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Phase formation regularities in nanometer powders of Al-Cu-X ternary alloys prepared by gas evaporation process ①

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[Abstract] The phase formation regularities in nanometer powders of ternary alloys prepared by gas evaporation process with induction current as the heating source were investigated. Al-Cu-Mn and Al-Cu-Cr were chosen as the samples in this study. The experimental results show that the necessary conditions for forming ternary compound phases in a ternary alloy nanometer powder are that they must be able to form in a conventional bulk alloy. To obtain a ternary compound phase in nanometer powders, composition of the master alloy must be in a certain range. Changing the composition of the master alloy and evaporation temperature can control the kinds of the phases and their relative amount in the alloy nanometer powders. In present study, AlMn, Al_8Mn_5 and $AlCu_2Mn$ phases formed in as-prepared nanometer powders of Al-Cu-Mn alloy, and the highest relative amount of $AlCu_2Mn$ phase is about 99%. In nanometer powders of Al-Cu-Cr alloy, Cu_9Al_4, Al_2Cu_3, $AlCr_2$ and $Al_{13}Cr_2$ phases formed without any ternary compound phase.

[Key words] nanometer powder; gas evaporation; alloy powder
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1 INTRODUCTION

Nanometer alloy powders are expected to possess novel properties, which can not be obtained in pure metal nanometer powders and conventional bulk materials[1,2]. Many investigations on the preparation and properties of nanometer alloy powders have been carried out. Several technologies have been developed to prepare alloy and composite nanometer powders, such as gas evaporation process in inert gas with reduced pressure using induction electron current as heating source, liquid chemical reduction process, plasma evaporation technology[3~5], and many research achievements have been reported[1,6~11].

It is of significance to study the phase formation regularities in nanometer alloy powders for controlling the phases in the powders. In present study, ternary alloys of Al-Cu-Mn and Al-Cu-Cr are chosen to investigate the formation regularity of the compound phases in nanometer powders.

2 EXPERIMENTAL

The experimental apparatus was the same as mentioned in a previous paper[10]. Al-Cu-Mn master alloys were evaporated at 1673 K in reduced Ar atmosphere with purity of 99.995% in an alumna crucible with dimensions of d 110 mm × 100 mm. The purity of Al, Cu, Mn, and Cr were about 99.95%. Temperature of the melt in the crucible was examined by an optical pyrometer and controlled by adjusting the input power of the device. The argon gas was supplied from the bottom of the evaporation chamber. A deactivating oxidation treatment was applied to the as-prepared powders before being exposed to air. Pressure of the gas atmosphere and temperature fluctuation of the melt didn't arise during the evaporation procedure.

The kinds of the phase and morphology of the particles were examined by X-ray diffractometry (XRD) and transmission electron microscopy (TEM). The relative amount of each phase was calculated by computer software. Lattice parameter (a_0) of Al phase in the as-prepared nanometer powders were examined by X-ray diffractometry.

3 RESULTS

3.1 Effects of Cu on phase formation regularity

To obtain the AlCuMn ternary compound phase in as-prepared powders, a master alloy with the composition of $Al_{79.5}Cu_{15}Mn_{5.5}$ (mole fraction, %) were firstly evaporated at 1673 K and 1773 K respectively in an argon atmosphere of 1.0×10^3 Pa to identify the proper composition range of the master alloy. The X-ray diffraction patterns of as-prepared powders is shown in Fig. 1. The results show that only binary Al-Mn compound phase formed in the powders, with-

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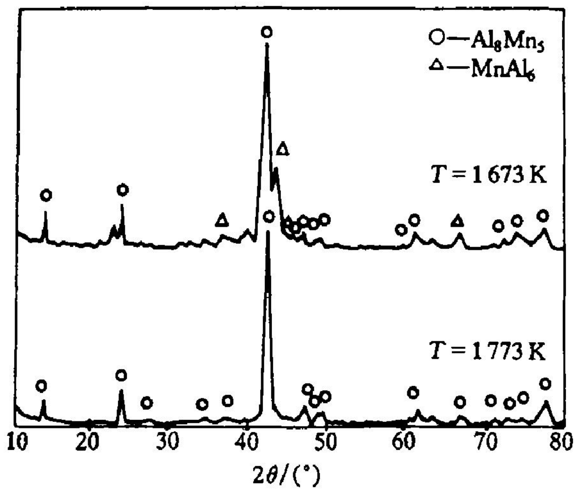

Fig. 1 X-ray diffraction patterns of $Al_{79.5}Cu_{15}Mn_{5.5}$ alloy powders

out any ternary AlCuMn compound phase. Referring to the result given in Ref. [10], it can be concluded that the reason why AlCuMn ternary compound phase did not form in the powders is that the amount of Cu element in the master alloy is not high enough.

A series of master alloys with the composition of $Al_{97-x}Cu_xMn_3$(x = 45, 50, 58, 65) were respectively evaporated at 1 473 K in an argon atmosphere of 1.0×10^2 Pa. X-ray diffraction patterns of the as-prepared powders are shown in Fig. 2.

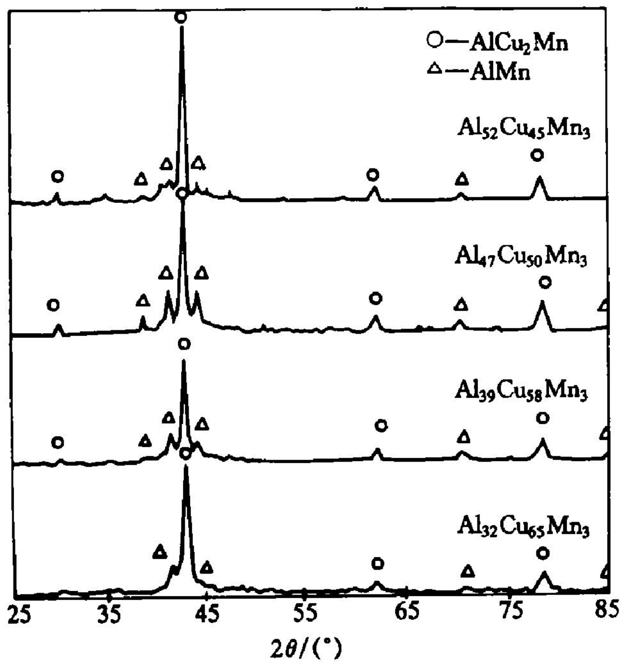

Fig. 2 X-ray diffraction patterns of $Al_{97-x}Cu_xMn_3$ alloy powders
(T = 1 473 K, p_{Ar} = 1.0×10^2 Pa)

As identified in Fig. 2, the resultant powders are composed of $AlCu_2Mn$ and AlMn phases whose relative amounts vary, which can be judged by the change of diffraction peaks intensity of each phase, with the composition of the master alloys. With increasing Cu content in the master alloys, the relative amount of $AlCu_2Mn$ phase varies from 95% to 87% by the semi-quantity calculation of computer software.

3.2 Effect of Mn on phase formation regularity

Several master alloys with the composition of $Al_{35-x}Cu_{65}Mn_x$(x = 3, 5, 7) were respectively evaporated at 1 673 K in the argon atmosphere of 1.0×10^2Pa to examine the effect of Mn on the formation regularities of the phases in as-prepared nanometer powders. The X-ray diffraction patterns of the powders are shown in Fig. 3.

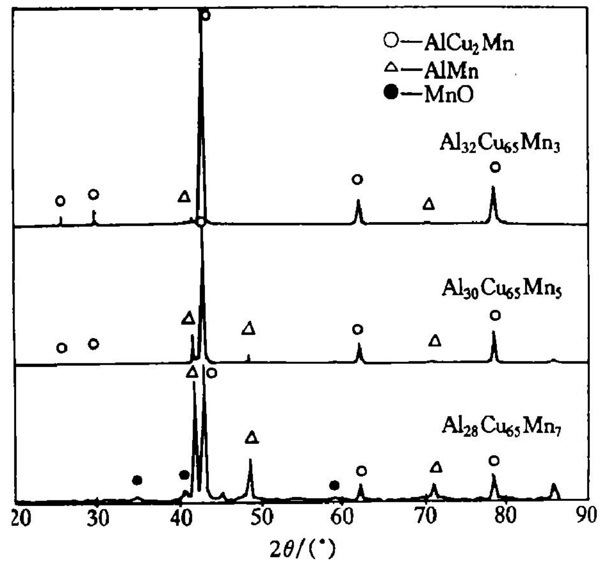

Fig. 3 X-ray diffraction patterns of $Al_{35-x}Cu_{65}Mn_x$ alloy powders
(T = 1 673 K, p_{Ar} = 1.0×10^2 Pa)

As identified in Fig. 3, $AlCu_2Mn$ and AlMn phases occurred in the powders when the amount of Mn element in the master alloys ranged from 3% to 5% (mole fraction). When the amount of Mn was up to 7%, pure Mn phase formed. Because metal Mn tends to oxidate in air, the resultant phase is MnO. The relative amounts of $AlCu_2Mn$ and AlMn phases vary with increasing Mn content in the master alloys. For $Al_{32}Cu_{65}Mn_3$ master alloy, the relative amounts of $AlCu_2Mn$ and AlMn phases are 98.3% and 1.7% respectively; for $Al_{30}Cu_{65}Mn_5$ alloy, they are 91.8% and 8.2%; but for $Al_{28}Cu_{65}Mn_7$ alloy, they are 67% and 33%. It is clear that increasing Mn content in the master alloys helps the AlMn phase and pure Mn to form.

3.3 Influence of evaporation temperature on phases

A series of master alloys with the composition of $Al_{97-x}Cu_xMn_3$(x = 45, 50, 58, 65) were respectively evaporated at 1 673 K in the argon atmosphere of 1.0×10^2 Pa. The X-ray diffraction patterns of as-prepared powders are shown in Fig. 4.

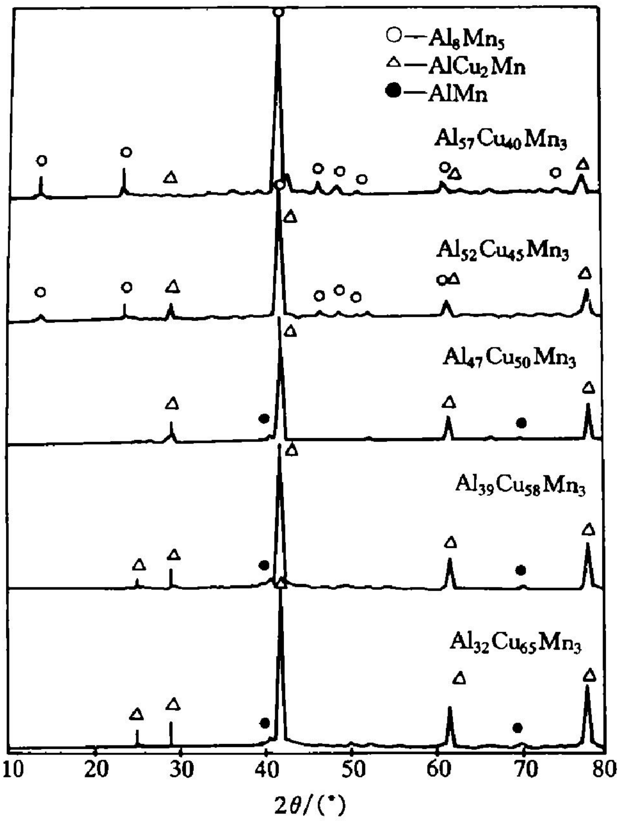

Fig. 4 X-ray diffraction patterns of $Al_{35-x}Cu_{65}Mn_x$ alloys powders (T = 1 673 K, p_{Ar} = 1.0×10^2 Pa)

As identified in Fig. 4, when the evaporation temperature is elevated to 1 673 K, the phase formation regularities are different from the results shown in Fig. 2. When the master alloy of $Al_{97-x}Cu_xMn_3$ is evaporated, the main phase in the resulting powder is Al_8Mn_5 with a little amount of $AlCu_2Mn$. With increasing Cu content in the master alloy, the relative amount of $AlCu_2Mn$ phase gradually increases. When Cu content in the master alloy is up to 50%, the main phase in the as-prepared powder is $AlCu_2Mn$. When Cu content in the master alloy ranges from 50% to 65%, the relative amount of phase $AlCu_2Mn$ is between 98% and 99%. Referring to the results shown in Fig. 2, it can be concluded that the relative amount of $AlCu_2Mn$ phase can be increased by elevating evaporation temperature. Under certain evaporation conditions, a nanometer powder with single phase of $AlCu_2Mn$ may be obtained.

4 PARTICLE MORPHOLOGY

The TEM morphologies of two kinds of alloy powder particles are shown in Fig. 5. The particle size is 20 ~ 50 nm in spherical and near spherical shape. Referring to the results shown in Fig. 2, it is obvious that the particles with less amount of AlMn phase have regular spherical shape without inhomogeneous diffraction contrast. The existence of AlMn phase in particles leads to irregular shape and the oc-

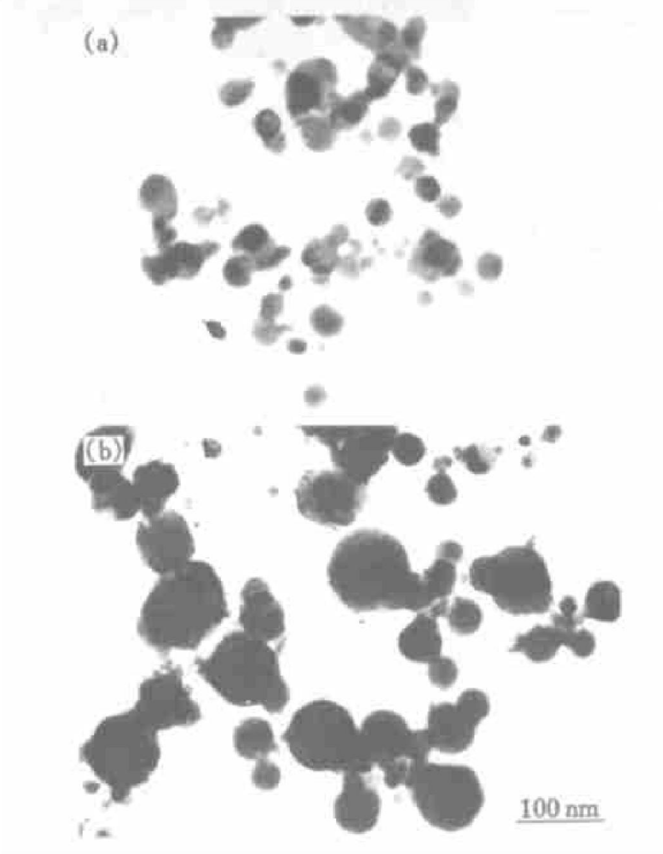

Fig. 5 TEM micrograph of nanometer Al-Cu-Mn alloy powders

currence of inhomogeneous diffraction contrast.

5 DISCUSSION

During the evaporation procedure of a ternary alloy, the composition of a mixed vapor of the metals is controlled by the saturation vapor pressure of each metal at given evaporation temperature and the interaction among various atoms in the master alloy. The composition can be calculated if the activities of the components in master alloys are known. The formation mechanism of alloy nanometer powders with ternary compound phases is similar to that of nanometer powders of binary alloys which include nucleation and growth procedures[1]. But these procedures in the mixed vapor of a ternary alloy are much complicate than that of a binary alloy. A serials of complex reactions may occur in the following several ways: formation of the pure metal, binary or ternary nucleus, reactions among various kinds of nucleuses; growing of these nucleuses or particles by absorbing metal vapor atoms in the mixed vapor and so on. It is nearly impossible to make it clear how the compound phase forms during the evaporation procedure, which is mainly a dynamics problems. But the basic formation regularity of alloy phases in nanometer powders can be qualitatively concluded from the experimental results.

For Al-Cu-Mn ternary alloy, Al atom can compound with Cu atom and Mn atom to form compound phases, and Cu atom and Mn atom can form solid-solution phase according to the binary phase diagrams of Al-Cu, Al-Mn and Cu-Mn alloys[12]. Referring to the formation regularity of the phases in a binary alloy, it can be concluded that if ternary compound phases can form in conventional bulk alloys, they can form in nanometer powders of corresponding ternary master alloy, too. In present study, the formation of $AlCu_2Mn$ phase can confirm it.

To further confirm the conclusion, another ternary alloy Al-Cu-Cr was chosen as a sample. The master alloys with two kinds of compositions were respectively evaporated at 1773 K in Ar gas at 1.0×10^2 Pa. The X-ray diffraction patterns are shown in Fig. 6.

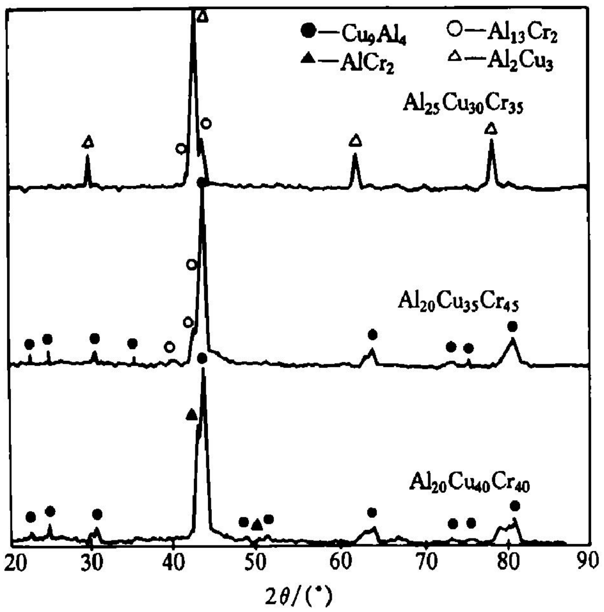

Fig. 6 X-ray diffraction patterns of nanometer Al-Cu-Cr alloy powders
($T = 1673$ K, $p_{Ar} = 1.0 \times 10^2$ Pa)

As identified in Fig. 6, AlCuCr ternary compound phase did not form in as-prepared nanometer powder; but Al-Cu and Al-Cr binary compound phases formed. For Al-Cu-Cr alloy, Al atom can compound with Cu atom and Cr atom to form compound phases, but Cu atom and Cr atom can not form solid solute phases or compound phase according to the phase diagrams of Al-Cu, Al-Cr, Cu-Cr alloys[12]. This result coincides with the above results.

6 CONCLUSIONS

1) The necessary conditions for forming ternary compound phases in a ternary alloy nanometer powder are that they must be able to form in the conventional bulk alloy.

2) To obtain a ternary compound phase in nanometer powders, composition of the master alloy must be in a certain range. Changing the composition of the master alloys and the evaporation temperature can control the kinds of the phases and their relative amount in as-prepared alloy nanometer powders.

3) AlMn, Al_8Mn_5 and $AlCu_2Mn$ phases formed in the nanometer powders of Al-Cu-Mn alloy, and the highest relative amount of $AlCu_2Mn$ phase was about 99%. In nanometer powders of Al-Cu-Cr alloy, Cu_9Al_4, Al_2Cu_3, $AlCr_2$ and $Al_{13}Cr_2$ phases formed; but no any ternary compound phase formed.

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