NASA TECHNICAL MEMORANDUM

(NASA-TM-73691) CRYOGENIC PROPERTIES OF A NEW TOUGH-STRONG IRON ALLOY (NASA) 13 p HC A02/MF A01 CSCL 11D

N77-28269

Unclas
G3/26 39297

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ABSTRACT

A program was undertaken to develop an iron-base alloy having a fracture toughness of 220 MPa $\cdot$ m$^{\frac{1}{2}}$ (200 ksi $\cdot$ in$^{\frac{1}{2}}$) with a corresponding yield stress of 1.4 GPa (200 ksi) at -196° C. An Fe-12Ni alloy was selected as the base alloy. Factors considered included reactive metal additions, effects of interstitial impurities, strengthening mechanisms, and weldability. The goals of this program were met in an Fe-12Ni-0.5Al alloy strengthened by thermomechanical processing or by precipitate strengthening with 2 percent Cu. The alloy is weldable with the weld metal and heat affected zone in the post-weld annealed condition having toughness equivalent to the base alloy.

INTRODUCTION

Nickel-containing steels are frequently selected for use at cryogenic temperatures because of their excellent toughness and/or strengths. For example, 304 stainless steel containing 8 percent nickel is characterized by very high toughness at cryogenic temperatures, but it has relatively low strength. In contrast, 18Ni, 200 grade maraging steel is characterized by very high strengths at cryogenic temperatures, but with a penalty of reduced toughness. Similarly, 9Ni steel is characterized by good toughness and moderate strength at cryogenic temperatures, while 9Ni-4Co steel has greater strength, but substantially lower toughness in the same temperature range.

The purpose of an experimental program conducted at the NASA-Lewis Research Center was to identify a single Fe-base alloy that would combine the

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apparent divergent cryogenic properties of high toughness and high strength. A goal was set for this program to develop an alloy with -196° C properties of fracture toughness equal to 220 MPa $\cdot$ m$^{\frac{1}{2}}$ (200 ksi $\cdot$ in$^{\frac{1}{2}}$) and corresponding yield strength equal to 1.4 GPa (200 ksi). The initial work at the University of California, Berkeley Campus, on Fe-12Ni alloys [1] provided an impetus for the direction of our program. One method of achieving high toughness in iron-base alloys for cryogenic service is to keep embrittling interstitial element (C, O, and N) concentrations low by alloying with reactive ("gettering") elements which will precipitate them as innocous particles. A total of 11 reactive metals were added to the Fe-12Ni system including Al, Hf, La, mischmetal, Nb, Si, Ta, Ti, V, Y, and Zr. Preliminary studies [2] showed that Al, Nb, Ti, and V were most effective in improving toughness. Aluminum was chosen as the optimum reactive metal for more detailed investigations. Factors considered in the program were optimization of nickel content, optimization of Al content, the effects of interstitial impurities, strengthening mechanisms, and weldability.

EXPERIMENTAL PROCEDURE

Alloys were prepared by arc-melting 1 kilogram laboratory size ingots followed by hot rolling to a thickness of 7 millimeters. Rolling was normally at 1100° C, but temperatures of 650° and 25° C were also employed to investigate the effects of thermomechanical processing on toughness and strength. Aluminum and Ni optimization were studied in the Fe-Ni-Al alloy system with Al contents ranging from 0 to 4 atom percent and Ni contents ranging from 0 to 18 atom percent. Carbon was intentionally added to the experimental alloys to determine the maximum amount of this impurity that could be tolerated and still maintain high toughness. Carbon was also considered as a possible strengthener along with precipitate strengthening and solid solution strengthening. Weldability of the experimental alloys also was explored using the gas-tungsten arc (GTA) welding technique. Details of the welding techniques can be found elsewhere [3]. Evalua-

tion of toughness was by slow bend fracture toughness tests on precracked Charpy specimens using the Equivalent Energy (K_{Icd}) technique [4] to analyze the data. Comparison of results using this technique on Charpy specimens machined from valid K_{Ic} specimens has shown good agreement between K_{Icd} and valid K_{Ic} measurements [5].

RESULTS

Aluminum Optimization

Initial efforts were directed at improving the toughness of the Fe-12Ni base alloy as a result of adding reactive metals. The results of these studies [2] indicated that Al additions gave the best tradeoff in toughness and strength. The effects of Al additions on the cryogenic toughness of the Fe-12Ni base alloy are illustrated in figure 1. The three curves shown in this figure represent 2-hour anneals at temperatures of 550° C (single phase α region), 685° C (two phase $\alpha + \gamma$ region), and 820° C (single phase γ region). A maximum in toughness is achieved at an Al concentration of 0.5 atom percent for the two higher annealing temperatures and from 0.5 to 1.0 atom percent Al after annealing at 550° C. The improvement in toughness is attributed to the scavening of interstitial impurities as well as a reduction in grain size by about one-half with these dilute Al additions to the Fe-12Ni alloy. Because of the sharp drop in toughness for the Fe-12Ni-1.0Al alloy upon annealing at 685° and 820° C (fig. 1), 0.5Al was selected as the more promising Al concentration for further detailed studies.

Nickel Optimization

The effects of Ni content in the Fe-0.5Al system annealed at 550° C and tested at -196° C are shown in figure 2. An improvement in toughness with increasing Ni content is noted for Ni contents up to 12 atom percent. Beyond this amount a decrease in toughness occurs at Ni contents to 18 atom percent. Accompanying this drop in toughness is an increase in the amount of retained austenite (as determined by X-ray diffraction) in these alloys. The austenite level is nil in the

12Ni alloy but increases to as high as 18 percent in the 18Ni alloy. Scanning electron micrographs indicated that fracture at -196° C was primarily by cleavage at 10Ni, a mixture of cleavage and large dimpled fracture at 12Ni, while at 18Ni the fracture was almost planar and characterized by very fine dimples. The maximum in toughness occurring at the intermediate Ni content of 12 atom percent is believed to be due to the offsetting effects of Ni changing the fracture mode and promoting retained austenite at higher Ni contents. Toughness increased as the fracture mode changed from cleavage (which normally is characteristic of brittle, low toughness materials) at 8 to 10Ni to primarily dimpled rupture at a 12Ni content and above. Beyond 12Ni the presence of retained austenite lowered the yield strength which in turn reduced the toughness as austenite content further increased.

Effects of Welding

The weldability of the Fe-12Ni-0.5Al alloy is illustrated in figure 3, where the ratio of toughness for either weld metal or the heat affected zone (HAZ) to that of the base alloy are compared for the alloy in different conditions. The weld conditions shown are as hot-rolled, annealed at 550° C prior to welding, and hot-rolled, welded plus a post-weld heat treatment at 550° C. It should be noted that for the Fe-12Ni-0.5Al alloy a post-weld heat treatment is required to achieve toughness in the weld or HAZ comparable to that in the base metal.

The weldability of the Fe-12Ni base alloy containing two other reactive metal additions is also shown in figure 3. An Fe-12Ni-0.25Ti exhibits excellent toughness in the weld or HAZ under all test conditions. In contrast, an Fe-12Ni-0.25Nb alloy exhibits poor weldability under all test conditions. Electron microprobe scans showed that Nb segregation to dendritic cell boundaries in the weld and HAZ probably accounts for the poor weldability in the Fe-12Ni-0.25Nb alloy. From these results, Ti additions are considered to be most effective in providing good weldability in the Fe-12Ni alloy, while Al additions also produce excellent weldability if the alloy is given a post-weld anneal.

The results described above have shown that an Fe-12Ni-0.5Al alloy possesses cryogenic toughness in excess of the 220 MPa $\cdot$ m$^{\frac{1}{2}}$ goal set for this program. The yield strength of this alloy typically ranged from 0.9 to 1.1 GPa, or about 65 to 70 percent of the 1.4 GPa strength goal.

Effects of Carbon Additions

Research efforts were subsequently directly at strengthening the Fe-12Ni-0.5Al alloy. Solid solution strengthening was abandoned early in the investigation because this approach caused the ductile-brittle transition temperature to increase with a corresponding drop in toughness.

Figure 4 shows the effects of carbon additions on this alloy where strength and toughness are compared for alloys annealed in either the α or $\alpha + \gamma$ regions, 550° and 685° C, respectively. For both conditions, yield strength increases rapidly with small C additions (up to slightly less than 100 ppm C, by weight. Above this concentration there was a continual slight increase in strength with increasing C content up to the maximum level studied (1800 ppm C). At 550° C, the solutibility of C in α Fe is about 60 ppm C, approximately the level where rapid strengthening ceases in this alloy. Maximum toughness is achieved in this alloy at a carbon content of about 60 ppm, but the toughness goal of 220 MPa $\cdot$ m$^{\frac{1}{2}}$ can be met over a carbon range from 40 to about 600 ppm. The strength goal of 1.4 GPa was only achieved in this alloy at the maximum studied C content of 1800 ppm for the 685° C annealed material. However, at this C content, toughness dropped off to 60 MPa $\cdot$ m$^{\frac{1}{2}}$. Based on the rapid drop in toughness beyond 100 to 200 ppm C and the reduced strengthening rate with increasing C content beyond this concentration, an Fe-12Ni-0.5Al with about 150 ppm C is considered to be the optimum composition in this alloy series. Toughness exceeds the 220 MPa $\cdot$ m$^{\frac{1}{2}}$ goal and a yield strength of 1.1 GPa can be achieved for the 685° C anneal.

Effects of Thermomechanical Processing.

In addition to the standard 1100° C, hot-rolling procedure, ingots were rolled at 650° C (within the $\alpha + \gamma$ region) and at room temperature to evaluate thermo-mechanical processing effects on the strength of Fe-12Ni-0.5Al. Results showed that the 650° C rolled material was slightly stronger than the 1100° C hot-rolled material. The effects of the 1100° and 25° C thermomechanical treatments on cryogenic strength and toughness of Fe-12Ni-0.5Al are shown in figure 5 as a function of annealing temperature. Annealing the rolled materials within the temperature range of about 550° to 625° C resulted in maximum cryogenic toughness (i.e., -196° C toughness in excess of 220 MPa $\cdot$ m$^{\frac{1}{2}}$). Above about 625° C toughness for both rolled conditions decreased rapidly with increasing annealing temperature. In contrast, the cryogenic strength of 1100° C rolled materials was near 0.97 GPa over the annealing temperature range of 500° to 625° C and then increased to about 1.1 GPa at annealing temperatures of 650° to 820° C. The strength of cold rolled material was, in general, independent of annealing temperature over the range 500° to 820° C and was very near the strength goal of 1.4 GPa. Thus, the -196° C toughness and strength goals can both be met in cold worked Fe-12Ni-0.5Al annealed over the temperature range 550° to 625° C.

Effects of Copper Additions

A final method of strengthening Fe-12Ni-0.5Al involved precipitate strength-ening, copper was chosen as the precipitate material since it has very low solu-bility in Fe and precipitates as a copper-rich terminal solid solution containing a small amount of iron. Copper thus would not be expected to form an embrittling intermetallic compound. Copper additions ranging from 0.5 to 3.0 atom percent were investigated, with optimum strength and toughness being achieved at 2 per-cent Cu. Further results showed that annealing at 450° C gave the highest strength while still maintaining the toughness goal.

Strengthening in the Fe–12Ni system due to Cu is illustrated in figure 6. This figure summarizes the beneficial effects of both Al and Cu to the Fe–12Ni base alloy. Copper increases the strength of Fe–12Ni with little improvement in toughness, while aluminum dramatically increases toughness with an accompanying increase in strength. These two effects are synergistic, as shown for the Fe–12Ni–0.5Al–2Cu alloy. The -196° C strength of this alloy is 1.6 GPa with a corresponding toughness of 220 MPa $\cdot$ m$^{\frac{1}{2}}$. Copper is believed to be effective because of two possible mechanisms. First is the presence of very fine Cu precipitate particles, less than 400 A in diameter, as shown by the transmittion electron micrograph in figure 7. Dislocations are noted to bow around these particles, contributing primarily to strengthening in these alloys. Secondly, as Cu content was increased from 0.5 to 2 atom percent in the Fe–12Ni–0.5Al alloy, the martensite lath width decreased from 0.65 to 0.27 micrometer as shown for the 2 Cu addition in figure 7. This decrease in lath width further contributes to strength.

DISCUSSION

The results of this study have shown that it is possible to combine both high toughness and high strength in an Fe–12Ni–0.5Al alloy at -196° C. These results were achieved by optimizing the Ni and Al contents to achieve high fracture toughness, which reached a maximum value of about 310 MPa $\cdot$ m$^{\frac{1}{2}}$. This high toughness was developed in an alloy containing about 100 ppm C that was aged at 550° C to produce a microstructure consisting primarily of martensite and ferrite. High toughness is achieved by maintaining this microstructure with the absence of any austenite, which is inherrently weaker than the body centered cubic phases and in turn lowers the toughness as was demonstrated for higher Ni content alloys containing retained austenite. Results also have shown that toughness was greatly reduced in alloys containing oxygen as the predominant interstitial impurity. The primary role of the reactive metal Al is to scavenge such impurities to render them innocuous in the alloy. Grain size also decreased with Al additions, further

increasing strength and toughness. The Fe-12Ni-0.5Al can be welded by the GTA process and, after post-weld heat treatment, has toughness in the weld metal and HAZ comparable to that of the base metal. Strengthening in this alloy system was achieved by three methods, including the intentional addition of carbon, thermomechanical processing, and precipitated Cu.

A comparison of this series of experimental alloys with several commercial steels for cryogenic service is shown in figure 8. Toughness decreases with increase in strength for both the experimental alloy and the commercial steels. At a strength level near that for 200-grade maraging steel, the Cu-strengthened alloy has over a two-fold advantage in toughness. Further, at the toughness level for 304 stainless steel, the experimental alloys offer over a two-fold advantage in strength.

CONCLUSIONS

Based on a study of toughness and strength of Fe-Ni-Al alloys for cryogenic service, the following conclusions are drawn:

1. An optimum Ni content of 12 atom percent and an optimum Al content of 0.5 atom percent provide an alloy with exceptionally high toughness and moderate strength at -196° C.

2. The Fe-12Ni-0.5Al alloy can be further strengthened with Cu-rich precipitates to meet the 1.4 GPa (200 ksi) yield strength goal of this program and still maintain the toughness goal of 220 MPa $\cdot$ m$^{\frac{1}{2}}$ (200 ksi $\cdot$ in$^{\frac{1}{2}}$).

3. Thermomechanical processing and the addition of about 150 ppm carbon are additional effective methods of strengthening while maintaining high toughness.

4. Post-weld annealing of welded material produces toughness in the weld and heat affected zones comparable to that of the base alloy.

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3. J. H. Devletian, J. R. Stephens, and W. R. Witzke, Welding J., Res. Suppl., 56(4): 97-s (1977).

4. F. J. Witt and T. R. Mayer, ORNL-TM-3894 (1972).

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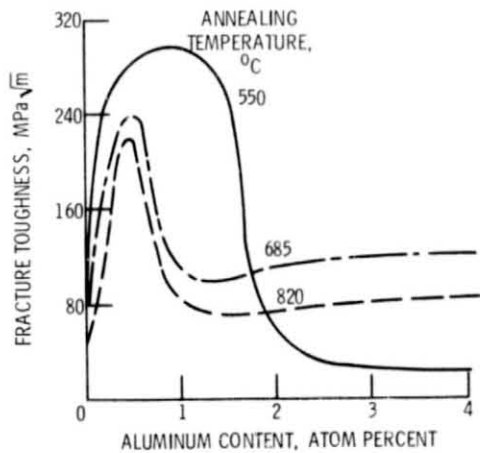
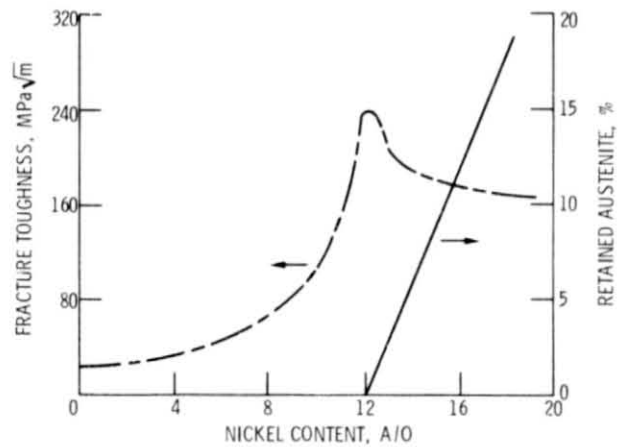Figure 1. - Effect of aluminum content on fracture toughness of Fe-12Ni-Al alloys at -196°C.

Figure 2. - Effect of nickel content on fracture toughness and retained austenite of Fe-Ni-0.5Al alloys annealed at 550° C and tested at -196° C.
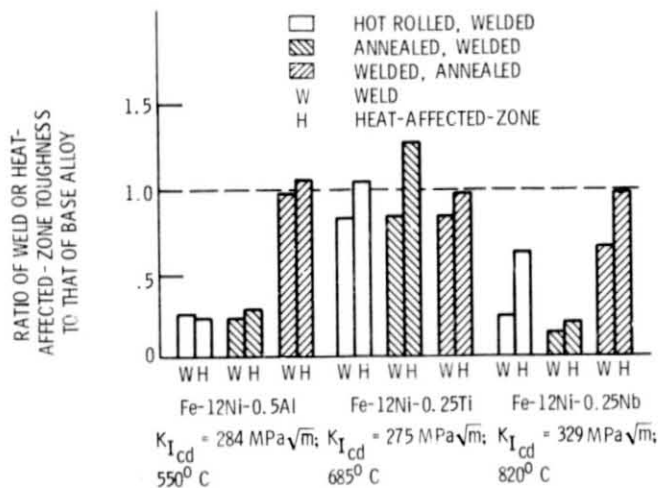
Figure 3. - Effects of pre- and postweld heat treatments on -196° C fracture toughness of welded Fe-12Ni alloys compared to base alloy heat treated for maximum toughness.
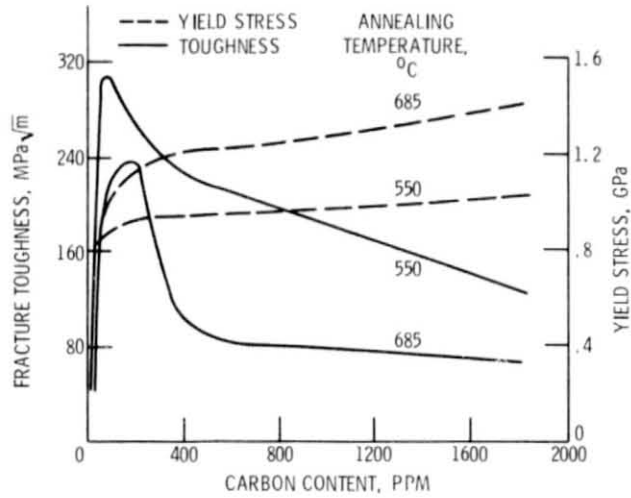
Figure 4. - Effect of carbon content on fracture toughness and yield stress of Fe-12Ni-0.5Al alloy at -196° C.

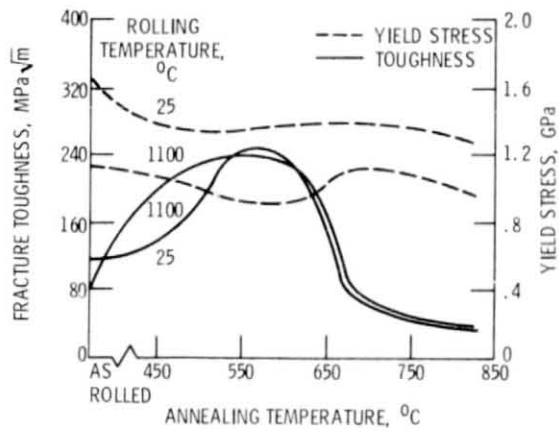Figure 5. - Effects of thermomechanical processing on fracture toughness and yield stress of Fe-12Ni-0.5Al alloy at -196° C.
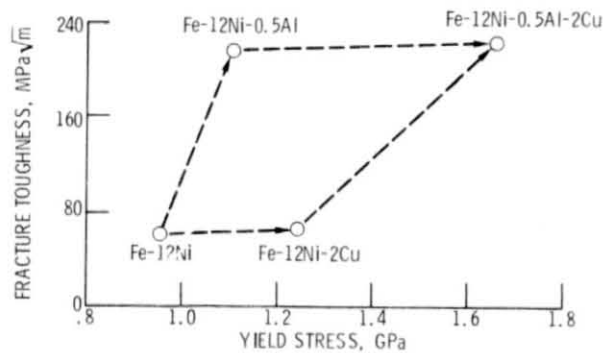

Figure 6. - Contributions of Al and Cu to toughness and strength of Fe-12Ni alloy annealed at 450° C and tested at -196° C.

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Figure 7. - Transmission electron micrograph of Fe-12Ni-0.5Al-2Cu alloy illustrating bowing of dislocation around less than 400 Å diameter Cu precipitates.
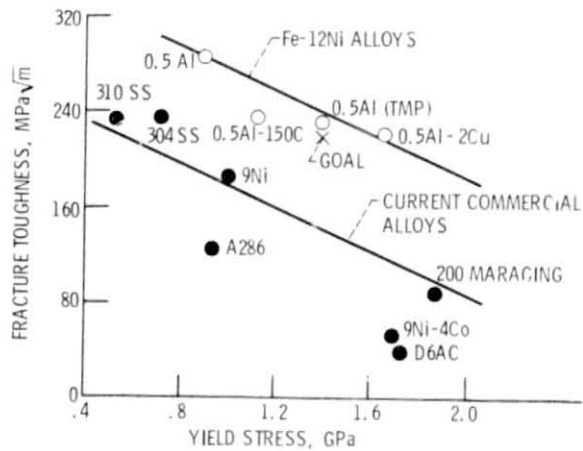

Figure 8. - Comparison of fracture toughness and yield stress of Fe-12Ni experimental alloys with commercial steels at -196° C.