EVALUATION OF HYDROGEN EMBRITTLEMENT WITH FRACTURE MECHANICS TESTS

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ABSTRACT

The influence of hydrogen embrittlement on the toughness of SA350LF3 steel was investigated. Two methods, the more traditional disk pressure test and a small specimen fracture mechanics tests, were applied. The quantitativ determination of a fracture toughness parameter on base material, weld metal and heat affected zone, can be executed for two loading conditions using the second method.

KEYWORDS

Hydrogen embrittlement, low alloy steel, disk pressure test, small specimen fracture mechanics tests, loading condition.

INTRODUCTION

In connection with the construction of a chemical plant a large number of pressure vessels, heat exchangers, pipes and pipe components had to be designed. Choice of materials, dimensioning and design of the components was carried out on the base of ASME Section II and ASME Section VIII Div. 2. Adequate toughness of the materials had to be demonstrated in order to achieve safety against brittle fracture under the most severe design conditions. Table 1 lists the scope of testing and the associated requirements for ferritic materials. Some of the plant components operate containing free hydrogen with contaminations of O_2 and H_2O of less than 2 vpm. For the materials used to construct these plant components, the loss in toughness due to the presence of hydrogen under pressure has to be evaluated. This paper deals with one low alloy steel used at operating temperature below 20 °C.

TABLE 1 Testing Procedure for Ferritic Materials

Test Object	Thickness	Type of Testing	Requirement
Material type (Heat, Welding procedure, heat treatment)	> 5 mm	KCV at T_{Design} K_{ID} at T_{Design}	50 Joule -
	>20 mm	Additionally: KCV-Transition curve NDT-Temperature	- T_{Design} -15 °C
	>50 mm	Additionally: K_{Ic} at T_{Design}	-

QUALITATIVE TEST METHODS

Two test methods have mainly been used up to now for the determination of susceptibility to hydrogen embrittlement:

Determination of the reduction in area after fracture on tensile test specimens.
The reduction in area after cathodic charging with hydrogen is compared with that occurring after the test in air. The decrease of reduction in area due to the effect of hydrogen shall be less than 30 % for a material to be acceptable for service under hydrogen conditions. This method however is not suitable for the evaluation of welded joints and in particular of heat affected zones. Furthermore there is no correlation available between the hydrogen take up under cathodic charging and under hydrogen pressure loading.

Determination of bursting pressure with the disk pressure test [1]. Thin disks are pressure loaded in a testing device. The bursting pressure is measured under hydrogen respectively helium. The degree of sensitivity of a material to hydrogen embrittlement is regarded as tolerable when the ratio of bursting pressures p_{He}/p_H is less than 2.

Disk Pressure Tests on SA 350 LF3 Steel

The SA 350 LF3 (3,5 % Ni) was used for the construction of a large number of plant components and vessels. Disk pressure tests on this steel grade were commissioned to two different laboratories.

Chemical analysis of the material investigated:

C	Si	Mn	P	S	Ni	Al	N_2	O_2
0,1	0,245	0,39	0,0085	0,009	3,34	0,024 [%]	90	60 [ppm]

Heat treatment:

Hardening 850 °C/10 h/water; annealing: 630 °C/8 h/air
Simulated stress relief annealing: 550 °C/9 h,

Mechanical properties:

R_m = 537 N/mm² R_e = 432 N/mm² A_5 = 33 % Z = 77 %

Charpy V at -33 °C 212 J; at -101 ° C 120 J

The disk pressure tests on the base material were carried out using disks with the dimensions Ø 58 x 0,75 mm, on the weld metal disks Ø 10 x 0,25 mm were used. The investigations were executed at room temperature with hydrogen of highest purity. The pressure was increased at a rate of 60 bar/min. The experimental results are summarized in Fig. 1. The main part of the tests on the base material produced results above the tolerable pressure ratio p_{He}/p_H of 2. Therefore the tested materials would be not acceptable for the intended use.

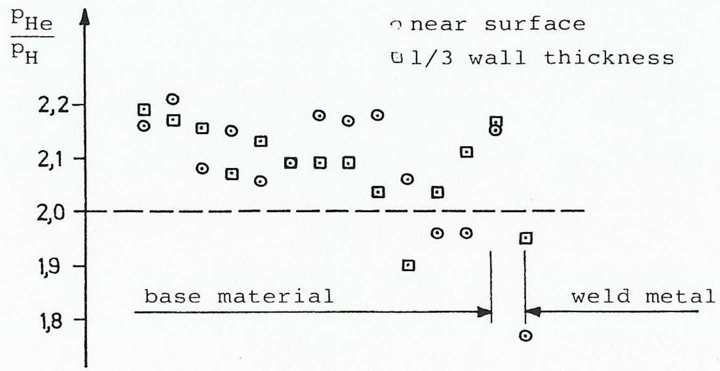


Fig. 1 Results of disk pressure tests

Tests with four disks of the same base material carried out in the second Laboratory yielded a pressure ratio of 2.6 at room temperature and with a pressure increase rate of 14,5 bar/min. It was not possible to clarify how far differences in the purity of the hydrogen or in the pressure increase rate have influenced the investigations.

Problems in connection with the disk pressure test method:
The disk pressure test gives no quantitative results concerning the safety of components against fracture. It does not account for the basic toughness in air of the materials. Due to the small thickness of the test specimens, local material inhomogeneities

can have a much bigger effect than in actual components. The manufacture of specimens out of heat affected zones is difficult.

QUANTITATIVE TEST METHODS

It was tried to find a suitable testing technique enabling quantitative evaluation of the sensitivity of materials to hydrogen embrittlement under service conditions. Two closely related methods are described, both of which provide a fracture mechanics evaluation of the change in toughness due to take up of hydrogen. The test specimen is in both cases the same and the expenses of both methods are within tolerable limits.

Test Specimens and Location of Specimens

The dimensions of the test specimens correspond to those of a standard Charpy-V specimen. The specimens were fatigue precracked to an average crack depth $\bar{a}$ of approximately 3,0 mm. All the specimens were taken from a segment of a welding procedure test ring (fig. 2).

The base material and weld metal of the ring is identical to that used for the disk pressure tests.

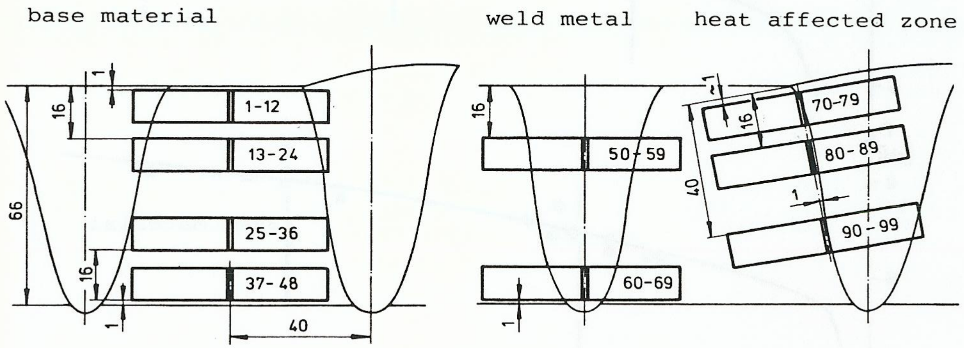


Fig. 2 Locations of test specimens

Testing of Preloaded Specimens

The ratio of the fracture toughness in air and in hydrogen K_{Jair}/K_{JH} was determined. For the tests, a number of precracked specimens were loaded in calibrated loading rings (fig. 3) up to different values of deflection. During the loading process a load versus specimen deflection curve (P = f(F)) was recorded.

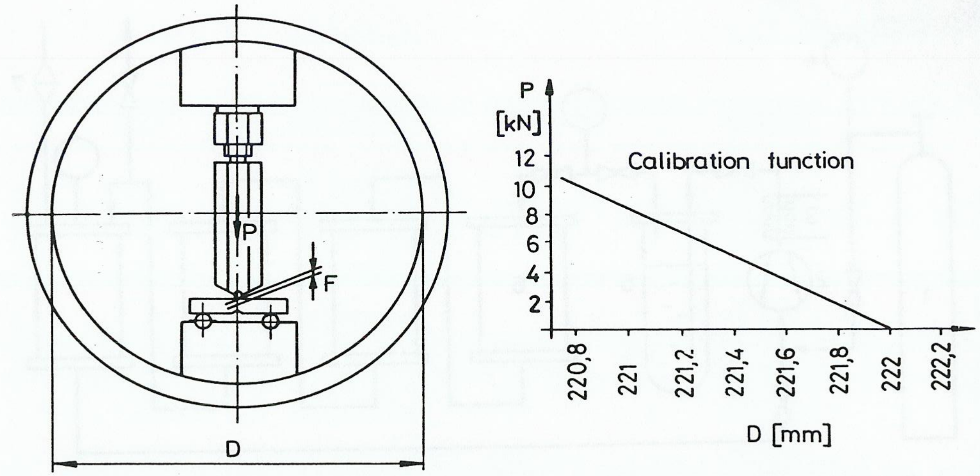


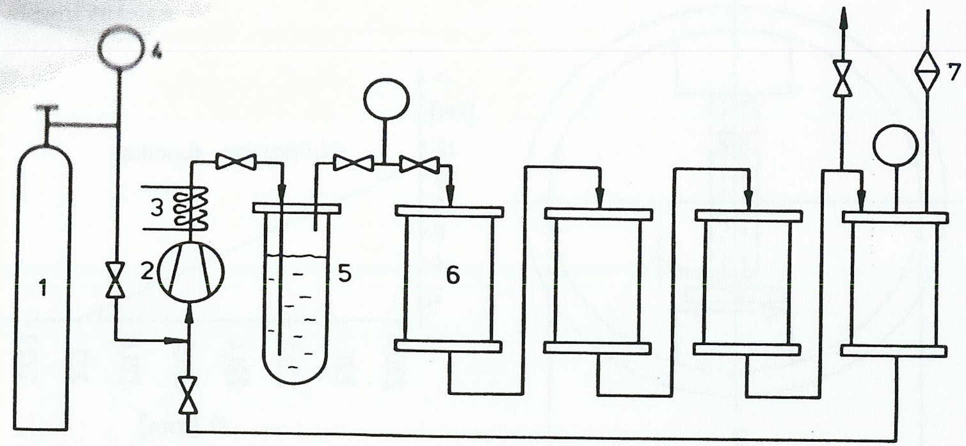
Fig. 3 Equipment for preloading

Tests in air:
For the tests in air the specimens were unloaded after the end of the relaxation process, usually after a holding time of a few minutes. The specimens were than thermally etched and broken open at low temperature. The fracture surfaces were examined under a light microscope (magnification up to 65 x) and in some cases with the scanning electron microscope for the presence of stable crack extension. In this way the critical point corresponding to the onset of stable crack growth could be defined on the load-deflection curve.

Tests in hydrogen:
For the tests in hydrogen the specimens were preloaded as described above. Four loading-rings with the preloaded specimens were located in a pressure vessel. A total of 16 specimens could be tested simultaneously. The specimens were subjected to the hydrogen gas for three days. The layout of the testing equipment is shown in fig. 4. The hydrogen pressure was 220 bar and the gas was passed approximately 450 times per day through an appropriate dessicant solution in order to remove O_2 and H_2O from the hydrogen gas, so that a purity equal to that of the hydrogen in the industrial plant was achieved.

After removal of the loading-rings from the vessels, load and specimen deflection was again measured in order to determine any relaxation. The specimens were than etched, broken open and examinated as described above.

The experimental results are summarized in fig. 5 and the points of onset stable crack growth are shown for the tests in air and in hydrogen.



1 Pressure bottle with H_2
2 Membrane compressor
3 Cooler
4 Manometer
5 Autoclave with dessicant
6 Pressure vessel with preloaded specimens
7 Bursting disk

Fig. 4 Layout of the experimental equipment

Sections of the fracture surfaces of two HAZ-specimens with signs of stable crack extension are shown in fig. 6 and fig. 7; scanning electron microscope, magnification 200 x.

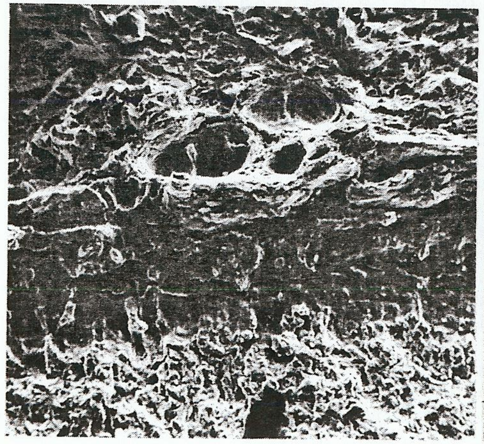


Fig. 6 Fracture surface specimen Nr. 81

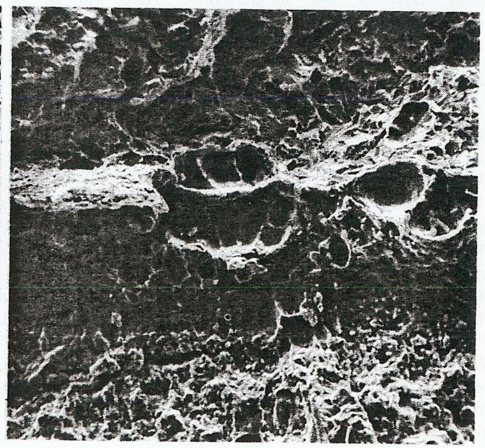


Fig. 7 Fracture surface specimen Nr. 89

The ratio of fracture toughness $K_{J_{air}}/K_{J_H}$ was obtained from the ratio of energy A used to reach the point of onset of stable crack extension on the load-deflection curve, $K_{J_{air}}/K_{J_H} = \sqrt{A_{air}/A_H}$.

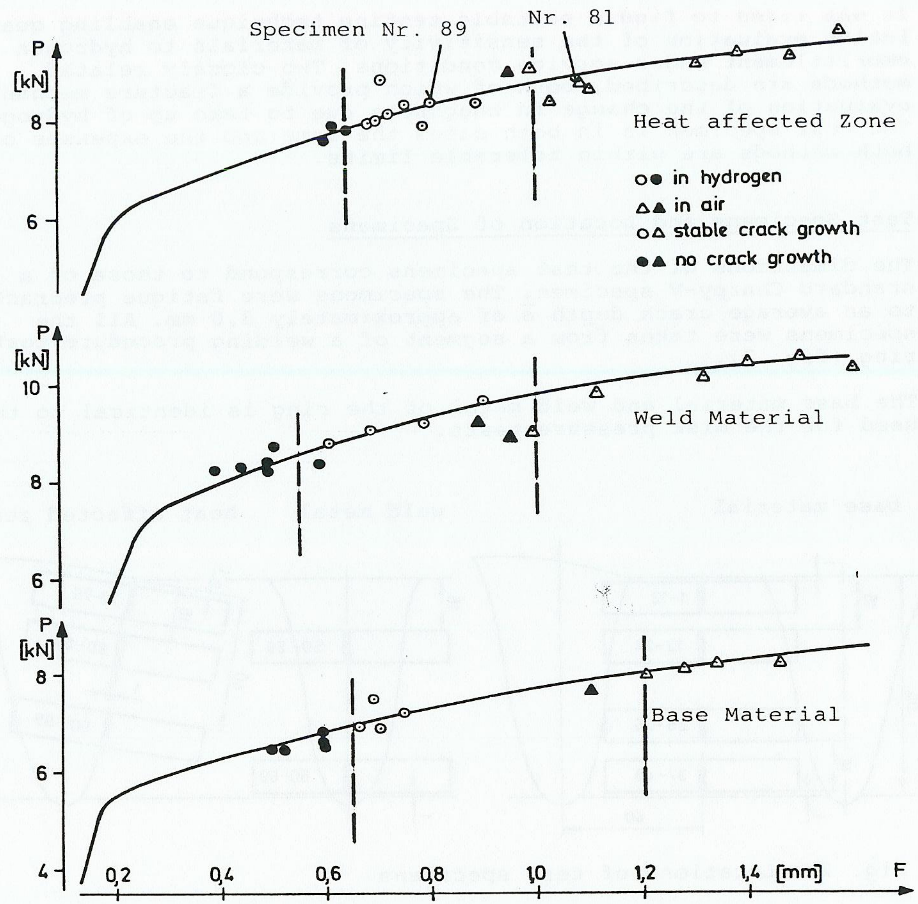


Fig. 5 Results of preloaded specimens

The basic fracture toughness values in air of the materials (base material, weld metal) were obtained from standard fracture toughness tests using CT-specimens, for the HAZ estimation from K_{ID} tests were done. The final results are shown in Table 2.

TABLE 2   Test Results of Fracture Toughness

Material type	$K_{J\,Air}/K_{J\,Hydr.}$ 20 °C	$K_{J\,Air}$ $[Nmm^{-3/2}]$ 20 °C, 2" CT	$K_{J\,Hydr.}$ $[Nmm^{-3/2}]$ 20 °C
Base material	1,49	9500	6360
Weld metal	1,52	5800	3800
HAZ	1,33	7700	5800

Testing of Specimens with Constant Deformation Rate

It is not yet known how the results of tests with specimens loaded at a constant deformation rate in hydrogen gas correlate with the results shown in fig. 5 and table 2. Therefore tests in hydrogen gas with a specimen deformation rate of 10^{-4} mm/sec were carried out. The loading device is shown in fig. 8, the deformation rate was regulated by means of a servo hydraulic testing machine. The conditions for the hydrogen gas were the same as for tests with preloaded specimens. The specimens were loaded up to different values of deflection, the onset of stable crack growth on the load-deflection curve was determined as for the preloaded specimens. A load-deflection curve for the HAZ is shown in fig. 9. The corresponding fracture surface is represented in fig. 10. For a maximum deflection of 0,35 mm, stable crack growth occurs. The onset of stable crack growth was found at 0,25 mm deflection. Up to now only a few tests have been carried out, the approximate results are summarized in Table 3.

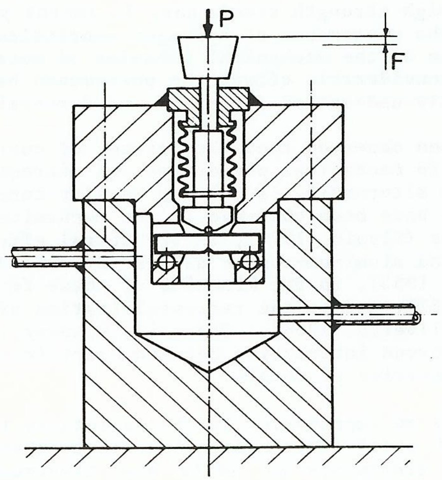


Fig. 8   Loading device for constant deformation rate

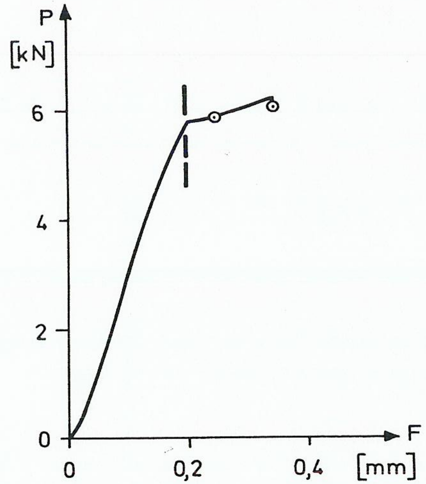


Fig. 9   Load-deflection curve

Fig. 10   Fracture surface HAZ magnification 50 x

TABLE 3   Test Results of Fracture Toughness

Material type	$K_{J\,Air}/K_{J\,Hydr.}$ 20 °C	$K_{J\,Air}$ $[Nmm^{-3/2}]$ 20 °C, 2" CT	$K_{J\,Hydr.}$ $[Nmm^{-3/2}]$ 20 °C
Base material	3,5	9500	2700
Weld metal	2,9	5800	2000
HAZ	3,5	7700	2200

The few test results show, that loading with a deformation rate of 10^{-4} mm/sec has a much stronger effect on hydrogen embrittlement than preloading. But it remains a certain nonlinear material behaviour before the onset of stable crack growth.
The tests will be continued and supplemented by other values of deformation rates. In order to find out to which extent an improvement in toughness can be established, a second heat of SA350LF3 will be tested for hydrogen embrittlement.

REFERENCE

[1] J.P. Fidelle et al. Disk Pressure Testing of Hydrogen Environment Embrittlement. ASTM STP 543.