JOURNAL OF MATERIALS SCIENCE 16 (1981) 2737–2744

The temperature dependence of the strength of pseudo-binary platinum-based $L1_2$ alloys with B-subgroup elements

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The notable effect of atomic radius ratio of the components of $L1_2$-ordered alloys on the temperature dependence of their strength has been demonstrated using suitable alloys and alloying elements. Pt_3In and Pt_3Sn were chosen and partial substitutions were made of the minor components with Ga and Tl, and with Ge and Pb, respectively, keeping the electron concentration of the system constant. The positive temperature dependence was generally found to be strengthened by the smaller substituting atoms and to be weakened by the larger atoms. On the other hand, the substitution of In was found to strengthen the anomalous behaviour of Pt_3Ga. Thus, a maximum anomalous strengthening is achieved with an In-rich $Pt_3(In, Ga)$ alloy, which forms a continuous solid solution with a $L1_2$ structure. All these effects are consistent with the model obtained previously by experiments on binary $L1_2$ alloys and are correlated to the radius ratio rule, which leads to the change of phase stability of $L1_2$ alloys.

1. Introduction

There have been frequent attempts to understand the effect of ternary additions on the positive temperature dependence of strength of Ni_3Al [1–7]. In most cases, ternary alloying was implicitly confined to the elements which might play an important role as alloying additions in the development of nickel-based superalloys. However, they do not provide a sufficiently detailed basis for a proper understanding of the anomalous mechanical behaviour and the constitutional effects of these additions are still complex. Little attention has been paid to the correlation between changes in phase stability and the characteristic mechanical behaviour in ternary $L1_2$-ordered alloys until recently. Recently [8], the present authors found that most of platinum-based $L1_2$ alloys with B-subgroup elements have a positive temperature dependence of strength as well as the nickel-based $L1_2$ alloys with B-subgroup elements. In addition, they have shown that, depending on the atomic radius ratio of the components, the magnitude of the positive temperature dependence of strength of binary $L1_2$ alloys is systematically changed at a constant electron concentration [9], (see Fig. 7 of [9], which is reproduced in Fig. 5). This scheme has been discussed in terms of phase stability of the $L1_2$ phase with respect to other geometrically close-packed crystal structures.

These results clearly indicate that systematic variation in the phase stability can be achieved by the partial replacement of one of the components in either direction, according to whether the average atomic radius is to be decreased or increased. It is hoped that the partial replacement would enable the anomalous strengthening behaviour of an $L1_2$-ordered alloy to be strengthened or weakened. The present work, motivated by this possibility, is mostly concerned with the effect of ternary additions on the mechanical behaviour of Pt_3In and Pt_3Sn. Both compounds are likely to be very suitable ones for demonstrating the drastic change in the mechanical property by a small amount of ternary additions.

0022–2461/81/102737–08$02.80/0

TABLE I Summary of heat-treatment of specimens used in this study and the lattice constant obtained.

Compound	Composition (at %)		Heat-treatments Temperature (K)/Time (ksec)	Lattice constant (10^{-1} nm)
Pt_3In	In 25.0	–	1373/345.6 → 1173/259.2	3.999
	In 22.9,	Ga 2.2		
	In 17.9,	Ga 7.5		
$Pt_3(In, Ga)$	In 12.9,	Ga 13.0		see Fig. 1
	In 8.1,	Ga 18.0	1273/259.2	
	In 3.0,	Ga 23.8		
Pt_3Ga	–	Ga 27.0		3.894
$Pt_3(In, Tl)$	In 24.5,	Tl 0.1	1373/345.6 → 1173/259.2	4.001
Pt_3Sn	Sn 24.75	–	1273/604.8 → 1173/86.4 → 1073/86.4	4.007
$Pt_3(Sn, Ge)$	Sn 24.25,	Ge 0.5		4.001
$Pt_3(Sn, Pb)$	Sn 24.25,	Pb 0.5		4.014

2. Experimental procedure

The starting materials used in this investigation had a purity of 99.99% or better. Alloy buttons of about 20 g were made by arc-melting on a water-cooled copper hearth using a tungsten electrode in a gettered argon atmosphere. The buttons were turned and remelted several times to achieve homogeneity. Most of ternary alloys were prepared with scraps and used specimens of binary alloys in order to save platinum metal. Chemical analysis of the resultant specimens was not performed because the evaporation loss was extremely small, with the exception of the alloy containing thallium. The heat-treatments employed are listed in Table I.

Rectangular compression specimens, 3 mm x 3 mm x 7 mm, were prepared for all alloys by a slitting wheel cutter. Specimens were strained in an Instron-type machine at a strain rate of approximately 1.4×10^{-4} sec^{-1}. As the phase diagrams of all these pseudo-binary systems are not known yet, careful inspection was needed to confine the alloys used to those in the $L1_2$ phase field. By X-ray diffraction and metallographic observation, all the ternary alloys of $Pt_3(In, X)$ were confirmed to be a single phase of $L1_2$ structure with very fine and fairly equiaxed grains of about 4 μm diameter. Since Pt_3Sn is a Daltonide-type compound, i.e. a line compound, inclusion of a small amount of second phase is inevitable. X-ray analysis was carried out using mostly MoKα and CrKα radiations using a Shimadzu diffractometer. The details of the experimental methods have been given previously [8–10].

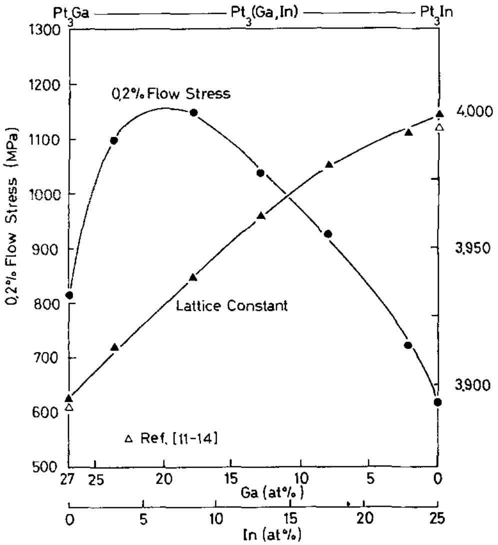

Figure 1 The composition dependence of the lattice constant and the 0.2% flow stress at 77 K of $Pt_3(In, Ga)$ solid solution with the $L1_2$ structure.

3. Results

3.1. Pseudo-binary platinum-based alloys with 3B-subgroup

It is expected that Pt_3In and Pt_3Ga form a continuous series of solid solutions with the $L1_2$ structure, because the atomic size factor seems to be favourable. This has been verified by metallographic observations and by the continuity of the lattice constant. The composition dependence of the lattice constant and the 0.2% flow stress at 77 K are shown in Fig. 1. Fig. 1 clearly shows that the lattice constant of $Pt_3(In, Ga)$ has a positive deviation from Vegard's law. The solution hardening by the substitution of Ga for In and that of In for Ga is seen to be prominent, while the maximum flow stress is not observed in the middle but at the Ga-rich portion of the system. In the case of Tl addition, the estimated composition

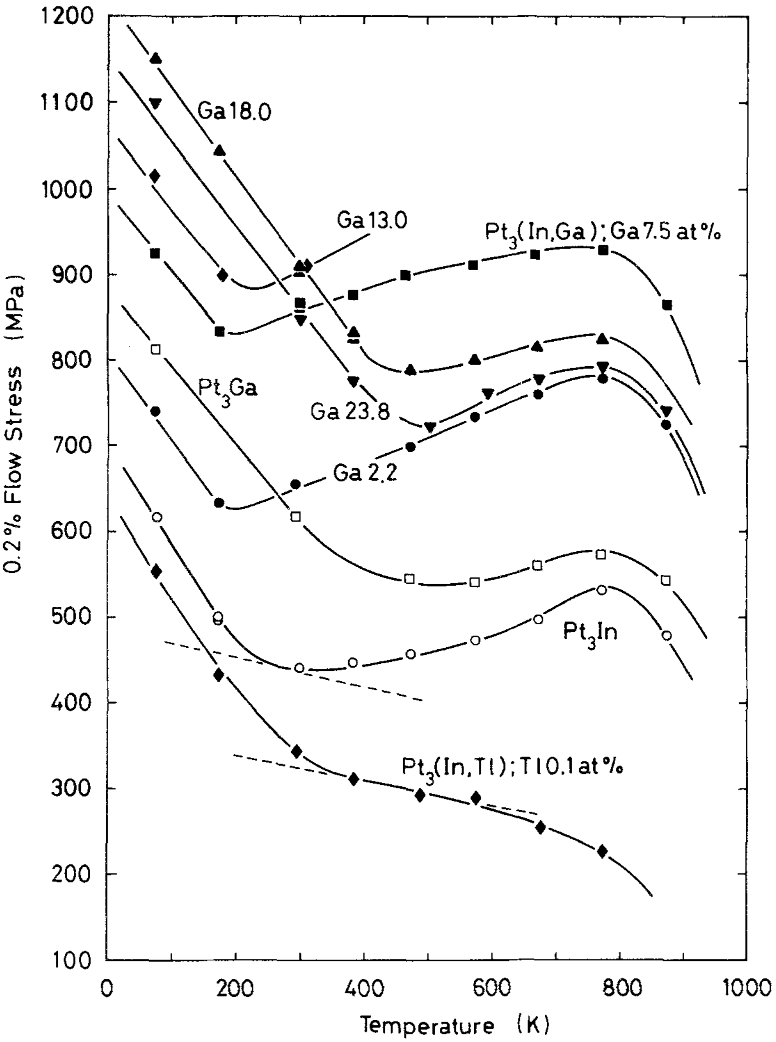

Figure 2 The variations in flow stress with temperature for $Pt_3(In,Ga)$ and $Pt_3(In,Tl)$ ternary alloys.

of $Pt_3(In, Tl)$ from the weight loss during the melting was at least 0.1 at% Tl apart from the intended composition, because of the higher vapour pressure of Tl. However, the lattice constant of the alloy changes to a somewhat larger value on addition of Tl, as shown in Table I. The solubility limit of Tl for Pt_3In could not be tested in this experiment.

The variations in flow stress with temperature for $Pt_3(In, Ga)$ and $Pt_3(In, Tl)$ are shown in Fig. 2, where the data of the binary alloys are taken from the previous work [9]. All the alloys shown in Fig. 2 have a marked flow stress–temperature sensitivity at temperatures below room temperature. This characteristic has been reported on most platinum-based $L1_2$ compounds [9].

The effect of Ga addition forms a remarkable contrast with that of Tl addition on the positive temperature dependence of strength of Pt_3In. Fig. 2 shows that the substitution of a small amount of Ga, being smaller in atomic size, causes an increase in the magnitude of the anomalous behaviour. This is verified by the fact that the bottom temperature (T_b) of Pt_3In, at which the flow stress shows a minimum, is reduced by the substitution of 2.2 at% Ga for In. The relationship between the anomalous behaviour and the bottom temperature will be shown later. On the other hand, the anomalous behaviour of Pt_3In is found to be removed by the substitution of a very small amount of Tl, being larger in size. Disappearance of the anomalous behaviour of Pt_3In is evidence that an appreciable amount of Tl was incorporated into Pt_3In.

In contrast to the above, the anomalous behaviour of Pt_3Ga appears to be strengthened by the substitution of In, being larger in size than Ga. Fig. 2 shows that the bottom temperature of $Pt_3(Ga, In)$ is lowered with increasing In contents

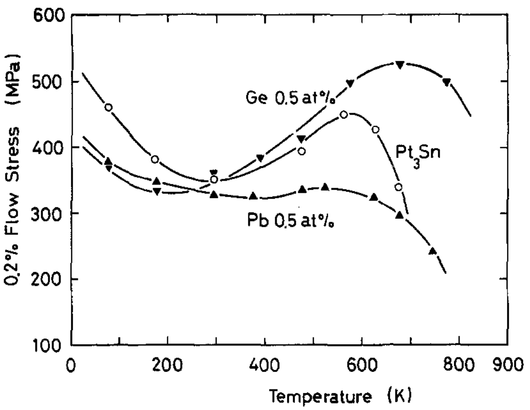

Figure 3 The variations in flow stress with temperature for $Pt_3(Sn, Ge)$ and $Pt_3(Sn, Pb)$ ternary alloys.

up to 17.9 at% In. These effects, however, are the expected variations in view of the previous result and will be discussed later in a quantitative manner, i.e. the change in activation constant.

3.2. Pseudo-binary platinum-based alloys with 4B-subgroup elements

The addition of Ge into Pt_3Sn results in a decrease in the lattice constant and the addition of Pb results in the opposite effect. The lattice constants of Pt_3Sn, $Pt_3(Sn, Ge)$ and $Pt_3(Sn, Pb)$ are shown in Table I. The formation of a continuous solid solution between Pt_3Sn and Pt_3Pb may be expected, whereas the extents of solubility of Ge and Pb in Pt_3Sn were not tested in this experiment.

Fig. 3 shows the effect of the alloying additions upon the temperature dependence of strength of Pt_3Sn. From Fig. 3, the positive temperature dependence of Pt_3Sn is apparently strengthened by the substitution of Ge, being smaller in size than Sn, and is weakened by the substitution of Pb, being larger in size than Sn. In this regard, Pt_3Pb is the only example having no anomalous behaviour among nickel-based and platinum-based $L1_2$ alloys with B-subgroup elements. It is obvious that the effects of Ge and Pb on Pt_3Sn act in the same way as those of Ga and Tl on Pt_3In. These are expected variations in view of the model reported previously [9].

3.3. Derivation of the activation constant

The transition from a positive to a negative temperature dependence of strength has been reported in a series of polycrystalline $(Ni, Fe)_3Ge$ ternary alloys with the $L1_2$ structure by the present authors [10]. In order to describe the weakly-positive temperature dependence of strength of $(Ni, Fe)_3Ge$ alloys, they have proposed a refined method for the derivation of the activation constant of the anomalous behaviour [9]. This will be briefly reviewed. The flow stress τ should be divided into two temperature-dependent components. One is dependent on temperature through the shear modulus change with temperature and the other temperature-dependent component is caused by the thermally-activated cross-slip of screw dislocation onto cube planes. The flow stress τ is then given by

$$\tau = \tau_0(1 - BT) + Ae^{-U/RT}, \qquad (1)$$

where τ_0 is the flow stress at 0 K, U is the activation constant, R is the gas constant, T is temperature, A and B are constants. B is defined as $[1-(G_2/G_1)]/(T_2 - T_1)$, where G_1 and G_2 are the shear moduli at temperatures T_1 and T_2. Under the condition of $d\tau/dT = 0$, a bottom flow stress τ_b and a bottom temperature T_b are introduced into a flow stress against temperature curve.

It has been shown previously that most platinum-based compounds show the considerably steeper, negative temperature dependence of strength below T_b than that expected from the temperature dependence of the shear modulus, B [9, 10]. In this case, τ_0 was estimated from the equation $\tau_0 = \tau_b/(1 - BT)$. The dotted lines in Fig. 2 indicate examples of hypothetical flow stress change of an alloy neither exhibiting the anomalous strengthening in higher temperatures nor the marked flow stress sensitivity at low temperatures below T_b. This procedure reveals that $Pt_3(In, Tl)$ has no anomalous behaviour, as shown in Fig. 2.

In addition, it has been shown that the bottom temperature T_b is a good index to evaluate the activation constant, U, when the temperature range from the bottom to the peak in flow stress is too narrow to estimate U by an ordinary Arrhenius plot [9]. According to this, T_b gradually shifts to higher temperatures at a constant stress level of the alloy, as the activation constant U increases. On the other hand, as pointed out previously [9, 10], the peak temperature T_p may not necessarily be a suitable index for describing the magnitude of the anomalous behaviour. The activation constants of the alloys tested were derived by the Arrhenius plot and/or by the T_b method. For the alloys for which there were enough data for an Arrhenius plot, ln $[\tau - \tau_0$

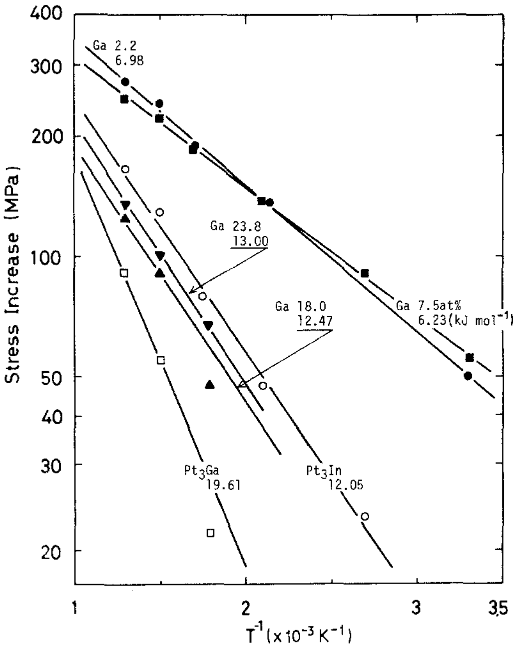

Figure 4 Logarithmic plots of the stress increment versus reciprocal temperature for Pt_3 (In, Ga) alloys.

$(1-BT)]$ is plotted against the inverse of temperature in Fig. 4. All the data shown in Fig. 4 are seen to obey the Arrhenius relationship.

Fig. 5 summarizes the relationship between the activation constant and the deviation from unity of atomic radius ratio, $(R_B - R_A)/R_A$, incorporating data for binary nickel-based and platinum-based A_3B alloys having the $L1_2$ crystal structure [9] and for the present pseudo-binary alloys. In Fig. 5, the reciprocal activation constant is used so as to make the value $U = \infty$ visible. In the case of pseudo-binary alloys, the weighted mean of the atomic radius of B-subgroup elements, R_B, is derived from the lattice spacing of the alloys, assuming that the atomic radius of platinum, R_A, is regarded as that of the pure f c c metal. In Fig. 5 some scatter is evident in the values of activation constant for both binary and pseudo-binary alloys. For this, there are several reasons other than the uncertainty of the determination of U. It has been well-known that the magnitude of the positive temperature dependence of strength is governed

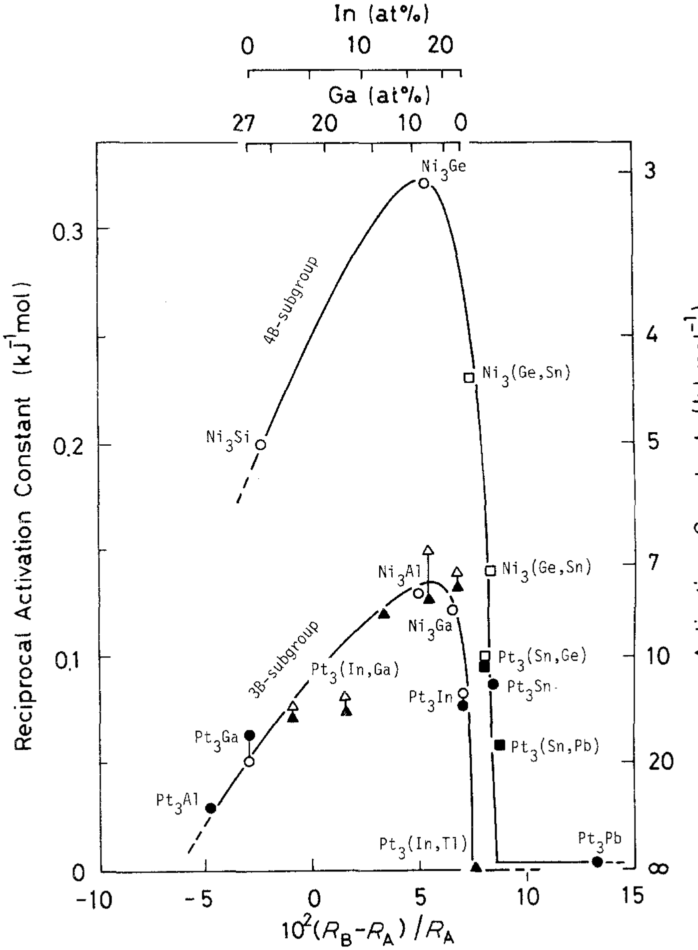

Figure 5 Activation constant against deviation from unity of atomic radius ratio for platinum-based and nickel-based $L1_2$ alloys with B-subgroup elements. Open marks are derived from the Arrhenius plot and solid marks from the T_b method.

by the crystal orientation [7, 15–19] and by the composition deviating from stochoimetry [4, 5, 7, 20]. In general, the cast material has columnar grains but not in most cases of this experiment. Furthermore, the present investigation clearly demonstrates that a very small amount of impurity may have a significant effect on the temperature dependence of strength of a binary alloy. Nevertheless, Fig. 5 shows a general trend for the relationship between the activation constant, the atomic radius ratio and the electron concentration. The values of activation constant for the alloys containing the elements of like valency lie within a band rather than on a well-defined line.

It can be seen that all the data of the present pseudo-binary alloys lie well within the bands and between the data of the binary alloys previously obtained. The drastic changes in the activation caused by the additions of Ga and Tl to Pt_3In apparently takes place for the same reason as for the changes caused by the additions of Ge and Pb to Pt_3Sn. The results of Pt_3In–Pt_3Ga $L1_2$ solid solutions satisfactorily show that the activation constant decreases to the level for Ni_3Al at a definite value of $(R_B - R_A)/R_A$ of approximately 0.05.

4. Discussion

The experimental results demonstrate that the magnitude of the temperature dependence of strength of platinum-based and nickel-based alloys with B-subgroup elements varies systematically with regard to the apparent atomic radius ratio, and that this scheme applies to all those substitutions in which the electron concentration remains constant. An application of this data to Ni_3Ge, an unpublished work [21], is added in Fig. 5. In this case, it is clear again that the substitution of Sn, being larger than Ge, also decreases the anomalous behaviour of Ni_3Ge. The above scheme suggests that the variation of phase stability of $L1_2$ phase with respect to other geometrically close-packed phases is closely related to the anomalous behaviour, as stated below. That is, the phase stability with respect to the $D0_{22}$, $D0_{19}$ and A1 phases are very important factors in view of the dislocation configuration in the $L1_2$ crystal structure [22–24].

An infinite value of the activation constant means that the $L1_2$ alloy has the ordinary, negative temperature dependence of strength, while a zero activation constant means that the $D0_{22}$ crystal structure or a long-period structure becomes the stable phase. This conclusion is based on the Kear–Wilsdorf mechanism, i.e. the thermally activated cross-slip of $a/2\ \langle 1\,1\,0\rangle$ dislocation from $\{1\,1\,1\}$ onto $\{1\,0\,0\}$ planes, because the $D0_{22}$ and other related structures are obtained crystallographically from the $L1_2$ structure by the periodic displacement of $\{1\,0\,0\}$ planes in the $a/2\ \langle 1\,1\,0\rangle$ direction [25]. Thus, the low phase stability with respect to the $D0_{22}$ phase results in a large anisotropy of antiphase boundary (APB) energy, i.e. a relatively low APB energy on the $\{1\,0\,0\}$ plane. Such an $L1_2$-ordered alloy is in accordance with Flimn's model, which takes into account only nearest-neighbour interactions [26]. This low $\{1\,0\,0\}$ APB energy is thought to be the driving force for the cross-slip onto $\{1\,0\,0\}$ planes [10, 27].

The dissociated structure of a superdislocation is characterized essentially by the APB energy and the stacking-fault (SF) energy (22–24]. Although the heat of formation or the melting point of the compound is a measure of $\{1\,1\,1\}$ APB energy, it is not possible in the present state of knowledge to estimate APB energies of the present pseudo-binary alloys. However, it has been already shown that the higher $\{1\,1\,1\}$ APB energy is not responsible for the anomalous behaviour [8–10]. Some of the Daltonide- or Berthoride-type compounds, in which the ordered structure is stable up to their melting points, show no anomalous behaviour [8, 10]. On the other hand, it has been suggested by the present authors that Cu_3Au is one of the ordered alloys having the anomalous temperature dependence of strength, although Cu_3Au has rather low phase stability with respect to a disordered f c c phase (A1) and hence has a relatively low $\{1\,1\,1\}$ APB energy [9. 10].

The SF energy of the $L1_2$ phase is closely related to the phase stability with respect to the $D0_{19}$ phase. It has been shown by van Vucht and Buschow [28] that geometric factors are very important in determining the type of geometrically close-packed structure in rare-earth trialuminides. The crystal structure of the rare-earth trialuminides changes from the cubic $L1_2$-type to the hexagonal $D0_{19}$-type with increasing the radius ratio of the rare-earth elements and aluminium. Moreover, based on the experiments of partial replacement of one of the components in various A_3B alloys, van Vucht [29] concluded that the radius ratio rule, as deduced from the rare-earth trialuminides, could apply to all substitutions as long as the valency of the system remained constant. In most cases, such a structure transition is found to be within a relatively narrow range from $(R_B - R_A)/R_A = 0.15$ to 0.25. It is obvious that the radius ratio rule can be applied to the present case, since the DO_{19} structure is known to occur in Ni_3In and Ni_3Sn, where $(R_B - R_A)/R_A$ is estimated to be 0.24 and 0.22, respectively.

In view of the above situation, it is assumed that with increasing radius ratio the SF energy of $L1_2$ alloys decreases continuously until it reaches a phase stability limit of $L1_2$ phase with respect to the DO_{19} phase. On reducing the SF energy the cross-slip becomes difficult. Thus the abrupt increase in the activation constant at a definite radius ratio is partly due to the lowering of SF energy. It is believed, however, that the main reason for this change is associated with enhancement of the phase stability with respect to the DO_{22} phase, although no quantitative explanation is given at present. This may be explained qualitatively by the optimization of higher crystal symmetry and the better packing density based on the hard sphere model [28, 29]. According to van Vucht [29], the packing density increases from the cubic $L1_2$ stacking to the hexagonal DO_{19} stacking and from the triangular-type layer ($L1_2$-type) to the rectangular-type layer (DO_{22}-type) with increasing atomic radius ratio of the component atoms.

The electron concentration effect on phase stability, which is thought to be the major factor controlling the type of structure, has been noted by many workers [30, 31]. The studies on various pseudo-binary system led to the conclusion that an increase in hexagonality of the stacking is associated with an increase in the electron concentration. In addition, it has been shown that on increasing the electron concentration the transition from the triangular-layer ordering to the rectangular-layer ordering takes place [30. 31]. The positive deviation from unity of atomic radius ratio and the increase of electron concentration seems to act in a similar fashion on reducing the phase stability of the $L1_2$ phase with respect to the DO_{22} phase. It may be concluded that the increase of electron concentration in the $L1_2$ phase gives rise to the reducing of the APB energy on the $\{100\}$ plane, which facilitates cross-slip onto $\{100\}$ planes.

There is additional experimental evidence to show that the electron concentration can reduce the stability of the $L1_2$ phase with respect to the DO_{22} phase and periodically modulated phases. The addition of a small amount of a polyvalent element, like Al, increases the electron concentration of Cu_3Au and makes the long-period structure more stable with respect to the $L1_2$ phase [32]. In the case of Au_3Cu, the long period structure can be created by the addition of a small amount of Al [33]. In order to explain their observations, Sato and Toth [34] suggested that the long-period structure gains its stability through the reduction of the Fermi energy by touching of Brillouin zone boundaries by the Fermi surface of the alloy.

It can be seen in Fig. 5 that the present case is consistent with the electron concentration effect. Noting that the zero activation constant implies the appearance of the DO_{22} structure, 4B-subgroup elements make the $L1_2$ phase less stable with respect to the DO_{22} phase than 3B-subgroup elements as one of the components in A_3B alloys. In view of this we can predict that a close-packed ordered phase with a 5B-subgroup element may form a periodically-modulated structure such as the DO_{22} structure. The only example is Pt_3Sb with the DO_{23} structure among the nickel-based and platinum-based ordered alloys with B-subgroups [8, 9]. The validity of the scheme shown in Fig. 5 may be further established by an experiment on the effect of B-subgroup elements with unlike valencies on the anomalous behaviour. These aspects of the problem are presently being investigated and will be reported later.

5. Conclusions

In conclusion, the validity of the model presented in a previous work, i.e. the correlation between the anomalous behaviour and the radius ratio of the alloys, could be confirmed for Pt_3In and Pt_3Sn by ternary additions. The appearance of the anomalous behaviour in Pt_3In and Pt_3Sn is very critical in the sense that the phase stability of the $L1_2$ phase is sensitively affected by a small change of the atomic radius ratio. A short transition from a positive to a negative temperature dependence of strength, in the cases of both compounds, takes place when varying the weighted mean of atomic radius of the primary and substituting B-subgroup elements. According to the scheme shown in Fig. 5, Ni_3Al (or Ni_3Ga) and Ni_3Ge correspond to the condition being least stable relative to the DO_{22} phase among the alloys with 3B-subgroup and 4B-subgroup elements, respectively. In contrast to the cases of Pt_3In and Pt_3Sn, it is concluded that a much greater amount of addition of a third element being larger in size, such as In for Ni_3Ga and Sn for Ni_3Ge, should be expected to weaken or remove the anomalous behaviour in these compounds, if the solubility of the substituting element is not restricted.

It is confirmed that a continuous series of solid solutions is formed in the Pt_3In–Pt_3Ga system. With an increase of In, the anomalous behaviour of Pt_3(Ga, In) increases gradually at first, reaches a peak value at the level of Ni_3Al with a further increase of In, and finally decreases suddenly into the value of the binary Pt_3In. The present result gives a clear image of how variation of the magnitude of the temperature dependence of strength can occur from alloy to alloy in terms of the atomic radius ratio rule. Further work is suggested on the electron concentration effect in collaboration with the present work.

Acknowledgements

The authors wish to thank Dr D. M. Wee of the Korea Atomic Energy Research Institute for many helpful discussions.

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Received 17 November 1980 and accepted 26 March 1981.