Properties and microstructure of stress-relieved submerged-arc weld metal containing niobium

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A review of previous investigations on the mechanical properties and microstructure of niobium micro-alloyed steel weldments indicated that there is a loss of ductility after post-weld heat treatment (PWHT). This effect has been ascribed to the precipitation of finely dispersed niobium carbonitrides. However, it can be shown that such precipitation is only observable when the niobium content in the weld metal exceeds 0.025 wt%. This limit appears to be confirmed in the current work by TEM observations on samples containing 0.03 wt% niobium, annealed for various times at 625° C.

1. Introduction

Post-weld heat-treatment (PWHT) is normally adopted for welded fabrications, when plate thickness exceeds 25 mm. The aim is to minimize residual stresses which can contribute to buckling and brittle failure. However, the removal of residual stress is accompanied by various microstructural changes during the heat treatment which may have opposing effects on mechanical properties, namely [1]:

(a) carbide spheroidization,

(b) carbide formation from enriched profiles in the ferrite, and

(c) dislocation recovery and ordering.

The combined effect of these factors upon toughness, K_{Ic}, and yield point, σ_y, of the stress-relieved material can be summarized using the vector approach of Garland and Kirkwood [2].

Fig. 1 illustrates the relative magnitude of these vectors for a typical C–Mn weld metal. It has been shown in previous studies [1] that the microstructure of the weld metal is highly dependent on the manganese content and that the as-welded structures can be divided into two major classes:

low manganese levels, proeutectoid ferrite (PF) and ferrite side plates (FSP); and

high manganese levels, acicular ferrite (AF).

The transition between the two kinds of microstructure has been identified by Tuliani [3] as being around 0.8 to 1.0 wt% Mn in C–Mn steel weld metals.

Thus, for microstructures composed primarily of AF, Mechanisms a and c will dominate and improved toughness will result after stress relief [4]. On the other hand, precipitation of carbides at grain boundaries and triple-points (Mechanism b) will dominate for microstructures typical of low manganese steel welds, and adverse effects on toughness will be produced by the introduction of easy crack paths.

The model presented by Farrar *et al.* [1] must be modified when micro-alloying elements such as niobium are present in the weld metal. Recent phase transformation work by Harrison who used continuous cooling curves [5] indicated that in low hardenability steel, niobium promotes FSP, whereas in high hardenability steel, niobium promotes AF and causes a general refinement of the structure.

Considering the Mechanisms a to c, the effect of niobium is as follows:

(a) removal or reduction of this effect;

(b) removal or reduction of this effect due to lower transformation temperature, but if many

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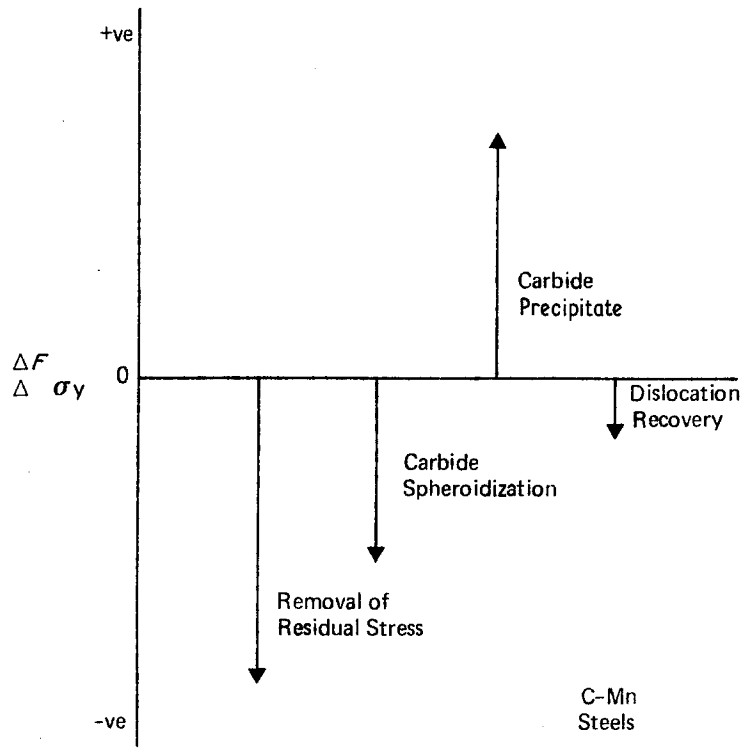

Figure 1 Vector diagram of factors affecting toughness in C–Mn weld metal.

FSP are present (low hardenability weld metal) the reverse can be true:

(c) still applicable.

However, the presence of niobium can introduce an additional factor, namely, the precipitation of niobium carbonitrides, Nb(CN).

From the above considerations the vector approach for C–Mn weld metal may be modified to account for the influence of niobium, as shown in Fig. 2. Once again it must be emphasized that the relative values of the vectors will depend on the precise microstructure of the as-welded sample.

2. Mechanical properties of stress-relieved weld metals containing Nb

There is still considerable disagreement in the literature concerning the precise effect of niobium on stress-relieved mechanical properties. Garland and Kirkwood [2] concluded that in thick sections, welds containing up to 0.025 wt% Nb were improved, whereas Watson [6] found that there was a general deterioration in properties when the weld metal contained between 0.02 and 0.03 wt% Nb.

Kirkwood [7] has recently attempted to rationalize these and many other results by pointing out the large variations in weld metal compositions, consumables and welding conditions employed by the various investigators. If we assume that the major detrimental effect is due to the precipitation of coherent to semi-coherent Nb(CN) [8], then the changes in observed properties will depend on the precise levels of niobium, carbon and nitrogen in the weld metal. There will also be a kinetic effect due to the cooling rates which apply in the precipitation temperature range. Recent work by Weatherly [9] who studied heat-affected zone properties has indicated the critical nature of the cooling rate between 800 to 500° C in controlling the precipitation of Nb(CN).

The propensity to form precipitates may be examined by considering the solubility product [10]

$$\log\,[\mathrm{Nb}]\,[\mathrm{C} + 0.86\mathrm{N}] = -\frac{6770}{T} + 2.26, \quad (1)$$

where T is the absolute temperature, which for a typical range of weld metals yields the solubility temperatures given in Table I.

Thus, at cooling rates typical for submerged arc welding (5 to $10^\circ\,\mathrm{C\,sec^{-1}}$) there is a very high possibility of producing some 50% supersaturation [11, 12] which can subsequently precipitate as Nb(CN) during PWHT. The data available in the literature on the changes in yield strength, $\Delta\sigma_y$, and fracture appearance transition temperature (FATT), ΔF, after stress relieving is summarized

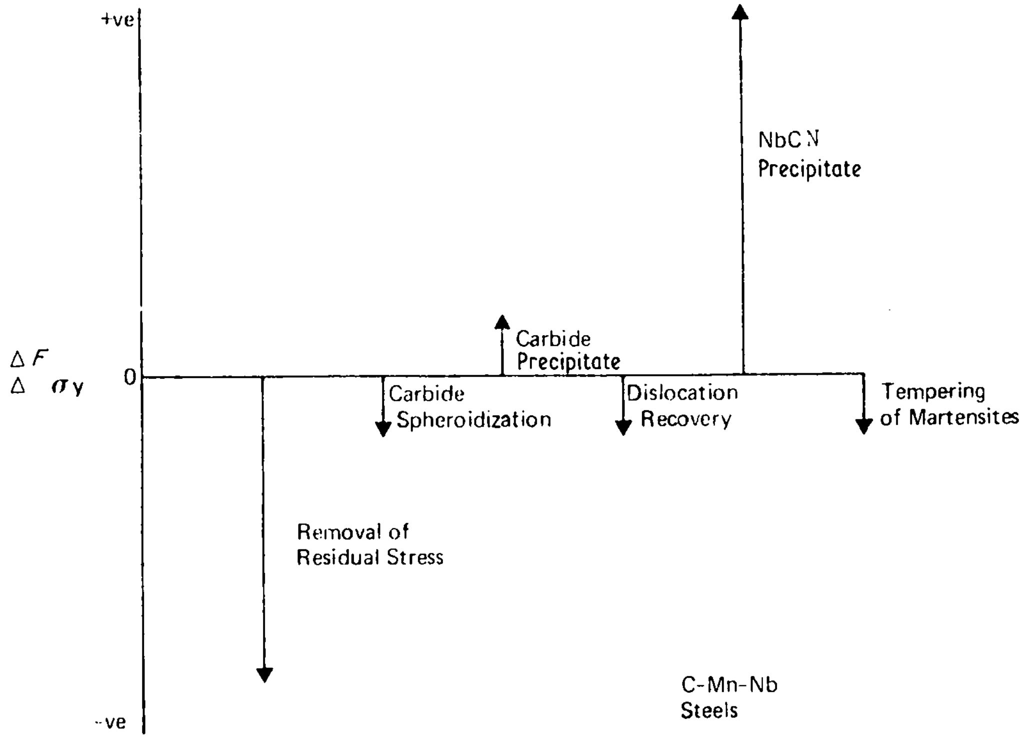

Figure 2 Vector diagram of factors affecting toughness in C–Mn–Nb weld metal.

in Fig. 3a and b for weld metals containing about 0.1 wt% C and 1.0 to 1.5 wt% Mn. The results plotted relate to welds which have been produced with plates thicker than 20 mm and using basic welding consumables.

The overall trends in the data appear to indicate that niobium is beneficial up to 0.025 wt%, but at levels above 0.03 wt%, the rise of σ_y causes a drop in K_{Ic}. Using the vector approach it seems reasonable to conclude that

(i) stress relief activates Mechanisms a to c,

(ii) stress relief causes precipitation of Nb(CN) to an extent which is dependent on the original niobium content, and

(iii) these mechanisms are competitive and precipitation gradually offsets the beneficial effects of Mechanisms a and c.

In fact, Fig. 3a clearly indicates a gradual increase of $\Delta\sigma_y$ with niobium content. If a precipitation mechanism is assumed, its extent will ultimately dictate the final properties of the stress-relieved weld metal.

TABLE I Weld metals and their solubility temperatures, determined from Equation 1

Composition (wt%)			Solubility temperature (° C)
C	Nb	N	
0.1	0.02	0.01	1080
0.1	0.05	0.01	1223
0.1	0.10	0.01	1330

3. Microstructures of stress-relieved weld metal

There is still limited evidence at the electron optical level of the presence of Nb(CN) precipitation in weld metals. Garland and Kirkwood [2] for instance, as well as Watson [6], were unable to confirm their existence by TEM, in spite of indirect evidence [12] which indicates a definite increase in σ_y after PWHT.

Table II summarizes the observations of Nb(CN) taken from the available literature. Considering these results three comments may be made:

(i) Successful imaging of Nb(CN) precipitates is always associated with high niobium content.

(ii) Annealing was carried out at temperatures above the standard stress-relief temperature of 625° C.

(iii) Cooling rates of the order of $1° C\,sec^{-1}$ are slower than the typical submerged-arc (SA) cooling rates and, hence, favour the kinetics of precipitation.

The fact that high annealing temperatures and

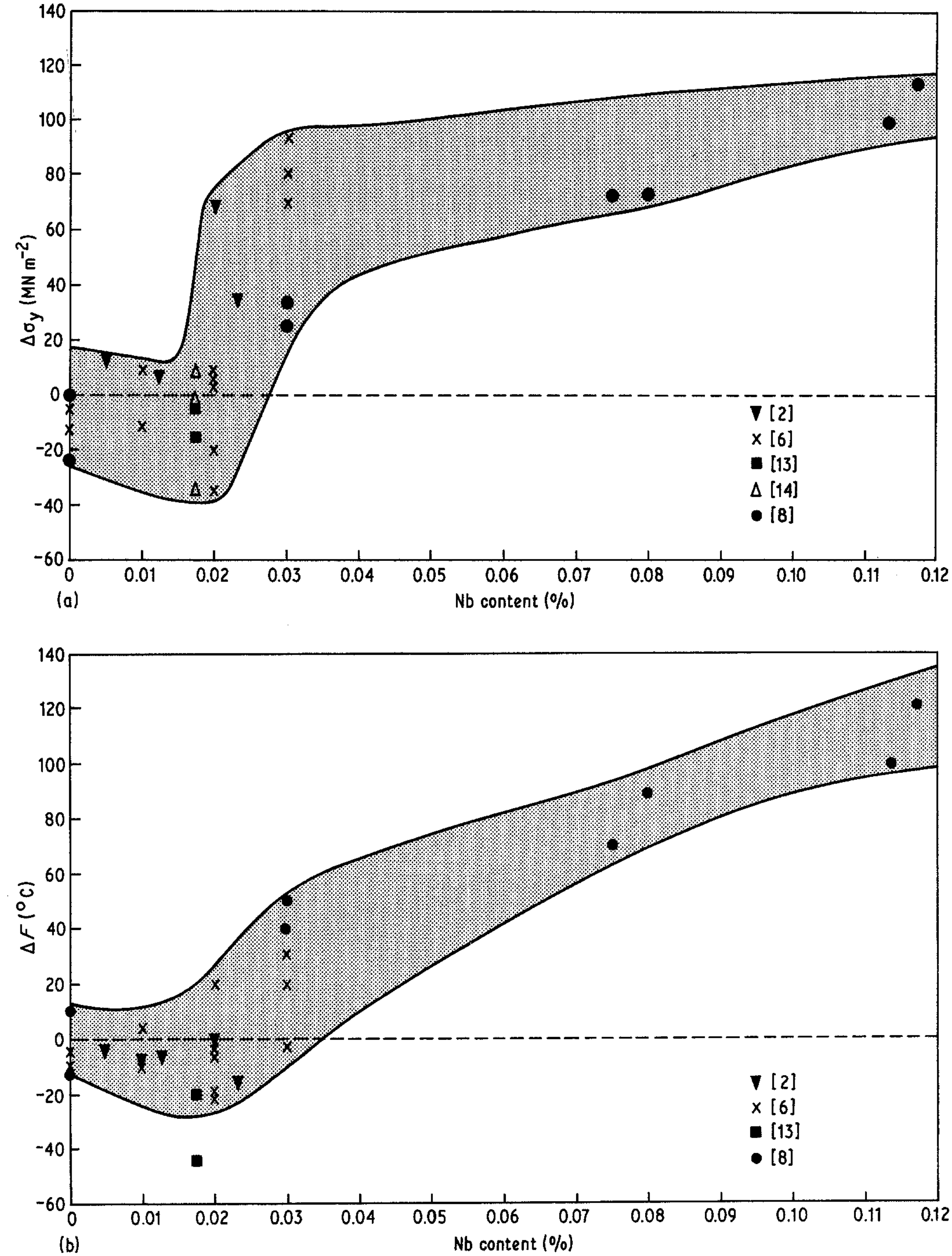

Figure 3 (a) Yield stress and (b) fracture appearance transition temperature dependence on niobium content in weld metal. Results are for typical HSLA steel plates thicker than 20 mm welded using basic fluxes and the submerged-arc technique.

very slow cooling rates are required to produce observable Nb(CN) precipitates suggests that it occurs by pre-precipitation on a fine scale and is followed by growth by the Ostwald ripening mechanism [9].

In the case of Bosansky [8], there was no observable precipitation of Nb(CN) for niobium content up to 0.075 wt%, suggesting that growth did not occur at the cooling rates employed in his studies. However, Bernard *et al.* [12] detected a

TABLE II Summary of the observations of Nb(CN)

Nb content (%)	PWHT		Cooling rate ($^\circ C\ sec^{-1}$)	Nb(CN) observed	Reference
	T (° C)	time (h)			
0.002	640	1	~ 1	No	[8]
0.032	640	1	1	No	
0.075	640	1	1	No	
0.113	640	1	1	Yes	
0.025	No PWHT		(Heat input = 3 kJ mm^{-1})	No	[12]
0.025	No PWHT		(Heat input = 7 kJ mm^{-1})	Very little	
0.027	550/650	1	1	Yes	[15]
0.04	900	0.5	Not known	Yes	[16]
0.02	625	1–12	~ 14	Little in PF regions	[17]

small amount of precipitation in the as-welded material, but it is important to notice that they used a heat input of 7 kJ mm^{-1} which would lead to very slow cooling rates.

Besides Nb(CN) precipitation, other microstructural modifications take place during PWHT. Carbide spheroidization has been observed by many investigators, as well as dislocation recovery [1, 2, 6, 15]. However, Hannertz [16] points out dislocation locking by Nb(CN) particles can lead to less dislocation recovery.

The overall result of the microstructural changes caused by PWHT seems to be a function of niobium content, as demonstrated by Fig. 3a and b. On the other hand, the results presented in Table II lead to the conclusion that investigations which aim to demonstrate the likelihood of Nb(CN) precipitation have employed relatively non-standard PWHT conditions. The purpose of this present paper is therefore to investigate the mechanism of precipitation of Nb(CN) in C–Mn–Nb weld metals and to determine the thermal conditions required for this precipitation.

4. Experimental details

4.1. Materials and methods

The full details of the welding conditions and consumables have been reported elsewhere [6] but may be summarized as

Base plate: HSLA 0.08 wt% niobium-bearing steel, 25 mm thickness;

Flux: Oerlikon OP41TT;

Wire: SD3 wire (0.07 wt% C, 1.80 wt% Mn, 0.47 wt% Si); and

Weld: SA, two-pass, laid down in 60° "V"-shaped grooves, no preheat and at about 100° C interpass temperature (conditions were 550 A, 28 V and a speed of 254 mm min^{-1} resulting in a nominal heat input of 3.6 kJ mm^{-1} and the cooling rate was about 15° C sec^{-1}).

After extracting suitable samples from the top run, simulated stress relieving was carried out at 625° C for between 1 h and 10 h. The samples were furnace-cooled to 400° C at 0.1° C sec^{-1} and then air-cooled to room temperature.

4.2. Metallography

Thin foils for electron microscopy were prepared from 3 mm discs by jet-polishing using a 10% perchloric acid–ethanol solution at −5° C. Observation was carried out on both as-welded and stress-relieved samples using a AEI 100 kV transmission electron microscope.

For optical microscopy, samples from the top run were prepared by standard techniques and etched in 2% nital.

5. Results and discussion

The chemical analysis of the material used was 0.076 wt% C, 1.27 wt% Mn, 0.28 wt% Si, 0.016 wt% Al and 0.031 wt% Nb. The microstructure of the as-welded sample is shown in Fig. 4. It consists primarily of acicular ferrite with small amounts of proeutectoid polygonal ferrite and side-plates at the prior austenite grain boundaries. At the optical level, no significant differences were observed between the as-welded and stress-relieved samples.

Fig. 5 shows an area containing both acicular and proeutectoid ferrite in an as-welded sample. Pearlitic carbides are present at grain boundaries but no Nb(CN) precipitation was detected.

Fig. 6 illustrates a sample which has been aged for 1 h. It shows one of the very few areas in which clear Nb(CN) precipitation could be detected.

Electron diffraction revealed the presence of faint spots of a cubic phase with a lattice parameter

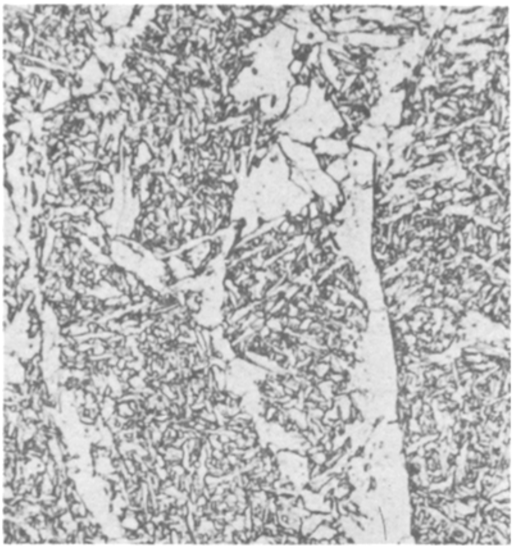

Figure 4 Microstructure of the weld metal 0.076 wt% C–1.27 wt% Mn–0.03 wt% Nb used in this investigation, 2% nital etch (× 500).

of 0.446 nm which was identified as Nb(CN), see Fig. 7. In general, the precipitation seemed to be associated with dislocations and was very rare. However, in samples annealed for up to 10 h, the precipitation became a more common occurrence, although not a general one. Figs 8 and 9a are a bright-field and corresponding diffraction pattern of a specimen stress-relieved for 10 h. The Nb(CN) spots are more intense; the extra spots have been identified as Fe_3O_4, which grows epitaxially on the specimen surface. This oxide forms with the

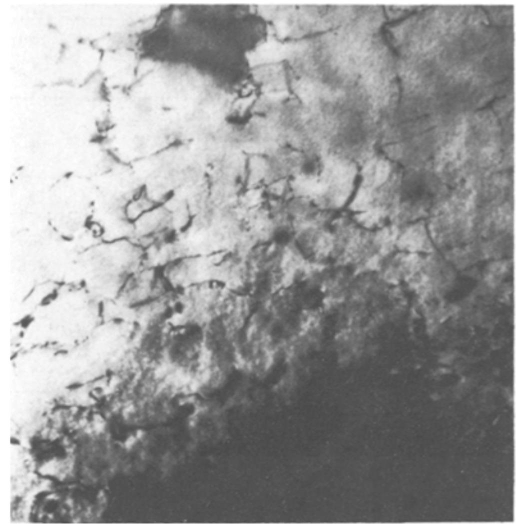

Figure 6 Precipitation of Nb(CN) in proeutectoid ferrite. Sample heat-treated at 625° C for 1 h (× 50 000).

Nishiyawa–Wasserman orientation relationship with the matrix (see Fig. 9b).

The above results confirm the occurrence of Nb(CN) precipitation for samples containing 0.03 wt% niobium. The degree of precipitation observed in Fig. 8 was not commonly observed and, in fact, it has been pointed out by many authors that precipitation in continuously cooled steels is very slow below 700° C [18, 19]. The transformation temperature of the weld metal used in this investigation has been measured as being around 630° C [5] and, as a consequence, rather slow precipitation kinetics are anticipated.

Figure 5 Typical microstructure of as-welded metal (× 16 000).

Figure 7 Diffraction pattern from sample heat-treated at 625° C for 1 h.

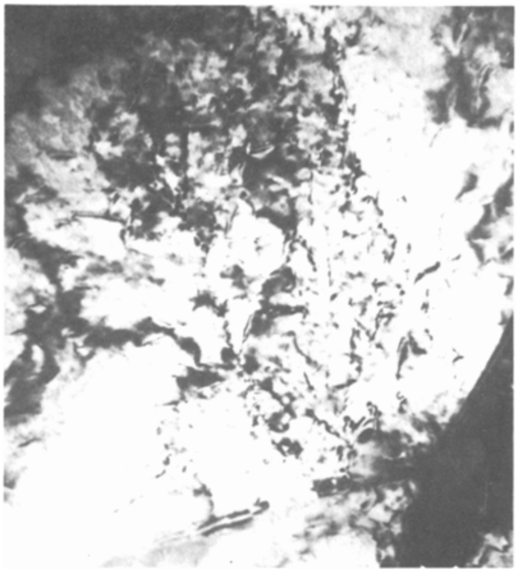

Figure 8 Precipitation of Nb(CN) in sample heat-treated at 625° C for 10 h (×50 000).

Another factor which may be responsible for the scarcity of Nb(CN) particles is the inhomogeneous distribution of niobium when in solid solution. In the present studies the precipitation seems to be confined within the proeutectoid ferrite and no obvious particles were detected within the acicular ferrite or original austenite boundaries.

This preference for proeutectoid ferrite can be explained by kinetics considerations, that phase being the first to form when austenite transforms, and is supported by recent microhardness studies by Farrar *et al.* [17]. These results show that in a weld metal containing 0.02 wt% niobium, hardness variations due to PWHT occur only in the FSP and PF areas, AF being unaffected. The overall effect of stress relief is a decrease on hardness, meaning that precipitation, if any, is easily off-set by mechanisms such as carbide spheroidization (see Fig. 10) and dislocation recovery.

In this context it is useful to remember that

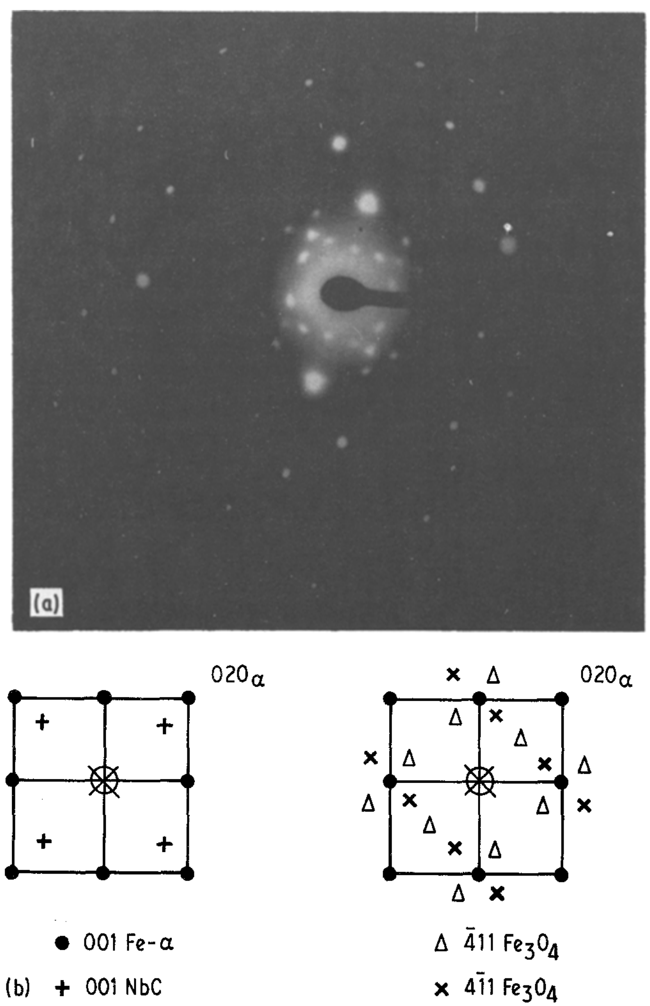

Figure 9 (a) Diffraction pattern from sample heat-treated at 625° C for 10 h. (b) Nb(CN) and Fe_3O_4 diffraction spots are present.

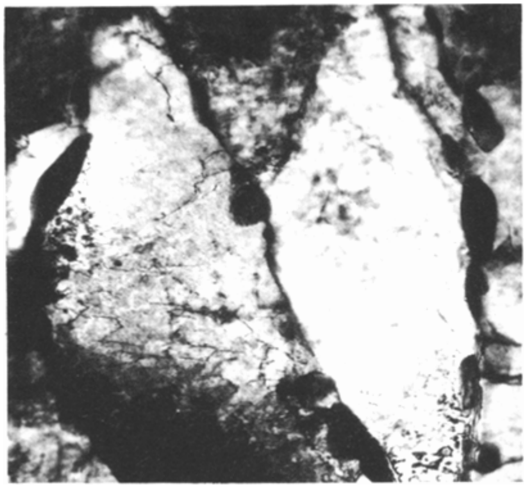

Figure 10 Occurrence of carbide precipitation along grain boundaries. Sample annealed 10 h at 625° C (× 20 000).

niobium decreases the amount of proeutectoid ferrite and ferrite side plates whilst favouring acicular ferrite in high hardenability weld metals [5]. This could well result in a decrease of the reaction rate of Nb(CN) precipitation.

6. Conclusions

(a) The main effects of PWHT of niobium-bearing steel weldments are Nb(CN) precipitation and dislocation recovery and ordering. Carbide spheroidization and precipitation has also been observed.

(b) PWHT is definitely beneficial up to a 0.025 wt% niobium content in the weld metal. Above this level, the increase of σ_y causes a gradual drop of K_{Ic}.

(c) Most of the literature concerning the effects of PWHT seems to overemphasize Nb(CN) precipitation. This is due to the extreme conditions of supersaturation and heat cycles employed, far higher and more severe than those expected in commercial practice.

(d) Samples containing 0.03 wt% niobium, stress-relieved for 1 and 10 h exhibit negligible precipitation which seems to be confined within the proeutectoid ferrite phase. No precipitation was detected in the as-welded samples.

(e) Spheroidization and precipitation of carbides at ferrite grain boundaries have been detected.

Acknowledgements

The financial assistance of the Science Research Council is gratefully acknowledged in funding the welding programme. The authors wish to thank Dr M. N. Watson for the use of unpublished data. One of the authors (MF) wishes to thank the Royal Society, London for a scholarship whilst at the Department of Mechanical Engineering, University of Southampton. Also, CNPq and Universidade Federal de São Carlos, Brazil are acknowledged.

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Received 15 December 1981
and accepted 18 January 1982