

# The Ni-Pt (Nickel-Platinum) System

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## Equilibrium Diagram

The liquidus and solidus of the assessed Ni-Pt phase diagram (Fig. 1) are based on thermodynamic calculations from this work. The system exhibits a minimum in the liquidus and complete solid solubility at high temperatures. At low temperatures, two ordered phases appear at NiPt and Ni<sub>3</sub>Pt, with critical temperatures of 645 and 580 °C, respectively. The variation of Curie temperature with composition in the Ni-rich solid solution follows an approximately linear relationship:  $T_C = 354.3 - 9.413 X_{Pt}$  (where  $T_C$  is in °C and  $X_{Pt}$  is at.% Pt). The melting points of the elements are taken from [Melt].

### Liquidus/Solidus

No new experimental data have been published since the assessment by [Hansen], and Fig. 1 differs from that of [Hansen] in that a minimum of the liquidus has been established on the basis of a thermodynamic calculation. Data from the thermal analyses, electrical measurements, and optical microscopy work of [33Kur] and X-ray measurements of [38Kus] and [44Esc] were referred to by [Hansen]. The solidification data points of [33Kur] are shown in Fig. 1, and the calculated liquidus and solidus from this work (see

“Thermodynamics” section) are in general agreement with these data. [81Mes] modeled the liquidus and solidus thermodynamically, employing a technique described below (see “Thermodynamics” section), and derived phase boundaries in reasonable agreement with Fig. 1, with a shallow minimum located at around 22.5 at.% Pt and 1437 °C. The data of [33Kur] were somewhat scattered and only extended up to 50 at.% Pt. Consequently, the liquidus and solidus of the assessed diagram are taken from the thermodynamic calculations in this work.

### Solid-State Equilibria

The presence of a superstructure of the AuCu (*tP2*) type in NiPt alloys below 645 °C and around the stoichiometric composition was reported first by [44Esc], on the basis of a series of X-ray and conductimetric measurements. The ordering temperature was reported later by [60Woo] as 603 °C, but [78Ste] used X-ray diffraction and micrography to confirm [44Esc]. Data points from [44Esc] and [78Ste] are shown in Fig. 1, and the phase boundaries are drawn accordingly.

[44Esc] did not observe an ordering reaction at the Ni<sub>3</sub>Pt composition. However, [38Kus] and [49Kus] noted superstructure lines in alloys of 25.4 and 31 at.%

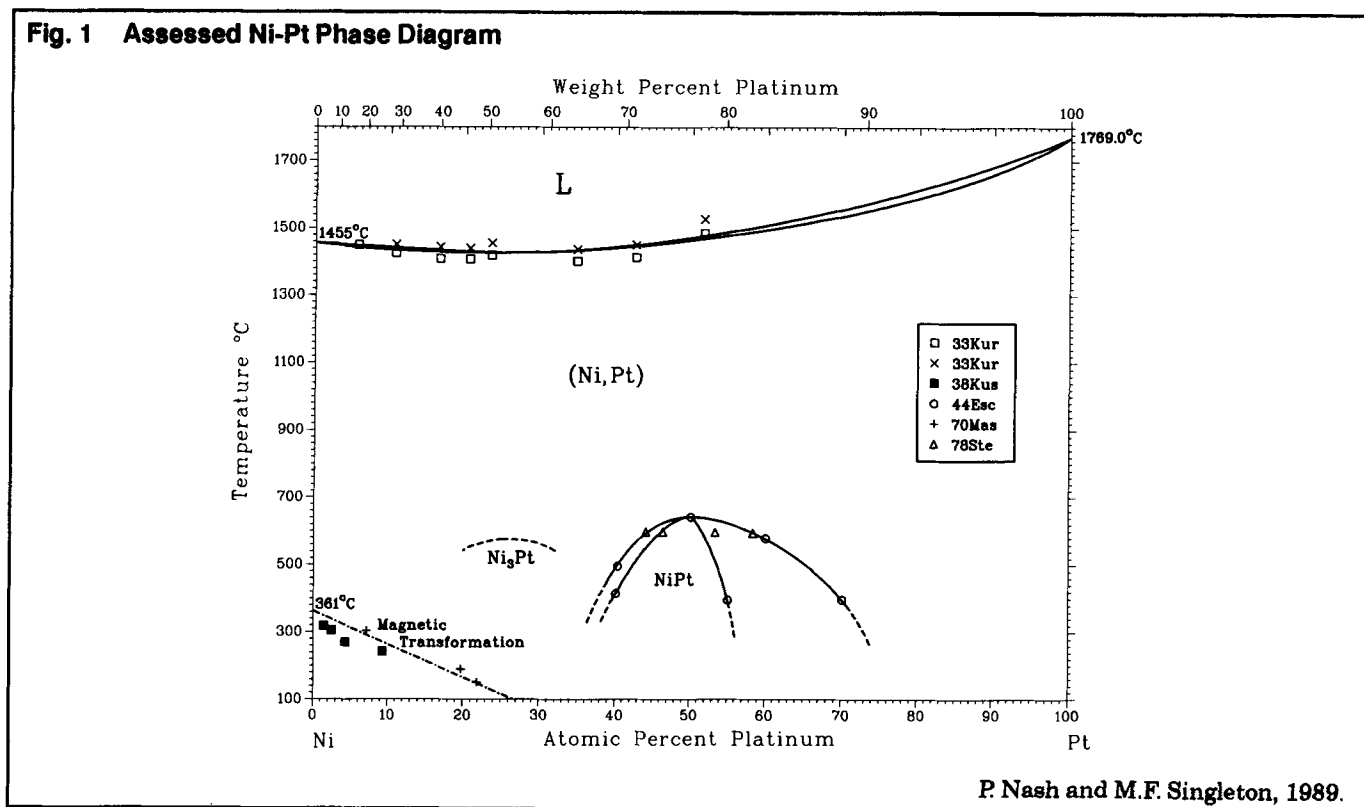


Table 1 Ni-Pt Crystal Structure Data

Phase	Composition, at.% Pt	Pearson symbol	Space group	Strukturbericht designation	Prototype	Reference
Ni.....	0 to 100	<i>cF4</i>	<i>Fm</i> $\bar{3}$ <i>m</i>	A1	Cu	[King1]
Ni <sub>3</sub> Pt.....	~ 18 to 30	<i>cP4</i>	<i>Pm</i> $\bar{3}$ <i>m</i>	L1 <sub>2</sub>	AuCu <sub>3</sub>	[53Ori]
NiPt.....	~ 40 to 55	<i>tP4</i>	<i>P4/mmm</i>	L1 <sub>0</sub>	AuCu	[44Esc]
Pt.....	0 to 100	<i>cF4</i>	<i>Fm</i> $\bar{3}$ <i>m</i>	A1	Cu	[King1]

Table 2 Ni-Pt Lattice Parameter Data

Phase	Composition, at.% Pt	<i>a</i>	Lattice parameters, nm <i>c</i>	<i>c/a</i>	Reference
Ni.....	0	0.35241	...	...	[King1]
Ni <sub>3</sub> Pt.....	25.07	...	...	...	[53Ori]
NiPt.....	50	0.3873	0.3589	0.939	[44Esc]
Pt.....	100	0.29233	...	...	[King1]

Pt, indicating that an ordering reaction occurs between 500 and 620 °C. [51Boz] gave 600 °C as an approximate critical temperature. To obtain a more precise value for the disordering temperature of Ni<sub>3</sub>Pt, [53Ori] measured temperature resistivity curves of a 25.07 at.% Pt alloy before and after a 26-day heat treatment in argon at 475 °C. An inflection in the resulting curves indicated with reasonable accuracy ( $\pm 5$  °C) that the disordering reaction takes place at 580 °C, the value employed in the construction of Fig. 1.

Investigations of the ordering reactions that occur in the Ni-Pt system were undertaken by [53Ori], [65Yam], [72Gre], [72Ive], [76Sil], [78Kat], [78Lak], [78Ste], and [81Kue]. An early X-ray diffraction analysis of ordering in single crystals of NiPt by [65Yam] revealed that in the early stages (at 500 °C), the ordered domains are lamellar in shape and oriented parallel to the {100} planes of the matrix. A similar observation was made by [72Ive].

Short-range order ( $\alpha$ ) was observed in a Ni-2 at.% Pt alloy by field ion microscopy [68Gol]. The observed order came from a deficit in the number of nearest-neighbor Ni atom pairs relative to a random solid solution, and the chemical interaction energy was determined to be  $-3178 \pm 1171$  J/mol.

## Crystal Structures and Lattice Parameters

Little work has been carried out on the crystal structures of phases in the Ni-Pt system. Available data are summarized in Tables 1 and 2.

## Thermodynamics

### fcc Solid Solution, ( $\alpha$ )

Thermodynamic investigations of the Ni-Pt solid solution were undertaken by [65Sch], [67Bor], and

[70Wal]. Those of [65Sch] and [70Wal] were analyzed subsequently by [81Mes]. [67Bor] used local X-ray spectroanalysis to investigate mutual diffusion in the Ni-Pt solid solution and found that in the range 1000 to 1400 °C, the diffusion coefficients of the elements in the alloys are strongly dependent on concentration. The sign of this dependence indicates that the Ni-Pt system exhibits negative deviations from ideality based on the analysis of [48Dar].

[81Mes] selected the thermodynamic data of [70Wal] (calculated from vapor pressure measurements obtained with a torsion-effusion apparatus and liquid-tin solution calorimetry) over the less complete results of [65Sch] as a basis for his treatment of the system. The  $H^{\text{ex}}$  values measured by [70Wal] were relative to ferromagnetic Ni at 298 K as a standard state and referred to ferromagnetic alloys for  $X_{\text{Pt}} \leq 0.3$  (where  $X$  = atom fraction of Pt). However, [70Wal] measured high-temperature activity data relative to paramagnetic standard states. To correlate the two sets of data of [70Wal], it was necessary to convert the enthalpy data to paramagnetic values. This was done by estimating the enthalpy of magnetic disordering from the Heisenberg theory and applying the correction [81Mes].

The corrected  $H^{\text{ex}}$  data were fitted to the Redlich-Kister equation [75Smi] for a series representation:

$$H^{\text{ex}} = x(1-x) \sum_{i=0}^m a_i (1-2x)^i \quad (\text{Eq 1})$$

where  $i$  is the number of parameters required to represent the solution. A regular solution requires only the parameter  $a_0$ , and a random subregular solution, only  $a_0$  and  $a_1$ . [81Mes] employed four enthalpy parameters in his treatment to allow for experimental uncertainty in the measured thermodynamic data of [70Wal]. These parameters are given in Table 3, along with four corresponding entropy parameters ( $b_i$ ) used in the Redlich-Kister equation for  $S^{\text{ex}}$ :

# NI-Pt

**Table 3 Gibbs Energy Parameters for Ni-Pt Phases (see Eq 1 and 2) [81Mes]**

Phase	Quantity	Parameter:			
		0 J/mol	1 J/mol · K	2 J/mol	3 J/mol · K
α(fcc).....	$H^{ex}$	-40 849	-3 960.6	10 917	4 706.9
	$S^{ex}$	-0.98514	0.60967	4.4774	2.9968
L.....	$H^{ex}$	-43 928	-6 237.2	4 974.8	0
	$S^{ex}$	0	0	0	0

**Table 4 Experimental Excess Heats of Mixing of Ni-Pt Alloys Compared with Thermodynamic Model**

Composition, $X_{Pt}$	Excess heats of mixing ( $H^{ex}$ ), J/mol	
	[70Wal, 81Mes]	This work
0.1.....	-3 014	-3 600
0.2.....	-6 153	-6 400
0.3.....	-8 554	-8 400
0.4.....	-9 905	-9 600
0.5.....	-10 036	-10 000
0.6.....	-9 650	-9 600
0.7.....	.....	-8 400
0.8.....	-5 434	-6 400
0.9.....	-3 341	-3 600

**Table 5 Lattice Stability and Interaction Parameters Used in Present Work**

$G^0(Ni,L) = 0$
$G^0(Pt,L) = 0$
$G^0(Ni,fcc) = -17 614 + 10.209 T$
$G^0(Pt,fcc) = -19 651 + 9.623 T$
$G^{ex}(L) = -45 300 X_{Ni} X_{Pt}$
$G^{ex}(fcc) = -40 000 X_{Ni} X_{Pt}$

$$S^{ex} = x(1-x) \sum_{i=0}^n b_i (1-2x)^i \quad (\text{Eq 2})$$

The entropy parameters ( $b_i$ ) in Eq 2 were found by first combining partial molar enthalpies obtained from the corrected calorimetry results of [70Wal] (assuming  $\Delta C_p = 0$ ) with  $G^{ex}(Ni)$  data from the mid-points of the (generally scattered) torsion-effusion results of [70Wal] to obtain values of  $S^{ex}(Ni)$ . These points were then fitted to Eq 2 to yield the parameters ( $b_i$ ).

The deviations from ideality range from slightly positive for  $x \leq 0.3$  to slightly negative for  $x \geq 0.3$ .

Integral excess heats of mixing in the solid solution phase at 1273 K are listed in Table 4 [70Wal, 81Mes]. The activity data of [65Sch] were consistently higher than the predicted values of [81Mes] (as in a similar treatment of the Ni-Pd system). However, the Gibbs energy equation fitted the data of [70Wal] well at the temperature midpoints, except at  $x = 0.1$  and  $0.9$ , where the equation gave activities about 0.03 higher.

### Liquid and α/Liquid Phase Boundaries

[68Alc] measured the activities of Ni in liquid alloys of Pt, using the Knudsen effusion method across a range

of compositions from a mole fraction of Pt equal to zero up to 0.6. Although the data were too sparse [81Mes] to permit reliable evaluation of an excess Gibbs energy equation, it was noted that they showed deviations from Raoult's law on the same order of magnitude as the (α) solid solution data. [77Che] measured the activity of Ni in liquid Pt and observed a positive deviation from ideality. Because this was not in agreement with the phase diagram, the data of [68Alc] are preferred.

[81Mes] estimated the thermodynamics of the liquid from the thermodynamic behavior of the solid and the measured phase relationship (cooling curve data for the liquidus and solidus) of [33Kur]. His approach was first to estimate the position within the (α + L) field of the  $T_0$  vs  $x$  line along which  $G(\alpha) = G(L)$ . Along this line,  $G^{ex}(L)$  is related to  $G^{ex}(fcc)$  by

$$G^{ex}(L)/x(1-x) = G^0_{Ni(fcc)}/x + G^0_{Pt(fcc)}/(1-x) + G^{ex}(fcc)/x(1-x) \quad (\text{Eq 3})$$

where  $\mu_0(i)$  is the standard chemical potential of element  $i$ .

[81Mes] then obtained  $G^{ex}(L)/x(1-x)$  values at regular composition intervals along the line and fitted them, using a multiple regression program, to obtain  $H^{ex}(L)$  and  $S^{ex}(L)$  equations of the type shown in Eq 1 and 2.

The approximate  $G^{ex}(L)$  equation (Eq 3) was then used in conjunction with  $G^{ex}(fcc)$  to back calculate the phase boundaries by means of a computer program that determines the common tangents to the Gibbs energy curves. Comparison of the calculated and measured boundaries generated a new  $T_0$  vs  $x$  line. The process was then repeated until the derived liquid Gibbs energy equation (Eq 3) interaction with the α solid solution equation in such a way as to best fit the measured data of [33Kur]. A three-parameter Gibbs energy equation (see Eq 1 and 2), for the liquid was thus derived, with the parameters given Table 3. The assumption that  $S^{ex}(L) = 0$  was considered reasonable by [81Mes], in view of the near-ideal entropy of the α solid solution.

The derived solidus and liquidus demonstrate reasonable agreement with experiment, with the exception of the liquidus point at  $x = 0.51$ . The measured solidus points reflected considerable undercooling in this system, as observed above, and the calculated solidus generally lies above these points. Predicted activity curves at 1873 K were compared with the

averages of the measured data of [68Alc]. The agreement between them lends weight to the accuracy of the evaluation procedure of [81Mes] and the predicted phase boundaries.

The Ni-Pt system also was modeled in this work using the method of [70Kau]. The present model was obtained by selecting an excess heat of mixing from the data of [70Wal], corrected by [81Mes] for the magnetic transition, and adjusting  $H^{\text{ex}}(\text{L})$  to fit the experimental liquidus/solidus data points. Regular-solution thermodynamics were assumed, because the correction for  $S^{\text{ex}}$  was small [70Wal]. Table 4 compares the calculated  $H^{\text{ex}}$  with those from [70Wal], and Table 5 lists the lattice stability and interaction parameters used in the present model. The lattice stability parameters are based on the melting points [Melt] and the enthalpies of fusion [83Cha] of the pure components. The liquidus and solidus of the assessed diagram (Fig. 1) were calculated from the model derived in this evaluation, and show excellent agreement with the experimental data taking into account the undercooling observed for the measured solidus data.

## Magnetism

The Ni-Pt solid solution exhibits ferromagnetism at Ni-rich compositions and paramagnetism at Pt-rich compositions [74Alb, 74Bei, 74Woh], with the critical concentration for the disappearance of ferromagnetism at approximately 42.5 at.% Ni.

The variation of the Curie temperature with composition was measured by [36Man], [37Mar], [38Kus], and more recently by [70Mas]. [Hansen] noted that alloys between 21 and 31 at.% Pt that were annealed at 450 °C prior to Curie temperature measurement gave values which were 20 to 40 °C lower than those of quenched alloys. This was thought to be due to the formation of the ordered Ni<sub>3</sub>Pt phase. The Curie temperatures of [38Kus], who quenched from 900 °C prior to measurement, were therefore used in the evaluation of [Hansen]. Although [70Mas] annealed their alloys at 1000 °C for 1 h and slow cooled at 300 °C/h prior to Curie temperature measurement, their results were in fair agreement with those of [38Kus] and are included along with [38Kus] in Fig. 1. The variation of Curie temperature with composition (atomic percent, Pt) is represented approximately by  $T_C = 354.3 \text{ °C} - 9.413X_{\text{Pt}}$ .

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\*Indicates key paper.

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## The K-Li (Potassium-Lithium) System

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### Equilibrium Diagram

K-Li is a simple system (Fig. 1) with three phases: (1) pure solid K; (2) pure solid Li; and (3) the liquid, L, which is miscible only at much higher temperatures (>1500 °C) where the vapor pressure becomes appreciable. According to the thermal analyses of [39Boh], Li and K do not alloy and there is no evidence for either intermediate compound formation or solid solutions.

The solubilities of Li and K in the K-rich and Li-rich liquid phases, respectively, have been investigated by [70Dot] and [74Smi]. The crystal structures of the pure metals taken from [83Cha], [King1], and [King2] are given in Table 1. There are no other reported phase equilibria measurements in the K-Li system.

The seven data points of [70Dot] are listed in Table 2. The measurements were obtained by equilibrating 377 g of K with 274 g of Li for over 16 h in a 304SS-type

container maintained in an inert atmosphere. The metals had been purified by filtration, hot trapping, and vacuum distillation. A complete chemical analysis revealed a total impurity content of less than 500 ppm wt.%, including 31 and 6 ppm of oxygen in the Li and the K metals, respectively. The authors stated that their results were in fair agreement with the unpublished data of Tepper at 540 °C (Mine Safety Appliance Research Co., Pennsylvania).

The data of [74Smi] were obtained by equilibrating 15 g of Li with 75 g of K in a Mo crucible held in an inert atmosphere. The Li had been purified by treating it with Zr in a Ti crucible at 800 °C for more than 100 h. The K had been vacuum distilled at 250 °C. Although complete impurity levels were not reported, chemical analyses showed the Li and K to have oxygen concentrations of 62 and 51 ppm, respectively. The results of [74Smi] were reported in a small diagram similar to Fig. 2 and were not published in tabular form.