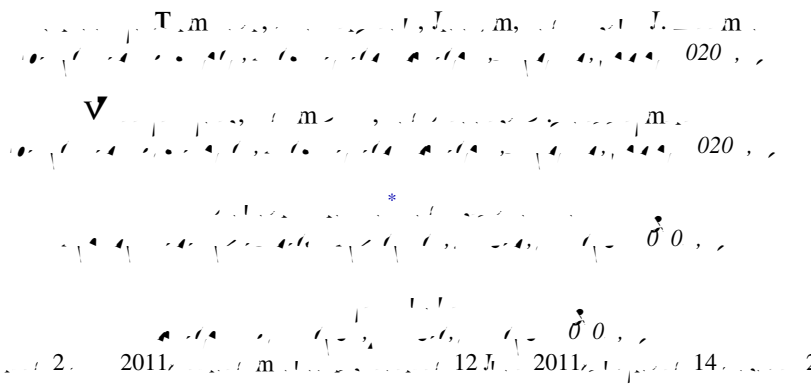


**Using design principles to systematically plan the synthesis of hole-conducting transparent oxides: Cu<sub>3</sub>VO<sub>4</sub> and Ag<sub>3</sub>VO<sub>4</sub> as a case study**



The synthesis of hole-conducting transparent oxides (TCOs), such as ZnO, SnO<sub>2</sub>, and In<sub>2</sub>O<sub>3</sub>, is a challenging task because of their wide bandgaps and low carrier concentrations. In this work, we have systematically designed and synthesized hole-conducting transparent oxides (TCOs) based on the design principles of the synthesis of hole-conducting transparent oxides (TCOs). The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>. The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>. The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>. The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>. The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>.

**I. DESIGN PRINCIPLES OF p-TYPE TRANSPARENT CONDUCTING OXIDES**

The synthesis of hole-conducting transparent oxides (TCOs) is a challenging task because of their wide bandgaps and low carrier concentrations. In this work, we have systematically designed and synthesized hole-conducting transparent oxides (TCOs) based on the design principles of the synthesis of hole-conducting transparent oxides (TCOs). The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>. The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>. The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>. The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>. The design principles are based on the synthesis of hole-conducting transparent oxides (TCOs) with a bandgap of 3.0 eV and a carrier concentration of 10<sup>19</sup> cm<sup>-3</sup>.





**m** **f** **m** **f**  
**m**

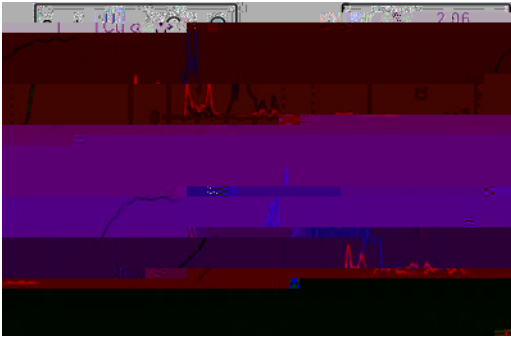


Fig. 4. (Color online) Band structure of  $\text{Cu}_2\text{O}$  and  $\text{Ag}_2\text{O}$  along high-symmetry directions. The conduction bands (CB) and valence bands (VB) are shown. The energy gap is indicated by the vertical line. The conduction band minimum (CBM) for  $\text{Cu}_2\text{O}$  is at the  $\Gamma$  point, while for  $\text{Ag}_2\text{O}$  it is at the  $L$  point. The valence band maximum (VBM) for both is at the  $\Gamma$  point. The energy gap for  $\text{Cu}_2\text{O}$  is approximately 2.1 eV, and for  $\text{Ag}_2\text{O}$  it is approximately 2.7 eV.

**C. Band-structure properties of  $\text{Cu}_2\text{O}$  and  $\text{Ag}_2\text{O}$ : Optical properties of the binaries**

Figure 4 shows the band structure of  $\text{Cu}_2\text{O}$  and  $\text{Ag}_2\text{O}$  along high-symmetry directions. The conduction bands (CB) and valence bands (VB) are shown. The energy gap is indicated by the vertical line. The conduction band minimum (CBM) for  $\text{Cu}_2\text{O}$  is at the  $\Gamma$  point, while for  $\text{Ag}_2\text{O}$  it is at the  $L$  point. The valence band maximum (VBM) for both is at the  $\Gamma$  point. The energy gap for  $\text{Cu}_2\text{O}$  is approximately 2.1 eV, and for  $\text{Ag}_2\text{O}$  it is approximately 2.7 eV.

The structure of  $\text{Ag}_3\text{VO}_4$  is based on  $\text{VO}_4$  tetrahedra. The  $\text{VO}_4$  tetrahedra are arranged in a chain along the  $c$ -axis. The  $\text{Ag}^+$  ions occupy the interstitial sites between the  $\text{VO}_4$  tetrahedra. The structure is described by the space group  $C2/m$  (No. 12). The unit cell parameters are  $a = 0.456$  nm,  $b = 0.456$  nm,  $c = 0.456$  nm. The structure is shown in Figure 4.

The structure of  $\text{Cu}_3\text{VO}_4$  is based on  $\text{VO}_4$  tetrahedra. The  $\text{VO}_4$  tetrahedra are arranged in a chain along the  $c$ -axis. The  $\text{Cu}^{2+}$  ions occupy the interstitial sites between the  $\text{VO}_4$  tetrahedra. The structure is described by the space group  $C2/m$  (No. 12). The unit cell parameters are  $a = 0.456$  nm,  $b = 0.456$  nm,  $c = 0.456$  nm. The structure is shown in Figure 4.

**C. Crystal structures of  $\text{Ag}_3\text{VO}_4$  and  $\text{Cu}_3\text{VO}_4$**



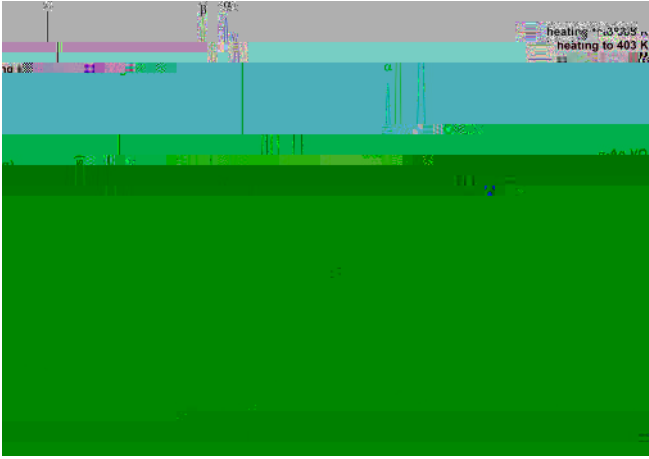


Figure 1 shows the XRD patterns of  $\text{Cu}_3\text{VO}_4$  synthesized at different temperatures. The patterns are indexed to the  $\text{Cu}_3\text{VO}_4$  phase (JCPDS card no. 03-065-0322). The patterns show the presence of multiple phases of  $\text{Cu}_3\text{VO}_4$  with different crystallite sizes. The heating cycles to 330 K and 403 K are indicated in the legend. The pattern at 330 K shows a broad peak at  $2\theta = 30^\circ$ , which is attributed to the presence of  $\text{Cu}_2\text{O}$ . The pattern at 403 K shows a broad peak at  $2\theta = 30^\circ$ , which is attributed to the presence of  $\text{Cu}_2\text{O}$ .

**B. Intrinsic defects, hole generation, and hole density in  $\text{Cu}_3\text{VO}_4$  and  $\text{Ag}_3\text{VO}_4$**

$$\text{Cu}_3\text{VO}_4 \rightleftharpoons (\text{Cu}_{3-x}\text{V}_1\text{O}_{4-x}) + x\text{O} + x\text{V}_2\text{O}_5$$





m f m f  
 f m m f m m  
 m f f f m f  
 f f ( ) f m  
 m m 32,33 \*

(

$f$   $m$   $f$   
 $4$   $m$   $f$   $m$   
 $0$   $m$   $f$   
 $m$   $f$   $m$   
 $( )$   $f$   $m$   
 $f$   $m$   $f$   
 $( )$   $f$   $m$   $f$   
 $f$   $m$   $f$   $m$   $f$   
 $( )$   $f$   $m$   $f$   $m$   $f$





