THE INTERGRANULAR-TRANSGRANULAR TRANSITION OF SCC IN TEMPERED LOW ALLOY STEELS

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A model of the microstructural micromechanisms present in the cracking processes during stress corrosion in low alloy steels has been defined. The model establishes that stress-corrosion cracking occurs because of a series of isolated and unstable local fractures inside the plastic zone, controlled by local embrittlement due to absorbed hydrogen.

The most important event in the SCC behaviour of these steels is the great change associated to the alteration in the fracture path from IG to TG, and produced in a very short tempering temperature range. The model establishes that the IG-TG transition is dependent on grain size and also explains how the environmental conditions affect it. These assessments have been experimentally verified by determining the SCC behaviour of tempered samples with different grain sizes in a solution with a pH value of 3.2, comparing this with the one obtained in simulated sea water, pH of 5.8.

INTRODUCTION

The stress corrosion cracking (SCC) behaviour of low alloy steels is controlled by the material microstructure and the environmental conditions (1-4). As a reference to the global behaviour of this type of steels, the SCC behaviour in simulated sea water of an AISI 4135 steel, under different heat treatments classified in six different series, has been obtained by using DCB samples and determining the corresponding threshold conditions (K_{Iscc}) and crack propagation rate at stage II (5). The obtained results showed an important variation in SCC behaviour where the effect of microstructure is conditioned by the strength level and the type of fracture mechanisms involved:

-At high strength levels, the dislocated and low tempered martensites had a very low SCC resistant intergranular cracking behaviour with minor changes controlled by microstructural differences at crack paths (grain boundaries), such as precipitation and grain size.

- At moderate strength levels, the increasing presence of bainite, mostly of the lower type, as a substitute of martensite, increases the SCC resistance, associated to transgranular cleavage processes.

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- In the same strength range the optimum SCC behaviour is obtained for martensites tempered over a critical temperature where intergranular-transgranular transition occurs. For these a very ductile transgranular TTS type path has been observed.

Considering SCC as a local fracture process, the models for general fracture (6) have been taken into account in order to establish a model that justifies and explains all the complex behaviour obtained for the 4135 steel, associated to its different treatments. The model (7) establishes that crack growth due to stress corrosion can be thought of as a process controlled by microstructure and hydrogen distribution, which occurs at the region ahead of the crack tip of a cracked material, which is loaded and affected by an appropriate aggressive environment. Critical cracking conditions are achieved if the strain, ε^p, at a particular original position, L*, reaches a critical value, ε_H, that depends on the profile of hydrogen concentration, variable with loading and microstructural distribution for each environmental condition. Under such conditions crack nucleates at L*, grows unstably and arrests.

Therefore, two complete profiles of strain fields along the plastic zone, r_Y should be compared: the applied one due to loading conditions (8), ε^p (X/δ), and the critical one ε_H (X/δ), associated to hydrogen concentration and microstructural singularities. As represented in Figure 1, the first one is independent of loading and geometry when represented with reference to the distance from the crack tip and measured relatively to the CTOD δ value, X/δ.The second one varies with loading and hydrogen concentration. So, by increasing the external load or the aggressivity of the environment a crack may reach the critical instability conditions and establish a subcritical advance.

Observed intergranular (IG) and transgranular (TG) behaviour differences, through fracture micromechanisms and microstructural hydrogen trapping competitivity have been explained by the model. But, the most important event in the SCC behaviour of these steels is the great improvement in cracking resistance associated to the change in the fracture path from IG to TG, which is produced in a very short tempering temperature range. As the cracking criterion has been formulated without limits, it must also explain why the IG processes are no longer achieved and why TG processes occur. It seems reasonable to assume that cracking will be transgranular if crack nucleates inside the grain and will be intergranular if it nucleates at the grain boundary, Figure 2. Therefore, the distance L*, limited by the model inside the plastic zone, may be compared with the grain facet length, defined theoretically as d/2.3, to establish the following complementary condition:

$$L^*_{IG} < \frac{d}{2.3} \quad \text{and} \quad L^*_{TG} > \frac{d}{2.3} \qquad (1)$$

Working with Eq. (1), it can be deduced that threshold values, K_{Iscc} and δ_{Iscc}, are limited in IG or TG SCC behaviour depending on microstructural conditions either directly, grain size, d, and indirectly through their mechanical characteristics,

$$\text{IG:}\quad \delta_{Iscc} < \frac{d}{2.3} \qquad \text{or} \qquad K_{Iscc} < 0,85\sqrt{\sigma_y E d} \qquad (2)$$

$$\text{TG:}\quad \delta_{Iscc} > \frac{15}{2.3}\frac{\sigma_y}{E} d \qquad \text{or} \qquad K_{Iscc} > 3.3\,\sigma_y\sqrt{d} \qquad (3)$$

These equations show that, as grain size increases then the intergranular behaviour range of these steels extends, and the tempering temperature for IG-TG transition increases.

The model also explains how the environmental conditions affect the IG-TG transition. When environmental aggressivity increases with a lowering of pH the hydrogen concentration increases in the plastic zone and the load necessary to achieve the critical conditions for crack nucleations, decreases. Under such circumstances lower δ_{Iscc} values are obtained and IG behaviour is mantained. So, as the environment gets more aggressive, the tempering temperature for IG-TG transition increases.

The aim of this work is to validate the model by determining experimentally the effects of both grain size and environment on the IG-TG transition of SCC in tempered low alloy steels.

EXPERIMENTAL PROCEDURE

In order to study the influence of grain size on the SCC IG-TG transition in quenched and tempered low alloy steels a series of heat treatments were performed on a AISI 4135 Cr-Mo steel based on differences in austenitizing temperature with the aim of obtaining different austenitic grain sizes in each one. The following three series of heat treatments were studied:

Series IB. Austenitizing at 825°C, oil quenching and tempering at 400, 420, 450, 480, 500 and 525°C.
Series IIB. Austenitizing at 900°C, oil quenching and tempering at 400, 425, 450, 475, 500 and 550°C.
Series IIIB. Austenitizing at 1000°C, oil quenching and tempering at 400, 420, 450, 475, 500 and 550°C.

In all cases metalographic analysis was carried out using optical and electronic microscopy (SEM and TEM) to characterize the obtained microstructure, tempered martensite. Mechanical strength was determined through the hardness obtained with a Vickers microhardness tester. Charpy impact tests were also performed.

To characterize the SCC behaviour of these steels, fatigue precracked DCB samples were used to determine the rate of crack propagation da/dt against stress intensity factor, K_I, in stages II and I of propagation and threshold respectively. Simulated sea water with a pH of 5.8 was used as the environment.The fractographic observation of the crack surface caused by SCC was carried out using the scanning electron microscope.

The influence of environment has been studied using the previous SCC test as a reference and by performing a new experimental phase in a more aggressive solution, composed of 50g of NaCl and 5g of acetic acid disolved in 945g of water,with an initial pH of 3.2. The solution was periodically changed during the SCC test to prevent the pH from passing 4.5. This SCC characterisation was carried out on previously heat treated samples from two of the three series presented above:

Series IB*. Austenitizing at 825℃, oil quenching and tempering at 450, 500 and 550℃.
Series IIB*. Austenitizing at 900℃, oil quenching and tempering at 550 and 600℃.

RESULTS

Effect of grain size

The values for the average austenitic grain size, variable with austenitising temperature, were 30 μm for series IB, 75 μm for series IIB, and 150 μm for series IIIB. In Table 1 the complete results of the mechanical and fractographic characterisations of these series are presented. The fracture paths observed were intergranular or transgranular, this latter caused by microvoids coalescence, except in some specimens in which both fracture types were present. These have been associated to IG-TG transition.

TABLE 1- Mechanical characterisation and SCC behaviour in simulated sea water of series tested to study the grain size effect.

	HV	σ_Y (MPa)	K_{Iscc} (MPa.m$^{1/2}$)	da/dt (m.s^{-1})	d/r_Y	FRACTURE TYPE
SERIES IB (d=30 μm) Tempering temperature (°C)						
400	447	1315	19.8	1.3 10^{-6}	3.3	IG
420	432	1270	22.5	0.3 10^{-6}	2.4	IG+TG
450	413	1215	75	4.2 10^{-8}	0.195	TG
480	396	1165	100	4.0 10^{-8}	0.103	TG
500	383	1127	130	4.0 10^{-8}	0.056	TG
525	375	1105	160	2.2 10^{-8}	0.036	TG
SERIES IIB (d=75 μm) Tempering temperature (°C)						
400	438	1290	20	1.2 10^{-6}	7.8	IG
425	421	1240	23	1.2 10^{-6}	5.5	IG
450	405	1192	38	1.0 10^{-6}	1.6	IG+TG
475	392	1155	71	5.0 10^{-8}	0.5	TG
500	380	1118	94	3.0 10^{-8}	0.27	TG
550	358	1055	163	2.0 10^{-8}	0.079	TG
SERIES IIIB (d=150 μm) Tempering temperature (°C)						
400	432	1270	21	1.2 10^{-6}	13.9	IG
420	417	1227	25	1.2 10^{-6}	9.14	IG
450	400	1177	34	1.1 10^{-6}	4.54	IG
475	390	1150	39	0.9 10^{-6}	3.29	IG+TG
500	375	1105	72	3.6 10^{-8}	0.84	TG
550	355	1045	112	2.7 10^{-8}	0.33	TG

The results show that improvements in the SCC resistance, high K_{Iscc} values, are accompanied by a decrease in the mechanical resistance of the material. The IG-TG transition behaviour was produced at different levels of mechanical resistance for each series, decreasing from 430 to 390 HV with the increase in grain size, from series IB to series IIIB. No tempering embrittlement phenomena were observed at the transition temperatures in the energy fracture results obtained with Charpy tests.

Figure 3 represents the SCC resistance value, K_{Iscc}, against the tempering temperature. The three heat treatment series possess a similar SCC behaviour evolution, with two clearly differentiated zones.

The first zone, with an IG fracture type, associated to low tempering, up to a variable temperature over 400℃, gives a gentle positive slope which represents a progressive improvement in SCC resistance with the increase in tempering temperature. This improvemment has been explained by, firstly, the loss of tensile strength, and, secondly, the evolution of tempering precipitation that increases continuously with temperature.

The end of this zone corresponds to a sudden improvement in SCC resistance, observed in every series, within a short tempering temperature interval of 25℃. This improvement is not associated to any significant or sudden change in the mechanical behaviour or in the microstructure, but is related to a change from IG to TG fracture types, the latter corresponding to higher K_{Iscc} values. As grain size increases, 30-75-150 μm, the IG-TG transition, measured by the minimum tempering temperature with TG behaviour, increases, 450-475-500℃.

According to the model this variation in the transition temperature is associated to the fact that IG fractures continue as the grain grows because of bigger plastic zones and, therefore, bigger K_I values still satisfy the IG condition established above. For the three series, the values of the d/r_Y ratio at threshold are less than one in the TG fracture types, but are over 2.3 for the IG fracture types.

The influence of grain size also produces important changes in SCC resistance when the fracture type is TG, the second SCC behaviour zone, shown in Figure 3, having a much better behaviour with small grains, K_{Iscc} from 130 MPa $m^{1/2}$ in series IB to 72 MPa $m^{1/2}$ in series IIIB at 500℃ of tempering, that may be explained through the differences in hydrogen trapping associated to the evolution of the specific surface of grain boundaries.

Figure 4 represents the crack propagation rate in stage II (logarithmic scale) against the tempering temperature. A general tendency is observed in which the low tempering temperatures have the highest propagation velocities, 10^{-6} ms^{-1} in IG regime, and the high ones have the lowest velocities, 10^{-8} ms^{-1} in TG regime. Therefore, an important change in propagation rate is observed at IG-TG transition whose value is greater than one order of magnitude. Considering very close values for the apparent diffusivity of the microstructures tested the model establishes, as has been experimentally shown, that the crack propagation rate is proportional to the inverse of the length of the crack advance, related with plastic zone size.

Effect of aggressive environment

The results of the SCC behaviour in the new aggressive solution (pH of 3.2) for the two quenched and tempered series, IB* and IIB*, are presented in Table 2.

TABLE 2- Mechanical characterisation and SCC behaviour in a solution with a pH of 3.2 of series tested to study the environmental effect.

	HV	σ_Y (MPa)	K_{Iscc} (MPa.m$^{1/2}$)	da/dt (m.s^{-1})	d/r$_Y$	FRACTURE TYPE
SERIES IB* (d=30 μm) Tempering temperature (°C)						
450	413	1215	22	1.0 10^{6}	2.2	IG
500	382	1125	32	0.8 10^{-6}	0.93	IG+TG
550	345	1015	101	0.1 10^{6}	0.076	TG
SERIES IIB* (d=75 μm) Tempering temperature (°C)						
550	353	1040	75	0.4 10^{6}	0.36	TG
600	315	930	115	3.8 10^{8}	0.12	TG

The obtained K_{Iscc} values in both tested environments, plotted against tempering temperature, are shown in Figure 5 for series IB and IB*. The two clearly different zones mentioned above, one IG and one TG, were observed in the two environments used. The sudden change in SCC behaviour corresponding to the IG-TG transition varies appreciably with the environment. For sea water, the transition appears at 450°C, with K_{Iscc} values varying from 22.5 to 75 MPa m$^{1/2}$, and for the solution with a pH of 3.2 the transition is produced at the tempering temperature of 550°C, K_{Iscc} changing from 32 to 101 MPa m$^{1/2}$.

Moving away from the transition zone a clear effect of the increased environmental aggressivity can be observed which, as the model suggests, lowers the SCC resistance of the material.

The SCC propagation rate values in stage II for both environments, plotted against tempering temperature, for series IB and IB*, are presented in Figure 6. An overall increase can be observed in propagation rate values for greater concentrations of hydrogen.The greater the acidity of the solution, the greater the hydrogen supply which leads to an increased propagation rate in TG fracture types. As in the IG fracture type the propagation rate stays constant, the change in propagation rate at the IG-TG transition decreases for the more aggressive environments.

CONCLUSIONS

The transition from intergranular to transgranular fracture types in stress corrosion cracking processes present in tempered low alloy steels takes place, as has been shown, in a short interval of tempering temperature, less than 25°C, for any grain size. In this interval a small change in microstructure and hardness produced by tempering, means that certain critical conditions are reached which radically change the mechanisms of the fracture.

The developed model of the SCC behaviour of these steels predicts that the transition takes place at a characteristic value of the nucleation distance in relation with the grain facet size, and the experimental results confirm it. The fact that this value is a characteristic of the process leads to explain that bigger grain sizes admit a bigger plastic zone at the transition temperature and, therefore, larger K_{Iscc} values. So, when the austenitic grain is bigger the IG brittle behaviour extends to higher tempering temperatures.

The SCC susceptibility of these steels increases with environmental aggressivity, justified by the greater quantity of hydrogen supplied to the microstructure in low pH environments. According to the presented model the tempering temperature at which the transition IG-TG is produced increases as the hydrogen concentration difussed to the crack tip increases.The obtained results show an increase of 100°C for the IG region when the environmental pH changes from 5.8 to 3.2.

ACKNOWLEDGEMENTS

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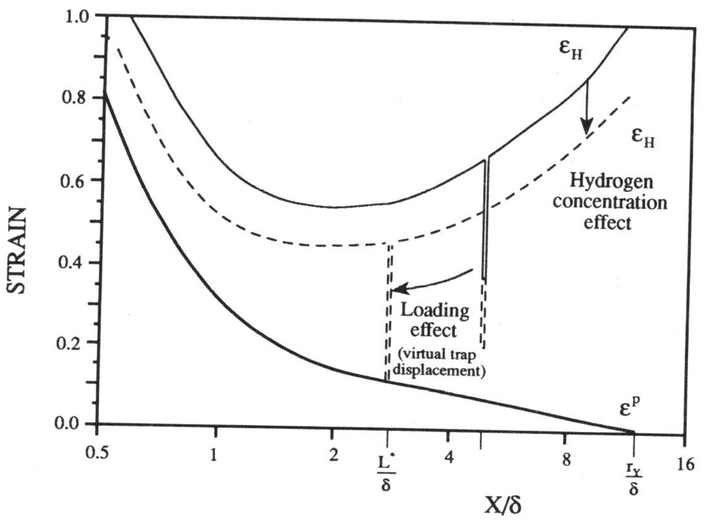

Figure 1- Application of SCC criterion. Loading and hydrogen concentration effects.

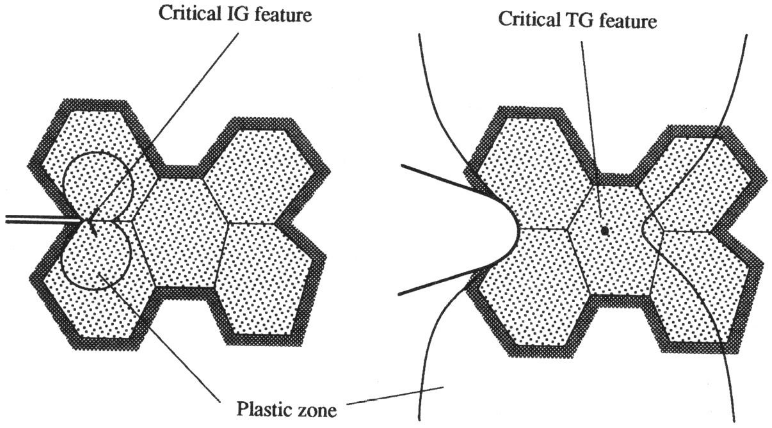

Figure 2- Position of crack nucleation at a critical feature for IG and TG cracking paths.

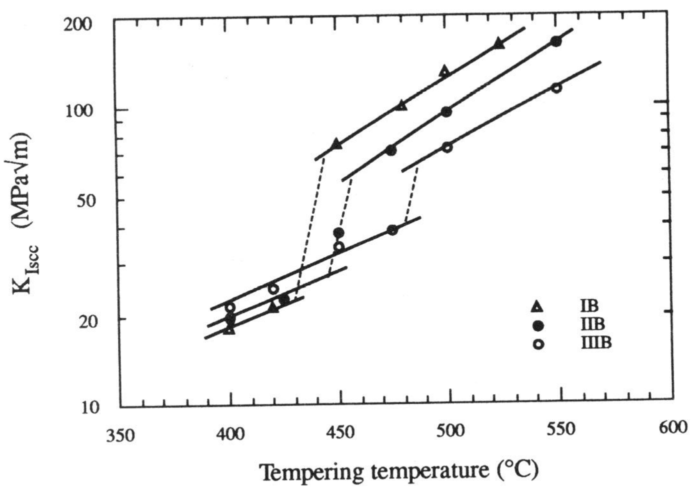

Figure 3- K_{Iscc}-tempering temperature relationship. Grain size effect.

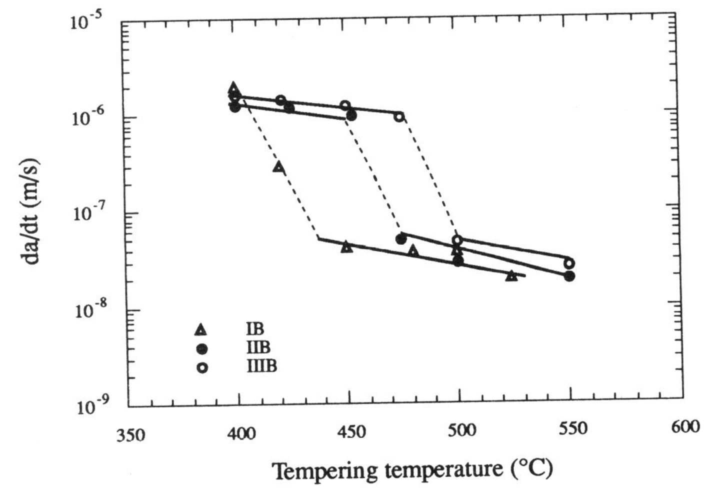

Figure 4- Crack propagation rate-tempering temperature relationship. Grain size effect.

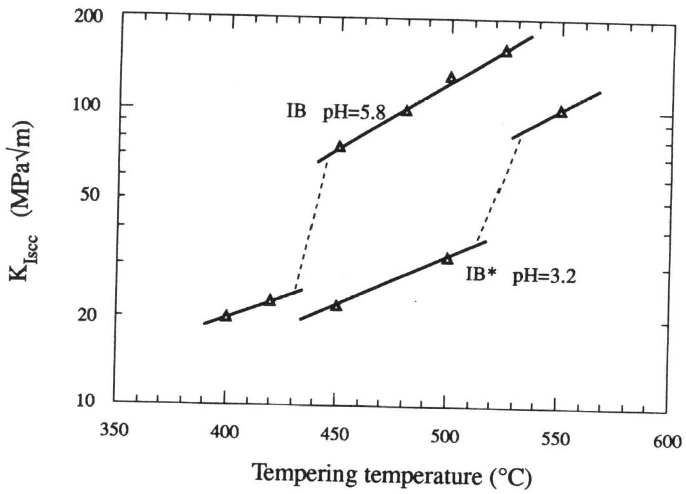

Figure 5- K_{Iscc}-tempering temperature relationship. Environmental effect.

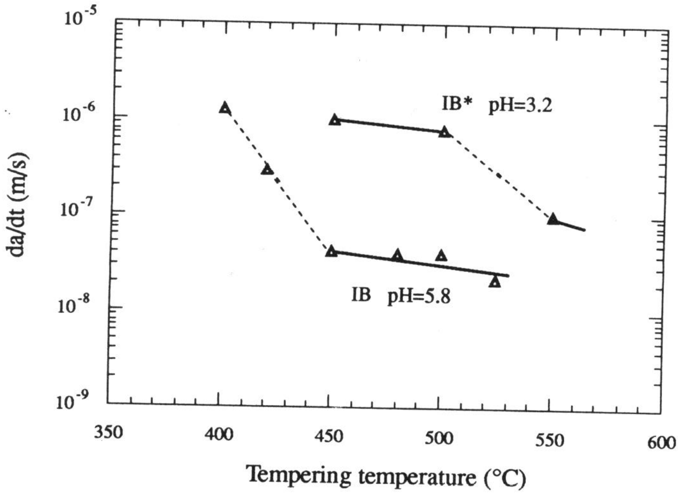

Figure 6- Crack propagation rate-tempering temperature relationship. Environmental effect.