PRESSURE. THE DASHED LINE REPRESENTS THE VALUE OF HOOP STRESS FOR INTERNAL PRESSURE OF 0.86 MPA (125 PSI) WITHIN THE IPS 6 DR 11.5 PIPE SECTION.

Similar to any other material, the fatigue life of MDPE is greater for lower applied stress for all tested conditions. However, the detrimental effects of gaseous H_2 on the fatigue life of MDPE was not observed. More interestingly, the fatigue life appears to slightly increase in the presence of gaseous H_2 compared to air for all tested stresses. This result may be attributed to the lack of moisture in the dry gaseous H_2 compared to laboratory air. Alternatively, the greater fatigue life in hydrogen may be related to microstructural changes resulting from the hydrostatic pressure of the gaseous environment. The latter explanation, however, would also suggest a pressure effect, which is not apparent in these data: the effect of pressure on the fatigue life was not distinguishable for this limited number of tests. Additional evaluation would be necessary to establish a mechanistic explanation for this observation, such as testing in other dry pressurized gases (such as nitrogen or helium). However, we can conclude the hydrogen does not degrade the fatigue life of MDPE for these fatigue test conditions.

3.2 Fracture resistance

In the case of metallic materials, concurrent exposure to gaseous H_2 is generally reported to decrease the material's fracture resistance. Hence, in the current study, the effect of gaseous H_2 on the fracture resistance of MDPE was investigated using the compact tension geometry and ASTM D5045. The fracture resistance is calculated using [12]:

$$K_Q = \left(\frac{P_Q}{B\sqrt{w}}\right) f(x) \qquad (1)$$

$$f(x) = \frac{(2+x)(0.886+4.64x-13.32\,x^2+14.72\,x^3-5.6\,x^4)}{(1-x)^{\frac{3}{2}}} \qquad (2)$$

where K_Q is the conditional fracture toughness, B is specimen thickness, w is specimen width, and x is fractional crack length at the beginning of the test (a_o/w). For tests in gaseous hydrogen, the term fracture resistance is used to distinguish the value as an environmentally dependent value. P_Q is defined as the value of load at the intersection between the load-displacement curve and the construction curve representing a compliance 5% greater than the measured compliance (compliance is defined as the actuator displacement divided by the applied load). A representative load-displacement curve obtained from a test conducted in air is provided in Figure 4, showing P_Q, the measured compliance slope (red solid line) and the construction line representing the compliance value 5% greater than the measured compliance (green dotted line).

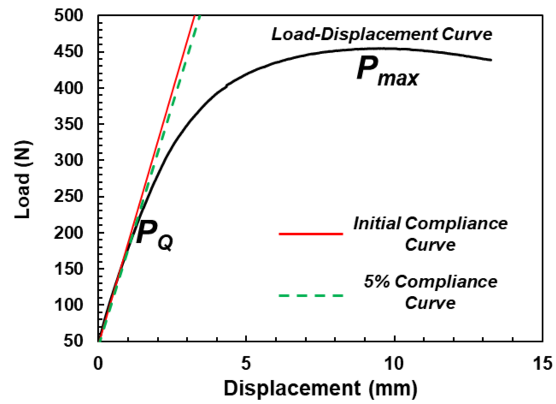

FIGURE 4: REPRESENTATIVE LOAD-DISPLACEMENT CURVE OBTAINED FROM FRACTURE TEST CONDUCTED ON MDPE IN AIR.

The average values of fracture resistance (two tests were performed for each condition), determined based on the load-displacement curve, the value of P_Q, and Eqns. (1) and (2) were calculated to be 1.34 MPa√m and 1.11 MPa√m in air and in gaseous H_2 (3.4 MPa), respectively. The effect of gaseous H_2 on the fracture of MDPE is not significant as the value of K_Q in air and in gaseous H_2 were observed to be similar. In addition, these values are similar to fracture toughness reported in the literature for MDPE tested at room temperature in laboratory air [13]. It is also important to note that ASTM D5045 defines a test to be valid if the ratio of maximum load (P_{max}) to P_Q is less than 1.1. Obviously from Figure 4, P_{max}/P_Q is much greater than 1.1 for the current tests. This result may be attributed to large plastic deformation at the crack tip arising from high ductility of MDPE at room temperature. Hence, the fracture toughness (in air) and fracture resistance (in H_2) do not represent valid plane-strain fracture toughness values. However, these measurements still allow for comparison between the two environments.

3.3 Failure assessment diagram

In the presence of a long crack and internal pressure, a section of pipe may fail due to unstable crack growth or plastic collapse or a combination of these failure modes. Avoiding these failure scenarios is an important consideration in structural integrity management of pipeline systems. Failure assessment diagrams (FAD) can be used to assess whether a structure is subjected to these failure modes or not. Here, a FAD was constructed based on the equations defined in [14] and given as:

$$f(L_r) = K_r = (1 - 0.14(L_r^P)^2) * (0.3 + 0.7\,exp(-0.65\,(L_r^P)^6)) \qquad (3)$$

$$K_r = \frac{K_I^P + \phi K_I^{SR}}{K_{mat}} \qquad (4)$$

$$L_r^P = \frac{\sigma_{ref}^P}{\sigma_{ys}} \qquad (5)$$

Equation 3 defines a general form for the FAD master curve where unstable crack growth is characterized by K_r, defined as the ratio of applied stress intensity factor to material fracture toughness (K_{mat}) and plastic collapse is characterized by L_r^P, defined as the ratio of primary reference stress (σ_{ref}^P) to the material yield strength (σ_{ys}). Furthermore, the applied stress intensity factor resulting from primary loads and the ones attributed to the secondary loads/residual stresses is termed as K_I^P and K_I^{SR}, respectively. For the current analysis, only the effect of primary loads was considered; secondary loads/residual stresses were not considered.

The values of K_I^P and σ_{ref}^P were calculated for an infinitely long, internal surface crack with depth equal to 20% of wall thickness for a pressurized cylindrical shell with an internal pressure of 0.41 MPa (60 psi). Several standard IPS pipes sizes were considered as shown in Table 2 from [15]. The relationships for calculating each of the terms in Eqn. (4) and (5) can be found in [14] for the infinitely long surface crack in the longitudinal direction located in the inside of the pipe (stress intensity factor and reference stress solutions identified as KCSCLL1 and RCSCLL1, respectively). The fracture toughness (K_{mat}) and yield strength (σ_{ys}) of the MDPE piping material were assumed to be 1.1 MPa√m and 20 MPa, respectively. It is also important to note that for the same defect size and configuration, the value of stress intensity factor for an external crack was smaller compared to the value calculated for an internal crack.

Table 2: DIFFERENT IPS PIPE DIMENSIONS [15] USED FOR CONSTRUCTING FAD.

Nominal Pipe Size (IPS)	*DR*	*Outside Diameter (mm)*	*Wall Thickness (mm)*
1	9.33	33.2	3.6
2	9.33	60.4	6.5
3	9.33	88.8	9.5
4	9.33	114.2	12.2
5	11	141.4	12.9
6	11.5	168.2	15.3

The FAD is shown in Figure 5 along with the calculated values of K_r versus L_r for the conditions described above and all of the piping listed in Table 2. The dotted line is the cutoff point defined by the API 579-1/ASME FFS-1 document for the materials whose strain hardening behavior is not well defined [14]. The region under the curve is considered a safe region, where failure resulting from unstable crack growth and plastic collapse will not occur. On the other hand, the region above the master curve is considered to be an unsafe region, where the crack may grow in an unstable manner or the system may fail due to plastic collapse. Even assuming an infinitely long internal crack with a modest depth of 20% of the wall thickness and relatively high internal pressure of 0.41 MPa (60 psi), MDPE piping remains well within the safe region of the FAD.

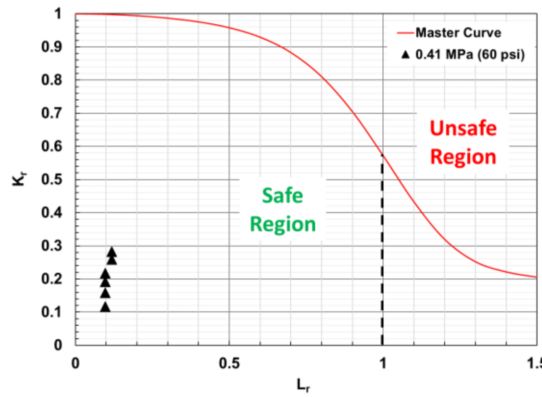

FIGURE 5: FAILURE ASSESSMENT DIAGRAM DEVELOPED FOR MDPE CONSIDERING AN INTERNAL CRACK EQUAL TO 20% OF WALL THICKNESS AND 0.41 MPA (60 PSI) INTERNAL PRESSURE FOR ALL THE PIPING DIMENIONS LISTED IN TABLE 2.

4. CONCLUSIONS

In the current study, the fatigue life and fracture resistance of MDPE were evaluated in pressurized gaseous H_2. In addition, a failure assessment diagram is constructed to evaluate the structural integrity of the MDPE piping, which assess the failure in the context of unstable crack growth and plastic collapse. Based on the obtained results, the following conclusions were drawn:

1. The fatigue life of MDPE was not degraded by testing in gaseous hydrogen at pressure up to 21 MPa. Indeed, the fatigue life was improved, which may be associated with either the absence of moisture in the H_2 testing environment or possibly a change of the MPDE microstructure due to imposed hydrostatic pressure.
2. Similar to fatigue behavior, fracture resistance of MDPE was not significantly degraded in gaseous hydrogen.
3. Using the tensile and fracture properties of MDPE, a failure assessment diagram was constructed for typical IPS pipe dimensions and a large defect, showing that the structure remains within the safe range for typical service conditions of natural gas distribution pipeline systems. Of course, this result is expected since hydrogen did not reduce the fracture resistance of the material.

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