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Short Notes

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Liquid-Solid Equilibrium Phase Diagrams for III-V Ternary Solid Solutions with Miscibility Gap

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The existence of miscibility gaps (MG) in III-V ternary and quaternary solid solutions is a general feature, as is known from the calculated unstable regions (UR) bounded by spinodal curves.¹⁾ However, its relationship to liquid-solid equilibrium phase diagrams has not been clearly elucidated. This note shows a calculated liquid-solid equilibrium phase diagram in the MG region, taking ternary $\text{In}_{1-x}\text{Ga}_x\text{As}$ as an example.

Liquid-solid equilibrium for III-V ternary systems is described by a pair of equations between solid and liquid component chemical potentials, whose expressions have been given on the basis of the strictly regular solution approximation.²⁾ These equations can be solved for liquidus temperature T_L and equilibrium solid composition x , when the liquid compositions are specified.

The result for the In-Ga-As system in the In-rich region is shown in Fig. 1. Necessary thermochemical data^{2,3)} are listed in Table I. As the Ga and As atomic fraction in the liquid decreases, (iso-concentration) solidus lines become dense, while the liquidus temperature rapidly decreases. The solidus line for $x=0.5$ terminates at $T_L=482^\circ\text{C}$. This temperature is the calculated critical temperature T_C for solid-phase stability, and it is given by $\alpha_{\text{InAs-GaAs}}/2R$, where $\alpha_{\text{InAs-GaAs}}$ is the

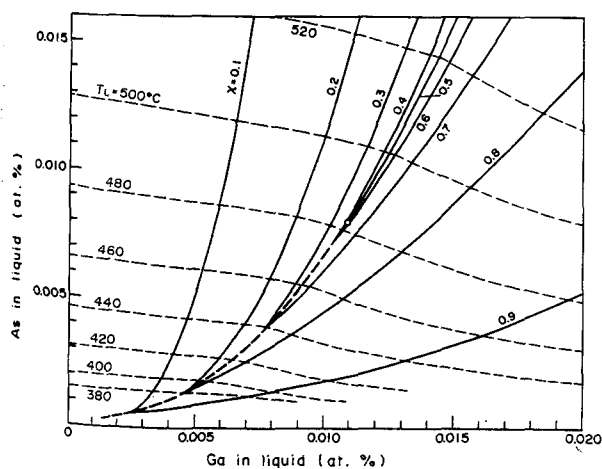


Fig. 1. Calculated liquid-solid equilibrium phase diagram for In-Ga-As system in In-rich region, showing existence of solid miscibility gap. Solid lines and thin broken lines represent iso-concentration solidus and liquidus lines, respectively. Bold broken line designates trace of binodal points. Open circle corresponds to critical point for solid-phase stability.

Table I. Thermochemical data used in the calculation for the In-Ga-As system.²⁾ T^F and ΔS^F are the temperature and entropy of fusion, respectively, α^L is the liquid interaction parameter, and α is the solid interaction parameter.

T_{InAs}^F	1215 K
T_{GaAs}^F	1511 K
ΔS_{InAs}^F	14.52 cal/mole·K
ΔS_{GaAs}^F	16.64 cal/mole·K
$\alpha_{\text{In-As}}^L$	3860-10.07 cal/mole
$\alpha_{\text{Ga-As}}^L$	5160-9.16T cal/mole
$\alpha_{\text{In-Ga}}^L$	1850 cal/mole*
$\alpha_{\text{InAs-GaAs}}$	3000 cal/mole

*Data from ref. 3.

solid interaction parameter and R is the gas constant.⁴⁾ For $T_L < T_C$, other solidus lines successively coincide with their conjugate composition solidus lines, terminating at the coincident point, where the two solid compositions can coexist. It is clear that, once the solidus line for x terminates at $T_L = T$, a solid solution with the composition x cannot exist in equilibrium any more at $T_L < T$, which suggests the existence of an MG.

An MG, if it exists, is defined by a set of binodal points, which are the tangent points of a double-tangent straight line to the free energy curve.⁵⁾ The solid solution component chemical potentials are equal for the set of conjugate compositions on the binodal points. In fact, the terminal point for each solidus line is determined by the binodal points, which are given by⁶⁾

$$RT = \frac{1-2x}{\ln\{(1-x)/x\}} \alpha_{\text{InAs-GaAs}} \quad (1)$$

Note that the MG extent defined by eq. (1) is slightly wider than the UR extent defined by a set of spinodal points, which are given by¹⁾

$$RT = 2(1-x)x\alpha_{\text{InAs-GaAs}} \quad (2)$$

Since the solidus lines are very dense just above the critical point (0.0109 at. % Ga and 0.0081 at. % As), precise control of the solid solution composition will be very difficult for such liquid composition ranges. This fact may be regarded as a precursor effect for the immiscibility.

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