Hydrogen Assisted Fracture of Inconel 600 Charged at High Temperature

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ABSTRACT

Effect of hydrogen charging from the gas phase at 350-600°C and 30-45 MPa as well as subsequent ageing at room tenperature on fracture mode and plasticity of Inconel 600 has been studied. No change in microstructure has been produced by charging below 550 and 40 MPa whereas exposure at higher temperature and pressure caused hydrogen attack. Ageing at RT changed the distribution of hydrogen in metal and thus modified the fracture mode and plasticity. Hydrogen present in solid solution decreased plasticity much more than that segregated around inclusions.

KEYWORDS

hydrogen embrittlement, hydrogen attack, Inconel 600, fish eyes

INTRODUCTION

Fracture of Ni and Ni base alloys is modified by the introduction of hydrogen: intergranular, transgranular cracking and modified microvoid coalescence can be observed. In the appearence of certain fracture mode, difference in impurity segregation (Jones et al., 1983; Heubaum et al, 1981) inclusion size and volume fraction (Thompson, 1982) and test conditions (Heubaum et al., 1981) play an essential role. In case of Inconel 600 and Inconel 625, the slightly modified fracture of microvoid coalescence (Thompson, 1982) and intergarnular , transgranular and unchanged ductile fracture (Price et al., 1983) has been observed, depending on charging mode. In above works, only charging conditions have been determined, hydrogen content and its destribution being unknown.

The aim of present work is to correlate the fracture mode and plasticity of Inconel 600 with hydrogen content by the comparison of above properties for specimens subjected to RT ageing (desorption) after thermal hydrogen charging.

EXPERIMENTALS

Tensile specimens (gauge length 20mm, width 2.5mm) were cut from Inconel 600 tube (0.052C,72.3Ni,15.4Cr,9.6Fe,0.001S,0.01P wt % and 2ppm H) along the rolling direction. Unstrained and prestrained in tension to ε=0.2 specimens were charged with hydrogen in high pressure chamber. Temperature and pressure of hydrogen gas were kept stable during experiment or changed as shown in Table 1.

Table 1. Hydrogen charging conditions

temperature, °C	pressure, MPa	time, hrs
350	40	280
450	40	280
550^x	40	280
550	30	400
600	35-45^xx	1300

^x specimens covered with transparent blue layer after charging
^xx pressure increased steadely during experiment

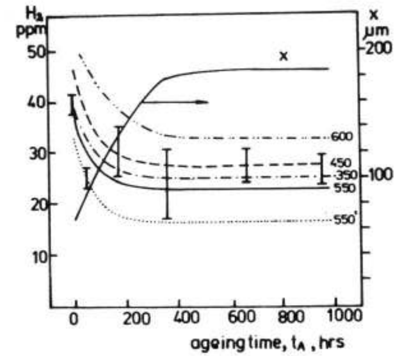
After removing of specimens from chamber (about 10 hrs elapsed between the beginning of cooling down and specimen removing) hydrogen content measurements (using LECO determinator) and tensile test ($\dot{\varepsilon}$=5 x 10^{-5} s^{-1}) have been repeadetly performed during ageing at RT for up to 2300 hrs. The surface of hydrogen charged specimens as well as fracture surface of each tensile specimen were examined using SEM. The mapping of whole fracture surface was done at magnification 300x, and the density of features seen on fracture surface at higher magnification was estimated by means of quantitative metallography. The density was expressed either by parameter $\underline{N}$- the number of interseptions of features per unit length of test line, or by the total number of features on fracture surface. Precipitates and matrix were analyzed using EDAX .

RESULTS

In Fig.1, the change of hydrogen content during ageing at RT is shown for specimens charged under different conditions. In all cases hydrogen content decreases rapidly during ageing for about 30 to 50 hrs, then further steep decrease is observed. Ageing for longer than 300-350 hrs does not affect hydrogen content. The higher hydrogen gas temperaure and pressure, the higher hydrogen content after charging and during ageing for similar time. The only exceptions are specimens charged at 550°C and 40 MPa. However, it was a doubt concerning the stability of conditions during experiment in that case, cf. Table 1

In Fig.2, the change of elongation to fracture during ageing is shown for differently charged specimens. Hydrogen charging causes decrease in plasticity as compared with as received condition (marked in Fig.2). Ageing at RT for about 350 hrs does not affect plasticity of hydrogen charged specimens. The recovery of plasticity occurs only after ageing for longer time. It should be also noted that on stress-strain curves of some hydrogen charged and aged for less than 350 hrs specimens, the serrated

yielding appears which has not been observed for as received and aged for longer time specimens. From comparison of results shown in Figs 1 and 2 follows that the higher hydrogen content in metal caused by given charging conditions, the more pronounced decrease in plasticity.



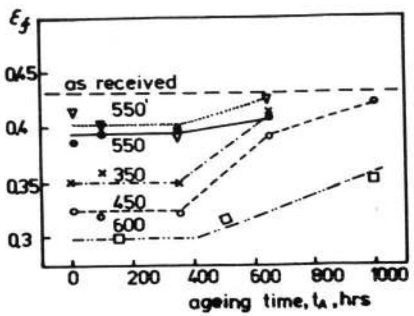


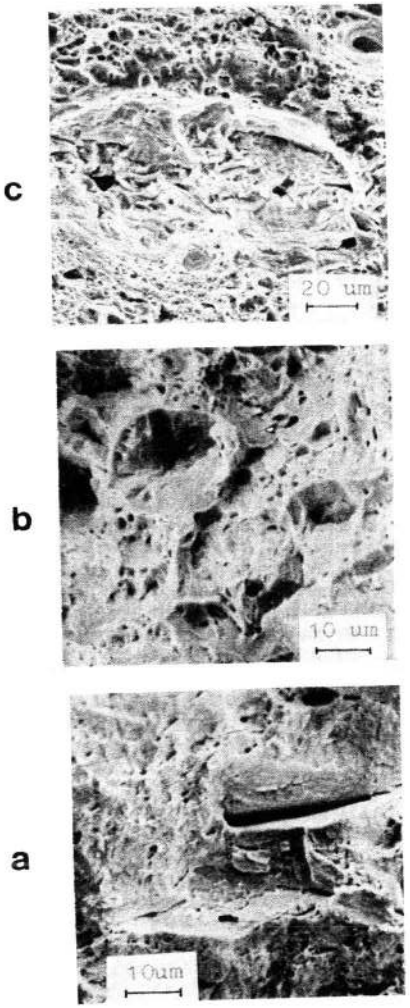
Fig.1 Hydrogen content vs ageing time for differently charged specimes; x - depth of ductile outer layer on fracture surface.

Fig.2. Elongation to fracture vs ageing time for differently charged specimens
(—·—) 350°C, 40 MPa, 280 hrs
(— — —) 450°C, 40 MPa, 280 hrs
(———) 550°C, 40 MPa, 280 hrs
(........) 550°C, 30 MPa, 400 hrs
(—··—) 600°C, 30 to 45 MPa, 1300 hrs

Microstructure of used material reveals the grain size of 30 um and fine carbide precipitates on grain boundaries and in bands. Some large (about 5 um) particles are also present. To check the effect of hydrogen charging at high temperature and pressure on microstructure, the surface of hydrogen charged specimens was carefully studied using SEM. No microvoids or other surface defects have been found either on unstrained or on prestrained specimens, the latter being known to be more susceptible to hydrogen attack (Clugston et al., 1983).

Fracture surface of as received specimens is fully ductile with shear dimples On fractur surface of specimens containing hydrogen no typical brittle fracture is observed. However, hydrogen charging modifies the fracture. Two areas can be distinguish on fracture surface of specimens hydrogen charged and aged for different time. The outer layer reveals a fully ductile fracture. The thickness of that layer (x) changes with ageing time for all charging conditions in a similar way as shown in Fig 1. The inner part of fracture surface exhibits some specific features not observed in as received specimens:

1. intergranular crevices situated along grain boundaries normal to the fracture surface with a mean length of 20 um (Fig.3a)



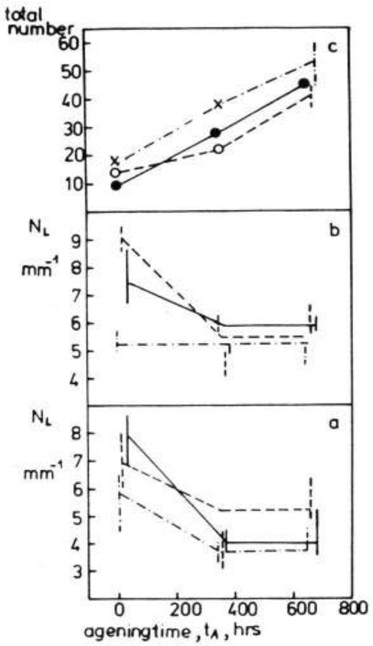


Fig. 3. Features on fracture surface of charged and aged specimens; a - integranular crevices; b - conical dimples; c - fish eyes

Fig.4. Density of intergranular crevices (a), conical dimples (b), and fish eyes (c) on fracture surface vs ageing time for specimens hydrogen charged under pressure of 40 MPa for 280 hrs at temperature
(–·–·–) 350°C
(– – – –) 450°C
(———) 550°C

2. conical dimples with a mean diameter of about 30 um (Fig.3b)
3. fish eyes - quasi cleavage areas extended for several grains around large particles (fig. 3 c).

The change in density of above features on fracture surface during ageing at RT is shown in Fig.4 for specimens charged under hydrogen pressure of 40 MPa at 350°C, 450°C and 550°C for 280 hrs. For all charging temperatures, the density of conical dimples and intergranular crevices decreases during ageing for about 360 hrs and remains unchanged for longer time, cf.Fig 4 a and 4b. Number of fish eyes increases steadely with ageing time, cf.Fig.4c. It should be noted that also size of fish eyes increases with ageing time from about 50 um at the beginning to about 150 um after 700 hrs.

Fracture surface of specimens charged at 600°C and pressure 30-45 MPa for 1300 hrs exhibits grain boundary separation not observed for specimens charged at lower temperature, cf. Fig.5. That type of fracture cannot be described as usual intergranular fracture since grains underwent heavy plastic deformation manifested itself by grain elongation and slip lines on grain boundaries(Fig.5a). However, separation of grains by the formation of voids along grain boundaries and their linkage is obvious, cf.Fig.5b. No significant change of above picture has been observed during ageing.

In Fig.6, the EDS analysis for matrix (a), particle in a center of fish eye (b) and for material on the bottom of a hole seen after removing the particle (c) are shown. Intensity of Cr and Fe sygnals for particle is higher than that for matrix and material surrounded the hole. For particle as well as for material on the bottom of hole, the sygnal of sulphur appears which has not been detected in matrix.

DISCUSSION

Effect of RT ageing of Inconel 600 specimens hydrogen charged at 350-550°C on plasticity, hydrogen content, thickness of outer layer on fracture surface and density of intergranular crevices and fish eyes is shown schematically in Fig.7. Two stages of geing can be destinguished. During ageing for about 350 hrs, decrease in hydrogen content, decrease in density of intergranular crevices, increase in thickness of ductile layer and change of plasticity have been observed. Longer ageing causes no effect on hydrogen content, thickness of ductile layer and density of intergranular crevices, whereas recovery of plasticity and further increase in fish eyes number occurs.

Ageing at RT of hydrogen supersaturated Ni alloy causes both the hydrogen desorption from specimen and the hydrogen segregation at traps. As a result of desorption, the total hydrogen content decreases and hydrogen depleted zone is formed seen on the fracture surface as a ductile outer layer. Desorption is complited during first period of ageing and for longer ageing only hydrogen segregation takes place without change in total hydrogen content.

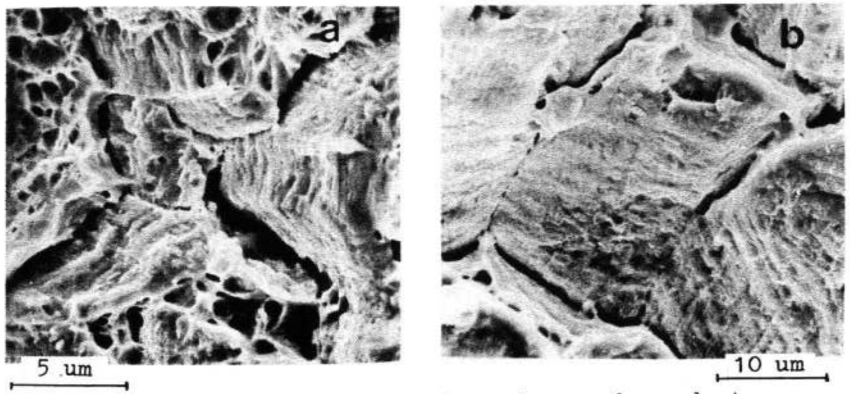


Fig.5. Fracture surface of specimens charged at 600°C , 30 to 45 MPa for 1300 hrs.

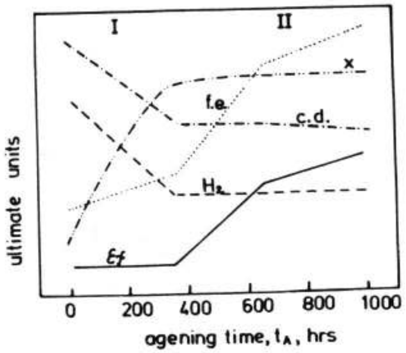


Fig. 6. EDS analysis for matrix (a),particle around which fish eye was formed (b) material in bottom of hole after particle removing (c).

Fig. 7. Change of elongation to fracture (——), hydrogen content (– – –),density of intergranular crevices (—·—),fish eyes (·······), thickness of ductile layer (—··—) vs ageing time of hydrogen charged specimens.

Since parameters affecting the fracture mode and plasticity of hydrogen charged Ni alloys (impurity segregation, inclusion size and volume fraction, loading mode and strain rate) are not altered in present experiments, the observed change of fracture mode and plasticity during ageing can be attributed to the difference

in hydrogen content and in its distribution.
Described features of fracture surface of hydrogen charged Inconel 600 (conical dimples, boundary crevices, fish eyes) are commonly observed in iron and steel Therefore,the explanation of their formation should not involve hydrogen effects specific for certain materials as hydrogen induced softening of iron or hydride formation in Ni base alloys.

At the beginning of ageing (first period), modification of plastic deformation processes by hydrogen dissolved in matrix could be mainly expected. That assumption is supported not only by the decrease in elongation to fracture (in comparison to as received material) but also by the appearence of serrated yielding for hydrogen containing specimens. Hydrogen induced modification of plastic deformation processes should also affect the fracture mode of material. The main process involved in the formation of ductile fracture of ferrous and nonferrous metals is the dislocation cross-slip which assists in void nucleation, development and coalescence (Goods et al.,1979).Suppression of cross-slip and thus inhibition of secondary slip and promotion of refiniment and planarity of slip by hydrogen established in Fe (Lunarska et al.,1982,) and Ni (McInteer et al.,1980) should alter ductile fracture. Constriction of cross-slip promotes the lateral expansion of voids and glide plane separation which leads to the appearence of conical dimples on fracture surface. On the other hand, suppression of cross-slip prevents the relaxation of stress concentration formed at sites with high plastic incompatibility as boundaries at which slip bands from adjacent grains impinge; grain boundary decohesion occurs seen on fracture surface as IG crevices.

Redistribution of hydrogen during ageing causes the effect of hydrogen in solid solution to be less pronounced after long time than that of hydrogen segregated at inclusions.

As seen from EDS analysis (Fig.6), the particles around which fish eyes nucleate are the chromium carbides probably covered with iron sulphide shell surrounded by the material with a high segregation of S.This implies the weak cohesion between particle and matrix (Brooksbank et al.,1971) which may be further decreased by hydrogen.The low cohesion interfacesserve as hydrogen collectors during ageing.Plastic deformation of such metal causes the strain induced dissolution of accumulated hydrogen (Zurek et al.,1980) and hydrogen enrichment of material within slip bands formed at inclusions.As a result,transgranular cracks occur due to nucleation and coalescence of microvoids within slip bands (Nagumo et al.,1980)or due to hydrogen induced decohesion along heavely strained material. The cracks are seen on fracture surface as fish eyes.It should be noted that in present case, the effect on metal plasticity produced by hydrogen modification of deformation processes are found to be more detrimental than that of hydrogen accumulated at particles, cf. Fig. 7.
Specimens charged with hydrogen at 600°C and pressure 30-45 MPa for 1300 hrs exhibit on fracture surface the sites with grain boundary separation shown in Fig.5. Since that feature has not been observed for specimens charged at lower temperature and

pressure, the assumption can be made that in this case the hydrogen attack takes place. The formation of bubbles at Cr carbides has been reported for austenitic steel exposed to hydrogen at 600°C and 14 MPa for 480 hrs (Yacaman et al., 1985). Presented in above work analysis of possible hydrogen attack shows that the hydrogen charging conditions used in present work (600°C, 30-45MPa, 1300hrs) may be sufficient for the formation of microvoids at grain boundary carbides during charging, which are plastically expanded during tensile testcausing grain boundary separation.. No significant effect of RT ageing on that process could be expected.

CONCLUSIONS

1. Hydrogen charging of Inconel 600 at temperature up to 550°C and pressure 30-40 MPa produces no change in microstructure. Hydrogen attack can, however, occur due to exposure at higher temperature and pressure.
2. Redistribution of hydrogen during ageing at RT causes change in fracture. Modification of deformation processes due to hydrogen present in solid solution promotes the formation of grain boundary crevices and conical dimples. Accumulation of hydrogen at particles with high S segregation causes the formation of fish eyes.
3. Effect of hydrogen present in solid solution is more detrimental on metal than that segregated at large particles

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