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DEVELOPING HIGH IMPACT TOUGHNESS IN Fe-Ni
ALLOYS AT LIQUID NITROGEN TEMPERATURE

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ABSTRACT

By reheating interstitial-free Fe-Ni alloys to just above the A_s temperature, a pronounced improvement in the impact toughness, measured at liquid nitrogen temperature, was achieved. The useful reheating range for optimum impact toughness was extended to higher temperatures by preheating for a short period below the A_s or by reducing the heat up rate in the temperature range below the A_s . Cold working the specimens prior to reheating also extended the optimum reheating temperature range. Reheating, whether preceded by cold working or a preheating treatment, significantly grain refined the prior austenite grain structure and was the major cause of the improved impact toughness. For the 12% Ni alloy a critical grain size of 15 μm , or a lath packet size of 7.0 μm , was needed in order to achieve optimum impact toughness at liquid nitrogen testing temperature. The grain size requirement for high toughness in the 8% Ni alloy were more severe, requiring a grain size less than 5 μm , whereas the 16% Ni alloy required only a 30 μm grain size. The toughness of Fe-Ni alloys was thus found to be a function of both the Ni content and grain size.

INTRODUCTION

Brophy and Miller¹ were the first to report the unusually high toughness that could be obtained in low carbon 8 to 10% nickel steels. The improved toughness was developed in such steels by a heat treatment which consisted of austenitizing and then holding in the $\alpha + \gamma$ two phase field for suitable lengths of time. These original investigators suggested that austenite retention might be an important factor in producing the high impact toughness. Although the exact mechanism of austenite toughening was not explored, it is now known that the presence of either a stable fcc phase which exhibits no low temperature brittleness or an unstable one which can transform to non-brittle martensite during deformation can each have a positive effect on the toughness of the primarily bcc structure.²

Marschall et al.,³ in a further study of the same alloy system, suggested a different reason for the improved toughness. They felt that the small volume of austenite formed during reheating in fact acted as solute sinks for carbon and other deleterious elements, thereby improving the low temperature toughness of the remaining ferritic matrix. In Marschall's view, the mechanical properties of the retained austenite per se did not contribute significantly to the increased toughness. In support of this view, there is considerable evidence² to show that reducing the concentration of certain interstitial and metalloid elements in a ferritic lattice can have a pronounced effect on reducing the onset of brittle failure to much lower temperatures. Furthermore, Ooka et al.,⁴ in their more recent work on 9% Ni-0.1% C steels, found that no quantitative relationship existed between the relatively small volumes of retained austenite and the resulting low temperature impact toughness.

In conformity with Marschall's suggestion, it was decided to investigate the impact toughness of Fe-Ni lattices that are essentially free of the interstitials and metalloids normally found in commercial Fe-Ni steels. The method chosen for achieving such an alloy, was to use Ti as a scavenger of the undesirable elements.* Alloys of electrolytic Fe and Ni (8,12 and 16%), each with 0.5% Ti added, were selected for study.

The elements commonly known to cause poor toughness in ferritic lattices, besides C, include interstitial elements such as N, H, O and several non-metallic elements--principally S and P. Leslie and Sober,⁵ using internal friction measurements, found that as little as 0.15 wt% Ti was adequate in removing C, N and O from solution in concentrations normally found in high purity 99.9+% Fe. Also, Fishel et al.⁶ found Ti produced a similar effect with respect to sulfur.

The fact that nickel additions produce improved low temperature toughness in steels is well known. Only manganese and possibly several elements of the platinum group have comparable effects on iron. But the exact role Ni plays in terms of dislocation mechanisms and fracture mechanics is far from understood.² Brophy and Miller, for example, have determined that additions of the order 8 to 10% Ni were needed in overcoming notch impact sensitivity in an 0.1% C alloy containing up to 1% Mn. But besides its inherent toughening effect on iron, nickel also

* Note: In addition Ti could be used as a strengthening agent via solute and precipitation hardening in a further development of the alloy.

widens and depresses the temperature limits of the two phase $\alpha + \gamma$ field. This property of Fe-Ni alloys permits a great deal of thermal-mechanical processing flexibility which results in a variety of complex microstructures. For example, recently Miller⁷ has shown that an ultrafine grain microstructure of the order of several microns could be developed in nickel steels by annealing in the two phase $\alpha + \gamma$ region after first cold rolling. Other investigators^{8,9} have shown that hot rolling in the low temperature end of the austenite phase field could also produce considerable grain refinement. In each of the above cases, use is made of the ferrite-austenite transformation to refine and stabilize the grain structure. The expected improvement in yield strength through such grain refinement as suggested by the Hall-Petch relationship was observed for most of these alloys. In addition, past studies^{9,10} have also shown that notch toughness as measured by Charpy V-notch impact testing could also be increased by austenite and/or ferrite grain refinement, perhaps through a mechanism such as the one suggested by Cottrell.¹¹

In the present study, the impact toughness of Fe-Ni lattices to which titanium had been added is examined with special emphasis given to the 12% Ni alloy. The study includes a detailed investigation of the changes in impact toughness and microstructure brought about through reheating in both the two phase $\alpha + \gamma$ temperature range and in the higher, austenite temperature range.

MATERIALS AND EXPERIMENTAL PROCEDURES

The compositions of the Fe-Ni alloys used are listed below in weight percentages:

Alloy Designation	Ni	Ti	Mn	C	Si	S	N	P
8 Ni	8.07	0.52	<0.005	0.001	0.003	0.004	0.010	<0.005
12 Ni	12.07	0.32	<0.005	0.004	0.003	0.004	0.004	<0.005
16 Ni	15.50	0.34	<0.005	0.003	0.003	0.004	0.002	<0.005

Three-inch diameter ingots were cast in copper chill molds after induction melting 99.9+% Fe, Ni and Ti in zirconium oxide crucibles under first a 10^{-3} mm Hg vacuum and then an argon atmosphere. The ingots were next vacuum homogenized for 72 hr at 1100°C, furnace cooled, reheated to 1100°C, and then upset forged and air cooled.

One-half in. square cross section bars, 3 in. long, were next heat treated (50C°/min heat-up rate) in air furnaces controlled to $\pm 5C^{\circ}$ and then either ice brine quenched (100C°/sec quenching rate) or air quenched (2C°/sec). Charpy V-notch impact specimens or 0.25 in. diameter $\times$ 1.0 in. gauge length tensile specimens were carefully machined from the heat treated blanks.

Impact tests were carried out as specified by ASTM procedure E-23-64 on a 225 ft-lb capacity impact testing machine. In each case at least two specimens per heat treated condition were tested. Tensile properties were determined on an 11,000 lb capacity Instron Testing Machine using a crosshead speed of 0.04 in./min. Hardness measurements were made using at least 4 impressions for each specimen tested.

The kinetics of the phase transformation during both heating and cooling were studied by dilatometry. Measurements were made on 1 in. long by 0.25 in. diameter specimens. Heating and cooling rates between 8 to 50C°/min were used. The retained austenite content was determined using a Norelco diffractometer with $\text{Cu-K}_{\alpha 2}$ ($\lambda = 1.54\text{\AA}$) radiation and a graphite monochromator. The specimen surface was carefully prepared to remove any distorted material. The scan was run from $2\theta = 40$ to 83° including the (111), (200) and (220) fcc reflections.

The microstructure was examined by both light and electron microscopy. Specimens were etched with a 1:1 Kalling-5% nital mixed etchant. For better resolution, direct carbon replicas were taken from the optical mounts and examined at 60 kV in a Siemens 1A electron microscope. Fracture surfaces of broken Charpy specimens were examined with a Jeolco JSM-U3 scanning electron microscope operated at 25 kV via secondary electron emission.

RESULTS AND DISCUSSION

A. Improvement in Impact Toughness from Reheating

A phase transformation diagram generated by dilatometry heating and cooling measurements were determined for the series of Fe-Ni-Ti alloys. The austenite start and finish temperatures (A_s - A_f) on heating, and the martensite transformation start and finish temperatures (M_s - M_f) on cooling are shown in Fig. 1. Annealing at intermediate temperatures between the two temperature bands resulted in the slow formation of an $\alpha + \gamma$ duplex structure which will be described in detail in a later section. Curve A of Fig. 2 shows the impact energy absorbed for the 12% Ni alloy at -196°C by standard Charpy V-notch specimens. These specimens were initially austenitized at 900°C and air cooled to room temperature and then reheated to temperatures spanning both the equilibrium $\alpha + \gamma$ and γ phase fields, held for 1 hr, and water quenched. The typical lath martensite microstructure of the as-austenitized condition as viewed by two different optical techniques, are shown in Fig. 4a and 4b. The cleavage fracture resulting from impact testing at -196°C is shown in Fig. 4c. From a minimum absorption of 10 ft-lbs for this microstructure, the energy rose to a maximum of 110 ft-lbs and failure by ductile dimpled rupture when reheated between 675 and 725°C . It can be seen from Fig. 1 that this temperature range falls in the lower temperature region of the γ phase field.

Reheating to lower temperatures in the equilibrium $\alpha + \gamma$ two phase region produced only slight improvement of impact toughness over the as-austenitized condition, whereas reheating too high in the upper γ phase field likewise produced low toughness. This behavior is in marked contrast to that obtained by Marschall et al. in Fe-9% Ni low carbon

steels, where they found that maximum toughness from reheating occurred well below the A_s temperature in the $\alpha + \gamma$ two phase field. When reheated to higher temperatures near the A_s temperature they found a rapid drop off in toughness.

When austenitized specimens of the present 12% Ni alloy are reheated in the temperature range below the A_s temperature (e.g., 600 or 650°C), α transforms to γ initially at the preferred high energy sites at prior austenite boundaries and at lath packet boundaries, as shown in Figs. 5a and 6a. With additional annealing time or at higher temperatures, further transformation occurs, resulting in a $\alpha + \gamma$ microduplex structure as shown in Figs. 5b and 6b. The size of the micro-constituents in the structure is about 1 micron. But regardless of how long the specimens are annealed below the A_s temperature, they retain the relatively low toughness of the prior 900°C austenitized samples and show outlines of the prior austenite grain boundaries in their fracture surfaces. This can be seen by comparing, for example, the fractographs of Figs. 4c and 5c.

It should also be pointed out that the γ which forms during reheating appears to completely transform back to α when cooled to room temperature. No retained austenite was detected by X-ray (sensitivity ~2%) or by selected area electron diffraction. It, therefore, seems appropriate to conclude that retained austenite played no role in the improved toughness achieved in the present alloys.

As mentioned earlier, if the reheating is carried out above the A_s temperature, a pronounced improvement in toughness results. This improvement is found to correspond to the reversion of α to the γ phase via formation of new smaller grains of γ . Curve A of Fig. 3 shows how the new austenite grain size* varies with reheating temperature, and Fig. 7 shows how the Charpy energy increases with time at the optimum reheating temperature of 700°C. Examination of the microstructural changes which take place during isothermal annealing reveals that the high impact toughness corresponds to austenite grain refinement resulting from the α to γ transformation. This is illustrated in the series of micrographs and fractographs of Fig. 8.

During the heat up stage of the 700°C reheating treatment (Fig. 8a), a microduplex structure of $\alpha + \gamma$ (the dark areas of Fig. 8a) similar to the structure formed at 650°C forms while the specimen is heating up through the temperature range just below the A_s . At this point, the impact toughness has improved only slightly (~30 ft lb). With additional holding at 700°C, small areas from 3-10 microns in diameter of γ (the light areas of Fig. 8a) form at the prior austenite boundaries. Larger areas of γ eventually consume most of the $\alpha + \gamma$ duplex structure after 1 hr, with a corresponding improvement in the impact toughness (Fig. 8b), and after 2 hr nearly all of the duplex structure has disappeared (Fig. 8c).

* Note: The austenite grain size was found by Sasaki and Yokota¹² to be related to the lath packet size by a factor of 2.1 (e.g., austenite grain size of 15 μ would have a packet size of 7 μ).

Comparing the results of Brophy et al. and Marschall et al. with the above observations, it seems clear that for an iron-nickel alloy which has been gettered of interstitial elements by Ti additions, the two phase $\alpha + \gamma$ reheating step produces only slight improvements in toughness and is greatly overshadowed by the large toughness increases resulting from grain refinement. Furthermore, austenite retention does not appear to contribute to the peak toughness, because no measurable amounts were detected in any of the high toughness specimens.*

B. Improvement in Impact Toughness from Two-Step Reheating

Reheating to temperatures much above the A_f temperature (e.g., 750 and 800°C) produced progressively larger austenite grains, as was shown in curve A of Fig. 3. Correspondingly, the α to γ transformation that occurred during heating and holding at these higher temperatures took place with little or no $\alpha + \gamma$ duplex formation. In order to test the role, if any, that the $\alpha + \gamma$ duplex structure might play in the grain refining process, specimens were initially given an intermediate treatment at 650°C for several hours before heat treating at the higher 750 and 800°C temperatures. The results of such a treatment on the impact toughness and grain size are shown in curves B of Figs. 2 and 3. Reheating after the additional 650°C treatment produced finer grain sizes

* NOTE: With regard to the role of retained austenite, it should be pointed out that in all of the alloys investigated in this paper, the martensite transformation results in massive or lath martensite and a bcc structure, whereas commercial Fe-Ni alloys containing C often form plate or acicular martensite and a bct structure. Retained austenite is usually associated with the bct structure and is normally not found in massive martensite structures.

and a corresponding extension of the high toughness reheating range from 725°C to 775°C. A similar improvement of grain size could be accomplished by simply slowing the rate of heating through the $\alpha + \gamma$ phase region.

Examination of the dilatometric response for specimens initially heated in the 650°C range proves instructive. A comparison of curves A and B of Fig. 9 shows that the 650°C pretreatment has produced, when the specimen is subsequently reheated, a broadening of the $A_s - A_f$ temperature range. This lowering of the A_s and raising of the A_f temperatures suggests at least one possible reason for the more effective grain refinement of the two-step procedure; namely, that during the 650°C preheat treatment, Ni partitions between the α phase and the γ phases. When such a specimen is reheated again, regions that are higher than normal in nickel transform to γ at lower temperatures, whereas regions low in nickel remain α until higher temperatures are reached. In terms of the grain refinement produced, regions high in nickel serve as nuclei for γ transformation, whereas smaller regions low in nickel which remain α until higher temperatures are reached, act to restrict grain boundary motion, thereby restricting grain growth.

A second consequence of introducing an intermediate 650°C reheating step is to prevent, at least on a massive scale, the reverse shear of martensite back to austenite during reheating. If the transformation to austenite should take place primarily by such a process, little or no grain refinement would be expected. In effect, the original austenite grain boundaries and orientations would be reformed that were present prior to the quenching and reheating step. Conversely, such a reverse

shear process cannot take place if an intermediate step is introduced which destroys the original colony and grain boundaries as well as the former orientations. Destruction of these elements of structure can be accomplished by either diffusion (e.g., the 650°C intermediate step) or by mechanical deformation (e.g., an intermediate cold working step).

In order to test the occurrence and consequences of a reverse shear process on grain refining, thin-section specimens were prepared (~0.010 in.) and up-quenched into a salt bath at various temperatures above the A_f . Heating rates of the order of 1000°C/min, compared to the standard 50°C/min heat up rate used in all other heat treatments, were achieved by this process. The much larger grain sizes produced by the rapid up-quench (curve D of Fig. 3), are in marked contrast to the relatively finer grain sizes produced by the standard heat up rate, (curve A) and the interrupted 650°C heat up treatment, (curve B). As discussed above, such a grain size response for the rapid up-quenched reheating would be expected from a reverse shear transformation to γ .

C. Improvement in Impact Toughness from Cold Working Prior to Reheating

The $\alpha \rightarrow \gamma$ transformation which takes place during 700°C reheating, (Figs. 8a and 8b), occurred preferentially at the prior austenite and lath packet boundaries. These initially transformed areas in effect dominate the final average grain size achieved upon completion of the $\alpha \rightarrow \gamma$ transformation. To produce finer grain sizes, it follows that the uniqueness of these prior austenite boundaries as preferred sites for γ -nucleation must be altered so that a greater number of nucleation sites are possible. The one obvious method is by cold working,

and curve C of Fig. 3 shows how effective it is on the grain refining process. The improved grain refinement of this method has the effect of shifting the high impact toughness temperature region beyond the 800°C reheating treatment. In other words, it represents an 100°C increase in the high toughness range over that of simple reheating using no intermediate step. The dilatometry results (curve C of Fig. 9) shows γ forming at lower reheating temperatures than for an as-austenitized specimen. This is probably a result of the higher number of low energy nucleation sites produced by the cold working step, and the cause for the improved grain refinement.

A second factor aiding in a finer grain structure is that cold working also destroys the simple orientation relationships of the original austenite structure and the transformed martensite. This should definitely prevent transformation by the reverse shear process, as discussed earlier, making necessary a nucleation and growth diffusion controlled process to cause transformation to austenite.

The above results show that a large improvement in the impact toughness can be achieved by reheating to a temperature where fairly rapid $\alpha \rightarrow \gamma$ transformation takes place. This was found true of the 12% Ni alloy as well as the 8 and 16% Ni alloys. However, for the 8% Ni alloy this temperature is such that very rapid grain growth also occurs. In any case, in each alloy a much refined austenite grain size results and, when cooled to room temperature, transforms completely back to the α (bcc) structure. The simple one-step reheating treatment produces an optimum energy absorption peak with a 50°C-wide temperature range for the 12% Ni alloy.

When a thermal or mechanical pretreatment precedes the reheating step, this optimum reheating temperature range can be extended by nearly 100°C. The extension of the reheating temperature range resulted from the higher degree of grain refinement and/or grain boundary stabilization achieved by introducing the thermal or mechanical pretreatments. For the 8% Ni alloy, the temperature range where optimum low temperature toughness is achieved is reduced over the 12% Ni alloy, whereas the 16% Ni alloy produces a wider temperature band of high toughness.

The relationship between the refined austenite grain size and the resulting impact toughness can be illustrated by Fig. 10, which plots the impact energy vs the grain size for the several grain refining treatments described above. Such a plot shows that whatever method is used to refine the grain size, an austenite grain size less than 15 μm is needed for impact toughness to exceed 110 ft-lb at liquid N testing temperature. This 15 μm grain size can be considered a critical grain diameter for ductile behavior at liquid N temperature for the 12% Ni alloy investigated in this study. Likewise the 8% Ni and 16% Ni alloys have critical grain sizes for ductile behavior at liquid N temperature. Figure 11 shows how the austenite grain size requirements for ductile behavior vary with the Ni content of the alloy. For the 8% Ni alloy the critical grain size lies somewhere less than 5 μm , whereas the 16% Ni alloy has a critical grain size of 30 μm .

CONCLUSIONS

1. By reheating a 900°C austenitized Fe-Ni-Ti alloy to just above the A_s (austenite start) temperature, a pronounced improvement in the impact toughness was achieved.
2. The useful reheating temperature range for optimum impact toughness was extended to higher temperatures by preheating for a short period below the A_s or by decreasing the heating rate in the temperature range below the A_s .
3. Cold working prior to reheating also extended the optimum reheating temperature range.
4. Reheating, preceded by a cold working or preheating treatment, grain refined the prior austenitized grain structure and was the major microstructural cause of the improved impact toughness.
5. A critical grain size of 15 μm , or packet size of 7.0 μm , was required in the 12% Ni alloy in order to achieve the maximum impact toughness of over 110 ft-lb at liquid nitrogen testing temperature.
6. The grain size requirements for high toughness in the 8% Ni alloy were much more severe ($<5 \mu\text{m}$), whereas high toughness in the 16% Ni alloy required only a 30 μm grain size.

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FIGURE CAPTIONS

- Fig. 1. Phase transformation diagram showing austenite start and finish temperatures (A_s - A_f) on heating and martensite start and finish temperatures (M_s - M_f) on cooling at a rate of 50°C/min.
- Fig. 2. Relationship between Charpy energy absorption and reheating temperature for (a) single step reheating, (b) two-step reheating and (c) cold working plus reheating treatments.
- Fig. 3. Relationship between grain size and reheating temperature for (a) one step reheating, (b) two-step reheating, (c) cold working plus reheating and (d) rapid reheating treatments.
- Fig. 4. (a) Bright field light micrograph of 900°C-air cooled austenitized structure. (b) Same area as in (a) but viewed by Nomarski interference contrast. (c) SEM fractograph of Charpy bar broken at -196°C.
- Fig. 5. Light micrograph of 900°C austenitized plus 650°C reheated structure (a) after 2 hr, (b) after 30 hr and (c) SEM fractograph of 650°C reheated Charpy bar broken at -196°C.
- Fig. 6. Direct carbon replica of 650°C reheat structure (a) after 2 hr (b) after 30 hr.
- Fig. 7. Development of Charpy impact toughness by reheating above the A_s (i.e., 700 and 800°C) and below the A_s temperature (i.e., 650°C).
- Fig. 8. Microstructure vs fracture mode transformation during reheating at 700°C (see Fig. 7).

Fig. 9. Dilatometric responses during (a) single step reheating,
(b) two step reheating and (c) cold working plus reheating.

Fig. 10. Relationship between Charpy impact toughness and grain size
developed by one step reheating, two step heating and cold
working and reheating.

Fig. 11. Relationship between austenite grain size and nickel content
for ductile-brittle behavior at -196°C for interstitial-free
Fe-Ni alloys.

XBL 743-5888

Fig. 1

Fig. 2

Fig. 3

XBL743-5889

Fig. 4

XBB 747-4725

XBL 7311-5597

Fig. 7

$t = 0.5 \text{ hrs}$

$t = 1.0 \text{ hrs}$

$t = 2.0 \text{ hrs}$

Fig. 8

XBB 747-4726

XBL73II-5598

Fig. 9

XBL743-5890

Fig. 10

XBL 743-5887

Fig. 11

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