

## STUDIES ON THE THERMIONIC VACUUM ARC DISCHARGES IN THE VAPORS OF Cu-Ag AND Cu-Sn ALLOYS

T. Akan<sup>\*</sup>, N. Ekem, S. Pat, R. Vladioiu<sup>a</sup>, G. Musa<sup>b</sup>

Department of Physics, Osmangazi University, Eskisehir, Turkey

<sup>a</sup>Department of Physics, Ovidius University, Constanta, Romania

<sup>b</sup>National Institute for Physics of Laser, Plasma and Radiation, Bucharest, Romania

The thermionic vacuum arc (TVA) discharges were studied in the vapors of various metals (Zn, Pb, Ag, Sn, Cu, Au, Co, Ti) and Cu-Ag and Cu-Sn alloys. Cu-Ag and Cu-Sn alloys in various mass ratios were prepared by using the TVA, and the TVA discharges were generated in the vapors of these alloys. The volt-ampere characteristics of the TVA discharges generated in the vapors of these alloys were investigated with respect to the ratio of Ag in the Cu-Ag alloy and Sn in the Cu-Sn alloy. Cu-Ag alloy thin films with various mass ratios were deposited onto the glass substrates by using TVA discharges. The ratios of Cu and Ag in the thin Cu-Ag alloy films were found using the SEM-EDX microanalyses. The breakdown voltages of the TVA discharges depend on the boiling temperature of the material used to produce metal vapors.

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### 1. Introduction

Copper and its alloys have a huge variety of uses that reflect their versatile physical, mechanical, and chemical properties. The properties of alloys depend on the chemical composition and impurities which are related to the used technology. Alloys are usually made by conventional melting and powder processing routes. Several rapid solidification processing methods, such as atomization, melt spinning, spray forming, etc. have been examined in recent years [1-3].

TVA is a plasma source generating pure metal vapor plasma and consists of a heated cathode emitting thermoelectrons and an anode containing material to be evaporated [4-7]. We studied the TVA discharges in the vapors of Zn, Pb, Ag, Sn, Cu, Au, Co, and Ti. We also used Cu-Ag and Cu-Sn as anode material and produced their alloys by electron bombardment in the vacuum vessel before TVA discharge studies in alloy vapors. Dependence of the breakdown voltage values of TVA discharges in Cu-Ag and Cu-Sn vapors on the percentage (in weight) of Ag and Sn were investigated. Cu-Ag alloy thin films with various mass ratios were deposited onto the glass substrates by using TVA discharges. The ratios of Cu and Ag in the thin Cu-Ag alloy films were found using the SEM-EDX microanalyses.

### 2. Experimental

A schematic presentation of the TVA electrode arrangement for an interelectrode angle  $0^\circ < \varphi < 90^\circ$  is shown in Fig. 1. The electrodes (cathode and anode) can be arranged in various relative angular positions  $\varphi$  and distances  $d$  with respect to the anode.

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\* Corresponding author: akan@ogu.edu.tr

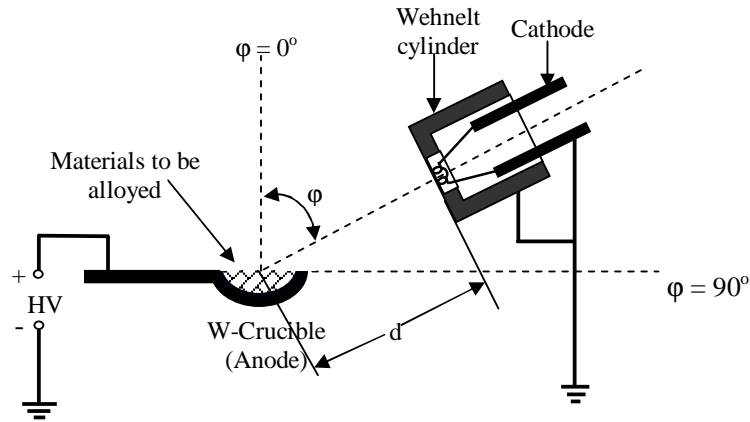


Fig. 1. Schematic diagram of the TVA electrodes arrangement for  $\phi > 0^\circ$ . “HV” indicates the high voltage power supply.

The TVA consist of a directly heated cathode, which is a filament formed of 4 loops of 0.4mm diameter wolfram wire wound to a 1mm diameter rod, and an anode. This filament is mounted inside a molybdenum Wehnelt cylinder with a 5 mm diameter hole at the front. The crucible, which is the anode containing the materials to be evaporated, is made of 0.2 mm thick wolfram and has a shape of spoon with an upper side diameter of 10 mm. The two electrodes are mounted on a table which is placed in the center of a vacuum cylindrical stainless steel chamber having a volume of 65 lt. The vacuum was obtained by a conventional pumping system, which consists of a rotary pump with a capacity of 1.027 lt/s, and a diffusion pump with a capacity of 135 lt/s. A base pressure of  $2.5 \times 10^{-6}$  mbar was achieved before the experiments. The residual gas pressure during the experiments was in the range of  $(2-5) \times 10^{-5}$  mbar. The electrical system consists of two power supplies (low and high voltage), a voltmeter, an ammeter, and a ballast resistor (300  $\Omega$ ). An a.c. variable low voltage power supply with maximum value of 1kW, 0-27V was used to heat the cathode and a d.c. variable high voltage power supply with maximum value of 10 kW, 0-5 kV was used to accelerate the electrons from the cathode to the anode.

### 3. Results

The TVA discharge is produced under vacuum conditions between a heated cathode emitting thermoelectrons and an anode containing the material to be evaporated. The accelerated electron beam, incident on the anode, heats the crucible together with its content to high temperature. The material at the anode first melts and, after some time, starts to evaporate if the energy input is kept at a sufficiently high level. Consequently, a steady-state density of the evaporating metal atoms is established in the interelectrode space. If the voltage applied between anode and cathode is high enough, a bright discharge ignites between the electrodes. Continuous operation of this discharge requires two basic conditions [6]: (1) a sufficient high vapor production at the anode to ensure a sufficient high vapor density in the interelectrode space, (2) a sufficient high charge carrier production due to inelastic electron-atom collisions within the interelectrode space to compensate the continuous loss of charge carriers due to ambipolar diffusion and recombination.

Condition 1 can be described by a power balance equation. The power dissipated at the anode is roughly determined by the product of the electron current  $I_e$  and the breakdown voltage  $U_b$  (voltage at which the TVA discharge ignites for a given electron current, electrode geometry, and anode material), and by the arriving electrons to anode. Unlike common gas discharge, the TVA discharge continuously replaces the loss of the discharge sustaining gas, i.e. metal vapor, due to the continuous evaporation of the material connected to the anode. Consequently the meaning of breakdown is different from those currently used. Indeed, for ignition, the applied voltage must generate in vacuum conditions the metal vapors in the interelectrode space. Power losses occur due

to anodic evaporation, radiation, thermal conductivity, and ionization of vapor on the front of the anode. This leads to the expression

$$I_e U_b = qn + c_1 \epsilon \sigma T^4 + q_{\text{cond}} + c_2 \alpha N_e - c_3 N_e \Phi, \tag{1}$$

where  $q$  is the evaporation energy per atom;  $n$  is the number of the metal atoms evaporated from the unit anode surface area in unit time;  $\epsilon \sigma T^4$  is the energy loss per unit time from the anode due to blackbody radiation;  $q_{\text{cond}}$  is the energy loss from the anode due to thermal conductivity;  $\alpha$  is the ionization energy of the metal atoms;  $N_e$  is the electron density;  $\Phi$  is the work function;  $c_n$  are constants.

The TVA discharges were generated in the vapors of Zn, Pb, Ag, Sn, Cu, Au, Co, and Ti for fixed electron current ( $I_f=15A$ ) and fixed electrode geometry ( $d=6mm$  and  $\phi =50^\circ$ ). The values of the breakdown voltages of these TVA discharges and some material parameters are given in Table 1. When these breakdown voltage values was established, we waited 30s for each 15 V applied between electrodes.

Table 1. Some material parameters of various metals [8] and, the breakdown voltages of TVA discharges generated in the vapors of Zn, Pb, Ag, Sn, Cu, Au, Co, and Ti for the following working condition,  $I_f=15 A$ ,  $\phi = 50^\circ$  and,  $d = 6mm$ .

Material	$T_{\text{melt}}$ (°C)	$T_{\text{boil}}$ (°C)	Thermal Cond. (W/cmK) at 373 K	Density (g/cm <sup>3</sup> ) at 20 °C	TVA Breakdown Voltage (V)
Zn	419.5	907	1.12	7.13 (25 °C)	280
Pb	327.5	1740	0.34	11.35	400
Ag	961.9	2212	4.26	10.50	645
Sn	231.9	2270	0.48-0.63	5.75	780
Cu	1083.4	2567	3.95	8.96	810
Au	1064.4	2808	3.13	19.30	880
Co	1495	2870	0.89	8.90	1185
Ti	1660	3287	0.2	4.54	1515

In Table 1 are given some physical properties of the used material for the TVA breakdown studies. From the examination of these data, we can see a nearly linear relation between the TVA breakdown voltage of considered material and its boiling temperature. This dependence is given Fig. 2. Similar curve was obtained between TVA breakdown voltage of considered material and the temperature for 0.1 torr saturated vapor pressure and 1 torr saturated vapor pressure.

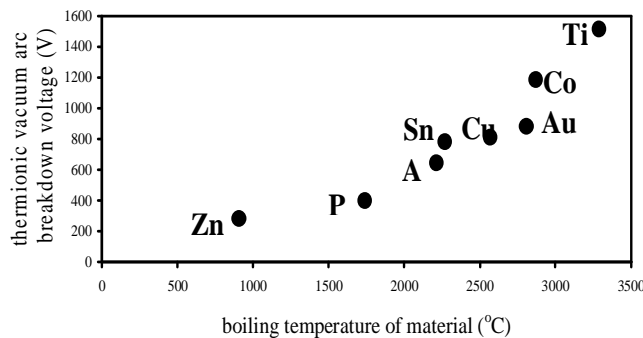


Fig. 2. Dependence of TVA breakdown voltage on boiling temperature of the material.

We first used the TVA for alloy processing [9,10]. For this purpose, different selected material pieces of 2-4 mm size (copper-silver and copper-tin pieces in our study) are put into the anode crucible. After applying a current  $I_f$  to the cathode filament and ensuring thermoelectronic emission of the cathode, a high voltage is applied across the electrodes. The impact of accelerated electrons heats the anode material. At a certain value of the applied high voltage, material pieces diffuse into each other and become a ball of alloy. In this way, alloy is produced by using the TVA.

Cu-Ag alloys containing 25 wt. % Ag, 50 wt. % Ag, 72 wt. % Ag, and 85 wt. % Ag were made by using the TVA, and the TVA discharges were generated in the vapors of these alloys. Cu-Ag alloys containing 72 wt. % Ag has the minimum melting point shown in the phase diagram of Cu-Ag alloys. The experimental conditions were kept identical for each experiment. The TVA discharges for each run were established for the following conditions: (i) Cathode filament heating current  $I_f = 14A$ , (ii) interelectrode angle  $\phi = 50^\circ$ , (iii) interelectrode distance  $d = 5mm$ .

From the volt-ampere characteristics of the TVA discharges generated in the vapors of Cu-Ag alloys with various mass ratios, it is seen that the breakdown voltages of the TVA discharges depend on the ratio of Ag in the Cu-Ag alloys. The dependence of the breakdown voltages of the TVA discharges in Cu-Ag alloys vapors on the percentage (in weight) of Ag is given in Fig. 3. The breakdown voltages of the TVA discharges for pure Cu and pure Ag are also given in Fig. 3.

For each given set of alloy data represented various symbols we repeated the experiment three times. The spread of the points between the given alloys sets shown various marks in Fig. 3 due to use for each set of data a new filament. It is hard to believe that the filament was mounted in the identical position. As seen in Fig. 3, the breakdown voltage of Cu-Ag alloy TVA discharges decrease until it reaches a value, i.e. ratio of 72 wt. % Ag in the Cu-Ag alloy, they increase after this minimum value until the breakdown voltage of the TVA discharge in the Ag vapors. The breakdown voltages of the TVA discharges for Cu-Ag have a similar shape with the phase diagram of the Cu-Ag alloys [9].

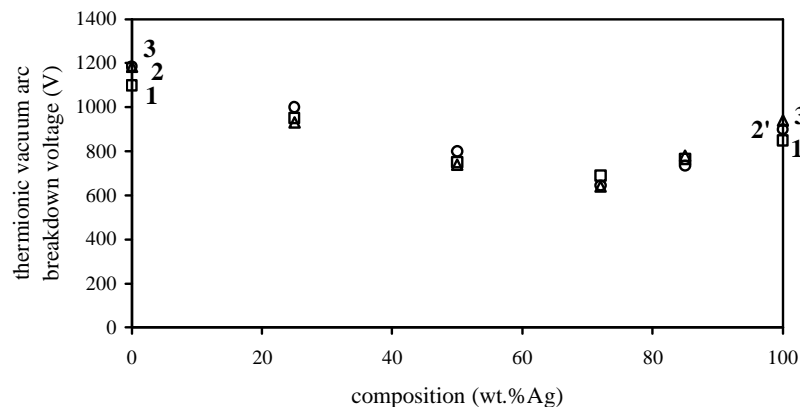


Fig. 3. Dependence of the breakdown voltages of the TVA discharges Cu-Ag alloys vapors on the percentage (in weight) of Ag. The points marked with 1,2,3 and 1',2',3' represents the breakdown voltages of TVA discharges for pure Cu and pure Ag, respectively

Cu-Sn alloys with mass ratios of 20 wt. % Sn, 40 wt. % Sn, 50 wt. % Sn, 60 wt. % Sn, 80 wt. % Sn were obtained in the crucible by using the TVA, and their TVA discharges were ignited increasing the value of the applied voltage. The experimental conditions were kept identical for each experiment. The electrode arrangements for the experiments of Cu-Sn alloys were the same as those of Cu-Ag alloy experiments but cathode filament heating current was 15A. The breakdown voltages on the volt-ampere characteristics of the TVA discharges generated in the vapors of pure Cu, Cu-Sn alloys with various mass ratios, and also pure Sn were investigated. The dependence of the breakdown voltage of the TVA discharges for Cu-Sn alloys on Sn percentage in alloy is given in

Fig. 4. The breakdown voltages of the TVA discharges only in the vapors of Cu or Sn are given in Fig. 4 as in Fig. 3.

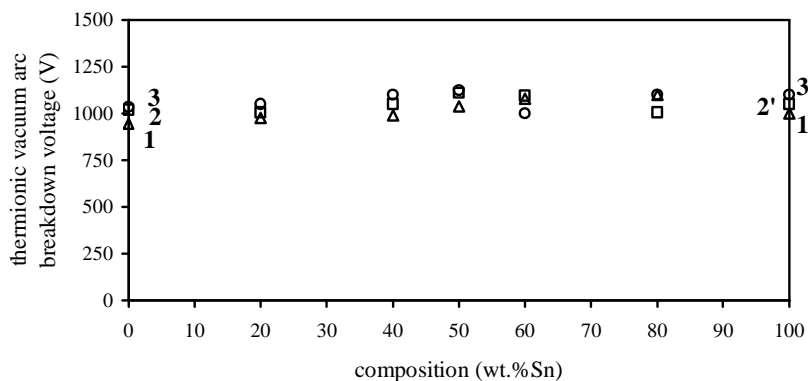


Fig. 4. Dependence of the breakdown voltages of the TVA discharges in Cu-Sn alloys vapors on the percentage (in weight) of Sn. The points marked with 1,2,3 and 1',2',3' represents respectively the breakdown voltages of TVA discharges for pure Cu and pure Sn.

As seen in Fig. 4, the breakdown voltages of Cu-Sn alloy TVA discharges are nearly the same for each discharge while the phase diagram of Cu-Sn alloy decreases until the melting point of Sn. The melting points of Cu and Sn are relatively different; however, their boiling temperatures are nearly the same. Furthermore, the melting points and the boiling temperatures of Cu and Ag are different. We concluded that our experiments show that the breakdown voltage depends on the boiling temperatures of the alloys.

Thin Cu-Ag alloy films with various mass ratios were deposited onto the glass substrates using the Cu-Ag alloy evaporation by TVA discharge. Deposition time was 45s and discharge power was 350W. Total duration time to evaporate all crucible material (0.2g) was 150s. EDX microanalyses these films were established via SEM-EDX (Scanning Electron Microscope-Energy Dispersive X-ray Spectrometry) system. The results of SEM-EDX microanalyses of Cu-Ag alloy thin films deposited during 45s using TVA are given in Table 2.

Table 2. The results of SEM-EDX microanalyses of Cu-Ag thin films deposited during 45s using TVA.

Ag ratio inside the crucible before Cu-Ag alloying (wt % Ag)	Ag ratio inside the deposited thin Cu-Ag alloy film (wt % Ag)
15	3.58
50	28.54
75	90.93

If Ag content in the crucible increases, Ag ratio inside deposited Cu-Ag alloy thin films becomes higher than the initial alloy material charged in the crucible. Because the boiling temperature of Ag is lower than that one for Cu, we may expect Ag atoms to evaporate faster than Cu. However, in case of low Ag content in the crucible, Ag atoms can not get out due to the high density of Cu atoms.

#### 4. Conclusions

Breakdown voltage of TVA discharge for more than eight metals has been measured. TVA was used also for alloy processing. Cu-Ag and Cu-Sn alloys were prepared by using the TVA.

The breakdown voltage of alloy TVA discharge dependence on the boiling temperature of materials used for alloy has been established experimentally.

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