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# 硝酸亜鉛六水和物と硝酸銅(II)三水和物を用いた溶液成長法による ZnO/CuO ヘテロ接合の形成

# Fabrication of ZnO/CuO Heterojunctions by Chemical Bath Deposition Using Zinc Nitrate Hexahydrate and Copper (II) Nitrate Trihydrate

愛媛大院理工[1]，マレーシア工科大 MJIIT[2]，愛媛大工[3]

○寺迫 智昭[1]，ヌル・アシキン・ハンバリ[2]，ヌルル・アズヤティー・ジャヤ[2]，一ノ谷 光[3]，
平松 知晃[3]，福井 一平[3]，真鍋 豪[3]，脇坂 俊也[3]，アブドル・マナフ・ハシム[2]，白方 祥[1]

Grad. School Sci. & Eng., Ehime Univ.[1], MJIIT, Univ. Teknol. Malaysia[2], Fac. Eng., Ehime Univ.[3]

○Tomoaki Terasako[1], Nur Ashikyn Hambali[2], Nurul Azzyaty Jayah[2], Hikaru Ichinotani[3],
Tomoaki Hiramatsu[3], Ippei Fukui[3], Goh Manabe[3], Toshiya Wakisaka[3],
Abdul Manaf Hashim[2], Sho Shirakata[1]

E-mail: terasako.tomoaki.mz@ehime-u.ac.jp

Introduction　Zinc oxide (ZnO) with a hexagonal wurtzite structure has the band gap energy of 3.37 eV, which is transparent to visible light, and exhibits *n*-type conduction. On the other hand, cupric oxide (CuO) with a monoclinic structure has the band gap of ~1.35 eV and exhibits *p*-type conduction. Recently, Saji *et al.* have reported the successful growth of the CuO/i-ZnO/ZnO:Al heterojunction solar cell with the energy conversion efficiency of $3\times10^{-4}$ % by rf magnetron sputtering [1]. In our previous study, the cone-shaped ZnO nanorods (NRs)/CuO heterojunctions were successfully grown on the Au seed layers by chemical bath deposition (CBD) using zinc acetate dihydrate and copper (II) nitrate trihydrate [2]. The current density (J) – voltage (V) curves for the heterojunctions exhibited rectifying behaviors, but the photovoltaic effects were negligible weak. In the present paper, the fabrication of *n*-ZnO/*p*-CuO heterojunctions by CBD growth technique using zinc nitrate hexahydrate [$Zn(NO_3)_2 2H_2O$] and copper (III) nitrate trihydrate [$Cu(NO_3)_2 3H_2O$], and controllability of their structural, optical and electrical properties will be discussed.

Sample preparation　The commercial Au/Ti/Si(100) wafer was used as a substrate material. The CuO layers were prepared by CBD using 0.05 M aqueous solution of $Cu(NO_3)_2 3H_2O$ whose pH value was adjusted to be approximately 10 by the use of ammonia solution. For the CuO growth, the bath temperature ($T_B$) was 90 ℃. After the deposition process, the CuO films were annealed in the air at 250 ℃ for 10 min. The CBD growth of the ZnO layers was done by using the mixed aqueous solution of $Zn(NO_3)_2 2H_2O$ and hexamethylenetetramine (HMT). The concentration of the aqueous solution of $Zn(NO_3)_2 2H_2O$ was varied in the range of 0.01-0.1 M and the molar ratio of HMT to $Zn(NO_3)_2 2H_2O$ was 1:1. The $T_B$ for the ZnO layers was maintained at 90 ℃. Growth time was changed in the range of 60-180 min.

Results and Discussion　Fig. 1 shows cross-sectional SEM images of the heterojunctions with the ZnO nanorods (NRs) layers grown from the aqueous solutions of $Zn(NO_3)_2 2H_2O$ with the different concentrations. At the concentration of 0.01 M, the surface of the CuO layer is studded with the cone-shaped ZnO NRs. For the concentrations higher than 0.05 M, however, the topmost surfaces of the heterojunctions are covered completely with the ZnO NRs. With increasing the concentration of the aqueous solution, the shapes of the ZnO NRs change from cones to cylinders and the intervals between the neighboring NRs become smaller, resulting in the formation of the vertically aligned NRs. On the other hand, no remarkable change can be observed among the heights of the ZnO NRs grown from the solutions of 0.05, 0.07 and 0.1 M. Therefore, we can expect that the concentration of the aqueous solution is one of the important factors for controlling the growth rate in the radial direction rather than that in the axis direction.

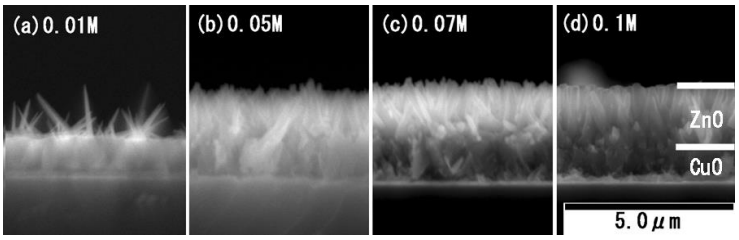


Fig. 1. Cross-sectional SEM images of ZnO/CuO heterojunctions. The ZnO NRs layers were grown from the aqueous solutions with different concentrations (a: 0.01M, b: 0.05M, c: 0.07M and d: 0.1M). Growth time was 60min.

References　[1] K. J. Saji *et al.* Phys. Status Solid A 210 (2013) 1386., [2] T. Terasako *et al.*, The 61st JSAP Spring Meeting, 17p-PG3-29 (2014).