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Study of the influence of silver addition on the order-disorder transformations in Cu-Au system

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Abstract

The order disorder transition takes a very important place because of its influence on the physical and mechanical characteristics of metals. The copper-gold system provides a classical example of structure which has been classified as long period superlattice. The addition of silver in Au-Cu binary system leads to the appearance of ordered regions and two-phase decomposition region. Thus, the ternary Au-Ag-Cu system undergoes different transformations such as ordering, precipitation and spinodal decomposition depending on the composition of alloy. In this work we study the effect of addition of Ag on order-disorder transformation in Au-Cu system. We have choose the following alloys: Au-25 wt. % Cu and Au-50 wt. % Cu as reference alloys and Au-18 wt. % Ag-7 wt. % Cu and Au-35 wt. % Ag-15 wt. % Cu, To characterize the structural evolution, we have used dilatometry and differential scanning calorimetry (DSC). The obtained results enable us to show that the Ag addition leads to the decrease of ordering temperature and the apparition of other transition in comparison with the reference alloys.

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Keywords: *order disorder transition, Au-Ag-Cu system, superlattice, dilatometry, differential scanning calorimetry.*

1. Introduction

The ternary Au-Ag-Cu alloys represent one of the most important systems studied extensively in various domain of research, even as nano-structured materials. This system is composed of relatively noble metals whose properties are well known, it displays a variety of diffusional phase transformations including compositional demixing and ordering [1], because it is characterized by ordered regions and a two phase decomposition region. Thus, the hardening mechanism of the ternary Au-Ag-Cu alloys is

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thought to be due to phase transformation such as ordering, precipitation and spinodal decomposition [2-7], depending on the composition of the alloy and on the ageing temperature [8].

The ternary phase diagram of the gold-copper-silver alloys [9] displays complete solid solubility at temperatures above about 700 °C and exhibits a miscibility gap for compositions near the center of the isothermal phase diagram [1, 9]. In this region, the solid-state decomposition of α (solid solution) → α_1+ α_2 occurs, where α_1 (silver-rich phase) and α_2 (copper-rich phase) are two solid solution fcc phases with different compositions [1, 9, 10]. The α_2 phase may undergo further ordering reactions with decreasing temperature, depending on its composition.

In the present paper different phase transitions are studied in Au-18wt. % Ag-7wt. % Cu and Au-35wt. % Ag-15wt. % Cu alloys which have respectively the following phases: α_1+Cu_3Au and α_1+$AuCu_I$ at 300°C and α solid solution at 775°C according to the ternary equilibrium diagram.

The different phase transitions in these ternary alloys are compared with those of the two known binary alloys; Au-50wt. % Cu and Auy-25wt. % Cu (used as reference) to deduce the effect of the presence of Ag addition.

2. Experiment

The alloys used in the present study were prepared by fusion using very pure materials: Au-18wt. % Ag-7wt. % Cu and Au-35wt. % Ag-15wt. % Cu. Dimensions of cylindrical specimens for dilatometric analysis and differential scanning calorimetric (DSC) testing are respectively; 20 mm x5 mm and 5mm x 5mm corresponding to a weight of approximately 200 mg. the studied alloys were homogenized at 775 °C for 1h under vacuum and then quenched into water to obtain a supersaturated solid solution.

Differential scanning calorimetric and dilatometric analyses were made to obtain general information on the structural transformations. Thus, the homogenized specimens were heated from room temperature to 665 °C and 775 °C (v= 5 °C/min) under Argon atmosphere, using respectively SETARAM DSC 131 and ADAMEL LHOMERGY dilatometer.

3. Results and discussion

DSC results:

Fig. 1(a) and (b) shows the DSC scans obtained for heated 1h at 775 °C and quenched in water Au-35wt. % Ag-15wt. % Cu and Au-18wt. % Ag-7wt. % Cu alloys. One can see during the heating of the first alloy three peaks:

- a first endothermic peak with a maximum at 127 °C, linked to the formation of $AuCu_3$ ordered phase,
- a second endothermic peak more important than the first one, related probably to the precipitation of Ag-rich α_1 phase in addition to the already present $AuCu_3$ phase,
- an endothermic peak in the temperature interval [515,628 °C], with a minimum situated at 598 °C due to the transformation of $AuCu_3$ + Ag-rich α_1 phases to α solid solution.

For Au-18wt. % Ag-7wt. % Cu alloy, the heating segment shows two exothermic peaks and one endothermic peak. The first exothermic peak situated at 133 °C is linked to the AuCu I ordered phase formation, the split endothermic peak is noted between 223 °C and 344 °C with two minima situated at 265 ° C and 303 °C corresponding respectively to the presence of two phases: the AuCu II and Ag-rich α_1. The second exothermic peak situated at 527 °C, is related to the transformation of: AuCuII + Ag-rich α_1→ α solid solution.

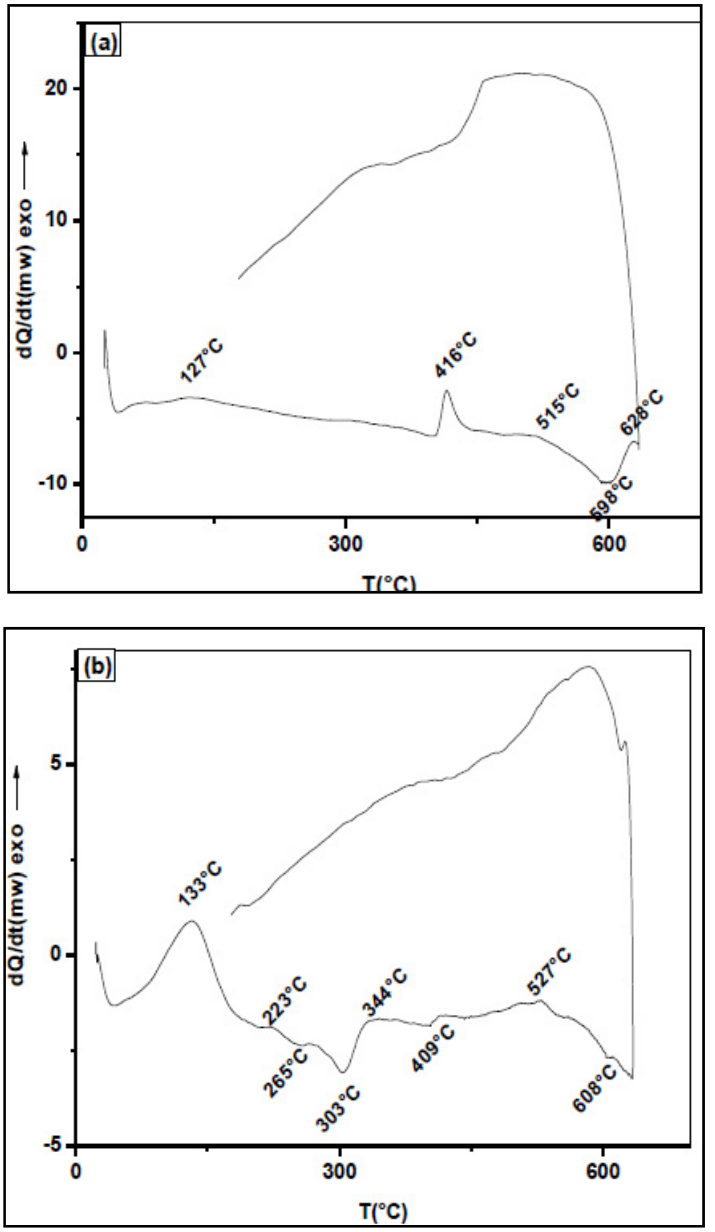

Fig. 1. DSC curves of heated 1h at 775 °C and water quenched Au-35 wt. % Ag-15 wt. % Cu (a) and Au-18 wt. % Ag-7 wt. % Cu (b) alloys.

Dilatometry results

Dilatometric curve of Au-35wt. % Ag-15wt. % Cu alloy (Fig. 2(a)) presents an important anomaly during heating from room temperature to 775 °C, which is composed of:
-a first contraction in the temperature interval [43-232 °C] due to the formation of $AuCu_3$ ordered phase from α solid solution,

-a first expansion with maximum of derivative curve situated at 403 °C, related to the precipitation of Ag-rich α_1.
-a split second contraction with two minima of derivative curve at 519 °C and 577 °C, probably due to the dissolution of $AuCu_3$ and Ag-rich α_1 phases respectively.
-a second expansion corresponding to the transformation of $AuCu_3$ and Ag-rich α_1 phases to α solid solution.

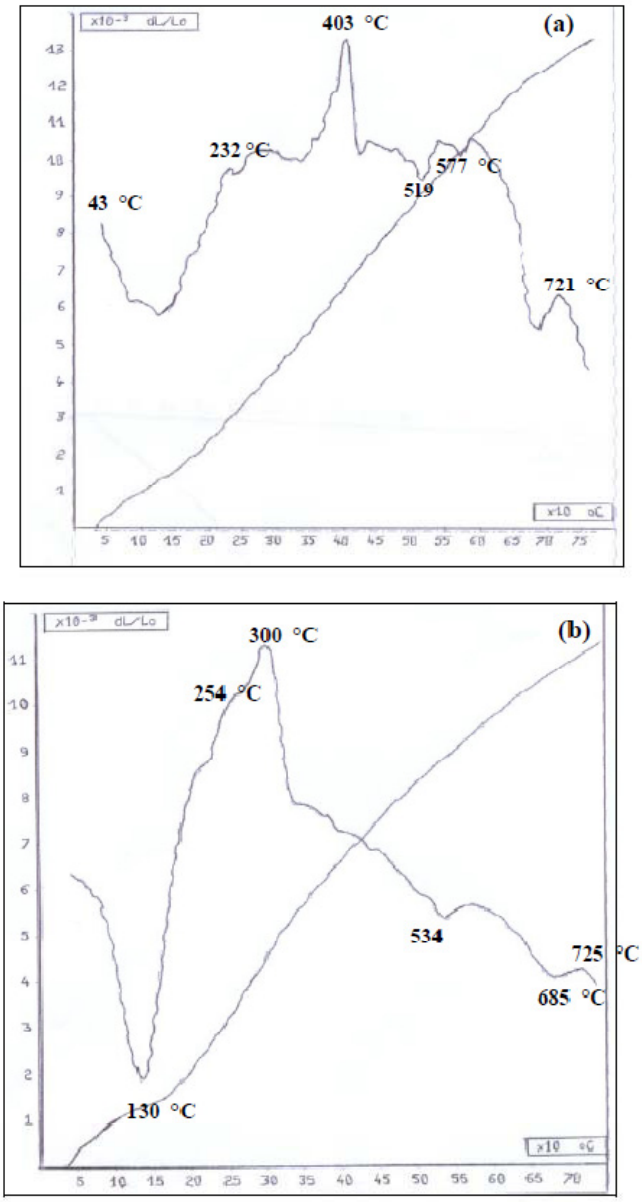

Fig. 2. Dilatometric curves of heated 1h at 775 °C and water quenched Au-35 wt. % Ag-15 wt. % Cu (a) and Au-18 wt. % Ag-7 wt. % Cu (b) alloys.

For Au-18wt. % Ag-7wt. % Cu alloy, heating segment (Fig. 2(b)), shows the existence of:
- a first important contraction situated at 130 °C related to the AuCuI phase formation,
- a split expansion with two maxima, the first one at 254 °C and the second one at 300 °C, corresponding respectively to the presence of AuCuII semi-ordered and A-rich α_1 phases.
- two other successive contraction situated at 534 °C and 685 °C linked to the AuCuII + α_1-Ag rich phases dissolution respectively, followed by an expansion corresponding to the transformation of AuCuII + α_1-Ag rich phases to α solid solution.

4. Synthesis of obtained results

The comparison between the DSC curves of the two studied alloys: Au-35wt. % Ag-15wt. % Cu and Au-18wt. % Ag-7wt. % Cu and the those of reference alloys: Au-50wt. % Cu and Au-25wt. % Cu, shows that the temperature corresponding to $AuCu_3$ and AuCuI ordered phases formation in the alloys with Ag addiiton is lower than that found by Hamana et *al.* [11, 12] in reference alloys. This behaviour is observed also on dilatometric curves where the temperature of the first contraction corresponding always to the formation of the ordered phases $AuCu_3$ and AuCuI of Au-35wt. % Ag-15wt. % Cu and Au-18wt. % Ag-7wt. % Cu becomes lower than that of reference alloys.

5. Conclusion

The obtained results suggest that the addition of silver atoms to the AuCu system provokes the diminution of ordering temperature. This behaviour is interpreted by the effect of silver presence which decreases the ordering energy. Thus the silver substitution in the gold lattice reduces linearly the ordering energy (the atomic radius of silver and gold are nearly the same).

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