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ELECTROMIGRATION IN d.c. ELECTROLUMINESCENT THIN FILMS OF ZnS

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Résumé - On propose d'expliquer la dégradation de l'émission de lumière par des structures ZnS électroluminescentes par la migration d'ions cuivre. Celle-ci affecte les propriétés de la couche au voisinage de l'électrode positive qui devient brillante. Des modifications de la surface de l'électrode et la précipitation de quantités importantes de cuivre ont été observées par microscopie optique et électronique.

Abstract - The degradation of the light emission from electroluminescent /EL/ ZnS structures is tentatively explained in terms of copper ion migration. The migration of copper ions affects the properties of the layer close to the positive electrode which appears bright. Some changes in the electrode surface and the precipitation of substantial amounts of copper were observed by means of the optical and electron microscopes.

Since many years the attention has been focused on problems related to failures of thin films devices because of their technological and economic importance.

The electromigration is one of the methods applied to investigate this problem. There are two basic types of measurements for thin film electromigration :

- direct measurements of the ion flux, or certain parameters, such as conductor lifetimes and resistance changes, which can be related to the mass transport,
- measurements of the change in the composition distribution caused by the passage of a direct current.

The experimental electromigration effects in semiconductors are smaller than in metals owing to the relatively small values of the diffusion coefficient and the electric field force known as the "electron wind force" which is restricted by the small number of charge carriers. Parameters such as the current density, the ambient temperature, the geometry of the conductors and the microstructure have an effect on electrotransport. Diffusion in thin films can cause serious problems in microelectronic devices. The net mass transport is usually manifested by the formation of voids and hillocks, which could be observed by the optical and electron microscope.

It is probable that the forming and aging processes of electroluminescent /EL/ devices are related to electromigration in d.c. field [1] The aim of this work is to show that the processes which are characteristic of electrotransport take place in EL devices. In other paper [2] we have shown that the stabilization with time of light emission from electroluminescent structures could be related with the electromigration effects. Microscope observations of the EL

* Madame G.M. Jakubowska n'a malheureusement pas été en mesure de venir au Colloque. Sa contribution a cependant été présentée et discutée à la Conférence, et le texte qu'elle nous a fait parvenir est publié dans ces comptes-rendus.

structure show that the brightness is inhomogeneous and that the light emission is localized on many spots of different sizes. We investigated the distribution of the brightness along the active region of the sample and its dependence on time. The localized emission disappears with increasing time, and the brightness of the sample becomes more homogeneous although its intensity is reduced. This degradation of the light emission is tentatively explained in terms of copper ion migration. The migration of copper ions affects the properties of the layer close to the positive electrode which appears bright. Some changes in the electrode surface and the precipitation of substantial amounts of copper are observed.

Experimental details

The structure of the EL device is prepared in few steps [2]. A thin film / $0.3 \mu\text{m}$ / of manganese - doped ZnS was evaporated onto a glass substrate maintained at 400°C in high vacuum / 10^{-6} Torr /. A Cu_2S layer was then produced by dipping the ZnS - Mn film into an aqueous solution of CuCl . The EL structure was completed by evaporating a second ZnS - Mn layer and then depositing aluminium strips as electrodes. These EL structures were examined using optical and electron microscopes. The light emitted was detected with a photomultiplier which was connected to an optical microscope and a photo - camera.

Results and discussion

The main observations of the processes related to the electroluminescence of thin ZnS films were performed with an optical microscope. The microscope image of the emitted radiation is composed of bright spots which confirms the existence of separate sources of light emission. The structure of the EL sandwich is non-uniform and the light emission is localized at small spots of different sizes. Light is not emitted simultaneously with the application of the electric field but is preceded by a forming process, i.e. by the initiation of conduction in the sample. Electrical measurements, which analysis would be published elsewhere later, show that the current increases with increasing voltage. An increase in the polarized voltage produces the increase in the electroluminescence and in the current intensity. However, when the voltage is kept constant the intensities of the emitted light and of the current decrease with time. At the same time the region of luminescence is extended and the number of light-emitting spots is increased. The light emission also becomes more homogeneous. This change in the emission is significant : the light-emitting spots are initially visible only in the microscope but later the light can be seen with the naked eye. Microscope investigations of the emitting surface show that initially the light emission is of high intensity and is located in spots, and that during continuous operation it decreases in intensity but extends over the whole surface. The changes in the surface structure of the electrodes can be seen clearly in the optical microscope observations. The microscope observations are reproducible in different samples. The results presented in this paper we have obtained from the experiments carried on at room temperature. Nowadays we performe as well the experiments in higher temperatures, increasing the rate of electromigration processes. The Fig.1 shows as an example a whole EL element after 2000 h of current operation.

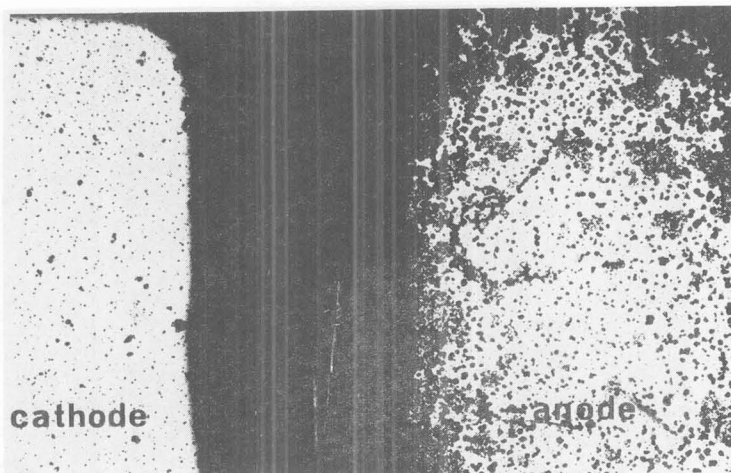


Fig. 1 - Optical micrograph of a whole EL element, after 2000 h of current operation, magnification 80x.

It can be seen the structure changes and growing whiskers on surfaces of two electrodes. A check with a computerized analyser connected to the electron microscope was made to confirm that the whiskers, seen in figure, contained substantial amounts of copper. One of these whiskers is shown in next Fig. 2, 3.

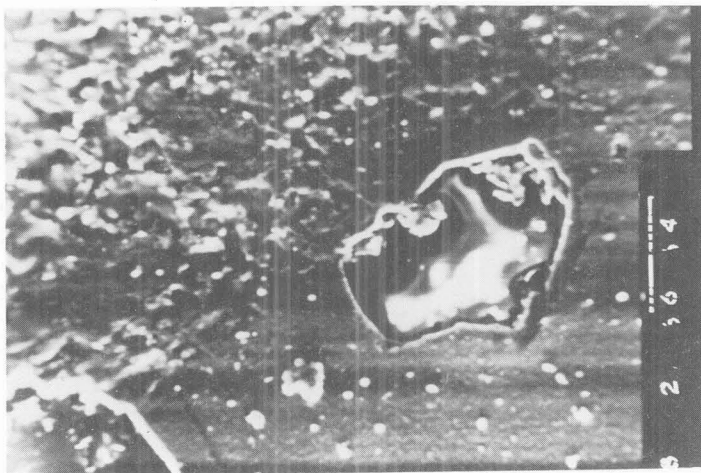


Fig. 2 - Electron micrograph of a part of whisker with the copper precipitation, magnification 1000x.

The assumption that these changes can be interpreted as the electromigration of copper is reasonable because it is known that copper diffusion is very fast in ZnS and because of our electrical measurements, which give the electromigration parameters for copper agreed with the literature data. If a voltage is applied between the

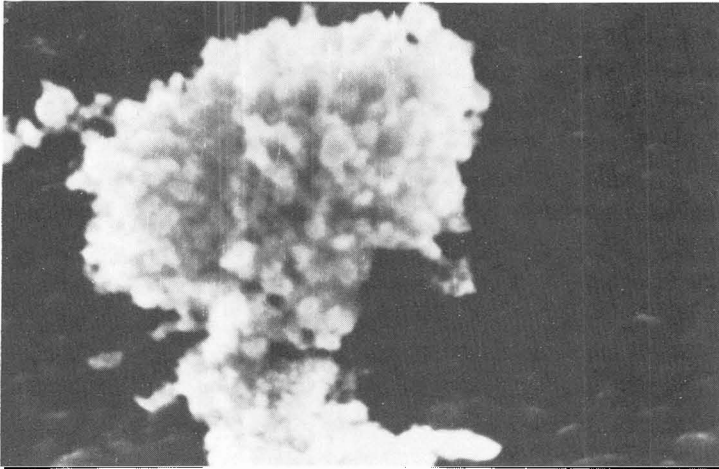


Fig. 3 - Electron micrograph of copper precipitation, magnification 3500x.

electrodes of the EL device a very large forming process current flows as the result of the low Cu_xS resistance. The Cu_xS short circuits the electrodes and a current flows but electroluminescence does not appear. The end of the forming process is indicated by a decrease in the current which is related to the appearance of electroluminescence and a change in the electrical resistance. These changes could be caused by the migration of copper which is manifested as material accumulation and depletion along the conductor as it is seen in Fig. 2 and 3. The highly resistive ZnS is located in the depletion region, it was checked by the electron-probe method. The spot emission of the light could be due to emission from the isolated sources which form ZnS/ Cu_xS heterojunctions. The

diffusion observed probably takes place mainly along the grain boundaries although diffusion into the grain interior is not excluded [3]. The increase in the area of the emitting surface could be related to the increase in the number of active sources due to the diffusion of copper.

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