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ARAŞTIRMA MAKALESİ/**RESEARCH ARTICLE**

AC ELECTRICAL PROPERTIES OF Cu(Zn,Cd)InS$_2$ FILMS PRODUCED BY THE SPRAY PYROLYSIS METHOD

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ABSTRACT

M-Cu(Zn,Cd)InS$_2$-M compound semiconductors in planar form prepared by the spray pyrolysis method. Frequency (ω) dependent conductivity (σ) of the films have been investigated and observed that at reasonably higher frequencies the conductivity varied as ω^s with s ranging from 0.8 to 1. In one of the specimen, the conductivity observed to vary with s=1.45 which is believed to be due to the lossy mechanism taking place. On the other hand s being less than 1 indicates that the conduction process is the hopping process.

Key Words: Ac Conductivity, Hopping Conduction, Spray Pyrolysis.

SPRAY PYROLYSİS YÖNTEMİYLE ELDE EDİLEN Cu(Zn,Cd)InS$_2$ FİLMLERİNİN AC ELEKTRİKSEL ÖZELLİKLERİ

ÖZ

M-Cu(Zn,Cd)InS$_2$-M bileşik yarıiletkenler planar formda "spray pyrolysis" yöntemiyle elde edilmiştir. Film iletkenliğinin (ω) frekansa (σ) bağımlılığı incelenmiştir. Yüksek frekanslarda iletkenliğin ω^s ile değiştiği ve s'nin 0.8 ile 1 arasında değerler aldığı gözlenmiştir. Numunenin birinde, iletkenlik s=1.45 ile değişmektedir. Bunun kayıp mekanizması nedeniyle olduğuna inanılmaktadır. Diğer taraftan, s'nin 1'den küçük olması iletimin hopping olayından kaynaklandığını göstermektedir.

Anahtar Kelimeler: Ac İletkenlik, Hopping İletim, Spray Pyrolysis.

1. INTRODUCTION

The I-III-VI$_2$ compounds and their alloys having the chalcopyrite structure attract much attention because of their technological importance as materials for solar cells, light-emitting diodes, and nonlinear-optical devices (Kuvahara, et al., 1994; Dhumure and Lokhande, 1994). The quaternary chalcopyrite alloys are very promising materials for photovoltaic applications (Gonzalez, et al., 1997).

The dc measurements provide information concerning conduction processes in metal-insulator-metal (MIM) and metal-semiconductor-metal (MSM) structures, and identification of electrode-limited and/or bulk-limited processes may be made. Such identification is made by varying several parameters including the type of metal electrodes, strength of the applied electric field, and sample temperature. Conversely, ac conductivity measurements provide information about the interior of the insulator or semiconductor which is a region of relatively low conductivity even when the conduction process is electrode-limited (Gould and Hassan, 1993).

In general, the conductivity decreases with increasing frequency in the case of band conduction process, while it increases with increasing frequency in

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the case of hopping conduction process. Hopping conduction mechanism is that a carrier can move from one molecule to another by jumping over the barrier via an excited state. The comparatively high conductivity and low activation energy of the intramolecular transfer do not appear in dc measurements but may appear in ac measurements. In fact, the available experimental results about the frequency dependence of ac conductivity have revealed a considerable similarity of behaviour for a very wide range of materials, ordered and disordered, conducting by electrons, holes, ions, polarons, etc., involving various types of chemical bonds and various electronic energy level structures (Kao and Hwang, 1979).

2. EXPERIMENTAL METHOD

The films were deposited by spray pyrolysis method. Spray pyrolysis is basically a chemical process, that is the spraying of the solution onto a substrate held at high temperature,

where the solution reacts forming the desired film (Br.Patent 632256, 1942; Chamberlin and Skarman, 1966; Lampkin, 1979; Afify, et al., 1991; Falcony, et al.1992; Nunes et al., 2002). In addition, the spray pyrolysis method is particularly attractive because of its simplicity and inexpensiveness (Riad, et al., 2001). The films in our study were deposited on glass substrates kept at temperatures 225 or 250°C. The substrate temperature was maintained to within ±5°C. The carrier gas was N_2 and the gas flow rate was kept at 2.5 ml min^{-1}. An aqueous solutions of 0.01M of CuCl2.2H2O, InCl3, H_2NCSNH_2, $CdCl_2.H_2O$ and $ZnCl_2$ containing the necessary elements in compound had been used as the solution.

The planar structures of the metal–semiconductor-metal systems had been produced by vacuum deposition of gold or silver electrodes on the semiconductors. All the electrical measurements were carried out in dark and at room temperature. The ac conductivities were measured, using a HP 4192A LF Impedance Analyzer, having the frequency range 5 Hz-13 MHz, amplitude ranging between 5 mV to 1.1 V.

3. RESULTS AND DISCUSSION

The frequency dependence of ac conductivity can be expressed by the generally accepted relation

$$\sigma_{ac}(\omega) \propto \omega^s \qquad (1)$$

where ω is the angular frequency, and the index s is not a constant for all substances, but is a function of the temperature, approaching unity at low temperatures and decreasing to 0.5 or less at high temperatures (Kao and Hwang, 1979; Mott and Davis, 1971; Hogarth and Naden, 1979; Lopez and Gould, 1995; Lopez and Gould, 1998; El-Samanoudy, 1995; Saleh, et al., 1993; Singh et al., 1996).

Curves in Figure 1 shown the variation of ac conductivity (σ) versus frequency (f) on a log-log scale, and the values of s were derived by calculating the slopes of the curves. In the figure for the Ag-$Cu_{0.5}Zn_{0.5}In_{0.5}S_{1..5}$-Au structure, the frequency-dependent conductivity was not observed until the frequency is reached to about 10^7 Hz. The reason could be the higher dc conductivity of this material compared to the other two. Although the range presenting the ac conductivity which is limited by the range of the impedance analyzer is not enough to conclude, nevertheless, it stills obeys ω^s law, with s 0.8-1. The conductivity of Au-$CuCd_{0.8}InS_{2.8}$-Au film is observed to be constant in the frequency range 10^3-$3x10^5$ Hz and varied with frequency with s=0.88 at higher frequencies. For Ag-$CuCd_{0.8}InS_{2.8}$-Au material, a rather different form of frequency-dependent conductivity has been observed. For this case, two different regimes were observed, the first showing a frequency-dependence with s=1, whereas the second showing with s=1.45. The exponent s being less than one is attributed to the hopping of the charge carriers between the localized states either near to the band edges or near to the Fermi level. Whereas s being greater than one is attributed to the presence of the lossy mechanism assumed taking place at higher frequencies.

4. CONCLUSIONS

The behaviour of the ac conductivity in disordered materials where the charge carriers (electrons, holes) proceed by discrete transitions between localized states exhibits a universal pattern consisting of a power-law

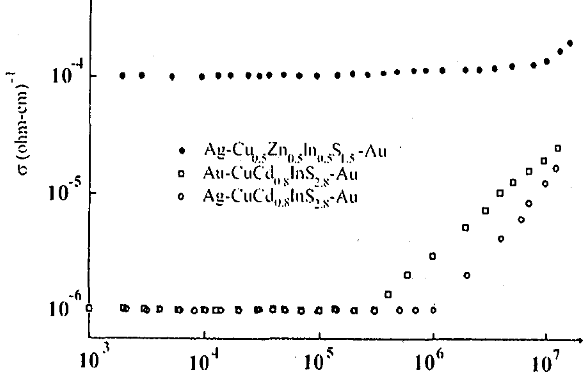

Figure 1. Frequency Dependence of Ac Conductivity σ_{ac}, for Different Compositions at Two Different Substrate Temperattiures: (•) 250°C, (□, o) 225°C.

domain at high frequency and a constant conductivity in the low frequency. In this paper, we studied the ac conductivities of $Cu(Zn,Cd)InS_2$ films in the frequency range of 10^3 Hz to 13 MHz. It has been observed that ac conductivity follows $\sigma(\omega) \propto \omega^s$ dependence where $0.5 < s < 1$. This behaviour was attributed to electronic hopping. This can be interpreted in terms of hopping conduction between the localized states either near to the band edges or near to the Fermi level. On the other hand the reason for the exhibition of s greater than 1 is not clearly understood, and also the causes for the higher values of s have not yet been classfied, we assume that the lossy mechanism is responsible for this behaviour.

Further studies of the high frequency and low temperature properties of the films are in progress.

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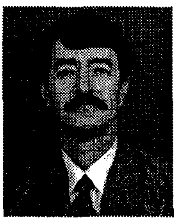

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