

論文 / 著書情報  
Article / Book Information

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## 論文要旨

THESIS SUMMARY

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Department of, Graduate major in	材料	コース
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申請学位 (専攻分野) :	博士	(工学, Engineering)
Academic Degree Requested	Doctor of	
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要旨 (英文 800 語程度)

Thesis Summary (approx.800 English Words )

Opto-electronic devices are one of the essential components supporting our daily lives, such as display, solar cell, smart window and so on. Especially, n-type transparent amorphous oxide semiconductors (TAOS) have been widely investigated and are already applied to drive pixels of FPDs represented by organic light emitting diode (OLED) displays. However, developing next generation displays is hampered by the lack of p-type TAOS arising from the nature of ionic oxides. Conventional organic luminescent materials have several serious issues, i.e., chemical instability in the ambient condition and short lifetimes originating from the intrinsic nature of organic materials. To resolve these issues, I propose a materials design concept and develop the copper(I) iodide-based compounds that possess promising opto-electrical properties such as high mobility p-type semiconducting nature or highly efficient luminescence. Copper(I) iodide (CuI) is already known as a simple binary compound consisting of two abundant and non-toxic elements. Cuprous ion ( $\text{Cu}^+$ ) has a closed (i.e.,  $d^{10}s^0$ ) shell configuration whose energy is almost comparable to those of the iodine 5p orbital. I 5p has much larger orbital radius ( $>200$  pm) than O 2p orbital ( $\sim 126$  pm). As a result, CuI exhibits a high hole mobility ( $\sim 40$   $\text{cm}^2/\text{Vs}$  in single crystal) with a direct band gap ( $\sim 3.1$  eV). Moreover, its crystallographic dimensionality of [Cu-I] tetrahedral site is easily controllable by introducing Cs ion, resulting in the adjustment of opto-electrical properties, such as PLQY, bandgap, mobility and carrier concentration. This thesis describes opto-electrical properties for application to next generation optoelectronics by design of copper (I) iodide-based compounds.

In order to obtain p-type transparent amorphous semiconductor (TAS), in Chapter 2, a material design concept for p-type TAS materials is proposed utilizing the pseudo s-orbital nature of spatially spreading iodine 5p orbitals and amorphous Sn-containing CuI (a-CuSnI) thin film is reported as an example. The resulting a-CuSnI thin films fabricated by spin coating at low temperature ( $140^\circ\text{C}$ ) have a smooth surface. The Hall mobility increases with the hole concentration and the largest mobility of  $\sim 9$   $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$  is obtained, which is comparable with that of conventional n-type TAOS.

In Chapter 3, this chapter describes a new strategy for p-type transparent semiconductor (TS) with low carrier concentration and new luminescent material with unique optical properties.  $\text{CsCu}_2\text{I}_3$  with one dimensionally connected  $\text{CuI}_4$  unit has smaller dispersion of VBM than CuI, whereas the formation energy

for Cu-vacancy has higher than CuI. Thus, I expected low carrier concentration suitable for semiconductor application would be realized. The material synthesized by inverse temperature crystallization (ITC) method showed  $\sim 2.34 \times 10^{13} \text{ cm}^{-3}$  with Hall mobility of  $\sim 0.34 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ . Furthermore, novel optical properties, such as broadband and large Stokes shift luminescent material with PLQY of  $\sim 8\%$  was obtained.

I found 1D  $\text{CsCu}_2\text{I}_3$  as a yellow luminescent material. However, 1D  $\text{CsCu}_2\text{I}_3$  is still insufficient phosphor due to low PLQY in solid state. To resolve this problem, in Chapter 4, I thought copper (I) iodide-based compound with zero-dimensional photoactive site,  $\text{Cs}_3\text{Cu}_2\text{I}_5$ , would be a new luminescent material with high PLQY.  $\text{Cs}_3\text{Cu}_2\text{I}_5$  synthesized by slow vapor saturation of an antisolvent (VSA) method showed a high PLQY of 90% and good air-stability. Each Cu-I photoactive site is separated by Cs ions; this arrangement may be considered a host-guest system. An embedded core/shell structure is effective for enhancing the PL by confining the exciton into the photoactive site; this accounts for the large exciton binding energy ( $\sim 490 \text{ meV}$ ). For practical applications, blue electroluminescence of Pb-free halides was demonstrated using solution-derived 0D  $\text{Cs}_3\text{Cu}_2\text{I}_5$  thin film. Furthermore, I succeeded in fabricating a white luminescent thin film by one-step spin-coating method using the mixed precursor solution of 1D  $\text{CsCu}_2\text{I}_3$  (yellow) and 0D  $\text{Cs}_3\text{Cu}_2\text{I}_5$  (blue) to achieve full coverage in the visible range of the spectrum.

In summary, I established new material design concepts to realize next generation display with high opto-electrical properties, environmental reliability and non-toxic. It was demonstrated that copper(I) iodide-based compounds have great potential as new opto-electrical materials for optoelectronic devices. Consequently, this thesis will open up the infinite possibility of copper(I) iodide-based compound for the high-end optoelectronic applications.

備考：論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。

Note : Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1 copy of 800 Words (English).

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