CRACK INITIATION IN HIGH DENSITY POLYETHYLENE PIPE RESULTING FROM CHEMICAL DEGRADATION

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Abstract. The fracture phenomena in engineering thermoplastics such as high density polyethylene (HDPE) resulting from chemical degradation is usually observed in the form of a microcrack network within a surface layer of degraded polymer exposed to a combined action of mechanical stresses and chemically aggressive environment. Degradation of polymers is usually manifested in a reduction of molecular weight, increase of crystallinity in semi-crystalline polymers, increase of material density, a subtle increase in yield strength, and a dramatic reduction in toughness. The critical level of degradation for fracture initiation depends on the rates of toughness deterioration and build-up of the degradation related stresses as well as on the manufacturing and service stresses. In this paper, the evaluation of chemical degradation induced stresses and modeling of fracture initiation in HDPE pipes is presented. The probability of the crack initiation is formulated using the formalism of statistical fracture mechanics (SFM) and chemical degradation induced stresses. The critical level of degradation is determined by using point-wise Weibull distribution of material toughness parameter. Experimental observations obtained from accelerated tests of HDPE pipe at an elevated temperature and chemically aggressive environment are compared with the results of the mathematical model of the described above processes.

Keywords: High density polyethylene, Fracture, Mechano-chemical degradation, Statistical fracture mechanics

1. INTRODUCTION

Stress corrosion cracking (SCC) is one of the main causes of premature failure of plastic pipes in potable water application. SCC results from a complex interaction between a chemical degradation of materials and the thermo-mechanical stresses.

Figure 1 displays examples of SCC in various material-environment systems. There are multiple circumferential cracks visible on axial cross sections of natural gas transmission steel pipe (a), cold water distribution polybutylene pipe (b) and acetal fitting (c). Multiple axial cracks are shown on the circumferential cross section of a potable water distribution polyethylene pipe (d). SCC in thermoplastic pipes usually starts as a microcrack network within a thin surface layer of degraded polymer adjacent to the pipe surface and thus exposed to combine action of mechanical stress and chemically aggressive environment. The microcrack network slow evolves into a colony of cracks illustrated on (Figure 2). Chemical degradation of the polymer is the first step of SCC, which have been studied by many researchers (Ivanova et al., 1995, Bishop et. al., 2000, White, 2006). However, the effect of chemical degradation on mechanical stresses in HDPE pipes is not adequately studied. There is a small number of papers on this topic in spite of a very significant impact of degradation induced stress on the plastic pipes lifetime.

(a) Steel pipe  
(b) Polybutylene pipe  
(c) Acetal pipe  
(d) Polyethylene pipe

Figure 1. Formation of SCC in various materials ((a), (b) and (c) are axial and (d) is the circumferential cross sections)
In this paper, the evaluation of chemical degradation induced stresses and modeling of fracture initiation in high density polyethylene (HDPE) pipes is presented. The probability of the crack initiation is formulated using the formalism of statistical fracture mechanics (SFM). The critical level of degradation is determined by using point-wise Weibull distribution of material toughness parameter. Experimental observations obtained from accelerated tests of internally pressurized HDPE pipe at an elevated temperature and chemically aggressive environment are compared with the results of the mathematical model of the described above processes.

3. EVALUATION OF CHEMICAL DEGRADATION RELATED STRESSES

Disinfectants, such as chlorine (Cl\textsubscript{2}) are commonly added to potable water. The chlorine in water undergoes hydrolysis and forms hypochlorous acid (HOCl\textsuperscript{+}), a strong oxidizing agent. In water, the hypochlorous acid, in its turn, dissociates producing the hypochlorite ion (OCl\textsuperscript{-}): HOCl \leftrightarrow H\textsuperscript{+} + OCl\textsuperscript{-}.

The oxidation of HDPE leads to a reduction of molecular weight (MW). An easy measurable degradation parameter can be expressed in terms of current and initial MW. Such parameter \( \omega(t) = 1 - M_w(t)/M_w(0) \), where \( M_w \) stands for the weight average molecular weight, has been proposed by Niu et al. (2000). The relationships between \( \omega(t) \) and crystallinity, density and toughness variations have been studied using a UV accelerated oxidation technique. Reduction of molecular weight, i.e., increase of the degradation parameter \( \omega \) leads to an increase of crystallinity in semicrystalline polymer as well as density, a subtle increase in yield strength, and a dramatic reduction in toughness.

![Figure 3. Formation of degradation induced stress with degradation layer of HDPE pipe](image)

Figure 3, presents a sketch of the cross section of a plastic pipe with degraded inner layer. Two layers, material I and material II, are defined due to the difference in basic properties of the original and degraded plastic. An interfacial pressure (\( P \)) is build up due to the incompatibility of two layers. The volumetric dilatation \( \Delta \) within the degraded layer
(material I) can be expressed in terms of radial and hoop components strain tensor: \( \Delta = \epsilon_r^I + \epsilon_\theta^I \). On the other hand, the volumetric dilatation \( \Delta \) is directly related to material density variation in process of degradation (Shames and Cozzarelli (1992)):

\[
\bar{\rho} = \frac{\rho}{1 + \Delta},
\]

where \( \rho \) and \( \bar{\rho} \) are the original and degraded materials density respectively.

The hoop and axial stresses associated with degradation as a function of the extend of degradation (degradation parameter \( \omega \)) are presented in Figures 4 and 6. The evaluation of stresses is performed as following.

\[
\frac{\sigma_{w,zz}}{\sigma_{0,zz}} = \frac{E}{2(1+\nu)} \left( \frac{\rho}{\bar{\rho}} - 1 \right)
\]

where, \( E \) is the Young’s modulus of HDPE, and the subscripts \( i \), \( d \) and \( o \) at the radius \( r \), represent inner, interface and outer radius of the pipe respectively.

The degradation induced stresses build up in material I as a function of the radius, \( r \), can be determined using interfacial pressure (2):

\[
\Delta \rho = \frac{E r_o^2 (r_i^2 - r_o^2)}{4 r_i^2 (r_i^2 - r_o^2)} \left( \frac{\rho}{\bar{\rho}} - 1 \right)
\]

where, \( \rho \) and \( \bar{\rho} \) are the original and degraded materials density respectively.
Radial stress: 
\[
\sigma_r = -\frac{E(r_d^2 - r_i^2)}{4(r_i^2 - r_o^2)} \left[1 - \left(\frac{r_i}{r_o}\right)^2\right] \left(\frac{\rho}{\rho} - 1\right) 
\]

Hoop stress: 
\[
\sigma_{\theta\theta} = -\frac{E(r_d^2 - r_i^2)}{4(r_i^2 - r_o^2)} \left[1 + \left(\frac{r_i}{r_o}\right)^2\right] \left(\frac{\rho}{\rho} - 1\right) 
\]

Axial stress: 
\[
\sigma_{zz} = \nu(\sigma_{rr} + \sigma_{\theta\theta}) = -\frac{E(r_d^2 - r_i^2)}{4(r_i^2 - r_o^2)} \left(\frac{\rho}{\rho} - 1\right) 
\]

where, \(\gamma\) is the Poisson’s ratio of HDPE.

It can be seen from (3), (4) and (5) that the major components of degradation induced stresses are hoop stress and axial stress. The radial stress can be neglected.

There are some parametric studies with degradation induced stresses. Figure 5 presents the effect of chemical degradation on normalized hoop stress (the ratio between the degradation induced hoop stress, \(\sigma_{\theta\theta}\), and the hoop stress at 60 psi of internal pressure without chemical degradation, \(\sigma_{\theta\theta,0}\) ) as a function of the inner radius of the pipe.

Figure 6, express the effect of chemical degradation on normalized axial stress (the ratio between the degradation induced hoop stress, \(\sigma_{zz}\), and the hoop stress at 60 psi of internal pressure without chemical degradation, \(\sigma_{zz,0}\) ). In Figure 7 shows the effect of degradation parameter on normalized axial stress.

2. THE CRACK INITIATION MODEL

According to numerous observations, the orientation of fracture and kinetics of fracture growth obey deterministic laws, whereas the time and location of individual microcrack initiation site are essentially random events. Thus, we consider an alternative to classical nucleation theory approach in modeling of fracture initiation by means of Statistical Fracture Mechanics as Chudnovsky (1973) and Chudnovsky and Kunin (1987).

We define Fracture Initiation (FI) as an event of spontaneous expansion of a localized fracture to a macroscopic scale (a crack) triggered by a random defect formed on a sub-microscopic scale. Certain disinfectants added to the water create a chemically aggressive for HDPE environment, which with time results in formation of a layer of degraded material of thickness \(l_0\) adjacent to the inner surface. The degradation process affects the fracture initiation in two ways: it causes a significant decay of specific fracture energy \(J(\omega)\) and a buildup of degradation related stress \(\sigma_{\omega}\) as a noticeable part of total stress \(\sigma_{tot}\). The fastest rate of degradation in this case takes place at the inner surface of the tubing, which is in direct contact with the flowing water. Therefore the first, sub-microscopic defect that triggers FI is, most likely, located at the inner surface. The micrograph displays a layer of degraded polymer adjacent to the inner surface of the tubing. A boundary that separates the degraded and original materials is also well visible due to the differences in color. Multiple fractures initiated at the inner surface of the tubing and extended through the degraded layer are also well visible. Figure 8(b) is a schematic representation of a sample set \(\Omega\) of random fracture paths, that
may be constructed considering a large number of cracks shown in Figure 8(a) shifted toward one origin, \( x_0 \). It can be seen on Figure 8(a) that only one particular path \( w(x) \) from the set \( \Omega \) of virtual paths is realized at each particular location. Moreover, the stress release resulting from fracture formation determines almost regular spacing between individual fractures.

Let consider a fracture path \( w(x) \) that starts at a point \( x_0 \) on the inner surface and terminates at a point \( X \) on the interface between degraded and original material. The condition of fracture extension form \( x_0 \) to \( X \) along \( w(x) \) is the requirement that Griffith condition is met at every point of fracture trajectory \( w(x) \). Here \( G_1 \) is the conventional in Linear Fracture Mechanics notation for the energy release rate (ERR), and the specific fracture energy \( \gamma(x) \) is considered to be a random field that reflects a random variation of material composition, morphology and the extend of degradation on microscopic and sub-microscopic scales. Point-wise distribution of \( \gamma \)-field along the fracture path \( w(x) \) follows Weibull distribution (with shift \( \gamma_{\min} \), scale \( \gamma_0 \) and shape factors \( \alpha \)) respectively, since the crack “selects” the weakest direction at each step of it advance. The above condition of FI, i.e., fracture extension form \( x_0 \) to \( X \) along \( w(x) \) is a natural generalization of Griffith condition for a heterogeneous media. A method to compute a probability of \( FI \), i.e., a probability of spontaneous crack propagation from \( x_0 = 0 \) to \( X = l_0 \) along \( w(x) \) has been proposed by Chudnovsky (1973) and Chudnovsky and Kunin (1987). In the first approximation (the roughness of fracture path is ignored) \( Pr\{FI\} \) can be expressed as:

\[
Pr\{FI\} = \exp\left[-\int_{0}^{l_0} \exp\left(-\frac{G_1(x, \sigma_{tot}) - 2\gamma_{\min}}{2\gamma_0}\right) dx\right],
\]

where \( r \) is the correlation distance of \( \gamma \)-field. A dimensionless shift parameter \( q = \gamma_{\min} / \langle \gamma \rangle \) is here for further analysis. It represents ratio of the shift factor and mathematical expectation \( \langle \gamma \rangle \) of statistically homogeneous \( \gamma \)-field. We also introduced a degradation parameter \( \omega \) which is discussed in the following section.

Energy release rate (ERR) \( G_1 \) for the problem at hand can be expressed as 

\[
G_1 = (1.12\sigma_{tot})^2 \pi \nu E^{-1}
\]

where \( E' = E(\omega)/(1-\nu^2(\omega)) \), \( E \) and \( \nu \) stand for the Young’s modulus and Poisson’s ratio weakly dependent on degradation parameter, and the total stress \( \sigma_{tot} \) comprises of residual (manufacturing related) stresses, service stresses \( \sigma_0 \), and degradation induced stresses \( \sigma_{\omega} \). \( G_1 \) as well as parameters of Weibull distribution depend on the degradation parameter. Therefore \( Pr\{FI\} \) depends on \( \omega \) and can be rewritten as \( Pr\{FI, \omega\} \). We can define a critical level of degradation \( \omega^* \), as the value of the degradation parameter at which fracture initiation takes place with certainty. Then cumulative distribution as well as the statistical moments of \( \omega^* \) can be obtained using eq. (1).

\[
F(\omega^*) = Pr\{FI, \omega\}|_{Pr\{FI\}=1} \geq < \omega^* >= \int_{0}^{\omega^*} dF(\omega^*), \quad \sigma^2(\omega^*) = \int_{0}^{\omega^*} (\omega^* - < \omega^* >)^2 dF(\omega^*),
\]

Figure 9. Critical degradation parameter dependence on internal pressure for various \( q \) values

Figure 10. Schematics of predicting \( FI \) by calculating the critical level of degradation
The original HDPE has sufficient toughness $2\gamma_0$ to make the crack nucleation practically an impossible event. However, there is a decrease of $\gamma(\omega)$ and an increase of $\sigma_{tot}$ with progression of degradation (increasing $\omega$). Thus, the probability of crack nucleation increases to the certainty ($P = 1$) with $\omega$ approaches a critical value. The kinetics of the process of degradation in time $\omega(t)$ together with the value of $\omega^*$ allows one to determine the time $t_i$ of fracture initiation.

The mathematical expectation $\langle \omega^* \rangle$ dependence on internal pressure is depicted in Figure 9 for various values of $q$ parameter ($q = \gamma_{\min}/\langle \gamma \rangle$). Apparently, the greater the applied internal pressure, the lesser level of degradation is required to initiate fracture. A simple procedure for evaluation of time to fracture initiation as the result of polymer degradation is presented in Figure 10. The schematic diagram of Figure 4 for the evaluation of the time to fracture initiation suggests that the onset of fracture in PB is primarily controlled by the degradation process (depletion of antioxidant) and weakly dependent on applied load.

5. CONCLUSIONS

In this paper, the mechanism of the stress corrosion cracking (SCC) initiation in high density polyethylene (HDPE) is investigated. Some conclusions could be drawn from the study:

1. SCC of high density polyethylene pipes is usually observed as multiple cracks with crack clusters formed on the side of the pipe exposed to an aggressive environment.

2. The initiation of SCC is defined by a combination of chemical degradation and mechanical stress. The kinetics of chemical degradation plays the main role in determining of fracture initiation time. The degradation significantly affects the specific fracture energy and creates noticeable built-up stresses, i.e. chemical degradation induced stresses, due to densification of the degraded material. The expressions for degradation related stresses are presented.

3. A formalism for evaluation of the critical level of degradation at the onset of fracture is presented. It is based on the formalism of Statistical Fracture Mechanics (SFM) combined with a point-wise Weibull distribution of material toughness parameter and calculated degradation induced stresses.

6. REFERENCES


7. RESPONSIBILITY NOTICE

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