Solid Solution Hardening of Ni_3Al with Ternary Additions

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An attempt is made to interpret the solid solution hardening of Ni_3Al alloys in terms of the elastic interaction between the strain fields of a dislocation and a solute. Ternary additions are made to the Ll_2 intermetallic compound with both B-subgroup and transition metal elements which substitute for Al-sites. Young's modulus of the ternary alloys is measured and the rate of change in elastic constant per one atomic per cent of solute (dE/dc) is determined. With the available data for the rate of solution hardening ($d\sigma/dc$) and the rate of change in lattice constant (da/dc), analyses are made on the relation between $d\sigma/dc$ and combined parameters. It is found that the solution hardening is best described by a combined parameter appropriate for the elastic interaction involving edge dislocations, where atomic size misfit plays a major role and the contribution of modulus misfit is minimal. However, the correlation between $d\sigma/dc$ and the combined parameter splits into two curves, one being for transition metal elements and the other for B-subgroup elements. It is found that for the additions of transition metal elements there is an extra hardening over what is expected from the treatment on B-subgroup elements.

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I. Introduction

The effect of ternary additions on the mechanical properties of the compound Ni_3Al has aroused a great deal of interest, not only because it exhibits the positive temperature dependence of strength as one of the Ll_2 type compounds but also because it has been utilized as precipitation strengthener in commercial Ni-base superalloys. The present authors have carried out a systematic investigation on the mechanical properties of the compound as

affected by ternary additions of B-subgroup elements and transition metal elements⁽¹⁾⁽²⁾. Although the emphasis in those works has been put on the effect of ternary elements on the positive temperature dependence of strength in the compound, it has been shown that the degree of solid solution hardening can be simply expressed by a linear relation with atomic size misfit for the additions of B-subgroup elements⁽¹⁾. Also it was pointed out that a unique relation does not hold for the whole sets of both B-subgroup and transition metal elements⁽²⁾. It is then felt that more detailed investigation is necessary for a comprehensive understanding on the solid solution hardening of the compound.

In terminal solid solutions, there have been a number of efforts on the interpretation of solid solution hardening of various solvents. Among them is an interpretation pioneered by Fleischer⁽³⁾⁻⁽⁵⁾ in which the solid solution hardening is governed by the elastic interaction between solute atoms and dislocations. The interaction is described as a combined parameter of atomic size misfit with modulus misfit⁽⁵⁾, which successfully accounted for the degree of solid solution hardening in binary copper, silver and gold alloys.

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It was first pointed out by Guard and Westbrook⁽⁶⁾ whether or not the solution hardening in one of intermediate phases, Ni₃Al, was able to be interpreted by the atomic size misfit based on the hardness measurements. Later, Rawlings and Staton-Bevan⁽⁷⁾ pointed out that for the additions of ternary elements the atomic size misfit governs the degree of solid solution hardening measured as flow stress increase. Aoki and Izumi⁽⁸⁾ applied the Fleischer-type concept in interpreting the solution hardening of Ni₃Al, although a limited number of solute elements were involved.

An attempt of describing the solid solution hardening of ternary Ni₃Al in terms of the "Fleischer-type" elastic interactions is made in the present work for a wide variety of both B-subgroup and transition metal elements, all of which substitute for Al-site in the compound. The rate of solution hardening per one atomic per cent of solute, $d\sigma/dc$, has already been measured as the 0.2% flow stress at 77 K by Ochiai *et al.*⁽¹⁾ and Mishima *et al.*⁽²⁾. In the present attempt, therefore, evaluations on the atomic size misfit and of the modulus misfit are inevitable. There has been a systematic investigation on the lattice parameter change in ternary compounds, in which the rate of lattice parameter change per one atomic per cent of solute (da/dc) was determined by Mishima *et al.*⁽⁹⁾. It is then necessary to evaluate the rate of change in elastic constant per one atomic per cent of solute, dE/dc .

II. Experimental

Alloying elements to form ternary Ni₃Al solid solutions in the present investigation are shown circled in Table 1 as a part of the periodic table. The additional elements in the table are those which will be included in the following work on the solution hardening of nickel. They are all known to substitute for Al-sites in the solid solution⁽¹⁰⁾. The purity of electrolytic nickel was 99.95% and that of other elements was the highest readily available. The additions of ternary elements were made within their maximum solid solubilities as determined by Ochiai *et al.*⁽¹⁰⁾. The base com-

Table 1 Ternary alloying elements added to Ni₃Al are shown circled as a part of the periodic table and are known to substitute for Al-sites.

4A	5A	6A	7A	8A			1B	2B	3B	4B	5B
									Al	Si	—
Ti	V	Cr	Mn	Fe	Co	Ni	Cu	—	Ga	Ge	—
Zr	Nb	Mo	—	Ru	Rh	—	—	—	In	Sn	Sb
Hf	Ta	W	—	—	—	—	—	—	—	—	—

position of Ni₃Al is slightly offstoichiometric, being 75.1–75.2 at%Ni, and the ternary additions were made to keep the Ni content constant.

All the alloys were prepared by arc-melting under an argon atmosphere. The weight loss after the melting was less than 0.1 mass% in all cases and, therefore, the nominal compositions were accepted. The ingot, approximately 30 g in weight, was vacuum encapsulated in a quartz tube and was homogenized at 1273 K for 432 ks followed by water quench. For the alloys with transition metal elements, homogenization was first at 1423 K for 432 ks and then at 1273 K for 3.6 ks. The grain size obtained through the above processes ranges from 300 to 400 μm .

Young's modulus of ternary Ni₃Al was determined at room temperature through the measurement of speed of sound in the specimen using Krautkramer USL32 ultrasonic transmitter with a 5 MHz quartz oscillator and VA2 receiver. Specimens of parallelepiped with dimensions of 10×10×30 mm³ were prepared by careful machining, grinding and lapping. Young's modulus, E , was obtained by the measured longitudinal wave velocity, v_l , according to the equation,

$$E = ((1 + \nu)(1 - 2\nu)/(1 - \nu))\rho v_l^2,$$

where ν is Poisson's ratio taken to be 0.35 and density, ρ , was calculated from the lattice parameter data previously reported⁽⁹⁾. Validity of the method was carefully checked through the measurements of pure metals of known Young's modulus such as Ni, W, Cu and Al.

III. Results and Discussion

1. Young's modulus of ternary Ni_3Al alloys

Young's modulus of ternary Ni_3Al solid solutions are shown in Fig. 1 for the additions of B-subgroup elements and in Fig. 2 for those of transition metal elements. Ono and Stern⁽¹¹⁾ reported the value of E for single crystalline Ni_3Al binary compound at ambient tempera-

Fig. 1 Changes in Young's modulus of Ni_3Al with ternary additions of B-subgroup elements.

Fig. 2 Changes in Young's modulus of Ni_3Al with ternary additions of transition metal elements.

ture and calculated that for polycrystals in forms of Voigt's and Reuss's averages being 214 and 162 GPa, respectively. Several methods of estimating Young's modulus for polycrystalline material using the data for single crystal has been proposed⁽¹²⁾⁽¹³⁾, and in any case the value falls in between those averages. Actually Davies and Stoloff⁽¹⁴⁾ have reported Young's modulus of polycrystalline Ni_3Al to be 179 GPa which is in good agreement with the present result. It can be seen that Young's modulus of the compound changes linearly with increasing solute concentration for all the ternary additions. The additions of B-subgroup elements are always found to decrease Young's modulus but those of transition metal elements except Hf to increase it. To be noted is that the result for Zr addition is not included in the figure because no clear correlation is found within the limited solubility of the element.

In Fig. 3, the rate of change in Young's modulus per one atomic per cent of solute, dE/dc , is then plotted against square of the valence difference between the solute and aluminum, $(\Delta Z)^2$, for the additions of B-subgroup elements. As can be seen in this figure, a linear relation holds for each group of the elements. On the discussion of the functional dependence of the change in various properties of noble metals due to alloying with B-subgroup elements, Köster⁽¹⁵⁾⁽¹⁶⁾ has pointed out that the properties such as lattice constant,

Fig. 3 Relation between dE/dc of Ni_3Al for ternary additions of B-subgroup elements and square of valence difference $(\Delta Z)^2$ between the solute and aluminum.

paramagnetic susceptibility and activation energy for diffusion are proportional to ΔZ while modulus of elasticity to $(\Delta Z)^2$. As for the changing rate of lattice constant, it has also been shown by Mishima *et al.*⁽⁹⁾ that the same relation holds not only in noble metals but also in transition metals such as Ni, Pd and Pt. Moreover, they have shown that it is also the case for ternary additions in intermetallic compounds, Ni₃Al and Ni₃Ga⁽⁹⁾. It will be shown in the forthcoming paper⁽¹⁷⁾ that the modulus change in nickel with binary alloying additions is also proportional to $(\Delta Z)^2$.

2. Interpretation of solid solution hardening in terms of elastic interaction theory

The solid solution hardening in binary alloys has been interpreted in terms of the elastic interaction arising from atomic size and modulus misfits between two species, the short range electrical interaction between solute ions and static electrical field around dislocations, and the chemical interaction between solute atom and the extended dislocation in fcc alloys. On the discussion of the solution hardening of copper with additions of B-subgroup elements, Cottrell⁽¹⁸⁾ has concluded that only a small fraction of the hardening can be attributed to the electrical interaction. Fleischer⁽⁵⁾ has also shown that the contribution of electrical interaction is only 1/6 to 1/3 of that of elastic interaction in binary copper alloys. Then Oren *et al.*⁽¹⁹⁾ have shown a calculation to find the contribution of chemical interaction being only about 1/5 of that of elastic interaction in copper alloys containing Si, Ge or Sn. All these are to suggest that the elastic interaction plays a major role in determining the degree of solution hardening, for which much of the effort on both theoretical and experimental analyses has been paid.

In the concept of elastic interaction to explain the solution hardening, the flow stress increase in a binary alloy, $\Delta\tau$, is generally expressed in a form,

$$\Delta\tau = Af_0^p c^q = A' \varepsilon^p c^q, \quad (1)$$

where A , A' , p and q are constants, c is solute

concentration, f_0 is the maximum interaction force between a solute and a dislocation. ε in the equation is termed "combined parameter" involving atomic size misfit and modulus misfit. The atomic size misfit, ε_a , is defined as $(1/a)(da/dc)$, where a is the lattice parameter of pure solvent metal. The modulus misfit, ε'_G , has been given in more complex forms but in general is a function of ε_G and often also of ε_a , where ε_G is defined as $(1/G)(dG/dc)$ and G is shear modulus of pure solvent metal. There have been reported a number of expressions for the combined parameter, ε , as to how these misfit parameters are combined. Such effort, touched off by Fleisher⁽³⁾⁻⁽⁵⁾, has been paid by many investigators to improve the expression to meet wider range of experimental observations and yet without losing too much of the physical meanings⁽¹⁹⁾⁻⁽²⁶⁾.

Gypen and Deruyttere⁽²⁴⁾⁻⁽²⁶⁾ most recently have proposed a new combined parameter with use of an improved expression for the modulus misfit parameter. For the elastic interaction involving edge dislocations, it is given as

$$\varepsilon_E = 1/2(|\varepsilon'_G + \alpha\varepsilon_a|^{p/q} + |\varepsilon'_G - \alpha\varepsilon_a|^{p/q})^{q/p} \quad (2)$$

with an expression for ε'_G (modulus misfit parameter),

$$\varepsilon'_G = (\varepsilon_G + A\varepsilon_a)/(1 + |\varepsilon_G + A\varepsilon_a|/2), \quad (3)$$

while for the elastic interaction involving screw dislocation it is given, using the same ε'_G , as

$$\varepsilon_S = |\varepsilon'_G + \alpha\varepsilon_a|. \quad (4)$$

In eq. (2), p , q and α are constants to be properly assigned, while the constant, A , in eq. (3) originally appears in the expression of modulus misfit parameter proposed by Takeuchi⁽²⁰⁾ and has been assigned as a materials constant for a particular solvent by various investigators. Using the above combined parameters, with a proper choice of constants, p and q , it has been shown that the solution hardening is better interpreted with eq. (2) in fcc Pb, Cu, Ag and Au alloys and with eq. (4) in bcc Fe alloys⁽²⁶⁾. In the following section, the solution hardening of Ni₃Al is analyzed through the above treatment.

3. Solid solution hardening of ternary Ni_3Al alloys

The previous investigations⁽¹⁾⁽²⁾ have shown that the relation between 0.2% flow stress, measured at 77 K, and the solute concentration is linear within the small concentration range for ternary additions of all the B-subgroup and transition metal elements examined. Then the value for $d\sigma/dc$ has been determined as the slope of the linear relation. With the results on the determination of dE/dc in the present investigation, we are ready to examine the solution hardening in Ni_3Al for a variety of ternary additions of both B-subgroup and transition metal elements through the treatment using equations (2) to (4). The conversion from uniaxial stress, σ , to the shear stress, τ , can be done by multiplying 1/2 according to the normal approximation, and that from $(1/E)(dE/dc)$ to $(1/G)(dG/dc)$ is unnecessary provided that the concentration dependence of Poisson's ratio being negligible. The modulus misfit parameter defined by eq. (3) is then obtainable taking the constant A as 3.8 which is the value assigned for nickel by Takeuchi⁽²⁰⁾. As to the constant α , which assigns the weight between ϵ'_G and ϵ_a , 3 is chosen for eq. (2) and 16 for eq. (4) according to the original treatment by Fleischer⁽⁵⁾.

Figure 4 shows the relation between $d\sigma/dc$ and the combined parameter defined by eq. (4) that is to represent the elastic interaction between solute atoms and screw dislocations. No correlation can be found for the additions of both B-subgroup and transition metal elements. In order to examine the validity of eq. (2) for the interpretation of solid solution hardening in Ni_3Al , the constants p and q should be properly chosen. In connection with eq. (1) in the preceding section, q can be taken to be 1 hereinafter. It has been proposed that the values for (p, q) are (1, 1) by Nabarro⁽²⁷⁾, (4/3, 2/3) by Labusch⁽²⁸⁾, (3/2, 1/2) by Fleischer⁽²⁹⁾ and (2, 1) by Suzuki⁽²³⁾, where in any cases the parameter p has been taken to be between 1 and 2.

Figures 5 and 6 show the relation between $d\sigma/dc$ and the combined parameter defined by eq. (2) assigning the parameters (p, q) to be

Fig. 4 Relation between rate of solution hardening per one atomic per cent of ternary element, $d\sigma/dc$, and a combined parameter appropriate for the elastic interaction involving screw dislocations, ϵ_S after Gypen *et al.*⁽²⁶⁾.

Fig. 5 Relation between rate of solution hardening per one atomic per cent of ternary element, $d\sigma/dc$, and a combined parameter appropriate for the elastic interaction involving edge dislocations, ϵ_E , after Gypen *et al.*⁽²⁶⁾ taking constants p and q being 1 and 1.

(1, 1) and (2, 1). In both cases a linear correlation is obtained for the additions of B-subgroup elements, and a separate non-linear correlation is found for those of transition metal elements. Comparing two figures it is seen that these correlations are nearly equally established regardless of the choice of (p, q) . Then the following implications are drawn; 1)

Fig. 6 Relation between rate of solution hardening per one atomic per cent of ternary element, $d\sigma/dc$, and a combined parameter appropriate for the elastic interaction involving edge dislocations, ϵ_E , after Gypen *et al.*⁽²⁶⁾ taking constants p and q being 2 and 1.

the solution hardening of Ni_3Al with additions of B-subgroup elements can be well interpreted by the combined parameter defined by eq. (2) for the elastic interaction between solutes and edge dislocations with the present choices for the parameters (p, q), 2) the contribution of modulus misfit to the solution hardening must be small, because the constant α in eq. (2) is 16 instead of 3. This is consistent with the fact that $d\sigma/dc$ for the additions of B-subgroup elements has been shown, in Fig. 10 in Ref. 1, to have linear relation with ϵ_a itself, and 3) with the additions of transition metal elements the degree of solution hardening exceeds over what is expected from the treatment.

It should be noted that a linear correlation could be obtained for the case of transition metal elements by an alternative choice of parameter p . It is more important, however, to recognize that the solution hardening in Ni_3Al can not be interpreted by a single correlation with the combined parameter for the whole sets of both B-subgroup and transition metal elements and that the anomalous hardening effect is exhibited by the additions of transition metal elements. The same anomalous hardening effect of transition metal elements has been notified first by Guard and Westbrook⁽⁶⁾ for

the additions of Mo and Cr in Ni_3Al and recently by Huang *et al.*⁽³⁰⁾ for those of Hf and Zr in the rapidly solidified Ni_3Al . It is interesting to note that several investigators have pointed out that there would be some additional factor other than atomic size misfit to interpret the solution hardening by the additions of transition metal elements in binary nickel alloys^{(31)–(33)}. The reason for the anomalous effect of transition metals is unknown at present, but the investigation is currently extended to the same type of treatment in binary nickel solid solutions to clarify whether it is inherent in the Ll_2 ordered structure of the compound or the transition metal solvent, nickel⁽¹⁷⁾.

4. Role of edge and screw dislocations in the deformation of Ni_3Al

The present result is to provide an original evidence for the solution hardening of intermetallic compound Ni_3Al being governed by the elastic interaction involving edge dislocations as in many other fcc terminal solid solutions. It would be reasonable because the Ll_2 ordered crystal structure of the compound is actually based on fcc. In turn, it is worthy of notice that the solution hardening of bcc alloys is generally attributed to the elastic interaction involving screw dislocations. This can be understandable due to the well-known fact that the Peierls stress for screw dislocation is substantially larger than that for edge dislocation in a bcc crystal structure⁽³⁴⁾⁽³⁵⁾.

Plastic behavior of Ni_3Al has often been characterized by the anomalous positive temperature dependence of strength which has been found in a family of Ll_2 intermetallic compounds⁽³⁶⁾. It is well known that the mechanism for the mechanical anomaly is attributed to the cross slip of segment of screw dislocations gliding on (111) planes onto (100) non-glide planes, by which the multiplication of dislocations on the (111) plane becomes suppressed. This is a thermally activated process which becomes frequent above room temperature. There have been a number of observations by transmission electron microscopy that most of dislocations are straight with screw component after high temperature deformation^{(37)–(43)} and, on the other hand, that they are of mixed type

having no pronounced directionality after deformation at 77 K⁽³⁷⁾⁻⁽³⁹⁾⁽⁴⁴⁾.

The present authors⁽¹⁾⁽²⁾ have treated the temperature dependence of flow stress in Ni₃Al to be the sum of two components in which the flow stress component being expressed by the Arrhenius type equation is added to the normal negative temperature dependent component. It is concluded that the former component is governed by the motion of screw dislocations and the latter by that of edge dislocations.

IV. Conclusion

The Young's modulus of ternary Ni₃Al alloys is measured and an attempt is made to interpret the solid solution hardening of the compound. The following conclusions are drawn:

(1) Young's modulus of the ternary compounds changes linearly with increasing solute concentration for additions of both B-subgroup and transition metal elements. Change in Young's modulus per one atomic per cent of solute, dE/dc , is determined from the slope of the linear relation for each ternary element.

(2) For B-subgroup elements, dE/dc is linearly related with the square of valence difference from aluminum, $(\Delta Z)^2$, for which they substitute in the compound.

(3) The solid solution hardening by the additions of B-subgroup elements is well interpreted by a combined parameter for elastic interaction between solutes and edge dislocations involving atomic size misfit and modulus misfit. The contribution of modulus misfit must be insignificant.

(4) The solid solution hardening by the additions of transition metal elements cannot be equally correlated with the same combined parameter as for B-subgroup elements. Transition metal elements provide anomalous hardening effect in Ni₃Al.

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