Photoelectric emission and work function of silver-zinc oxide Ag-ZnO (92/8) pseudo-alloy electrical contacts

M Akbi1,3 and M Cherif M’ziane2
1Université M’hammed Bouguerra de Boumerdes, Avenue de l’indépendance, Boumerdes, DZ-35000, Algeria
2Fundamental and applied Physics Laboratory, (FUNDAPL), Université Saad Dahleb Blida 1, PV 08, Route de Soumaa, B.P. N° 270, DZ-9000, Blida, Algeria
E-mail: akbim656@gmail.com; m.akbi@univ-boumerdes.dz

Abstract. Arc roots models require a good knowledge of the physical constants characterizing the contact material used in various kinds of switchgears. Electron work function (EWF) is one of the most important parameters of electron emission for metals and has not yet been studied in detail for pseudo-alloy contact materials. For pure metals, the values of these constants are well known and can easily be found in tables. This is not the case for the new silver-based industrial materials recently available on the market, which are more efficient without the manufacturers knowing the cause. The main goal of this paper is to understand better the photoelectron emission occurring in silver-metal oxide Ag-ZnO (92/8) electrical contacts, before and after several heat treatments from room temperature up to 813 K, under UHV conditions. The photoelectric work function of Ag-ZnO (92/8) pseudo-alloy electrical contacts has been measured by photoelectric effect, using both Fowler’s methods, i.e. isothermal curves method and linearized plots method. It was demonstrated that after several heating treatments in vacuum, the EWF of the silver–zinc oxide, Ag–ZnO (92/8), determined at T = 300 K, is kept within the following interval limits [\(\phi_{\text{Ag}}\), \(\phi_{\text{ZnO}}\)] = [4.26 eV, 4.68 eV]. This is due to the evaporation and/or diffusion phenomena which influences the surface composition of the pseudo-alloy electrical contact. Moreover, all the measurements carried out in this study give clearly a demonstrable proof that the EWF of the Ag-ZnO pseudo-alloy should be between the EWF values of each of the two components 4.26 eV for Ag and 4.64 eV for ZnO. Observations and surface analyses carried out under the scanning electron microscope illustrated the development of the surface composition of the Ag-ZnO contact material after several heating treatments in vacuum. Thus, surface analysis of two Ag–ZnO electrical contacts performed before and after VH treatments showed a significant increase of weight proportion for ZnO nanoparticles on the contact surface.

1. Introduction
Silver metal oxides Ag-MeO have found many applications in electrical and electronics industries. In particular, Ag-ZnO electrical contact materials are widely used in various kinds of switchgears, such as relays, contactors, engine starters and electrical switchers [1, 2]. The Ag-ZnO material is used for switching contacts in low-voltage (between 24 and 600 V) contactors (up to 1000 A) [1, 2]. In addition, high capacity and low-voltage electrical switchgear are widely used in industrial automatisms, in transport such as cars and railways, in lifts and forklift trucks, and in control and regulating devices, such as thermostats [3].
The Ag-CdO material has virtually dominated medium to high current switching applications (between 50 and 5000 amperes) for several decades after its debut in 1950 [4-5]. However, due to the toxicity of cadmium, environmentally friendly materials, such as Ag-SnO₂ [6, 7] and Ag-ZnO [8], have been developed as an alternative to Ag-CdO [9]. Ag-SnO₂ materials have progressively replaced Ag-CdO materials over the last four decades, as they present superior erosion resistance and anti-welding characteristics [10]. However, silver tin oxide shows a high temperature rise and its treatment is more delicate [11]. Due to these undesirable effects, the properties of silver and zinc oxide (Ag-ZnO) enabled it to effectively replace the contact material (Ag-SnO₂) [12].

To replace Ag-CdO contact material, Ag-ZnO was developed in the 1970s. Ag-ZnO contact material exhibits, comparatively with other contact materials, higher electrical and thermal conductivities, higher thermal stability, higher hardness and strength, higher welding resistance and low arc erosion. This new material is highly recommended when contacts are subjected to intense closing and opening cycles during which an electrical arcing occurs. Another favorable quality is that it is harmless and does not pollute the environment.

Ag-ZnO (92/8) innovative contact materials having multi-layered architecture were supplied by Metalor (France) and fabricated by powder metallurgy [3]. It should be emphasized that the performances achieved by Ag-ZnO contact materials have not yet been understood by the manufacturers. Therefore, the purpose of this study is to understand better the electron emission of the pseudo-alloy Ag-ZnO by examining the modifications of the photoelectron work function changes under various UHV heat treatments. The electron work function (EWF) of silver zinc oxide contacts was determined by photoelectric effect [13–20] using both Fowler’s methods: isothermal curves and linearized plots [21, 22].

In addition, to study the influence of vacuum heat treatments on the composition of Ag-ZnO cathode surfaces, on the surface distribution of ZnO nanoparticles and consequently on the electron work function of such electrical contacts, the microstructure of the Ag-ZnO contact surface has been observed by SEM and then analyzed by EDS.

2. Experimental

2.1. Materials

The electrical contacts carried out in the present study are manufactured by Metalor (Courville-sur-Eure, France). They have a cylindrical shape, a diameter of 8 mm and a height of 3.5 mm, with a radius of curvature of 16 mm. In this paper, a set of new experiments carried out with silver-zinc oxide Ag-ZnO (92/8) is highlighted. Numbers in parentheses represent the mass proportion of pure silver particles and granular ZnO particles, respectively. The grain size of ZnO is between 1 and 5 μm. It should be pointed out that the size of oxide particles distributed in the silver matrix plays a key role in diffusion processes which occur during vacuum heat treatment and in electron emission properties of the Ag-ZnO composite since it has been established that electron work function depends upon surface composition and microstructure [10] of silver-zinc oxide (Ag-ZnO) electrical contact. The conditioning processes of contacts have been given in much detail elsewhere [13–16]. However, the main process of cleaning the surface has been achieved by repeated heating cycles at 700 K under very high vacuum conditions. EWF measurements were carried out at low pressure (10⁻⁹ mbar), after repeated annealing cycles at 700 K, and for short periods (less than thirty minutes for one test). The experimental set-up has been reported in details in previous papers [13–20].

2.2. Experimental method

EWF data were obtained using Fowler’s theory of photocurrent. Fowler developed a theory of electron energy distribution, founded on the hypothesis that the free electrons of a metal satisfy Fermi-Dirac’s statistics [23]. In accordance with this theory, the emitted photoelectric current is:
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\[
I = \gamma \frac{4\pi e m (kT)^3}{h^3} f(x) = \gamma A \sqrt{T}^2 f \left( \frac{h(v-v_0)}{kT} \right) \tag{1}
\]

where:

\[
f(x) = e^x - \frac{e^{2x}}{2} + \frac{e^{3x}}{3!} - L \quad \text{ (} x < 0 \text{)}
\]

\[
= \frac{\pi^2}{6} + \frac{1}{2} x^2 - \left[ e^x - \frac{e^{-2x}}{2^2} + \frac{e^{-3x}}{3!} - L \right] \quad \text{ (} x > 0 \text{)}
\]

\[
A = \gamma \frac{4\pi e m k^2}{h^3} = 1.2 \cdot 10^6 \ A \cdot m^2 \cdot K^{-2} \text{ is Dushman thermionic constant.}
\]

Equation (1) is used to determine the photoelectron work function of the sample and the constant \( A = \gamma A_0 \) (\( \gamma \) is the probability of photo-ionization of the cathode surface) from experimental data since this equation can be written in the following form:

\[
\log \left( \frac{I}{T^2} \right) = B + F(x); \quad x = \frac{h(v-v_0)}{kT} \tag{2}
\]

where \( B \) is a constant independent of the frequency \( v \) of the incident radiation and \( T \) the absolute temperature of the emitting contact surface. The constant \( B \) represents the photoelectric surface yield; it depends on the optical absorption of the sample, the photoelectric efficiency of the emitting surface and the probability of escape of photoelectrons.

\[
F(x) = \log f(x) = \log f \left( \frac{h(v-v_0)}{kT} \right)
\]

is a universal function of \( x \); it is important to note that its graph has the same shape for all metals and alloys as well as for all temperatures.

More precisely,

\[
F(x) = \ln \left[ \frac{\pi^2}{6} + \frac{1}{2} x^2 - \left[ \exp(-x) - \frac{\exp(-2x)}{2^2} + \frac{\exp(-3x)}{3!} - L \right] \right] \tag{3}
\]

To assess the results of the measurements, the Fowler isothermal curve method can be used. In this method, photocurrents \( I \) are measured for different frequencies \( v \) at a given temperature \( T \) and as a function of the same number of photons of absorbed light. This technique requires a photon source of calibrated spectral intensity and variable energy in the range around the threshold frequency for emission \( v_0 \). The ultraviolet radiation device has been described in much detail elsewhere [13–16]. Photocurrent measurements (of order of \( 10^{-13} \) A) were performed using a picoammeter (Keithley’s model 466). The uncertainties in the EWF measures were \( \pm 0.03 \) eV, \( \pm 0.07 \) eV and \( \pm 0.08 \) eV at \( T = 300 \) K, \( T = 500 \) K and \( T = 800 \) K, respectively.

By plotting \( \ln \left( \frac{I}{T^2} \right) \) versus \( \frac{hv}{kT} \), we can compare the theoretical Fowler curve \( F(x) \) versus \( x \) to the experimental Fowler curve. A vertical offset of the data gives \( B \) which is nevertheless unimportant and a horizontal one gives the EWF \( \phi \). This is one of the most accurate methods for determining the photoelectron work function of metallic and allied materials, since in most cases both theoretical and experimental curves fit very well.

In addition, the evaluation of the EWF may as well be founded on the simplified theory of Fowler [21, 22] which establishes a linear relationship between the square root of the photoelectric current, \( \sqrt{I} \), and the incident photons energy \( h\nu \) in accordance with the relationship [14, 24]

\[
\sqrt{I} \approx (h\nu - \phi), \quad h\nu > \phi \tag{4}
\]

where \( \phi \) is the electron work function (EWF) of the sample surface.
The interception of $\sqrt{I}$ versus $h\nu$ is used to determine $\phi = h\nu_0$. We have assumed that the absorptivity of the surface is almost constant in the region where the photon energy, $h\nu$, and the quantum yield, $\sqrt{I}$ follow the Fowler curve.

3. Results and discussion

The reliability and service life of the components have been improved for recent electromechanical equipments containing electrical contacts with alternative materials such as silver-zinc oxide (Ag-ZnO) and silver-tin oxide (Ag-SnO$_2$). However, the physics of this phenomenon is still poorly known [4]. In addition, it should be stressed that there is no rational scientific explanation for the improved performance of new contact materials; It should also be noted that the new switching devices currently available on the market perform thousands of operations under load without any malfunction.

Notwithstanding that a large number of contact materials surveys and their application criteria have been published over the past decades, there is relatively little information on the properties of the innovative contact materials, principally in terms of electron emission mechanisms. Moreover, the theoretical models of the electric arc, mainly those of cathodic phenomena, considered today do not reflect the evolution of electron emissivity with the conditions on the arc feet and the nature of the material on which it originates. Theoretical studies conducted in recent decades have established that the lack of knowledge of the EWF of the emitted electrode causes a significant error on the physical quantities of the arc root (electric field, temperature, cathode spot radius...). For example, a variation of 0.1 eV of EWF causes a variation of 100 K in the cathode temperature for an electric arc conducting a 1000 ampere current.

For these reasons, this investigation tries to describe the work function measurements of these pseudo alloy contact materials using the photoelectric effect. Therefore, the objective of this work is to describe, quantify and explicate the phenomena of electron emission from both theoretical and experimental angles, in particular the effects of the annealing temperature on the EWF for silver-zinc oxide materials, Ag-ZnO (92/8), pure silver is the material reference. On the other hand, the effects of heating treatment in vacuum on the variations of EWF have also been extensively studied.

There are a variety of effective techniques, such as powder metallurgy and internal oxidation, to prepare the silver-zinc oxide materials used in circuit breakers. However, silver-zinc oxide contact materials containing mainly from 6 to 10% by weight of oxides including other small metal oxides, are produced exclusively by powder metallurgy. Figure 1 shows the microstructure of Ag-ZnO (92/8) before annealing. The higher magnification (x 8000) corresponds to the lower micrograph, while the higher one has a magnification (x 2000).

![Microstructure of Ag-ZnO (92/8)](image)

**Figure 1.** Microstructure of Ag-ZnO (92/8).

Notes: (a) SEM magnification x2000, (b) SEM magnification x8000.
3.1. EWF measurements of Ag–ZnO (92/8) electrical contacts

EWF is one of the most important physical parameters that characterizes the surface condition of metallic and allied contact materials, and checks their electron emission. It is defined as the minimum amount of energy required for an electron to release from a metal surface. Moreover, it depends upon both bulk and surface of the metal. The contribution of the contact volume can be described as the difference in energy between the Fermi level and the vacuum level. On the other hand, the contribution of the surface is due to the electrostatic dipole barrier which depends strongly on the surface conditions. Therefore, it can be said that EWF is a surface characteristic that is highly dependent on the microscopic details of the surface. Therefore, for the same metal, it is different from one microscopic surface to another.

The present investigation deals with the surface state modifications of the Ag–ZnO (92/8) contact pastille with annealing temperature under ultra high vacuum conditions. It is worth noticing that all the changes in surface electronic structures of a material are related with the movement of electrons which depends on Fermi level and electron work function (EWF) [27]. We demonstrated that annealing temperature in UHV environment affects the surface characteristics of Ag–ZnO contact materials, as for example microstructure, roughness and in particular photoelectron work function.

In order to determine the electron work function, the generated photocurrent was determined as a function of the energy of the incoming photons. The EWFs of the unpolished and virgin contact Ag–ZnO (92/8), that have been measured after 10 cleaning cycles by heating treatments and 8 days of outgassing, under UHV conditions at different temperatures, are summarized in Table 1.

Table 1. Electron work function of the virgin contact Ag–ZnO (92/8).
Studies on the influence of the heating temperature on the photoelectron work function for silver-zinc oxide contacts were carried out. For this purpose, photocurrent measurements have been extended to higher temperatures. As the temperature increases, the EWF measurements become rather difficult because the thermionic current is superimposed on the photoelectric one. As a result, the total current obtained fluctuates considerably, so that it was not possible to carry out precise photoelectric measurements. A method based on averaging was used, in which the total current was measured every 10 seconds at a given high temperature. Thus, twenty values were registered by a Keithley picoameter (model 486) for each wavelength of incident radiation. The photoemission was then interrupted by cutting off the power supply of the deuterium lamp and similar measurements were made for the thermionic current alone. The photoelectric current can be then deduced by subtracting the thermionic current from the total current.

When photocurrent readings were attempted at higher temperatures, large current fluctuations started to appear around 700 K and became more pronounced as the temperature was rising. Regrettably, these fluctuations have made it very difficult to measure the photocurrent. Typical photoelectron spectral distribution curves for silver-zinc oxide contacts at T = 297 K, T = 533 K and T = 823 K, and under ultrahigh vacuum, are shown in Figure 2.

![Figure 2. Spectral distribution curves.](image)

![Figure 3. Typical Fowler isothermal curves (Equation 1).](image)

Figure 3 compares the Fowler theoretical curve with the the experimental one which corresponds to the above measurements for a virgin Ag-ZnO electrical contact (92/8).

In addition, the evaluation of the EWF may as well as be based on the the simplified theory of Fowler [21, 22]. Consequently, the Fowler curve allows to determine the EWF \( \phi = h \nu_0 \) directly from an adequate extrapolation of the straight lines drawn in Figure 4; the intersections with the abscissa axis give I = 0 and consequently the electron work function \( \phi = h \nu_0 \).

### 3.2. Influence of heating treatment in vacuum

The EWF of the raw and virgin silver-zinc oxide contact, Ag-ZnO (92/8), before the UHV heating treatment of the sample and before the baking of the experimental chamber is equal to (4.62 ± 0.03) eV. The work function was taken to be 5.3 eV for the ZnO materials from T. Minami et al [25]. These authors have measured the work function of ZnO films by ultraviolet photoelectron spectroscopy operated in air. In our measurements, the EWF electrical contact of Ag-ZnO (92/8) began to decrease as soon as the annealing temperature increased more.

After five successive vacuum heating operations at 408 K, 473 K, 533 K, 713 K and 823 K for 3 h, 4 h, 16 h, 4 h and 3 h respectively on the fifth day of the measurements, a surface structure...
modification of the Ag-ZnO cathode surface occurs which explains the EWF variations in the contact surface. Thus, after these vacuum heat treatments, and in particular at the seventh cycle at 300 K, the EWF takes an intermediate value of 4.48 eV which indicates a surface covered with more nanoparticles of zinc oxide than silver atoms. In addition, after the sixth surface heat treatment, during a prolonged heating period of 18 h at 753 K, the EWF is 4.34 eV, which is quite close to that of pure silver. The surface of the contact should then be almost covered with silver atoms.

Figure 4. Typical linearized Fowler plots (Equation (4)).

Figure 5. Vacuum heat treatment effect on the EWF of an outgassed virgin Ag-ZnO (92/8) contact.

Obviously, it seems that the sample cleaned by heating treatments in vacuum has become exclusively covered with silver atoms since surface impurities have been removed by thermal desorption. Long-term vacuum heating has moved the EWF values of the pseudo-alloys silver-zinc closer to those of silver. This could be explained by the diffusion of silver atoms on the surface, which will then overwhelm the crystalline nanoparticles of zinc oxide. The latter hypothesis is attributable to the lower melting point of silver (961.8°C) compared to zinc oxide (1975°C), so that under the effect of temperature, the diffusion rate of Ag atoms is higher than that of ZnO nanoparticles. As a result, the EWF value of the Ag-ZnO contact is reduced, and becomes close to that of Ag.

This behaviour has already been observed and demonstrated in our previous work [15, 19], for Ag-Me multilayer contact materials, i.e. for silver alloys such as silver-nickel and silver-tungsten. However, this phenomenon is noticed for the first time for Ag-MeO pseudo-alloys, especially for Ag-ZnO (92/8) electrical contacts. Further investigations should be carried out subsequently for silver-pseudoalloys.

As the temperature of the Ag-ZnO (92/8) cathode increases with silver and zinc oxide, a gradual evaporation of the silver atoms of the surface layers is likely to occur. At the same time, the zinc oxide nanoparticles slowly and uniformly diffuse from the underlying layers to the contact surface. All experiments conducted in this study indicate that the EWF of the Ag-ZnO pseudo-alloy is confined between the EWF values of each of the two components, i.e. (4.26 ± 0.03) eV for Ag and 4.68 eV for ZnO.

During heating under ultra high vacuum, the two compounds present at different percentages on the surface of the contact evaporate therefrom at different speeds, which allow the zinc oxide nanoparticles to degass more slowly than the silver atoms, and the composition of the surface layer therefore varies considerably for the two complementary components. As shown in Figure 5, it is worth noting that the phenomena of diffusion and/or evaporation during the vacuum heating treatment, which affect the surface composition of the pseudo-alloy electrical contact, in a way limit the EWF of Ag-ZnO, measured at room temperature in the range \( \text{Ag} \), ZnO \\[ = \] 4.26 eV, 4.68 eV [.
The multilayer appearance of the Ag-ZnO contact material is at the origin of these outcomes: the two compounds present in the electrical contact do not mix as in a solution and therefore occupy successive layers [3].

4. Discussion

To study the influence of UHV heat treatments on Ag-ZnO pseudo-alloys, observations by SEM and surface analyses by EDS were carried out on three samples before and after the surface treatments (polishing and annealing). EDS line scans should be reported in a future article.

After several periods of 30h heat treatment at different temperatures within the range [299 K, 823 K], it was found that the EWF of the Ag-ZnO contact (92/8) was equal to (4.32 ± 0.08) eV at 823 K. It is very likely that this would be due to a significant increase in the atomic proportion of silver in the total composition of the contact surface of the Ag-ZnO contact after many UHV heating procedures (see Table 1).

Other series of vacuum heat treatments, with a total duration of 26 hours, significantly increased the value of the work function (i.e. 4.59 eV) of the Ag-ZnO electrical contact (92/8) measured at 813 K. It should be noted that the EWF value after these different heat treatments is quite similar to that of ZnO (4.68 eV) [26]. This substantial variation in the chemical composition of the contact surface that has undergone prolonged heating under vacuum is probably due to the diffusion of zinc oxide from the contact core towards its contact surface. This also implies that, during heating, the silver atoms evaporate from the surface layers of the electrical contact to the vacuum vessel, so that the surface of the sample becomes covered with a majority of zinc oxide nanoparticles.

After a heat treatment of 30 min at 623 K, electron microscopy observations show that the zinc oxide blue islets are much larger than before heating (Figure 6 (a) and Figure 6 (b)), while the silver grey areas decrease with increasing temperature.

With regard to the multilayer structure of silver-zinc oxide pseudo-alloys, the results obtained must therefore be well interpreted. In addition, the EWF of pure metals decreases with increasing temperature [13, 16], while an opposite result has been found for Ag-MeO pseudo-alloys, i.e. Ag-ZnO (92/8), and previously for Ag-Me alloys [15, 19].

![Figure 6](image_url)

**Figure 6.** EDS elemental mapping of (a) a polished Ag-ZnO (92/8) contact, (b) an annealed and polished Ag-ZnO (92/8) contact. Microscope magnification x 3000.

5. Conclusion

The EWF of a pseudo-alloy contact material is highly dependent on the condition of its surface. The main results obtained in this survey are as follows:
1. The multi-layer nature of the Ag-ZnO electrical contact (92/8) has been demonstrated. By heating a pseudo-alloy contact under ultra-high vacuum (UHV), significant variations in the EWF were observed, resulting from the vaporization of the material components by sheets.

2. The EWF of pure metals decreases with increasing temperature while an opposite result was found for Ag-McO pseudo-alloys, and previously for Ag-Me alloys. These opposite results are significant, because depending on the use of contacts (low voltage, high voltage, small arc duration...), we can predict the nature of the contact material which must be either a pure metal or an alloy or pseudo-alloy.

In conclusion, it could be inferred from this study that further research is needed to better elucidate these complex phenomena for which physics is still poorly known. Additional investigations will focus on the influence of vacuum heat treatments on the morphology, microstructure and EWF of silver-tin oxide Ag-SnO2 (88/12) and silver cadmium oxide Ag-CdO (88/12) electrical contacts.

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