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ANNEALING OF COLD WORKED HYPOSTOICHIOMETRIC Ni$_3$Al ALLOYS USING POSITRON LIFETIME SPECTROSCOPY

S. G. USMAR AND K. G. LYNN
Brookhaven National Laboratory, Department of Metallurgy and Materials Science, Upton, NY 11973

ABSTRACT

Hypostoichiometric Ni$_3$Al alloys of composition 76.2 Ni:23.8 Al containing impurity levels of boron and hafnium (supplied by Oak Ridge National Laboratory) were either cold rolled or pressed. Rolled and pressed samples were deformed by 20% and 10% thickness reductions, respectively. Samples were annealed isochronally at approximately 50°C intervals up to 1050°C. Two major annealing stages were apparent in all three alloys studied. These could be attributed to vacancy migration to sinks and annealing of dislocations and(or) recrystallization. The onset of vacancy migration occurred at approximately 200°C in all three alloys. Annealing of dislocations started at 650°C to 700°C and was complete at 1000°C for alloys which contained boron and or hafnium impurities. In the pure alloy the onset of dislocation annealing occurred at 800°C and was incomplete at the highest (1050°C) annealing temperatures reached.

INTRODUCTION

Recently the intermetallic alloy on Ni$_3$Al has come into prominence as a promising high temperature material. The pure alloy has a composition range extending approximately 2 at.% on each side of stoichiometry and undergoes intergranular brittle failure over this whole range. Impurity levels (100 to 500 wt ppm) of boron have been found to improve the ductility [1] of alloys in the hypostoichiometric (25 at.% >Al>23 at.%) composition range. Further hafnium has also been found to improve the high temperature mechanical behavior of these allcys [2].

With the above in mind, an investigation of the defect structures of Ni$_3$Al alloys manufactured by Oak Ridge National Laboratory (ORNL) from pure commercial grade raw materials was initiated. Positron annihilation spectroscopy was chosen as the investigative technique because of its ability to detect and differentiate submicroscopic defects [3].

EXPERIMENTAL

All alloys studied were prepared by drop casting arc melted raw materials into cold copper molds. Each casting then underwent a lengthy heat treatment (under a vacuum of 10^{-6} Torr) which both homogenized and annealed it. Alloys of composition:

(i) 76.2 at.% Ni:23.8 at.% Al (A)
(ii) 76.1 at.% Ni:23.9 at.% Al+0.24 % B (B)
(iii) 76.3 at.% Ni:23.2 at.% Al:0.5 at.% Hf+0.2% B (C)

where mechanically deformed. Alloy A was pressed with a thickness reduction of 10% whilst alloys B and C were cold rolled with a 25% thickness reduction. Samples suitable for positron lifetime spectroscopy were prepared from the deformed material.

Positron lifetime spectra were accumulated using a spectrometer employing "Fast-Fast" coincidence counting [4] with a timing resolution of approximately 165 ps FWHM. Isochronal annealing was accomplished by first sealing samples in an evacuated fused silica tube then heating the assembly in a furnace for 1/2 h.

To ensure cleanliness in the annealing the samples were sealed in the fused silica using a two stage process. The tube was first evacuated to 20 mT and back filled with argon gas three times; evacuated to 20 mT and sealed. Several strips of Ta foil were fired at 1000°C for >2.5 min and the samples sealed once more. Further samples were wrapped in Ta foil prior to being sealed in fused silica. At least one positron lifetime spectrum, containing $1.5x10^6$ counts, was accumulated, at room temperature, after each annealing stage.

RESULTS AND DISCUSSION

Positron lifetime spectra were analyzed, numerically, using an interactive form [5] of the computer program POSITRONFIT [6]. Initial analysis resulted in a "mean lifetime" τ_m temperature dependence shown in Fig. 1 for alloys B and C. Here it is evident that two annealing stages occur in both alloys. The low temperature stage, whose onset occurs at approximately 200°C, can be attributed to migration of vacancies to sinks in agreement with the observation of Wang et al. [7]. The high temperature stage is due to migration of dislocations and or recrystallization. An intermediate stage is apparent in C but not in B.

The mean lifetime data indicate that at least two lifetime components were present in spectra up to approximately 950°C. These components were associated with annihilation of positrons from the bulk and from vacancies or dislocations. In the temperature range 600°C to 700°C for B and 700°C to 800°C for C and A the mean lifetime (and therefore the annihilation parameters) remain constant. Here two lifetime components were present thus the pertinent spectra were fitted using two components. The

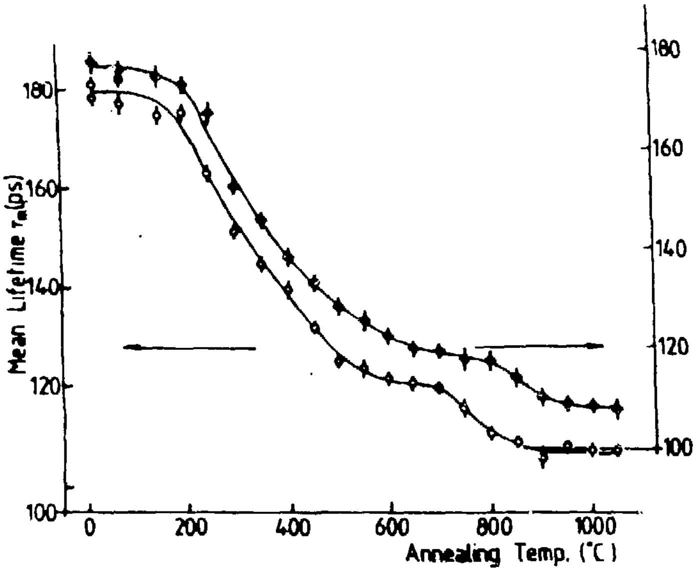

Figure 1. Mean lifetime vs annealing temperature for for cold rolled alloys; —o— :alloy B, —◆— : alloy C.

longest lived of these components had a magnitude of 135±5 ps suggesting it be associated with dislocaitons [8,9] in all samples. Now all other lifetime spectra were analyzed using two components, one of which was fixed at 135 ps. The results of these analyses are shown in Fig. 2(a) and (b). In these figures τ_1 (only present above 500°C) is the bulk lifetime; τ_2 the lifetime associated with positrons trapped at dislocations and τ_3 the lifetime associated with those trapped at vacancies. The switch, in the 2 component fit, from τ_3 to τ_1 occurred spontaneously (i.e., the initial guesses used when fitting the spectra in which τ_1 first appeared were values close to τ_3 180 ps). Two component fits, where necessary, were found to be better than single component fits and usually results in variances <1.2. No long-lived component which would suggest void formation was observed.

The detailed numerical analyses reveal that below approximately 450°C all positrons annihilated from either vacancies or dislocations, i.e., competative trapping occurred. Further, the initial ratio of vacancies to dislocations was the same in cold rolled alloys (c.f. Fig. 2(a)). At approximately 200°C vacancies become mobile and migrate to sinks (I_3 decreases) thus the fraction of positrons trapped at dislocations (I_2) increases. At 450°C I_3 has fallen to zero indicating vacancy annealing to be complete. A combination of dislocation density and trap strength conspire to allow some positrons to annihilate in the bulk.

A further increase of temperature resulted in annealing of dislocations. In alloys B and C dislocations become mobile in the temperature range 750°C to 800°C and annealing is complete at 1000°C. In this temperature range I_2 decreases and both I_1 and τ_1 increase in qualitative agreement with the two state trapping model [3].

A close inspection of the results for alloys B and C (c.f. Fig. 2(a)) suggest that the hafnium present in C may interact with both vacancies and dislocations. The solid and dashed lines in Fig. 2(a) emphasize the temperature range in which these interactions are evident. Obviously the effects are small but suggest, tentatively, that Hf stabilizes vacancies and pins dislocation. More work is planned to elucidate this point.

Annealing of A, the pure pressed alloy (c.f. Fig. 2(b)) indicates the defect structures in this sample to differ from those of cold rolled alloys. Pressing results in an initial vacancy: dislocation ratio much smaller than that observed in cold rolled sample. The vacancy annealing stage is, at least qualitatively, the same as that for rolled samples but vacancies persist to higher temperatures. Further dislocations are present up to the highest annealing temperature reached. This incomplete annealing is difficult to explain possibly the defects involved are not dislocations. In this respect a TEM study has been initiated. If the defects are dislocations then the presence of boron impurities in $Ni_{76.2}:Al_{23.8}$ would seem to enhance the mobility of dislocations, a effect not inconsistent with the known mechanical properties of these alloys. Evidently, more work is required to elucidate this behavior.

CONCLUSION

The cold rolled Ni_3Al alloys B and C studied here contained vacancies and dislocations which annealed in two distinct stages at 200°C and 750°C, respectively. Further as was found for carbon in iron [10], Hf may stabilize vacancies. Alloy A also contained, after pressing, both vacancies and dislocaions. Here, however, there may be other unknown defects, which persist at high temperatures, present. More work is planned to elucidate these possibilities.

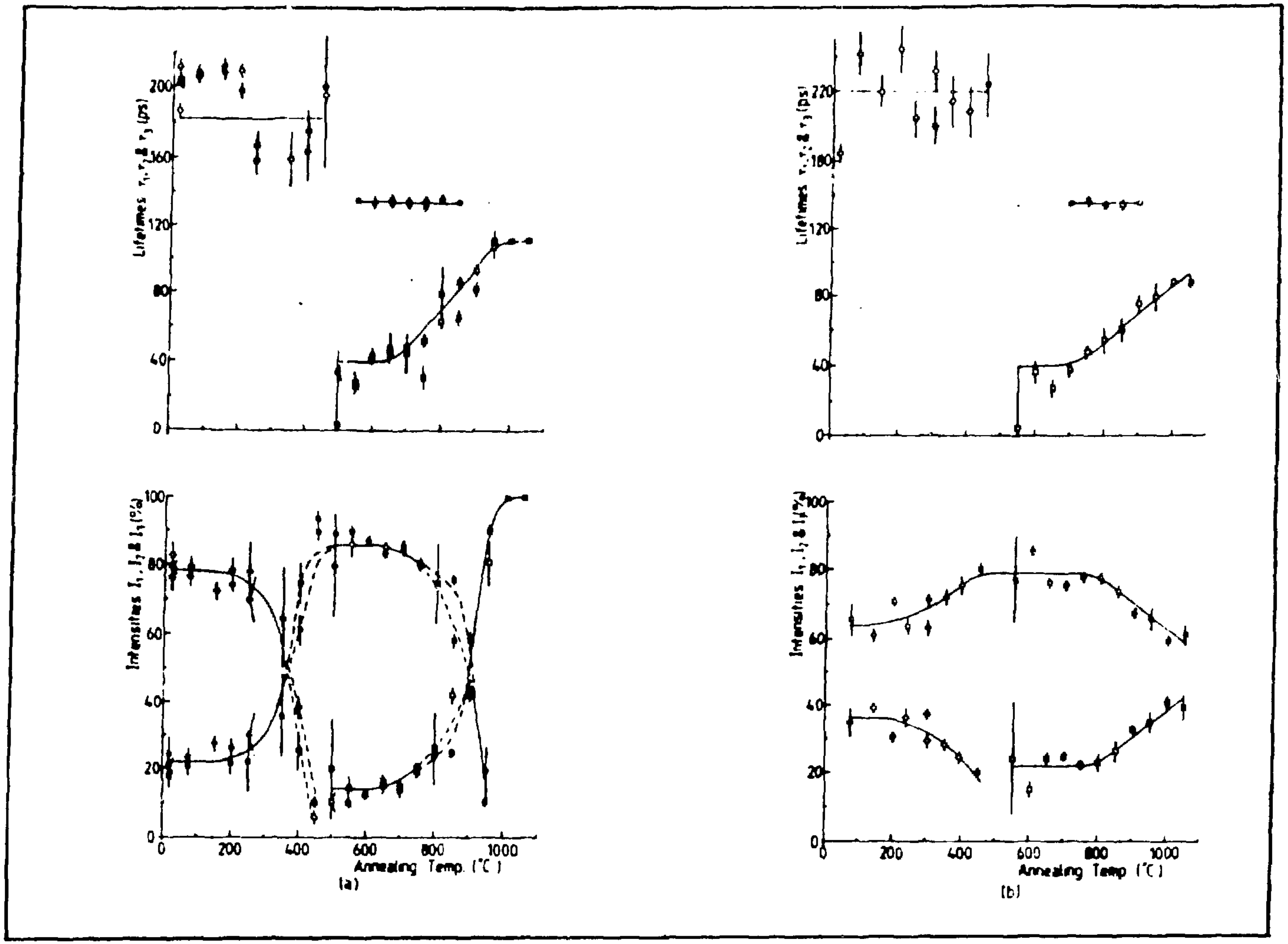

Figure 2. Positron annihilation parameters —□—: I_1, τ_1, —o—: $I_2 \tau_2$ and —◇—: I_3, τ_3 vs annealing temperature for, (a) alloys B(—□—) and C(—■—) and (b) alloy A.

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