Fracture of Bimetallic Structural Elements under Hydrogen - Temperature Interaction

O. Andreykiv, O. Hembara, V. Skalsky

Karpenko Physico - Mechanical Institute of the National Academy of Sciences of Ukraine, Ukraine

ABSTRACT: *A theoretico-experimental method for investigation of hydrogen-temperature fracture of bimetallic structural elements, operating in hydrogen-containing environments has been proposed. It was shown that fracture proceeds by coalescence of pores into a crack with its subsequent growth due to filling it with a gas that forms a high internal pressure. Using the theoretical investigation results a temporal dependence of the crack area formation and growth at the fusion boundary of $2.25Cr$-$1Mo$ steel with an austenite surfacing. A correctness of the theoretical ideas of the developed method for evaluation of hydrogen-temperature fracture of bimetals has been proved experimentally. For this purpose the methods of acoustic emission, ultrasonic testing and metallographic analysis have been used.*

NOMENCLATURE

$D = D(x,y,z,t)$ - diffusion coefficient of hydrogen in metal

$K_s = K_s(x,y,z,t)$ - factor of hydrogen solubility in metal

$C = C(x,y,z,t)$ - concentration of hydrogen in metal

$T = T(x,y,z,t)$ - temperature,

$f_{H_2} = f_{H_2}(t)$ - fugasity of hydrogen

R - universal gas constant

Q - heat of hydrogen transport in metal

V_H - partial molar volume of hydrogen in metal

σ - hydrostatic stress

$\vec{j}$ - flow of hydrogen in a body

μ - molar mass of hydrogen

t - time

f_v - the configuration function, which determines volume of hydrogen in crack at unit pressure

$\vec{n}_i$ - unit normal, which is directed inside of a crack

v - crack growth rate

σ_{YS} - yeld strength of the metal

ε - deformation

δ - crack opening displacement

δ_c - critical crack opening displacement

K_1 - stress intensity factor

r - radius of penny shaped crack

INTRODUTION

The equipment in nuclear, chemical, heat - power, and petroleum-refining, industry is protected against corrosion by applying various coatings and overlayers of corrosion - resistant materials.

In operation bimetallic structures there appear cracks. Under hydrogen-effect they cause (at first hidden) lamination of one material from the other along the connecting surface. As a result the unexpectedly failure of operating equipment, can occur even after technological process had stopped, when there was no contact with environment, loading and the equipment was cooled down [1,3]. This is a well known phenomenon. Since the fracture of bimetallic structures occurs in the metal bulk, it is difficult to investigate the process kinetics. An attempt has been done in this paper to model the process of delamination of one material from the other in a bimetal due to the effect of hydrogen and nonstationary temperatures. Literature and experimental data analysis concerning operation of industrial hydrocracking reactors showed that delamination of overlayers proceeds along the austenitic grain interface, that contains a great number of carbides. At high temperature and pressure, hydrogen interacts with carbide carbon forming methane. Methane fills in the micropores, as a result the much higher internal pressure than that caused by hydrogen is formed. At a certain stage the pores coalesce and a crack is initiated. This is the first, incubation period of fracture. It depends on the properties of the intermediate bimetal surface, velocities of its hydrogenation and cooling. The second stage is a new crack propagation. The pores at the crack are disloaded due to decrease of hydrogen concentration, (the hydrogen pours down into a crack) thus increasing a gas pressure. Therefore the physico - mathematical model of internal crack propagation in bimetals due to hydrogen filling them consists of: 1) equations of hydrogen diffusion in bimetal and filling with it the internal cracks as well as creation of overequilibrium pressure in them; 2) equations of crack propagation, accounting for the kinetics of hydrogen filling in the crack and hydrogen embritlment of the metal in the process zone.

PHYSICO - MATHEMATICAL MODEL OF INTERNAL CRACK PROPAGATION IN BIMETALS DUE TO HYDROGEN FILLING

The existing models that describe such events are not flawless, because they do not account for the kinetic of mass exchange between crack and

surrounding metal, thus, do not reflect the kinetic of pressure change in the defect which determines damage and life-time of structure as a whole. We propose numerical model [4] for estimation of hydrogen fugacity, which accounts for coupling of hydrogen diffusion in metal and hydrogen accumulation in a crack.

Let us assume that in metallic solid Ω with boundary $d\Omega$ certain plane area S is occupied by an internal crack (or several cracks), which sides we denote S_1 and S_2. Filling the crack with hydrogen is due to its diffusion from surrounding metal. Generally, when the non-uniformity of mechanical stresses, temperature field, phase and chemical composition are accounted for , the hydrogen flux J will be estimated by relation

$$\vec{j} = -D \cdot C \left\{ \nabla \ln \frac{C}{K_S \cdot exp \dfrac{V_H \cdot \sigma}{RT}} \Bigg|_T + \frac{Q_*}{RT^2} \nabla T \right\} \tag{1}$$

The diffusion equation can be represented traditionally

$$\frac{\partial C}{\partial t} = -div\vec{j} \tag{2}$$

The boundary and initial conditions are set in the form

$$C(x, y, z, 0) = C_0(x, y, z) \tag{3}$$

$$J\big|_{\partial\Omega_1} = k_{sd} \left(K_s \sqrt{P} - C \right)_{\partial\Omega_1} \tag{4}$$

$$C_{|\partial\Omega_2} = K_s \sqrt{P_{env}} \tag{5}$$

Since the rate of surface processes is quite high, we assume, that the equilibrium of hydrogen concentration along the crack side line and internal pressure in the crack is reached. In the other words, the Siverts law is valid (5). The pressure level can be estimated from the equation-of-state. Here the equation-of-state for ideal gas is employed

$$f_{H_2}(t) = \frac{RTM(t)}{\mu V(t)} \tag{6}$$

The hydrogen mass will be estimated by

$$M(t) = \int_0^t \int_S \left[\vec{J}_1 \cdot \vec{n}_1 + \vec{J}_2 \cdot \vec{n}_2 \right] dS dt \tag{7}$$

In framework of linear elastic fracture mechanics the crack volume is proportional to pressure level

$$V(t) = Pf_V \tag{8}$$

Taking into account relations (7) and (8), the hydrogen pressure in the crack can be presented as follows

$$f_{H_2}(t) = \frac{RT}{\mu V} \int_0^t \int_S \left[\vec{J}_{S_1} \vec{n}_1 + \vec{J}_{S_2} \cdot \vec{n}_2 \right] ds dt \tag{9}$$

Next, the methane pressure in the crack should be estimated. The traditional approach was used, which relate this parameter with fugacity of hydrogen, present in the crack, temperature, thermodynamic constants of the reaction, and chemical composition and structure of carbides

$$f_{CH_4} = K_0(T)(a_{Me_xC_y})^{1/y}(a_{Me})^{-x/y} f_{H_2}^2 \tag{10}$$

Next the crack propagation is controlled by pressure variation (from one side) and crack growth law (from the other side). For this the deformation criterion of failure is chosen. In the other words, we assume that the jump of the crack tip for the time increment Δt will be Δl, if under given loading condition F in the location x the following equality

$$\varepsilon(F, C_H) = \varepsilon_1(F) - AC_H \tag{11}$$

will be reached. Here we assume, that in the pre-fracture zone only small amount of hydrogen is accumulated and the linear relation between hydrogen concentration and drop of strain at failure is valid.
The averaged crack growth rate can be presented as follows

$$v = \frac{x_*}{t_*} = \frac{2\alpha\delta}{t_*} \tag{12}$$

The deformation in the pre-fracture zone is estimated using crack tip

opening displacement (CTOD). Thus $\varepsilon = h^{-1}\delta$, where h is certain proportional coefficient, temperature independent.

Then, the crack growth criterion can be written as

$$1 - \frac{\delta}{\delta_k} = BC(x_*, t_*) \tag{13}$$

We assume, that time increment for the crack jump is so short, that neither internal pressure, nor material properties ($D_i(t)$, $K_S(t)$) can change. Then the problem of hydrogen accumulation in pre-fracture region is reduced to the solution to equation of hydrogen diffusion under stresses field (1), (2) and conditions of continuity of hydrogen concentration and flux in the location of maximal stresses. Boundary and initial conditions are given as:

$$C(\xi,0) = C_0; \; C(0,\tau) = C_S; \; C(\infty,\tau) < \infty \tag{14}$$

The stress distribution on a line of crack trajectory extension can be approximated by piece-line dependency

$$\sigma = \sigma_T\{2.4 + 1.8[xH(x_m - x)/x_m + H(x_m - x)]\} \tag{15}$$

Making use of integral Laplace transform over the time scale, the method of asymptotically equivalent functions and method of limit interpolations the approximate relation was obtained which describe the hydrogen concentration in the pre-fracture zone.

$$C(x,t) = \frac{C_S \, exp \, 2m\zeta[\zeta\sqrt{\tau}C_0 + 2\sqrt{\pi}(C_S - C_0)exp(m\zeta - \zeta^2/4\tau)]}{\zeta\sqrt{\tau}C_S \, exp \, 2m\zeta + 2\sqrt{\pi}(C_S - C_0)exp(m\zeta - \zeta^2/4\tau)} \tag{16}$$

where $\zeta = \frac{x}{2\delta}, \tau = \frac{Dt}{(2\delta)^2}, m = V_H(\sigma_m - \sigma_0)/(2RT),$

$\sigma_m = 2.4\sigma_{YS}, \sigma_0 = \sigma_{YS}/\sqrt{3}, C_S = K_S\sqrt{P}.$

Substituting the hydrogen distribution into equation (13), the crack growth rate was estimated:

$$v \approx 2D(T)(\pi\alpha\delta)^{-1}(C_S - C_0)^2 \, exp(-2\alpha m(T)) \times \tag{17}$$

$$\times \left[B(T)C_S(P,T)exp(2\alpha m(T)) - 1 + \delta / \delta_k(T) \right]^2 \Big/ \left[C_S^2 \left(1 - \delta / \delta_k(T) - BC_0 \right) \right]^2$$

In the general case, when a crack is developing in one plane of a structural element, a kinetic equation is written as

$$\frac{\partial r}{\partial t} \left[1 + \frac{1}{r^2} \left(\frac{\partial r}{\partial \varphi} \right)^2 \right]^{-\frac{1}{2}} = v(K_I, C_H); \quad r\big|_{t=0} = r_0(\varphi) \qquad (18)$$

However, the direct numerical solution of such equation with a subsequent determining of the crack contour and SIF variation along the crack contour in the general case is so complicated that it is not be used for practical engineering assessments.

Effective solution of this problem is provided by the approximate method of integral evaluation of subcritical plane crack growth [5]. It uses the crack area as a main calculation parameter of defectiveness and reduction of the kinetic equation to the relationships that immediately describes the area variation during crack growth. The equation a more simple form, if instead of area we shall consider a linear parameter a_e - a radius of equal by the crack area circle

$$\frac{da_e}{dt} = v(K_I, C_H); \quad a_e(t=0) = a_0 = \sqrt{S_0 \Big/ \pi} \qquad (19)$$

Thus, a complete system of equation of the model of internal hydrogen crack development in metals during temperature variation is composed of equation of the gas-filled internal cracks and creation of pressure in them (1)-(10), and also the equation of crack growth (19). To have a complete system a relationship for determining SIF and crack volumes is needed.

MODEL APPROBATION

The pressure vessels for hydrocracking work in the hydrogen environment at temperature 723 K and pressure 15 MPa. They are manufactured from 2.25Cr-1Mo steel. To protect them against the hydrogen sulphate attack, the internal surface of vessel is clad by austenitic stainless

steel. After cladding the pressure vessel undergoes thermal treatment at 690C during 24 hours. During this process carbon diffuses from main metal into cladding, creating near boundary a brittle carbide interlayer. During the reactor service, the process of hydrogen attack of this interlayer takes place. The hydrogen react with carbon, creating methane. During the technological shut-downs or in the case of emergency the pressure drop to zero and the reactor' wall cools down with certain rate to ambient temperature. The thickness of main metal is 190 mm, but cladding is 8 mm thick. The hydrogen concentration on the welding line at ambient temperature reaches the maximum.

To verify the worked out model cylindrical specimens with an overlayer were tested in gaseous hydrogen-environments. Specimens prepared for tests before hydrogenation were subjected preliminary ultrasonic testing, that proved the absence of internal damages. Specimens were hydrogenated in gaseous hydrogen in a special autoclave in the conditions similar to those of the reactor operation. After saturation specimens were cooled down and gauges were fixed on them. A crack control was performed by acoustic emission signals (ASE). Further the ultrasonic and metallographic analysis was carried out that showed that spalling of the base metal from the overlayer occured. In this case spalling occurs along the boundaries of austenitic overlayer grains at the interface, that contains a great amount of carbide. Using to acoustic –emission model [6] according to temporal dependence of total AES amplitudes, a temporal dependence of variation of the crack area, shown in Figure 1 (curve 1), was plotted. It has been found during experiment that a crack nucleated after 10ks since cooling and reached the area of $3.8 \cdot 10^{-4} m^2$ after 140 ks. Using this data and solving a kinetic equation of crack growth (19) by Runge-Kutta method, a calculated temporal dependence of the crack area variation, shown in Figure 1 (curve 2) was obtained. Comparison of these curves proves the effectiveness and truth of the calculational model, presented above.

CONCLUSIONS

Therefore, the realisation of described above processes leads to hydrogen degradation inside metal. Under certain conditions this process can go even without applying of external loading. The necessary driving force for crack development and material tearing will be pressure of gases (hydrogen itself of its product of interaction with metal). Although the duration of such conditions for metal can be short, but their numerous repeat can cause dangerous damage of whole structure.
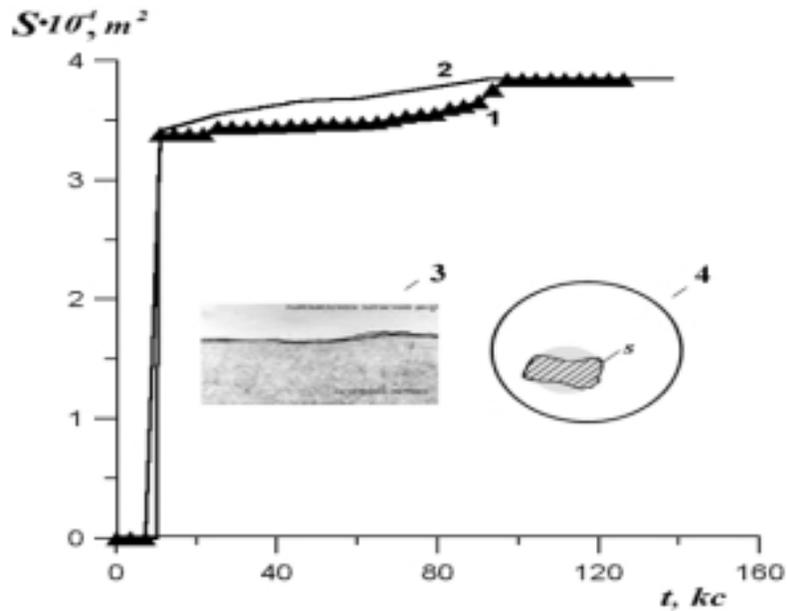
Figure 1: Prediction of crack formation in bimetallic structures during their cooling after high-temperature hydrogenation: 1 - acoustic emission method; 2 - calculation results; 3 - fractographic results; 4 - ultrasonic test results.

REFERENCES

1. Ohniski K, Chiba R., Watanabe Y. et. al. (1980) Russure Vessels Research Committee Meeting. New York, 1.
2. Imanaka T., Shimomura J., Nakano S., Yasuda K. (1985) Kawasaki steel Techn. Rep.13,109.
3. Ohniski K., Fuji A., Chiba R. et al. (1983)Quarterly J. Japan Weld Soc.3,75.
4. Panasyuk V., Andreykiv O., Hembara O. (2000) International Journal of Hydrogen Energy 25, 67.
5. Panasyuk V., Andreykiv O., Darchuk O.(1988) Physico-Chemical Mech. Mater. 6, 52.
6. Lysak M., Skalskyi V., Ivanitskyi Ya. (1989) Physico-Chemical Mech. Mater. 2, 79.