HYDROGEN EMBRITTLEMENT OF CAST ALLOY 718

EFFECTS OF

HOMOGENIZATION, GRAIN SIZE AND δ -PHASE

Göran Sjöberg* and Daniel Cornu**

* Materials Research and Development
Volvo Aero Corporation
461 81 Trollhättan
Sweden

** Laboratoire matériaux et procédé, Snecma Moteurs, Division Moteurs Fusées
B.P. 802, 27208 Vernon, Cedex
France

Abstract

Full size and sub-size tensile test specimens were machined from well characterized, specially cast, alloy 718 test material. Hydrogen was charged into the test specimens by the exposure to a pure 100 bar hydrogen atmosphere at 600 °C during 24 hours, leaving 35 ppm H in the full size test bars and 6 ppm in the sub-size. The evolvement of the fracture process was studied by, in SEM, in situ, testing of the sub-size bars polished on one side. Conventional tensile testing of the full size bars was performed at room temperature in air. Examination of the fracture surfaces was made in SEM.

Secondary phases are known to be important factors for hydrogen embrittlement, due to the trapping of hydrogen atoms. The effect of the Laves phase, always present in cast alloy 718, was examined by comparing a low temperature homogenization (1093 °C) with one made at higher temperature (1160 °C) where the phase partly dissolves. The better homogenization reduced the susceptibility to hydrogen embrittlement. The effect of the δ -phase was examined by deliberate precipitation and it was found that hydrogen reduces dramatically the elongation at fracture if δ -phase is present and if the hydrogen concentration is high – at lower only a minor effect is seen. The role of grain boundaries, also trapping hydrogen, was studied by comparing material from thick and from thin parts of the cast test blanks. The fine grain material loses proportionally more ductility than the material with the coarser grain structure. Also, a synergistic embrittlement effect of low hydrogen content and small amounts of δ -phase precipitates at grain boundaries is evident.

The study of the evolvement of the fracture, during the straining of the hydrogen containing material, reveals that the Laves phase fractures already in the elastic regime and that early grain boundary separation is important.

Introduction

Hydrogen is as a common fuel in rocket engines but due to the embrittlement it causes in many of the high nickel content materials used, alloy 718 being no exception, it is a hazard to the integrity of engine components [1,2]. One of the key, as well as controversial issues, seems to be how the hydrogen is trapped inside the material and the effect of such trapping on the premature failure [3]. Phase interfaces, e.g. between the bulk matrix and minor precipitated phases (the FCC-matrix and Laves phase, MC, δ -phase and gamma double prime in alloy 718), are sites where hydrogen accumulates and reasonably weakens the bond strength [4,5]. Less obvious is that the hydrogen may accumulate inside the secondary faces, due to the less packed structure of such less ordered phases, and might act as local internal high concentration hydrogen sources alleviating the crack initiation and propagation. Hydrogen may also severely embrittle the secondary phases per se.

In cast alloy 718, Laves and MC eutectics are formed between dendrites by segregation of the niobium during the solidification. The amount of Laves phase may however be reduced by subsequent high temperature homogenization when niobium is allowed to diffuse away from the eutectic areas. The effect of the degree of homogenization on the hydrogen embrittlement is therefore an important part of this study. The presence of the δ -phase in alloy 718 microstructures deteriorates ductility. In castings, this phase can however generally be eliminated by solution at high temperature. In wrought material the same heat treatment will, however, cause unacceptable grain growth. Often castings need to be joined with wrought material by welding. Although the cast part may be free from the δ -phase, the weld joint itself consists of newly cast material and can neither be homogenized nor be heat-treated to eliminate the δ -phase. For this and the reasons mentioned above the combined effect of δ -phase and hydrogen on ductility was investigated.

The embrittlement was studied by conventional tensile testing at Volvo; at Snecma by a special, SEM, in situ, sub size tensile testing in which the evolvement of the fracture could be studied in detail on polished test bars.

Experimental

Staircase shaped test blanks were cast in a symmetric tree to allow for consistent test material of various thickness as shown in figure 1.

Figure 1. Test blank (w 150 mm, thickest section 25 mm) and the investment cast tree arrangement.

The chemical composition of the material used in this investigation is shown in table I.

Table I Chemical composition, weight %

Ni	Cr	Fe	Nb/Ta	Mo	Ti	Al
53	19	17	5.2	3.1	1.0	0.68

Co	B	C	P	S	N
0.11	0.003	0.05	0.004	0.006	58 ppm

Careful characterization of the microstructure was made by comprehensive metallographic sectioning. In table II, the grain size and the Primary Dendrite Arm Spacing (PDAS) as well as the Secondary Dendrite Arm Spacing (SDAS) are shown for the different thicknesses of the test blanks. It should be noted that the PDAS numbers, although typical, are for practical reasons estimates only. Scattered microporosity was found but considered to have minor effect on mechanical properties.

Table II Grain size, Primary (PDAS) and Secondary Dendrite Arm Spacing (SDAS)

	1	2	3	4
Thickness, mm	25	12	7	3
Grain size, mm	2.9	1.5	1.0	0.6
PDAS, μm	300	200	150	100
SDAS, μm	52	36	31	22

The effect of homogenization and of the presence of δ -phase on the hydrogen embrittlement was investigated by exposing the tensile test specimens to different heat treatments as shown in table III. The low temperature homogenization heat treatment, 1093 °C/1.5 h, corresponds to the basic standard used by foundries. The higher temperature homogenization, 1160 °C/4 h, is commonly used as an integrated part of the HIP-processing applied to reduce porosity in castings for critical applications.

Table III Heat treatment schedule for the conventional tensile test specimens

Thickness	25 mm			7 mm
Homogenization	1093 °C/1.5 h	1093 °C/1.5 h 1160 °C/4 h		1093 °C/1.5 h 1160 °C/4 h
δ -phase H/T			940 °C/ 8 h	
Solution H/T	1050 °C/1 h	1050 °C/1 h		1050 °C/1h
Aging	720 °C/8 h 620 °C/8 h	720 °C/8 h 620 °C/8 h	720 °C/8 h 620 °C/8 h	720 °C/8 h 620 °C/8 h

The 1160 °C/4 h homogenization heat treatment is much more efficient than the 1093 °C/1.5 h treatment. From a theoretical estimate (based on the diffusivity of niobium in nickel) approximately one order of magnitude better. The effect of the homogenization heat treatment on the variation of the niobium concentration over the secondary dendrite arms was examined by EDS line analysis on polished samples. Although not accurate for absolute analysis, the variations are reasonably well reflected. In figure 2, the variations observed for the different heat treatments are shown by the curves normalized to 5 % niobium. By comparing the niobium distribution curves representing the two levels of homogenization with the one for the as cast material, the effectiveness of the high temperature homogenization is evident.

Figure 2: Effect of the homogenization heat treatment on the niobium concentration across secondary dendrite arms HT1: 1093 °C/1.5 h, HT2: 1093 °C/1.5 h , 1160 °C/4 h . For comparison reason, the three profiles have been normalized to 5 % niobium.

The δ -phase precipitation heat treatment, 940 °C/ 8 h, AC, was established by trial and error to produce a significant amount of δ -phase. Since the solution temperature is sensitive to the niobium levels and since the niobium content is higher in the center there is a significant variation of the amount of δ -phase across the thickness as indicated in figure 3.

For the SEM, in situ, sub size, tensile testing, a different δ -phase precipitation heat treatment, 940 °C/2 h, 10 °C/min, was used.

Tensile test specimens representing coarse and fine microstructure were excised from the test blanks at the positions as indicated in figure 4. The thickness of the rectangular cross-section tensile test specimens was 5 mm, mid-section width 8 mm, length 40 mm and with a total test bar length of 125 mm. Elongation at fracture was measured the aid of two indentation marks 32 mm apart.

Figure 3: Macroscopic δ -phase distribution. The dark areas illuminate inter-dendritic high niobium concentrations through the δ -phase precipitation.

Figure 4: Position of tensile test specimens in the test blanks. 6 test bars, three of which charged with hydrogen, were used to examine the influence of hydrogen embrittlement for each heat treatment as shown in table III.

The dimensions of the sub size bars for the SEM, in situ, tensile testing was 32 x 12 x 0.8 mm with a neck of 6 x 2.2 x 0.8 mm.

Strain rate for the full size bars was 0.005/min up to 2 % elongation in cloosed loop control. Above 2 % strain cross-head speed was 2 mm/min. The cross-head rate of the SEM, in situ, testing was 0.06 mm/min (a strain rate of 0.03/min on 2.2 mm active length)

Hydrogen charging

All test bars were charged in pure hydrogen as one batch in an autoclave at 100 bar, 600 °C and 24 hours. For the 5 mm thick test bars the results from the hydrogen analysis made on tested bars (after testing) are shown in the diagram of figure 5.

Figure 5. Hydrogen charging results for the four different specimen conditions, low temperature homogenization, high temperature homogenization, with δ -phase and with the finer microstructure in the test bars from the thin section material.

These results indicate that secondary phases may preferentially absorb hydrogen.

The sub-size, 1 mm thick, test bars, charged at the same time as the larger ones did, however, not retain as much hydrogen. The content was only 6 ppm. The reason for this is attributed to the fact that the autoclave charge was removed while still hot for rapid outside final cooling during which time the thinner test bars lost most of the charged hydrogen though retaining enough [7] for generating valuable results in this investigation.

Results and Discussion

Average hardness was measured by the Brinell method. The numbers are given in table IV. Evidently, better homogenization allows for higher hardness, as does the finer grain size of the thin section. It seems, however, questionable that the hardness is higher in the material containing δ -phase than in the same material without δ -phase since the matrix is drained of Nb through the δ -phase precipitation and consequently less Nb is left for the gamma-double prime hardening. One explanation could be general scatter. However, the same tendency is seen for the yield strength.

Table IV Brinell hardness numbers

Thickness (mm)	25			7
Heat treatment	LT Hom	HT Hom	HT Hom + δ	HT Hom
Hardness (HB)	342	358	365	374

Yield strengths for all tested bars are compared in figure 6. Three test bars of each variety were tested. The position of the bar in each group corresponds to the position from where the test bar was excised of the test blank (only for the three groups excised from the thicker part) as shown

in figure 4 above.. Thus, the test result bar in the middle of each group of three (except the fine grain group) in the bar chart corresponds to the material at the center of the blank.

Figure 6. Yield strength results for the four different material states as per table III. Two of the bars in the LT-homogenized hydrogen charged group did not render strength values due to gauge device slippage.

Hydrogen increases, although marginally, the yield strength for all material states. Presumably this is a Cottrell atmosphere effect on dislocation mobility. As is readily seen in fig. 6, the strength improves by better homogenization but above all by finer grain size. As with the hardness, the strength of the δ -phase containing material is unexpectedly higher than the non-containing material without δ -phase.

The severe hydrogen embrittlement of alloy 718 is not reflected in the ultimate tensile strength (fig. 7), although tendencies may be seen. The main reason for this is the flat stress strain-curve. A loss of ductility, as measured by elongation, does not materialize in a significant loss of the ultimate tensile strength.

The deleterious effect of hydrogen in alloy 718 on the elongation (fig. 8) is obvious, with more than half of the ductility lost for all of the four states of the material. For the δ -phase containing material the embrittlement effect of the hydrogen is even more dramatic. The center position test bar with its higher δ -phase content loses almost all the ductility which clearly demonstrates that the hydrogen preferentially interacts with the δ -phase in the premature fracture process. An interesting fact is that the fine grain material loses proportionally more of its ductility than the coarser grain material.

The ductility values observed in the SEM, in situ, sub-size, tensile testing (fig. 9) must be interpreted with care since scatter is larger in sub-size than in full size testing. However, the general trend of the full size testing is confirmed although hydrogen content is much smaller, 6 versus 35 ppm. With the full solution of the δ -phase at 1050 °C the embrittlement effect of hydrogen is smaller than with the presence of δ -phase.

Figure 7. Ultimate tensile strength results for the four different material states, as per table III.

Figure 8. Elongation at fracture, shown for the four different states of the material, as per table III. (The very low values for the right hand test bars in the low temperature and the high temperature homogenization groups are associated with local concentrations of micro-porosity found in the fracture surfaces of these bars.)

Figure 9. Elongation at fracture (average of three tests) at the SEM, in situ, sub-size, tensile testing. The hydrogen content is here only 6 ppm compared with approximately 35 ppm for the full size tensile test bars.

At the 940 °C, 1 hr, heat treatment (following on the 1050 °C full solution heat treatment) the δ -phase precipitation is already initiated (the TTT-diagram in [6] indicates that precipitation commences at a fraction of an hour at 940 °C). Since δ -precipitates nucleate at grain boundaries, the large reduction of ductility indicates a synergetic effect with grain boundaries.

Figure 10. Reduction of area for the four different states of the material, as per table III.

The reduction of area, (fig. 10) is a less sensitive measure for the hydrogen embrittlement than the elongation,. The embrittlement of the δ -phase containing material is, for instance, hardly noticeable as measured as reduction of area while as measured as elongation a very dramatic influence is revealed.

The progress of the fracture process on the polished surfaces in the SEM, in situ, tensile testing is illustrated in figure 11. Inter-dendritic sites are weak in the presence of hydrogen and the Laves eutectic phase fractures preferentially. Surprisingly, there are no evidence of separation at the interfaces between the matrix and the secondary faces (Laves- and δ -phase). In this investigation, this observation is consistent with the limited reduction of the elongation caused by hydrogen when the material contains significant amounts of δ -phase, figure 9. However, as has been reported [5], the interface between the δ -phase and the matrix is a favored in fatigue crack growth in high pressure hydrogen and at high stress intensities. In the full size tensile testing of δ -phase containing material, at the higher hydrogen levels, these interfaces are very clearly preferred crack path. This is evident from the fractographs shown in figure 12.

Examination of the fracture surfaces of the full size test bars at low magnification does reveal a striking similarity for all the four heat treatment states, with and without hydrogen. All surfaces appear, in one essence, as the ones representing the δ -phase containing material shown at low magnification in figure 12, in that the secondary dendrite arms are readily visible, which makes the fracture surfaces appear rough. This, however, hides the fact that the surfaces of the hydrogen embrittled material on a larger scale are smoother, a fact associated with material embrittlement. The smoother appearance can be seen either by the naked eye or better by stereo pair SEM-images. In the fine grain hydrogen charged material there is, however, an additional interesting feature consisting of smooth patches (covering 5 – 10% of the fracture surface, as illustrated in figure 13. These patches are most likely traces of early grain boundary separation similar to those observed at the lower hydrogen level, fig. 11b.

Figure 11: The fracture process as studied by SEM, in situ, of hydrogen charged cast alloy 718. Inter-dendritic separation, (a); early stage of grain boundary separation, (b); evolution of grain boundary fracture, (c) and (d); Laves phase fracture preference over δ -phase, (e) and (f). In these sub-size tensile bars the hydrogen level was low, 6 ppm.

As mentioned earlier, elongation at fracture is a sensitive measure of hydrogen embrittlement of cast alloy 718 due to the flat appearance of the stress strain curve. A small change of the

strain hardening results in a large effect on the elongation at fracture. The early fracture by the presence of hydrogen of secondary phases, as eutectic Laves phase, and the separations at the grain boundaries (fig.11b, 13) may well account for the observed moderate losses of ductility as the surprisingly strong embrittlement observed in the fine grain material compared with the coarser grain material (fig. 8).

Figure 12: Typical fracture surfaces of the full size tensile test bars of the δ -phase containing material at two different magnifications, without and with hydrogen.

Figure 13: Smooth patches on fracture surfaces of the fine grain hydrogen charged material interpreted as premature grain boundary separations.

However, at high hydrogen concentrations, the ductility is reduced to almost nil if a limited amount of δ -phase is present in the microstructure. This indicates that hydrogen severely embrittles the interface between matrix and the δ -phase or the δ -phase itself.

Interactions between hydrogen and secondary phases may indeed be very complex [3] and our observations explain, at best, only a part of the hydrogen embrittlement effects in cast alloy 718.

Conclusions

1. Reducing the amount of Laves phase through high temperature homogenization improves the ductility of hydrogen charged cast alloy 718.
2. At high hydrogen concentration the presence of δ -phase dramatically reduces the ductility.
3. Embrittlement of the fine grain cast material is larger than the coarse grain material due to grain boundary hydrogen sensitiveness.

Acknowledgements

The experimental work of graduate students, Jonas Ciardi, at Volvo in Sweden and David Pallois, at Snecma in France is here gratefully acknowledged. The supervisor role of Thomas Kruslind at the Volvo Aero Corporation, Materials Technology Laboratory is also acknowledged.

References

1. Leslie G. Fritzmeier and Willis T. Chandler, "Hydrogen Embrittlement – Rocket Engine Applications", Superalloys, Supercomposites and Superceramics (Academic Press, Inc., 1989), 491 – 524.
2. R. P. Jewett and J. A. Halchak, "The Use of alloy 718 in the Space Shuttle Main Engine", Superalloys 718, 625 and Various Derivatives, ed. E. A. Loria, (Warrendale, PA, The Mineral, Metals & Materials Society, 1991), 749 – 760.
3. I. M. Bernstein, "The Role of Hydrogen: Is the Story Any Clearer?", Hydrogen Effects in Materials, ed. A. W. Thompson and N. R. Moody, (The Mineral, Metals & Materials Society, 1996), 3 – 11.
4. B. G. Pound, "Hydrogen trapping in Precipitation Hardened alloys", *Acta metall. mater.*, 38 (12) (1990), 2373 – 2381.
5. S. Fukuyama and K. Yokogawa, "Effect of Heat Treatments on Hydrogen Environment Embrittlement of Alloy 718", Superalloys 718, 625 and Various Derivatives, ed. E. A. Loria, (Warrendale, PA, The Mineral, Metals & Materials Society, 1994), 807 – 816.
6. H. I. Eiselstein, "Metallurgy of Columbium-Hardened Nickel-Chromium-Iron Alloy", *Advances in the Technology of Stainless Steel and Related Alloys*, (ASTM STP No. 369, 1965), 62 – 77.
7. P. D. Hicks and C. J. Alstetter, "Internal Hydrogen Effects on Tensile Properties of Iron- and Nickel-base Superalloys", *Metallurgical Transactions A*, 21A (1960), 365 – 372.