

MEASUREMENT OF THE LIQUID-CRYSTAL EQUILIBRIUM DIAGRAM FOR HELIUM ISOTOPE SOLUTIONS

I. V. BOGOYAVLENSKIĬ, N. G. BEREZNYAK, and B. N. ESEL'SON

Physico-technical Institute, Academy of Sciences, Ukrainian S.S.R.

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The curves of the beginning and end of solidification of He<sup>3</sup>-He<sup>4</sup> solutions containing 53.6% or 76.5% He<sup>3</sup> are measured in the 1.4-4.0°K temperature range. In this way the width of the stratification region arising upon transition from the liquid to solid state of the solutions can be determined throughout the whole concentration range. The triple points connected with polymorphic transition to the solid state have also been measured for the solutions. An equilibrium diagram for the liquid and solid states of the He<sup>3</sup>-He<sup>4</sup> system is constructed. In the 50-140 atm range, the diagram is of the peritectic type.

IN an earlier paper [1] we plotted the general contours of the liquid-solid diagrams of state of the system of isotopes He<sup>3</sup>-He<sup>4</sup>. The lack of experimental data has made it necessary in many cases to indicate the lines of the diagram only qualitatively, using very general considerations, and made it impossible to determine the form of the diagrams quantitatively. In particular, in the temperature region from 1.4 to 4.2°K, the data

that are sufficiently complete are those for the curves of the start of solidification of solutions of different concentrations, making it possible to draw the "liquidus" line on the state diagram [1-3], whereas information on the width of the stratification region (curves corresponding to the end of the solidification of the solutions), which is necessary for the construction of the "solidus" line, was confined to measurements on a solution with approximately 25% He<sup>3</sup> [1].

In the present investigation we continue the measurements of the curves of the start and end of solidification of solutions of helium isotopes, which yield the width of the stratification region over the entire interval of concentrations. We measured the curves of the start and end of solidification of solutions with molar concentrations 53.6 and 76.5% of He<sup>3</sup> (the concentrations were determined with a mass spectrometer). We also determined the coordinates of the triple points connected with the polymorphic transition in the solid solution in equilibrium with the liquid. The data obtained are shown in Fig. 1 and are listed in the table<sup>1)</sup>.

The instrument used for this purpose differed from that described earlier [1] in having an additional coil to compensate for the thermal variation of the pressure transducer. This was done

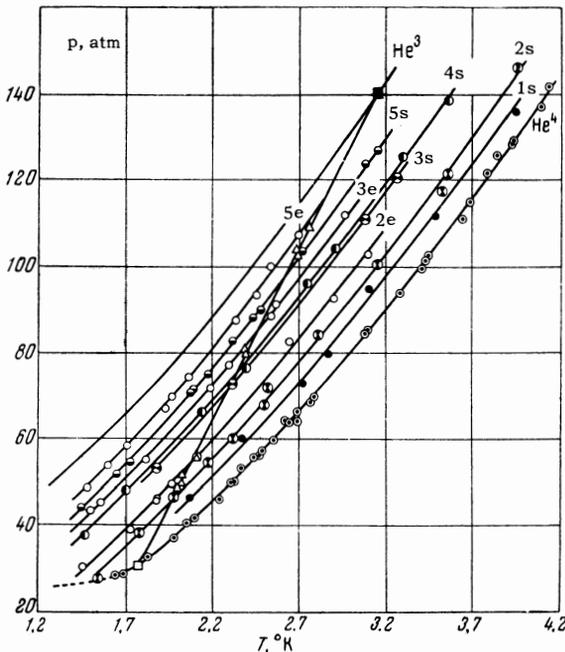


FIG. 1. Curve showing the start (s) and end (e) of solidification of He<sup>3</sup>-He<sup>4</sup> solutions with different He<sup>3</sup> contents: 1 - 11.2%; 2 - 25.6%; 3 - 53.6%; 4 - 54.7%; 5 - 76.5%. He<sup>3</sup> - results of Grilly and Mills<sup>[4]</sup>. Triple points: Δ - present work, □ - [1], ■ - [4].

<sup>1)</sup>As a result of a mass-spectroscopic analysis we have refined the concentrations of the helium isotope mixtures used in previous work.<sup>[4,2]</sup> The corrected data are presented in the table together with the data obtained in this work.

We have also refined the values of the pressure by the calibration of the transducer independently of the data of Swenson<sup>[5]</sup> for the solidification curve of He<sup>4</sup>.

Solidification temperatures and pressures of helium isotope  
with various He<sup>3</sup> contents

T, °K (scale of 1958 <sup>[6]</sup> )	P, atm	T, °K (scale of 1958 <sup>[6]</sup> )	P, atm	T, °K (scale of 1958 <sup>[6]</sup> )	P, atm	T, °K (scale of 1958 <sup>[6]</sup> )	P, atm
He <sup>4</sup>		11.2% He <sup>3</sup> , start of solidification		2.904	92.6	1.440	44.0
1.633	28.0			3.096	102.9	1.642	51.4
1.680+	28.2			54.7% He <sup>3</sup> , start of solidification		1.721	54.1
1.819	32.3	2.059	46.2			2.067	70.6
1.970	36.8	2.360	60.0			2.083+	71.4
2.045	40.1	2.719	72.7	1.460	37.5	2.167	75.0
2.091	41.3	2.866	79.7	1.695	47.9	2.313+	82.7
2.233+	45.9	3.104	94.9	2.133	66.1	2.434	88.1
2.301	49.5	3.488	111.7	2.394	76.3	2.471	90.0
2.320	49.7	3.949	135.8	2.747	96.2	2.709	103.5
2.361	52.9			2.912	104.2	3.082	123.9
2.434+	55.4	25.6% He <sup>3</sup> , start of solidification		3.300	125.8	3.157	126.9
2.463	55.9			3.569	138.5	76.5% He <sup>3</sup> , end of solidification	
2.483	56.8					1.473	48.5
2.548+	59.7	1.534	27.4	53.6% He <sup>3</sup> , start of solidification		1.591	53.6
2.619	64.1	1.769	37.8			1.701	58.2
2.640	63.8	1.974	46.3			1.923	66.7
2.681+	64.0	2.175	54.3	1.872	52.9	1.960	69.6
2.688+	66.1	2.317	59.7	2.314	72.9	2.055	74.2
2.758+	68.5	2.497	67.8	3.080	110.9	2.334	87.5
2.781	69.5	2.512	71.9	3.271	121.0	2.450	93.3
3.076	84.5	2.808	84.3	53.6% He <sup>3</sup> , end of solidification		2.534	100.0
3.091+	85.2	3.154	100.3			2.696	107.2
3.280+	93.8	3.526	117.6	1.490	43.1	Triple points	
3.411	99.8	3.565	121.7	1.552	44.9		
3.425	101.5	3.957	146.3	1.811	54.8	1.997	48.5
3.443	102.5	25.6% He <sup>3</sup> , end of solidification		2.180	71.6	2.007	49.4
3.646+	111.1			2.291	77.1	2.024	51.3
3.688+	115.1	1.444	30.1	2.536	88.6	2.107	55.5
3.789	121.7	1.720	38.7	2.566	91.2	2.386	80.3
3.843	125.8	1.870	45.3	2.964	111.9	2.388	79.7
3.922+	128.4	1.873	45.7	76.5% He <sup>3</sup> , start of solidification		2.684	103.9
3.932	129.2	1.960	49.3			2.693	102.8
4.093	137.2	2.107	55.5			2.758	109.2
4.140+	141.8	2.638	82.5				

by connecting into the bridge circuit, along with the measuring coil of the transducer, an analogous coil wound on a specially provided undeformed part of the calorimeter. If both coils are prepared by an identical technology and if their electric resistances are carefully chosen, there was no unbalance in the measuring bridge as the temperature ranged from 4.2 to 1.3°K, except when the pressure  $p$  inside the calorimeter was changed. The transducer was calibrated against a precision manometer. It must be noted that the gap in which the investigated helium was located was reduced in this calorimeter to 1 mm.

The temperatures and pressures of solidification were registered with equal ease both by determining the kinks on the  $T(t)$  heating (cooling) curves of the calorimeter, and the kinks on the iso-pycnal  $p(T)$  curves. Both methods give the same result, within the limits of experimental error.

The described construction of the instrument made it possible to obtain with sufficient accuracy data for the solidification curve of pure He<sup>4</sup>. These data lie within the range of published values [3-5], but differ somewhat from the data of

Swenson [5]. In this connection, we measured directly the temperature variation of the previously used transducer [1,2]. This made it possible to calculate the previously measured solidification curve He<sup>4</sup> [1,2], independently of Swenson's data [5], which were used in the earlier investigations [1,2] to introduce corrections for the temperature variation of the transducer. The He<sup>4</sup> solidification curves obtained with the aid of both instruments are in good agreement. The control experiments also yielded similar results for the solution with concentration 76.5% He<sup>3</sup> (the values obtained for He<sup>4</sup> and the solution with 76.5% He<sup>3</sup> using the old calorimeter are marked in the table by a plus symbol).

The present earlier results [1,2] have made it possible to plot the equilibrium diagram of the liquid and solid phases of the system He<sup>3</sup>-He<sup>4</sup> in the investigated region of temperatures and pressures, as shown in Fig. 2. In the 50-140 atm pressure interval this diagram is of the peritectic type. The width of the stratification region has a maximum value 0.11° (10-12% concentration) in the region of medium concentrations and is practically independent of the pressure in the interval

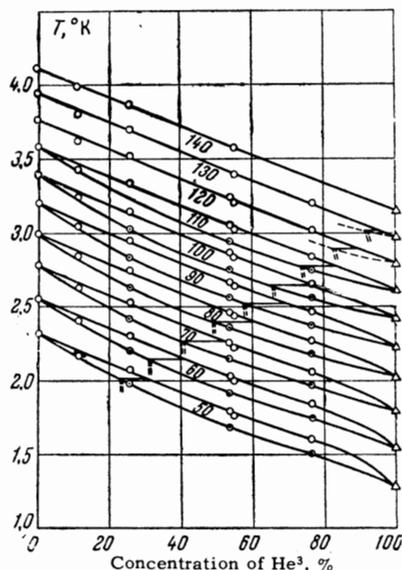


FIG. 2. Equilibrium diagrams of liquid and solid phases of the system of isotopes  $\text{He}^3\text{-He}^4$  (numbers near the curves give the pressures in atmospheres).

from 50 to 110 atm.

It must be noted that in accordance with the thermodynamics of phase transitions, the diagrams should reflect the kinks in the lines of the start of solidification, where the latter cross the peritectic transformation lines. However, these singularities are apparently too small to be noted on the diagrams so far. For the same reason, we cannot plot the details of the intersection of the lines of the end of solidification and the lines of

peritectic transformation, since the region of stratification into two solid phases is probably very narrow and has not yet been determined experimentally. Consequently, the lines of transition to the solid phase are marked dashed in the diagrams.

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