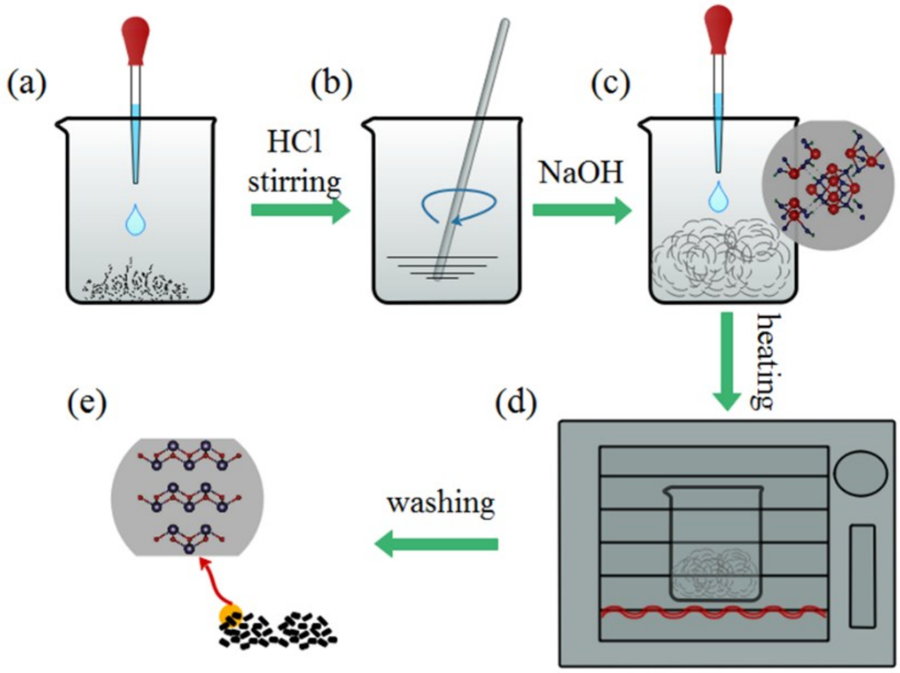


Fig. S1 Experimental flow chart of synthesis of large-size polycrystalline stannous oxide by sol-gel method

The white sol-gel intermediate is $Sn_6O_4(OH)_4$, as shown in Fig. S1c. and its crystal structure belongs to monoclinic system. Tuning the pH value of the solution at 0.8 by dropping hydrochloric acid into the solution can effectively inhibit the hydrolysis of $Sn^{2+}$. As the pH of the solution increases from 0.8 to 12.0, a white sol-gel product appears in the solution, which can be expressed by the reaction equation (1).

$$6Sn^{2+} + 8OH^{-} = Sn_6O_4(OH)_4 + 4H^{+} \qquad (1)$$

Then, by maintaining constant solution temperature, $Sn_6O_4(OH)_4$ is thermally decomposed to form tetragonal SnO, which can be expressed by the reaction equation (2).

$$Sn_6O_4(OH)_4 = 6SnO + 2H_2O \qquad (2)$$

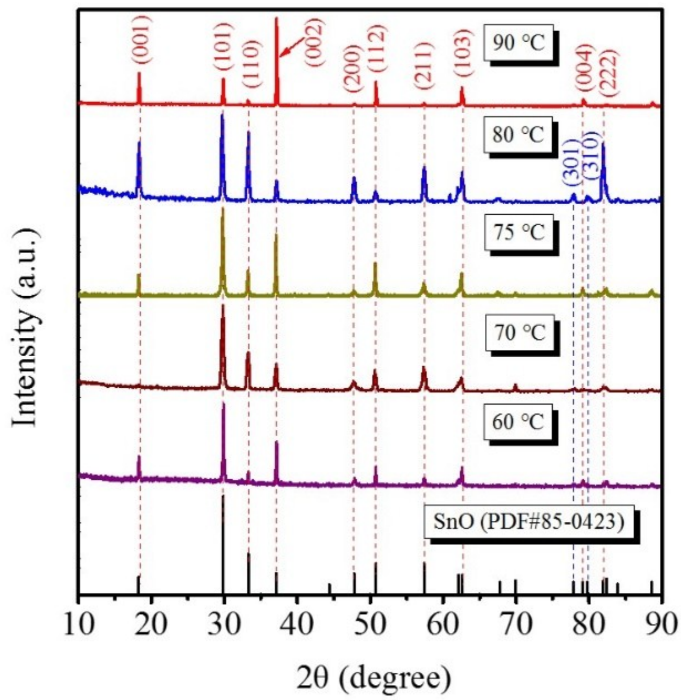

Fig. S2 XRD patterns large-size polycrystalline SnO which synthesized in different temperature.

Fig. S2 presents XRD patterns of large-size polycrystalline SnO which synthesized in 60 ℃, 70 ℃, 75 ℃, 80 ℃ and 90 ℃, respectively. The XRD peak position of the SnO crystal grown at each temperature corresponds to the peak position on the standard card (SnO, PDF#85-0423). The sharp peaks imply good crystallinity. Among them, at 75 ℃, the SnO crystal has the strongest preference growth along the (101) crystal plane.

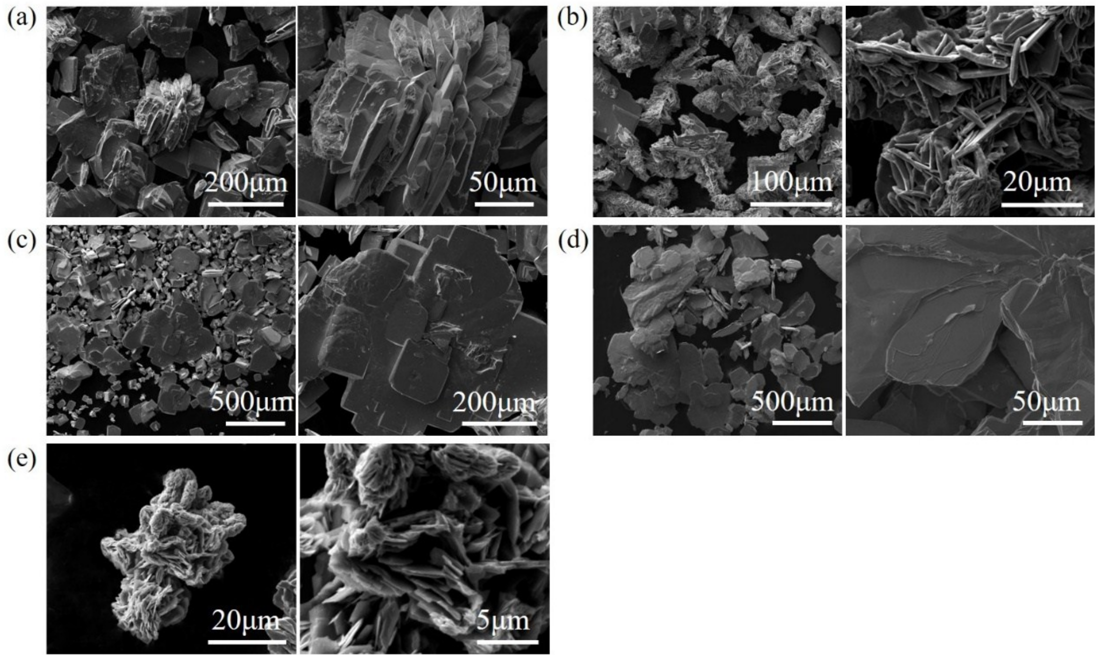


Fig. S3 SEM images of large-size polycrystalline SnO which, (a) synthesized in 60℃, (b) synthesized in 70 ℃, (c) synthesized in 75 ℃, (d) synthesized in 80 ℃, (e) synthesized in 90 ℃.

Fig. S3 presents SEM images of large-size polycrystalline SnO which synthesized in different temperature. Obviously, the size and morphology of SnO crystals are anisotropy at different temperature. Among them, at 75 ℃, the crystal size is exceeding 700 μm, which can reach the submicron level. Therefore, performance characterizations in the main text were performed using SnO crystals grown at 75 ℃.

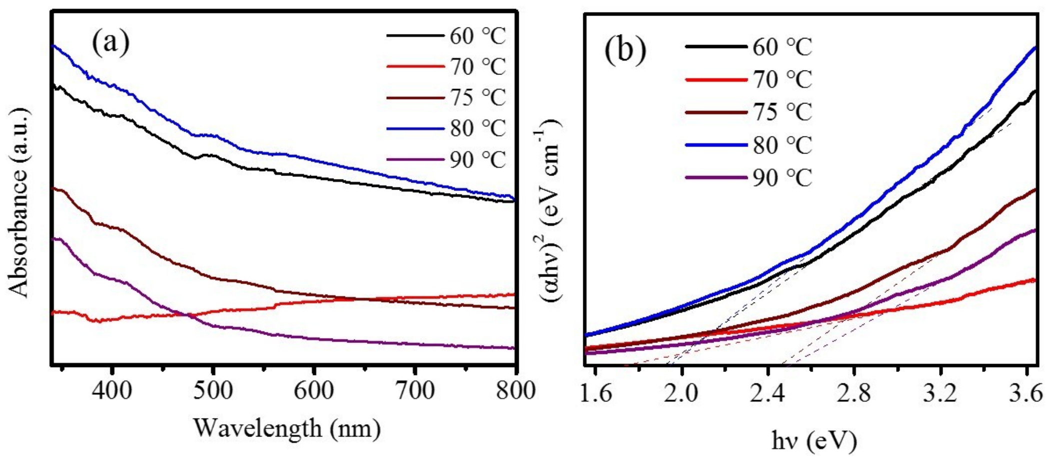


Fig. S4 (a) UV-Vis absorption spectra and (b) graphical evaluation of the obtained optical band gaps of large-size polycrystalline SnO which synthesized in different temperature.

Fig. S4 presents solid-state absorption spectra and graphical evaluation of the obtained optical band gaps of SnO in different temperature. As shown in Figure (a), the crystalline material has a clear absorption peak in the near blue light band, which its intensity tends to largest at 60 ℃, but shows no significant change at 70 ℃. As shown in Figure (b), the obtained optical band gaps of large-size polycrystalline SnO which synthesized in 60 ℃, 70 ℃, 75 ℃, 80 ℃, 90 ℃ is also different, which are 1.93eV, 1.78eV, 2.46eV, 1.97eV, 2.51eV respectively.