$(Cu_xZn_{1-x})Fe_2O_4$ FERRIMAGNETIC FILMS PREPARED BY ATMOSPHERIC MOCVD AT 360°C

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Chemical vapor deposition technique shows potential advantages in preparing magnetic ferrite thin films for communication device uses. Using highly volatile organometallic precursors, CVD can prepare crystalline ceramic films at temperatures below 500°C, avoiding inter-diffusion, and being compatible to multilayer devices manufacturing. During surface reaction step in CVD, elements present as individual adatoms (like two dimensional free flowing fluid), and being transported by surface diffusion to form crystalline films. Activation energy (Ea) of surface diffusion (<10 eV) is much less than the Ea of bulk diffusion required for sintering. However, modern multi element oxide film processes need various metal precursors, which make the CVD system complicate (1,2).

In this study, an MOCVD process has been successfully developed to deposit polycrystalline zinc copper ferrite, ($(Cu_{1-x}Zn_x)Fe_2O_4$) films at temperatures from 360 to 440°C, with zinc acetylacetonate, $Zn(acac)_2$, copper acetylacetonate, $Cu(acac)_2$, iron acetylacetonate, $Fe(acac)_3$, and oxygen as reactants, using a horizontal cold wall atmospheric reactor. Previous gas dynamics study shows that the least volatile precursor, $Cu(acac)_2$, have most chance to stick on surface, but the most volatile precursor, $Fe(acac)_3$, have less chance to stay on the surface, causing nonstoichiometry in films. Subsequent process developing efforts and experimental result show that stoichiometric $(Cu_xZn_{1-x})Fe_2O_4$ films can be deposit by introducing excessive amount of $Fe(acac)_3$ precursor in the vapor phase. We also found that $Zn(acac)_2$ is difficult to deposit upon copper oxide islands, but $Cu(acac)_2$ can adsorb and deposit easily on any kind of surface. The developed solution is replaced the original single stage precursor heating program (Fig.1) with three stage heating method (Fig2). AES and survey scan XPS results show deposited films have all elements in stoichiometric amount. XRD indicated that these CVD films were polycrystalline spinel ferrite, with grain orientations on (511), (400), (311), and (220) ($d_{spacing}$=1.61Å, 2.10 Å, 2.52 Å, 2.97 Å), with (311) being the preferential orientation for $(Cu_{1-x}Zn_x)Fe_2O_4$ films (Fig.3). The grain size varies with deposition temperature and achieves largest size for film deposited at 400°C (Fig.4). SQUID (Fig.5) results showed that CVD films are ferrimagnetic, with low coercive field, (2.5~8 Oe), initial magnetic permeability (μi) $3.2x10^{-4}$emu/Oe, saturation magnetization (Ms), $3x10^{-5}$ – $2.7x10^{-3}$ emu, and remanence magnetization (Mr), $1.6x10^{-6}$– $2.6x10^{-4}$ emu. SQUID results also show that both μi and Ms are related to grain size, and deposition temperature. μi and Ms achieve the largest value at 400°C film (Fig.6).

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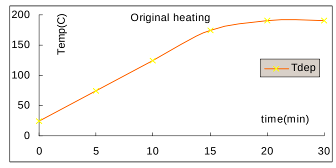
1. Yuneng Chang, C. Huang, P. Tsai, W. Wang, Z. Yiu , "Copper Zinc Ferrite Thin Films Prepared by Metal Organic Chemical Vapor Deposition Technology", INTERNATIONAL CHEMICAL CONFERENCE, TAIWAN, ICCT-2002, (2002).
2. Yuneng Chang, H. Tsen, M. Chen, and Menghsiu Lee,"A Study on The MOCVD Mechanism of Inverse Spinel Copper Ferrite Thin Films", 2001 MRS Meeting symposium U1.9, (2001).



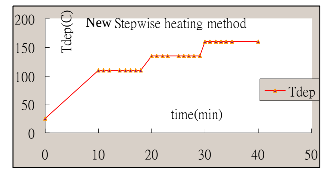


Fig. 1 original heating method Fig. 2 New heating method

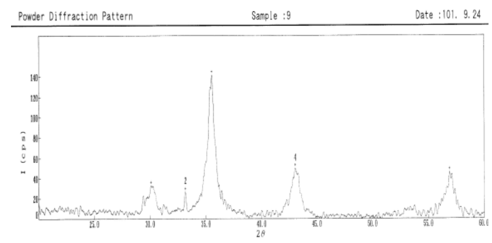
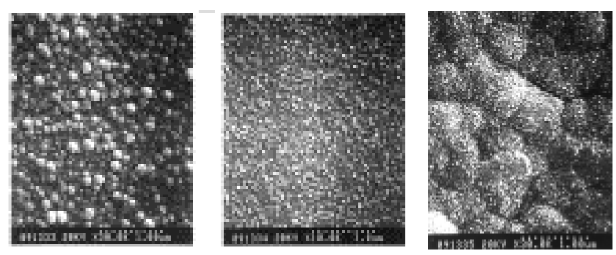


Fig. 3 XRD of $(Cu_xZn_{1-x})Fe_2O_4$ film at $[O_2]$ 50% 400℃



(A) high mag (A) low mag (B)
Fig. 4 SEM of $(Cu_xZn_{1-x})Fe_2O_4$ film at $[O_2]$ 50% and (A) 360℃ (B) 400℃

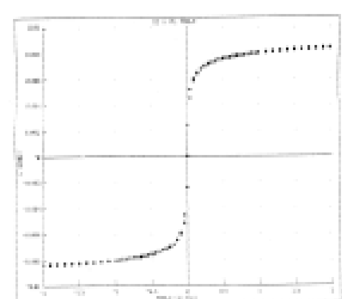


Fig.5 SQUID of $(Cu_xZn_{1-x})Fe_2O_4$ film at $[O_2]$ 50% 400℃

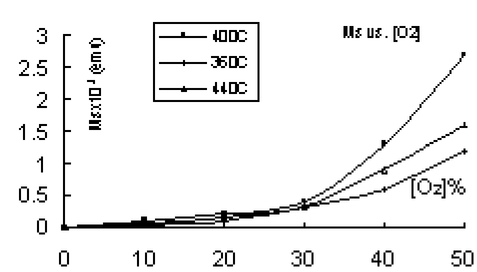


Fig.6 relationship between saturation magnetization of film and deposition temperature, oxygen concentreation

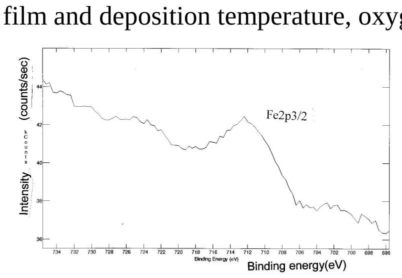


Fig. 7 Fe2p XPS from CVD film