

# The isothermal section of the phase diagram of the La–Ni–Cu ternary system at 673 K

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## Abstract

The isothermal section of the phase diagram of the ternary system La–Ni–Cu has been investigated by X-ray diffraction, differential thermal analysis, optical microscopy and electron microscopy techniques. It consists of 13 single-phase regions, 23 two-phase regions and 11 three-phase regions. At 673 K, the maximum solid solubility of Cu in  $\text{La}_2\text{Ni}_7$ ,  $\text{LaNi}_3$ ,  $\text{La}_7\text{Ni}_{16}$ ,  $\text{La}_2\text{Ni}_3$  and  $\text{LaNi}$  is about 2, 2, 3, 3 and 5 at.% Cu, respectively. The solid solubility of Cu in  $\text{La}_7\text{Ni}_3$  and Ni in  $\text{LaCu}_6$  is too small to observe. Cu and Ni can replace each other in  $\text{LaNi}_5$  and  $\text{LaCu}_5$ , the  $\text{LaNi}_5$  and  $\text{LaCu}_5$  form a continuous solid solution. The maximum solid solubility of Ni in  $\text{LaCu}_2$  and  $\text{LaCu}$  is about 15 and 4 at.% Ni, respectively. A new ternary compound  $\text{La}_{10}\text{Cu}_{85}\text{Ni}_5$  has been observed in the La–Ni–Cu ternary system.

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**Keywords:** Phase diagram; La–Ni–Cu ternary system; X-ray diffraction; Compound  $\text{La}_{10}\text{Cu}_{85}\text{Ni}_5$

## 1. Introduction

The lanthanum–nickel alloys are well established as hydrogen-storage alloys due to their excellent performance. The substitution of a small amount of Cu for Ni in  $\text{LaNi}_5$  alloys can change the property and hydrogen storage ability of  $\text{LaNi}_5$  alloys and decreases the cost of  $\text{LaNi}_5$  alloys and increases the stability of  $\text{LaNi}_5$ . The phase diagrams and thermodynamics are an important basis for the study of the phase transformation of the materials and the materials performance. Up to the present, the isothermal section of the La–Ni–Cu ternary system phase diagram has not been reported. In this paper, we studied the isothermal section of the La–Ni–Cu ternary system at 673 K.

The phase diagrams of the binary La–Ni, Ni–Cu, and La–Cu systems were reported in refs. [1–3], respectively. At 673 K, there are eight intermetallic compounds in the La–Ni system, namely  $\text{La}_3\text{Ni}$ ,  $\text{La}_7\text{Ni}_3$ ,  $\text{LaNi}$ ,  $\text{La}_2\text{Ni}_3$ ,  $\text{LaNi}_2$ ,  $\text{LaNi}_3$ ,  $\text{La}_2\text{Ni}_7$  and  $\text{LaNi}_5$ . There is no intermetallic compounds in the Ni–Cu system. There are four intermetallic compounds in the La–Cu system, namely  $\text{LaCu}$ ,  $\text{LaCu}_2$ ,  $\text{LaCu}_5$  and  $\text{LaCu}_6$ .

## 2. Experimental

In this work, the purities of lanthanum, nickel, and copper were 99.8, 99.99, and 99.9%, respectively. One hundred and fifty-two alloy buttons have been produced. Each sample was prepared with a total weight of 3 g. These alloy buttons were prepared in an argon atmosphere in a vacuum arc furnace or in a high-frequency induction furnace. Each arc-cast button had been melted two times and turned around after melting for better homogeneity.

After melting, the sample buttons were sealed in evacuated quartz tubes for homogenization heat treatment. The homogenization temperatures of the binary and ternary samples were chosen according to the phase diagrams of the La–Ni, Ni–Cu, and La–Cu systems and the results of differential thermal analysis (DTA) of some ternary alloys. The La-rich alloys containing more than 50 at.% La were homogenized at 693 K for 400 h. The Cu-rich alloys containing more than 85 at.% Cu were homogenized at 873 K for 300 h. The others sample buttons were homogenized at 773 K for 300 h. Subsequently, they all were cooled at a rate of 9 K/h to 673 K and kept at this temperature for 400 h, then quenched into liquid nitrogen.

X-ray powder diffraction analysis was mainly used in the present investigation. EPMA and DTA were carried out to check the results of X-ray diffraction analysis when needed.

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Samples for X-ray diffraction analysis were powdered and analyzed on a Rigaku D/Max 2500 V diffractometer with Cu K $\alpha$  radiation and graphite monochromator operated at 40 kV, 250 mA ( $20^\circ \leq 2\theta \leq 60^\circ$ ). The Materials Data software Jade 5.0 [6] and Powder Diffraction File (PDF release 2001) were used for phase analysis. By all these means, the phase relationships in the La–Ni–Cu ternary system were determined.

### 3. Results and discussion

#### 3.1. Binary system

##### 3.1.1. La–Ni system

According to Pan and Nash [1], eight kinds of La–Ni compounds, La<sub>3</sub>Ni, La<sub>7</sub>Ni<sub>3</sub>, LaNi, La<sub>2</sub>Ni<sub>3</sub>, LaNi<sub>2</sub>, LaNi<sub>3</sub>, La<sub>2</sub>Ni<sub>7</sub> and LaNi<sub>5</sub>, have been observed in the La–Ni binary system. But, according to refs. [4,8], La<sub>3</sub>Ni does not exist, and LaNi<sub>2</sub> is rather La<sub>7</sub>Ni<sub>16</sub>. In our work, our result obtained agrees with ref. [4].

##### 3.1.2. Cu–Ni system

The Cu–Ni binary system was reported in ref. [2], there is no compounds in this system. Cu and Ni can replace each other; they form a continuous solid solution phase (Cu–Ni).

##### 3.1.3. La–Cu system

In ref. [5], six compounds, LaCu, LaCu<sub>2</sub>, LaCu<sub>4</sub>, LaCu<sub>5</sub>, LaCu<sub>6</sub> and LaCu<sub>13</sub>, were reported. LaCu<sub>13</sub> is a high temperature phase. Under our present experimental conditions, we did not find the LaCu<sub>4</sub> and LaCu<sub>13</sub> at 673 K in the isothermal section of the phase diagram of the La–Ni–Cu ternary system, and the result obtained agrees with Chakrabarti and Laughlin [3].

#### 3.2. Ternary compound

Up to present, we have not found any reports about a ternary compound in the La–Ni–Cu system. But, a new ternary compound in the La–Ni–Cu ternary system was observed in our present experiment. By tentative analysis, its molecular formula is about La<sub>10</sub>Cu<sub>85</sub>Ni<sub>5</sub>. Its crystal structural is face centered cubic. Its space group is *Fm3c*; its lattice constant *a* is equal to 1.158 nm. The details of the new phase will be studied in another article.

#### 3.3. Solid solubility

At 673 K, the maximum solid solubility of Ni in LaCu and LaCu<sub>2</sub> is about 4, 15 at.%, respectively. The maximum solubility of Cu in La<sub>2</sub>Ni<sub>7</sub>, LaNi<sub>3</sub>, La<sub>7</sub>Ni<sub>16</sub>, La<sub>2</sub>Ni<sub>3</sub> and LaNi is about 2, 2, 3, 3 and 5 at.% Cu, respectively. Cu and Ni can replace each other in LaNi<sub>5</sub> and LaCu<sub>5</sub>, LaNi<sub>5</sub> and LaCu<sub>5</sub> form a single-phase region LaNi<sub>5–x</sub>Cu<sub>x</sub> ( $0 \leq x \leq 5$ ), they are a continuous solid

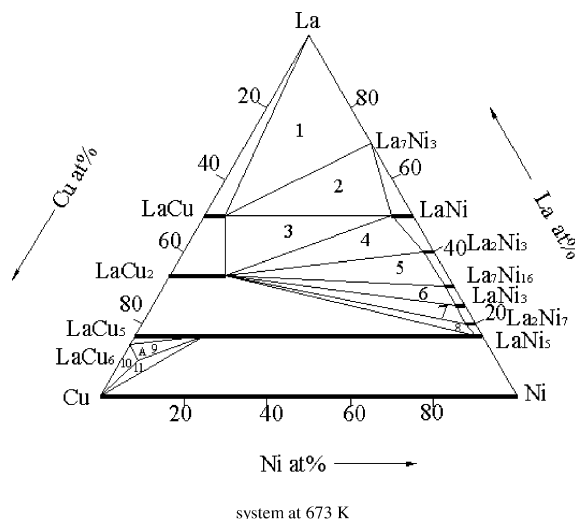


Fig. 1. The isothermal section of the phase diagram of the La–Ni–Cu ternary system at 673 K.

solution. The result obtained agrees with [7]. The solid solubility of other compounds is too small to be observed.

#### 3.4. Isothermal section

By comparing and analyzing the X-ray diffraction patterns of 152 samples, combined with metallography, DTA and EPMA, and identifying the phases presented in each sample, the 673 K isothermal section of the phase diagram of the La–Ni–Cu ternary system was determined (shown in Fig. 1). The isothermal section consists of 13 single-phase regions, 23 two-phase regions and 11 three-phase regions. Details of the three-phase regions are given in Fig. 1.

The 13 single-phase regions are: A (La<sub>10</sub>Cu<sub>85</sub>Ni<sub>5</sub>), B (La), C (Cu–Ni), D (LaCu<sub>5</sub>·LaNi<sub>5</sub>), E (La<sub>2</sub>Ni<sub>7</sub>), F (LaNi<sub>3</sub>), G (La<sub>7</sub>Ni<sub>16</sub>), H (La<sub>2</sub>Ni<sub>3</sub>), I (LaNi), J (La<sub>7</sub>Ni<sub>3</sub>), K (LaCu), L (LaCu<sub>2</sub>) and M (LaCu<sub>6</sub>).

The 23 two-phase regions are: C + D, D + E, E + F, F + G, G + H, H + I, I + J, J + B, B + K, K + L, L + D, D + M, M + C, C + A, A + M, A + D, L + E, L + F, L + G, L + H, L + I, K + I and K + J.

The 11 three-phase regions are: 1: B + J + K, 2: K + J + I, 3: L + K + I, 4: L + I + H, 5: L + H + G, 6: L + G + F, 7: L + F + E, 8: L + E + D, 9: A + M + D, 10: A + M + C and 11: A + C + D.

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