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Thermodynamics of Precipitation Systems

R. O. Williams

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CONTENTS

ABSTRACT	1
INTRODUCTION	1
ATTRIBUTES OF A USEFUL SYSTEM	2
THERMODYNAMICS OF ALLOY SYSTEMS	3
BINARY SYSTEMS	6
Miscibility Gaps	6
Binary Eutectic Systems	6
Compound Formation	9
Asymmetric Systems	10
HIGHER ORDER SYSTEMS	12
SUMMARY AND CONCLUSIONS	18
ACKNOWLEDGMENTS	18

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ABSTRACT

The particular combination of features that are required for a binary system to show useful precipitation hardening is examined. Because this combination is relatively rare, we are not optimistic that many useful binary systems remain to be discovered. When one goes to higher order systems one has much greater opportunities for developing useful systems because of the increased degrees of freedom. The difficulty of understanding what is happening is also correspondingly greater, but if it is possible to properly represent the thermodynamics of the system, then this problem can be resolved. Considerable potential is believed to exist for the development of new alloys containing three or more components.

INTRODUCTION

It is very well known that the first prerequisite for precipitation hardening is a decreasing solubility with decreasing temperature. Since this occurs universally as a result of the decreasing importance of the entropy of mixing with decreasing temperatures, one may well ask why there are not a large number of useful precipitation systems. In this paper we will examine what supplementary conditions are also necessary and show that the thermodynamics of useful systems is somewhat unusual. From this understanding we should be able to predict the usefulness of a specific binary system from its phase diagram and structural information. But we do not expect that many useful binary systems remain to be discovered.

When we go to systems with more components the situation takes on new complexity matched by expanded opportunities for precipitation hardening. The problem is in understanding these more complex systems in order to predict where useful compositions are to be found. This report shows how the problem can be addressed.

ATTRIBUTES OF A USEFUL SYSTEM

A useful volume of the second phase, typically in excess of 1%, must be able to form when the material is quenched from elevated temperatures and subsequently aged at some suitable temperature, normally a temperature above the desired service temperature. The precipitation must be sufficiently rapid that the optimum properties can be obtained in less than 24 h. The new phase must be finely dispersed, as in particles 10 nm in diameter. This requires that the particles and the matrix must be coherent because this degree of dispersion requires homogeneous nucleation and homogeneous nucleation of incoherent particles is unknown. In general, the two lattices will not match perfectly; thus the coherency will lead to some strain energy, which can be an important source of strengthening. This requirement of coherency limits the difference that can exist between the two structures. If the two lattices are basically the same, then a coherent precipitate may form if the difference between the lattice parameters does not exceed 10%. Further, a coherent hcp precipitate can form in an fcc lattice on the (111) habit plane provided the mismatch is similarly limited. This probably occurs on dislocation loops resulting from vacancy precipitation and thus is homogeneous precipitation in a restricted sense. Coherency between bcc and fcc or hcp lattices does not occur except possibly under extreme degrees of supersaturation.

The presence of order in the precipitate provides an important source of strengthening, and in the absence of coherency stresses this ordering is essential if useful strengthening is to be achieved. A further advantage of using ordered particles for strengthening rather than coherency strains is that such strains necessarily contribute to instability of the system; the presence of order does not.

THERMODYNAMICS OF ALLOY SYSTEMS

For our purpose the free energy of a solution having the α structure is representable as

$$G_{\alpha} = \sum_{i \neq j} \frac{X_i^2 X_j}{X_i + X_j} \phi_{ij} + \sum X_i (G_i + RT \ln X_i), \quad (1)$$

where

X_i = the atom fraction of the i th component,

ϕ = a heat of mixing parameter,

G_i = the free energy of pure i th component relative to its standard state.

Kaufman calls these the lattice stability parameters. This representation is known as the Kohler formula.¹

The serious reader should have a reasonably good grasp of the meaning of this equation. For this purpose we show in Fig. 1 a set of

Fig. 1. Free Energy Curves for a Binary System in Which the Structure of the Two Components is Different.

¹P. Kohler, *Monatsh. Chem.* 91: 738-40 (1960).

free energy curves, which might represent a binary eutectic system between terminal phases of different structures. The meaning of G_B is simply the free energy required to convert B from the stable β structure to the α structure. There is an analogous term for the component A and two similar terms for liquids A and B . Often the free energy curves are symmetric (when the lattice stability terms are ignored) such that $\phi_{ij} = \phi_{ji}$; such solutions are normally called regular. Frequently the equality does not hold, as is the case illustrated. More generally the ϕ s will have a temperature dependence representing a temperature-dependent heat of mixing and excess entropy. Lacking such a dependence our expression would correspond to an ideal entropy of mixing and the ϕ s represent the heat of mixing. Any temperature dependence beyond that included in Eq. (1) is too small to drastically change the free energy curves and need not be considered for our present purpose.

Our diagram also represents a useful observation. The free energy curves for the liquid and the three common solid structures are similar in the sense shown. Almost universally, the free energy curve for the liquid is somewhat more negative. We should, perhaps, restrict these statements to the kinds of solutions of interest here; in the case where the solubilities are very small because of a large positive heat of mixing and/or large lattice stability terms we are less sure of the true situation.

This correlation also extends to compound formation in that the more negative the heat of mixing the more likely is compound formation. Compounds are almost nonexistent in systems having a positive heat of mixing. For the case of asymmetry the compounds are more likely to occur near the composition where the heat of mixing is most negative. All of this means that the nature of the interaction between different kinds of metal atoms is not particularly structure sensitive.

The partial free energy for the i th component in this solution is given as

$$\begin{aligned} \bar{G}_{\alpha i} = & PT \ln X_i + G_i + \sum_j \left[\frac{X_i X_j}{X_i + X_j} \left(\frac{X_j}{X_i + X_j} + 1 - X_i \right) \phi_{ij} \right. \\ & \left. + \frac{X_j^2}{X_i + X_j} \left(\frac{X_j}{X_i + X_j} - X_i \right) \phi_{ji} - \sum_l \frac{X_j^2 X_l}{X_j + X_l} \phi_{jl} \right] . \quad (2) \end{aligned}$$

It is important to note that every term in the first sum in Eq. (1) is included in this expression. If we have a new phase consisting of X_i and X_j moles of i and j and having a free energy of G_{ij} , then this phase will form if $X_i G_i + X_j G_j$ is more positive than G_{ij} ; the precipitation from the α phase will cease when the equality is obtained. This criterion simultaneously applies to all possible new phases and can be extended to phases of more than two components.

It is well known that the coherency energy increases the solubilities; that is, the two-phase fields are reduced in extent. The way in which this effect is to be treated has recently been presented.²

A few sets of G are known experimentally, but usually they have to be estimated. Values have been provided by Kaufman and Bernstein³ and by Michaels et al.⁴ The ϕ s can be deduced from the thermodynamics and from the phase diagrams. A number of parameters have been published and the number should increase substantially in the future. At the present none of these parameters have been obtained from theory with useful accuracy.

²R. O. Williams, *Elastic Energy and Metastable Phase Equilibria for Coherent Mixtures in Cubic Systems*, ORNL-5509 (February 1979).

³L. Kaufman and H. Bernstein, *Computer Calculation of Phase Diagrams*, Academic Press, New York, 1970.

⁴K. F. Michaels et al., "Considerations on the Kaufman Approach to Binary Phase Diagram Calculation," *Metall. Trans.* 6A: 1843-48 (1975).

BINARY SYSTEMS

For binary alloys the simplest way to cover the important factors is to examine various types of binary systems.

Miscibility Gaps

When the structure of both members of a binary pair is the same and the heat of mixing is positive then there will exist a miscibility gap whose maximum, in Kelvins, is numerically equal to approximately the maximum heat of mixing in calories per mole (about one-fourth that in joules per mole). If the heat of mixing is too positive, the gap will be incomplete as a result of melting, as in the Cu-Ag system. For coherent precipitation the gap will be suppressed in proportion to the square of the difference in lattice parameters of the two components. In some cases, as the Au-Ni system, the amount of suppression can be very considerable. Within the coherent gap one can obtain coherent phase separation, a process termed spinodal decomposition. In some cases, as the Au-Pt and Fe-Cr systems, strength increases are observed.

Perhaps the most important defect of such systems is that the second phase is not ordered. Then, the coherency strains may be too small to strengthen or too great to permit precipitation. And further, the gap may extend to high temperatures and thus restrict the amount of the minor phase or else occur so low in temperature that the aging is too sluggish. In any case such binary systems are of little or no use.

Binary Eutectic Systems

Here we are concerned with the case where the structures of the two metals are different; otherwise they constitute a special case of the above. If the heats of mixing are again positive, Fig. 2(b), then one

ORNL-DWG 75-12628

Fig. 2. Possible Free Energy Curves for a Binary Eutectic System for Components with Different Structures. If, as in (b), the heat of mixing is positive, a metastable miscibility gap can exist. No gap can occur for negative heats of mixing, (c), but metastable compounds might form or, as suggested in (b), a metastable hcp phase might precipitate from the fcc phase.

will have metastable miscibility gaps, which can lead to decomposition, as considered above, with the possible important difference that the degree of supersaturation is reduced because of the metastability. The Fe-Cu system is a known case where this type of behavior occurs: one can precipitate bcc Cu in Fe or fcc Fe in Cu.

If the heats of mixing are negative, then no metastable gaps can exist, but metastable phases might form, as suggested in Fig. 2 (b and c), which indicates that an hcp phase could precipitate in the fcc phase or a CsCl structure could precipitate in the bcc phase.

Provided the degree of supersaturation is sufficiently great, it may be possible to precipitate coherent α in β or vice versa. The case of Cu-Cr may be such a case, although the Cr could be initially fcc or hcp.

The Ni-Cr system, Fig. 3, is a particularly interesting system of this type in which the heat of mixing for the bcc phase is positive, leading to a very high degree of supersaturation, which causes precipitation of Ni, presumably a bcc structure initially. The heat of mixing for the fcc Ni-rich phase is negative and leads to such a small degree of supersaturation that fcc Cr does not precipitate. There is an

Fig. 3. The Phase Diagram for the Cr-Ni System.

ordering reaction at lower temperatures in this phase, which does display some strengthening.

The reader should appreciate that what happens in systems of this type is dictated both by the heats of mixing and the magnitudes of the lattice stability terms. For a constant stability energy difference it is readily apparent that increasingly negative values of the heat of mixing increase the terminal solubilities. These higher values might be viewed as an asset, but, unfortunately, the solubilities change less rapidly with temperature, so the required degree of supersaturation may not be obtainable.

Compound Formation

If we have a system, as shown in Fig. 4, where a number of compounds have strongly negative heats of formation, then, in general, useful

Fig. 4. Phase Diagram and Free Energy Curves for a Case with Strong Compound Formation. This diagram is based on the Al-Ni system. For such systems it is normally possible to obtain supersaturation in the terminal solid solutions only with respect to the adjacent phase.

precipitation might occur in either terminal solid solution. It is likely that significant supersaturation can be obtained only with respect to the structure in equilibrium with the terminal phase. This results from the metastable solubility of the intermediate phases being large compared with the equilibrium solubilities. Whether or not such structures precipitate and are useful depends upon the structural similarity to the terminal solution. The system can still fail to provide useful age hardening compositions if either the amount of precipitate or the degree of supersaturation is too small.

Because the intermediate phases also show narrowing regions of stability with decreasing temperature, they might be precipitation strengthened. These possibilities are limited by the smallness of the degree of supersaturation and the inability to achieve supersaturation with respect to a suitable structure for homogeneous nucleation. The number of commercially important alloys based on intermediate phases is very limited, and no case comes to mind where the properties are modified through precipitation.

The Ni-Al system (Fig. 4) shows too little solubility for the Al-rich phase, and for the Ni-rich end the usefulness is rather marginal because the temperature dependence of the solubility is low.

Asymmetric Systems

In our analysis of binary systems so far we have assumed that the heat of mixing was more or less symmetric. In some important cases the heat of mixing is decidedly asymmetric, and this can have important consequences. The most important case, the Cu-Al system, is shown in Fig. 5 where we see that the heat of mixing is rather more negative on the Cu-rich end than on the Al-rich end. In the first place the Al-rich solution can become supersaturated relative to all the intermediate phases, and the one that forms first, Cu_3Al , most resembles the aluminum lattice. Supersaturation with respect to this phase requires a temperature somewhat less than that corresponding to Fig. 5. Further, the asymmetry leads to the rapid changes in the solubility in the Al-rich end, a feature that is in large measure responsible for the utility of

ORNL-DWG 76-12527

Fig. 5. Partial Phase Diagram and the Free Energy Curves for the Al-Cu System. Because of the strong asymmetry in the free energy of the fcc α phase it is possible to obtain supersaturation with respect to all intermediate phases in the Al-base solid solution. The initial precipitate (GP zones) results from either spinodal decomposition within the metastable miscibility gap or the formation of α' , an ordered phase akin to Cu_3Au .

the system. The degree of supersaturation in the Cu-rich end is much lower, and this, coupled with the low temperatures, makes precipitation unimportant in this end of the system. The Ni-Al system also shows similar asymmetry, but in that case the solubility of Ni in Al is too low to be of use.

The Al-Ag system is similar to the Cu-Al system. The Al-rich solution readily forms an Ag-rich precipitate, which is ordered below 190°C. After longer times an hcp phase forms; it is still coherent and retains useful levels of strengthening. Precipitation hardening is not known in the Ag-rich end of the system. The Al-Zn system is also asymmetric, but here the fcc Zn-rich precipitate is disordered and the strength levels are rather low.

In summary, the reasons why binary systems produce such a small fraction of useful precipitation systems is related to the following:

1. the lack of a suitable precipitate phase,
2. the inability to obtain the supersaturation required to precipitate the desired phase,
3. too little precipitate,
4. the smallness of the elastic strains and the absence of order, which do not provide useful strength increases.

These factors appear to be of similar importance.

HIGHER ORDER SYSTEMS

When we go to higher order systems a number of things can happen that considerably increase the possibilities for precipitation hardening. The following are known to be important.

The addition of a third element can drastically increase the extent of a miscibility gap; it can create a gap even when none existed in the binaries.⁵ This happens, for example, when the heat of mixing is rather

⁵J. L. Meijering, "Segregation in Regular Ternary Solutions, I," *Philips Res. Rep.* 5: 333 (1950); "... , II," *Philips Res. Rep.* 6: 183 (1951).

negative for one binary but ideal for the other two. One phase will consist of the solution having the negative heat of mixing and the other phase will be rich in the third component. A negative heat of mixing is associated with short-range order or compound formation, thus possibly giving enhanced strengthening. As we have seen, this happens in the binary case only for asymmetric systems; asymmetric binaries are not required in systems of three or more components.

The situation is complex, but in general the coherency strains are less in higher order systems.⁶ There are always possible sets of tie lines connecting phases of identical lattice parameter for which the coherency strains are zero. Further, the tie lines for coherent phases will tend to rotate from their unstrained equilibrium positions so as to reduce the coherency energy. This effect is impossible in binary systems.

The maximum entropy of mixing increases with the number of components, as shown by Eq. (1). This will increase the temperature dependence of the solubilities.

Precipitate phases that are impossible in the binaries can form. An Al alloy with Mg and Si is an example. Individually, Mg and Si impart limited age hardening capacity, but together they precipitate Mg_2Si , which gives enhanced properties. Ternary compounds are known with no counterpart in the binaries, but they are rather rare. In fact, while commercial alloys almost invariably contain three or more components, only two cases come to mind where ternary compounds are known to be important. These occur in aluminum alloys that contain Mg and Cu or Zn.

Supersaturated solid solutions can decompose by means of cellular or discontinuous precipitation, but properties are not enhanced because the resulting mixture is rather coarse. It has been proposed that large degree of elastic strain energy enhance this process by suppressing the coherent miscibility gap.⁶ Since this coherency energy can be reduced in higher order systems, it should be possible to reduce the importance of cellular precipitation in such cases.

⁶R. O. Williams, "Stability of the Body Centered Cubic Gamma Phase in the Uranium-Zirconium-Niobium System," *Journal of Nuclear Materials* (in press).

It is beyond the scope of this report to try to systematize the possibilities for obtaining useful precipitation in higher order systems apart from the above comments. It is useful, however, to consider specific systems and see how the above ideas apply.

As we have already indicated, aging in Ni-base alloys containing only Al is marginal, whereas the response in related commercial alloys is more substantial. This is a result of the increased entropy of mixing, at least in part.

While limited age hardening is possible in both the Cu-Fe and the Cu-Co systems, the addition of Ni to either system provides very extensive regions where spinodal decomposition can occur, a direct result of the higher mutual solubility of Ni with Fe, Cu, and Co. The coherency stress decreases with Ni content. Further, some compositions can be further strengthened by the formation of the ordered phases Ni_3Fe and Ni_3Co . The addition of Au to Cu-Ag alloys is very similar, and, in fact, there are many such ternary systems, all of which can be identified from the respective binary phase diagrams.

As further examples of these ideas, we wish to consider two practices in the glass industry. In making glass it is of utmost importance that no crystalline phase forms during cooling. The presence of a number of components in the liquid normally lowers the activity of each oxide, reducing the tendency to form crystalline phases. Further, it is interesting that in the case of oxides there are many ternary compounds (that is, structures containing three oxides), so little may be gained by going from a binary to a ternary composition, but one reduces the potential to form crystalline phases as one adds more components. Many factors go into determining the various glass compositions, but so far as we are aware, it has not been recognized that simply having many components will increase the stability of the glass relative to the crystalline phases.

Certain glasses develop interesting optical properties by the precipitation of small amounts of a crystalline phase, such as Cu or Au. The glass is made with a small, soluble addition of Au or Cu, cooled to a low temperature to nucleate the metallic phase, and then aged at an

elevated temperature to grow the particles to the size to interact strongly with light. That the component has a sparingly small solubility means that the heat of solution is so high that the high degrees of supersaturation required for nucleation can be obtained. Perhaps almost any material that has a small solubility could be similarly used.

A basic tenet of this report is that thermodynamic calculations provide a powerful approach in developing useful precipitation systems in higher order systems. One example is the calculation of the solubility of metastable phases. Such phases are often of concern during the early stages of precipitation, and experimental methods have a limited capacity to address this question. We have done thermodynamic calculations on the bcc phase in the U-Zr-Nb system and were rather more successful than previous investigators in understanding the complex transformations that occur in this system.⁶ In particular, we demonstrated that the accepted phase diagram is highly suspect. Our calculations included the effect of coherency. In a study on the Cu-Au system,⁷ we have shown the considerable care and effort that can be required to properly understand what is taking place. For this system the coherency and interfacial energies were responsible for the observed two-phase field being only one-fourth as wide as the true equilibrium field.

The accepted way to predict the occurrence of the sigma phase in superalloys is the Phacomp method.⁸ The electron-to-atom ratio of the matrix is calculated, and when it corresponds to positions to the left of Fe in the periodic table, sigma can form. Although this method is simple and useful, it cannot in principle be exact. In contrast, a calculation based upon the thermodynamics is in principle exact, although its implementation requires that we have adequate thermodynamic information. But having this information one can calculate the amount of the sigma phase as a function of temperature. Recently the problem

⁷R. O. Williams, unpublished study of Cu-Au system.

⁸C. T. Sims, *The Superalloys*, ed. by C. T. Sims and W. C. Hagel, Wiley-Interscience, New York, 1972.

has been reformulated in terms of a pseudoternary phase diagram.⁹ While this has been shown to give superior results compared with the Phacomp method it cannot be exact in principle.

In a way it is unfortunate that the Phacomp method works as well as it does because this leads some people to believe that it has some theoretical basis. It does not.

Recently Dreshfield and Wallace¹⁰ carried out an extensive experimental program to determine the amount of Ni₃Al in various superalloy compositions with six components and rationalize the results in terms of an equation. We do not question that such phase boundaries can be adequately represented analytically, but the selection of an adequate expression is difficult. Such expressions are also difficult to use. A representation based on thermodynamics will provide the phase equilibria, including metastable equilibria, as well as the activities of the components and the driving force for decomposition.

We are by no means suggesting that alloy design can be totally based upon thermodynamics. In the first place, the calculations will seldom be of sufficient accuracy. But what one expects is that the calculations will help to select changes in composition to modify the solubility, the coherency strains, and possibly the degree of order and to eliminate unwanted phases.

In this connection certain comments are warranted concerning precipitation. One may use any number of measurements to monitor the early stages of precipitation, also known as zone formation, preprecipitation, or spinodal decomposition, but the identification of the new phase remains difficult. Electron microscopy is of limited use because of the smallness of the initial precipitate particles. There must be changes in the diffuse x-ray scattering, but they may be too weak to detect, and in

⁹E. S. Machlin and J. Shao, "SIGMA-SAFE: a Phase Diagram Approach to the Sigma Phase Problem in Ni Base Superalloys," *Metall. Trans.* 9A: 561-68 (1978).

¹⁰R. L. Dreshfield and J. F. Wallace, "The Gamma-Gamma Prime Region of the Ni-Al-Cr-Ti-W-Mo System at 850°C," *Metall. Trans.* 5: 71-78 (1974).

any case their interpretation may well prove difficult. Nevertheless, x-ray diffraction remains the more definitive tool. An adequate thermodynamic representation can be a powerful tool. The precipitated phase may be classified as:

- A. an equilibrium phase,
- B. an equilibrium phase for the system but metastable at the given composition,
- C. a structure not found in the equilibrium system.

As we have already indicated, the precipitate is expected to be coherent in the early stages, and there will be an attendant compositional modification, but to refer to a coherent phase as a transition phase is, we believe, poor nomenclature. To call structures B or C transition structures can be misleading because, although with time these give way to structure A, there need be no structural relation between the successive stages. A case in point is the Al-Ag system, in which the first stage consists of the Ag-rich fcc phase being replaced by a coherent hcp phase, which presumably nucleates independently on dislocation loops. The structure that forms first is likely to be the one that most resembles the matrix and thus can nucleate most readily; this accounts for the occurrence of structures B and C. A reasonable number of structures of class B are known but for class C the situation is understandably less well documented because the structure cannot be studied in the bulk. The precipitation of Cr from Cu is believed to be class C since we doubt that the initial precipitate is bcc, but the actual structure and its composition have not been established.

To show how incorrect conclusions can survive a long time, consider the case of Al containing Cu. It is still generally believed that the initial stage consists of the formation of thin plates of pure copper to be followed by ordering. Yet in 1952 Meijering¹¹ concluded that the free energy curve is, as we have shown it in Fig. 5, such that the initial precipitate is a solid solution containing about 25% Al atoms. The precipitate is observed to order ideally as Cu_3Al . This order may exist

J. L. Meijering, *Rev. Metall.* (Paris) 49: 906 (1952).

in the initial precipitate but, if so, the smallness of the domains within each plate prevents its detection.

SUMMARY AND CONCLUSIONS

1. Only a small fraction of the binary systems possess the particular combinations of thermodynamics and structures that lead to useful precipitation hardening. In spite of the fact that much work on binary phases diagrams remains to be done, we expect few additional useful systems to emerge. This work has provided information about what kinds of systems should show promise.

2. While precipitation alloys containing three or more components have been developed, we believe that a vast potential still exists in this area. The difficulty is that without some guidance the scope of the problem can be overwhelming.

3. The thermodynamics of alloys is sufficiently well developed to provide this guidance. As one progresses to more complex systems, one must depend increasingly on thermodynamics if one is to retain any comprehension of what is happening.

4. We have shown that having many components in oxide melts contributes to the ease of retaining the melt as a glass. Should one want to form a dilute precipitate, one picks a substance of such sparing solubility that high degrees of supersaturation can be obtained.

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