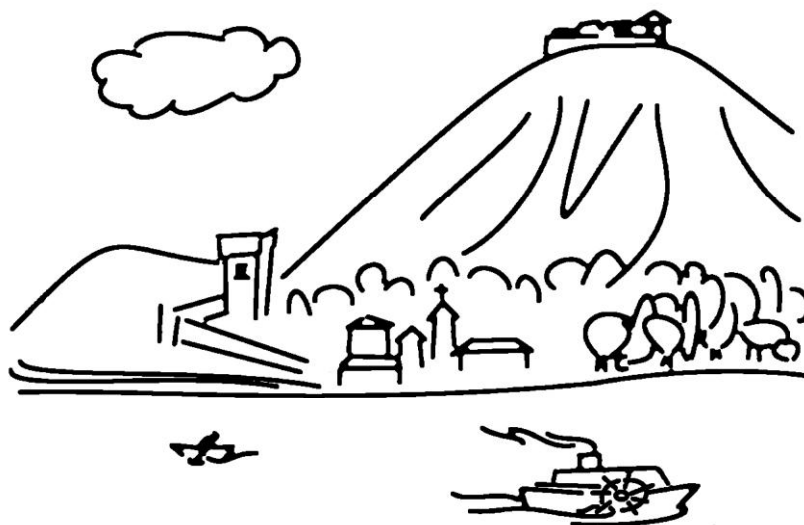
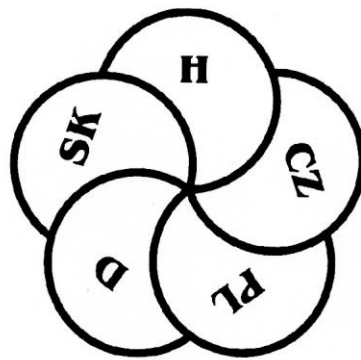




BUDAPEST UNIVERSITY OF TECHNOLOGY AND ECONOMICS

30th International Colloquium on
Advanced Manufacturing
and Repairing Technologies
in Vehicle Industry



22-24 MAY 2013, VISEGRÁD, HUNGARY

**30th International Colloquium on Advanced Manufacturing
and Repairing Technologies in Vehicle Industry,
22-24 May 2013, Visegrád, Hungary**

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Wetting Ability of Silver-Gold (Ag-Au) Alloys on Graphite Substrate

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The wettability of graphite by the silver-gold (Ag-Au) liquid alloy was measured using the sessile drop method at a temperature interval of 1273 – 1473 K. The measured alloys are in the solid solution range. The system is found poorly wetting with the contact angle at an interval of 112 ... 136 degrees. Typical alloying effects are found, as the Ag host metal is gradually replaced by Au alloying element. The results of the contact angle measurements were compared to the change of electrical resistance.

Keywords: wetting; contact angle; graphite; Ag-Au liquid alloy

1. INTRODUCTION

The significance of Ag-based alloys is important in brazing and welding processes [1]. The wetting phenomena between the filling alloy melt (brazing alloy) and the base material have a central role in obtaining sufficient quality, which serve the formation of steady joint between the parts. Alloying elements are also often used for this purpose in order to lower the melting point and also for increasing the wettability between the parts to be fixed. Nowadays the solders contain little amount of gold, because a few percent gold can increase the quality of the joint.

In earlier papers, where the theme of the investigation was the wetting angles of alloys with Ag host and Cd, In, Sn, and Sb solvent metals a systematic change determined in the solid solution range [2]. The electrical resistance shows similar changes with the wetting angles of the alloys in the solid solution range. The Ag-Au system creates solid solution on the whole concentration range. The melting point of the silver and gold are in the measuring range of the resistance-heated tube furnace and the materials susceptibility to oxidation are low. In the Mendeleev periodic table the silver and gold are in the same column, so the e/a (electron/atom) ratio is the same. The atomic diameter of silver and gold are almost the same ~ 2.88 Å and ~ 2.92 Å [3]. Both element have the same fcc crystal structure and there is no big difference between the electronegativities. Therefore the material pair meets the Hume-Rothery rules. The connection between the electrical resistance and the wetting properties can be verifiable in this system.

2. EXPERIMENTS

The investigated alloys were prepared from high purity (4N) Ag and Au, using induction melting in cold quartz crucible under inert (Ar) atmosphere. The graphite substrate was made from high purity, porosity free base material. The substrates were mechanically polished to $R_a=0.23\pm0.005$ µm, $r=1.009\pm0.0005$. Surface roughness was determined using a 3D laser profilometer (Rodenstock RM600 surface topography measurement system) [4].

Wetting experiments were performed in a sessile drop equipment [4], taking the methodological issues summarized by Sobczak [5] into account. The graphite substrate and the alloy were positioned into the furnace at ambient conditions. The pressure was then reduced to 0.1 Pa at room temperature in the chamber. The vacuum was replaced by a 10^5 Pa 99.999 % Ar gas. This procedure was repeated 3 times. Following this, the temperature was raised to 1000°C at a rate of about 4°C/s. Since only a small portion of the gas chamber is heated, the pressure around the droplet remains at about 10^5 Pa during the measurements. Measurements were made at 1000, 1050, 1100, 1150 and 1200°C.

The time dependence of the contact angle was reported in earlier paper [6]. Based on this, measurements after 2 minutes of holding time were used in this paper.

Contact angle values of the drop were measured directly from its images, processed by self-made automatic evaluation software in the Matlab environment. Although the uncertainty of this software was below 1 degree, the total uncertainty of the measured values was $\pm 3^\circ$.

After the last measurement at 1200°C, the furnace was switched off and was left to cool to ambient temperature. This took about 40 minutes. After that, the furnace was dismantled and the sample was removed. The alloy sample was weighed before and after each experiment. The average weight loss was below 0.5 % for pure Ag, Au and for all Ag-Au alloys.

The Ag-Au alloy compositions create homogeneous solid solutions on the whole concentration range. The measured alloy compositions were listed in Table 1.

Tab.1. Examined alloy compositions

Alloy composition	Liquidus temperature of the alloy [°C]
Ag	961.93
Ag ₉₇ Au ₃	984.69
Ag ₉₅ Au ₅	991.22
Ag ₉₀ Au ₁₀	1009.18
Ag ₈₀ Au ₂₀	1040.82
Au	1064.43

3. RESULTS

In the whole concentration range the melts solidify in the form of single phase solid solutions, with fcc crystal structure.

The wetting angles are illustrated as a function of the experimentally measured temperature. The Fig.2. shows that the wetting angles are rising while the temperature was increased. The shapes of the curves are still similar in every case. It is also remarkable, that the slope of each curve (except for the Ag₉₇Au₃) lower compared to the pure Ag.

The slope of $\Theta(T)$ for the pure Ag is positive on graphite. This indicates the decreasing wetting ability of graphite versus the temperature, i.e. the Ag/graphite interphase formation is energetically unfavorable, as the temperature increases (the interfacial energy increases) [7]. The influence of alloying element amount on $\Theta(T)$ is the similar to the previous results of wettability measurements of Ag based solid solutions.

As the phase-relation is very simple in this binary alloy system (single phase nature of liquids and the solid solutions), one can suppose that the alloying induced by the electron concentration has a detectable impact not only in the stabilization of liquid state (melting point suppression) but also in the surface energy of the liquid drop. The magnitude of the wetting angle depends only indirectly on the total surface energy, which is composed from the drop/gas and the drop/substrate interface components [8].

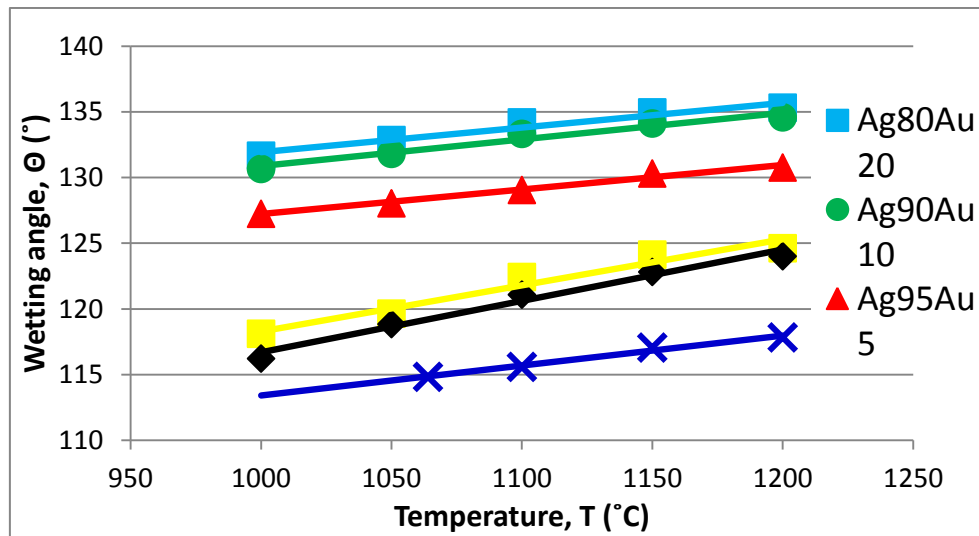


Fig.2. The temperature dependence of the wetting angle of pure Ag, Au and Ag alloyed with 3, 5, 10, 20 at.% of Au on graphite substrate

Fig 3. shows the electrical resistance changes of the Ag-Au alloy system. The changes follow through a typical tub curve. The pure materials have much better electrical conduction properties than the alloys, especially the medium alloyed ones. Correlation can be found between the wetting angle changes and the electrical conduction changes.

When the amount of alloying elements are increased the difference between the pure material and the alloy is bigger in both properties.

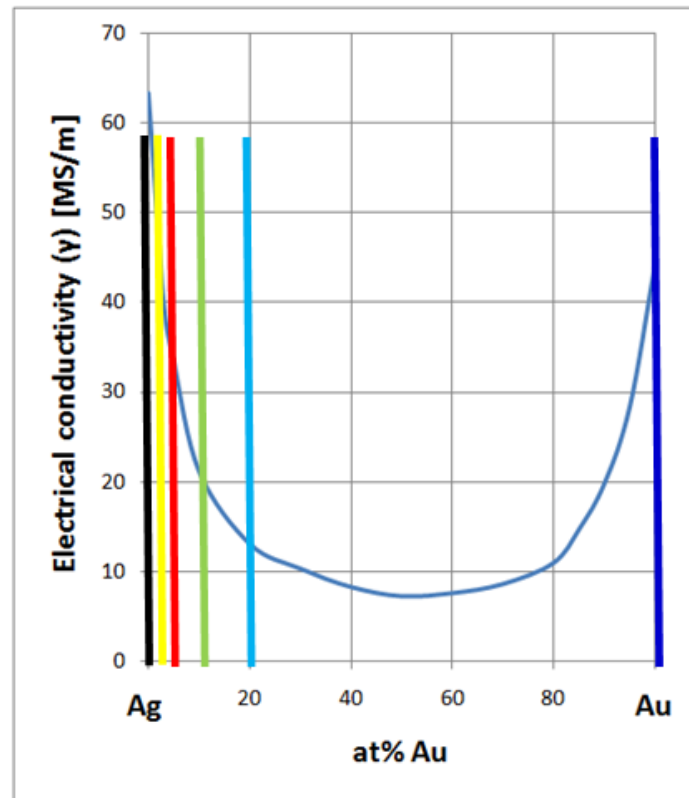


Fig.3. Electrical conduction of Ag-Au alloys [9]

The ratio of the electrical conduction and wetting angle changes are shown in Fig.4. The ratio were calculated from the difference between the pure Ag and the alloy compositions and correlated to the biggest difference. The rate of change of the electrical conduction and wetting angle changes seems to be similar in the $\text{Ag}_{95}\text{Au}_5$, $\text{Ag}_{90}\text{Au}_{10}$, $\text{Ag}_{80}\text{Au}_{20}$ samples. The results of $\text{Ag}_{97}\text{Au}_3$ show bigger difference. The change of the wetting angle is smaller than the change of the electrical conduction. The alloying effect is less relevant for the wetting angle in the case of $\text{Ag}_{97}\text{Au}_3$ alloy composition.

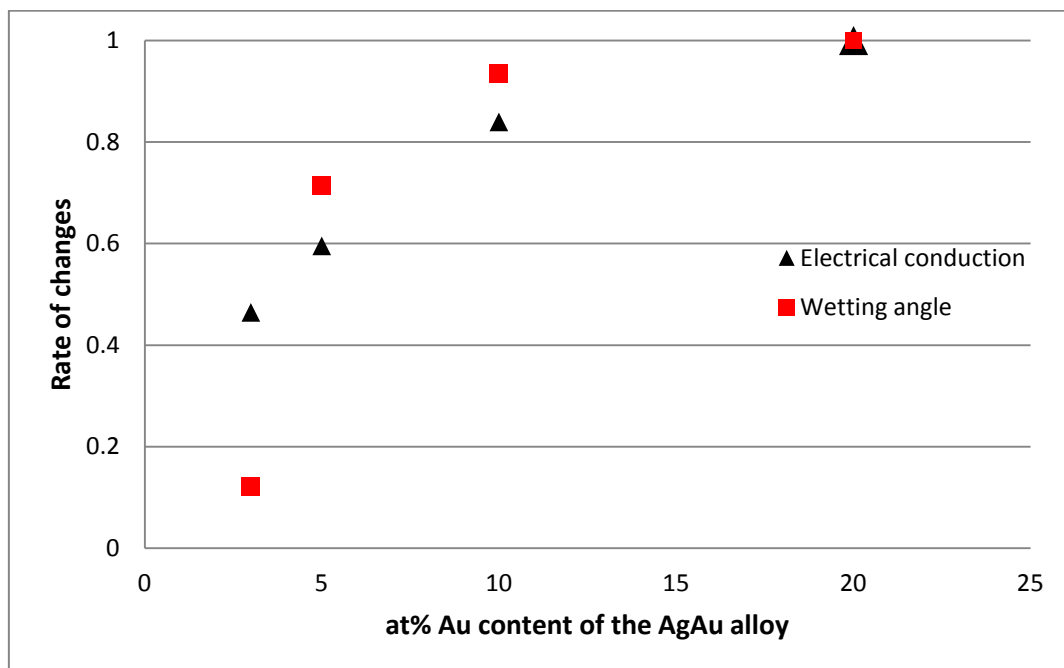


Fig.4. Property changes of the investigated alloys

4. CONCLUSION

The temperature dependence of wetting angle is measured on graphite for Ag-Au melts. In this system the wetting angle changes of the solid solutions are followed the rules that determined in earlier papers [2]. The increase of the alloying element in the alloy composition, increase the wetting angles. Connection determined between the changes of wetting angle and electrical conduction. The shifts of wetting angles are the same in the electrical conduction.

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