Supplementary Information

Compositional characterisations were firstly carried out on the as-deposited ZrO$_x$ films with O$_2$ flow rate ranging from 6 sccm to 20 sccm by EDX as shown in Figure S1. Films with a large thickness of 1 μm were deposited directly onto Si wafers to minimise the influence from the substrate. Sub-stoichiometric ZrO$_{2.4}$ films were obtained when the O$_2$ flow rate was below 12 sccm and the composition changes from ca. ZrO$_{1.2}$ to ZrO$_{1.8}$ with increasing flow rate from 6 sccm to 10 sccm. The O:Zr ratio then saturates when more O$_2$ is introduced into the chamber and stoichiometric ZrO$_2$ films were obtained.

Figure S1. (a) EDX quantitative O:Zr ratio of the as-deposited ZrO$_x$ films with different O$_2$ flow rates from 6 sccm to 20 sccm.

Figure S2 shows the XRD patterns of the ZrO$_x$ films deposited with different O$_2$ flow rate. Under the 6 sccm O$_2$ flow rate, the as-deposited ZrO$_x$ film is found to be amorphous, characterised by the large bump distributed in a wide range. Further oxidization of the ZrO$_x$ film resulted in higher film crystallinity and the films were gradually transformed into the tetragonal phase when the O$_2$ flow rate reaches 10 sccm. Another phase change to the monoclinic structure was also observed at higher O$_2$ flow rates (≥ 12 sccm) when stoichiometric ZrO$_2$ films were obtained.

Figure S2. XRD patterns of ZrO$_x$ films deposited at different O$_2$ flow rates from 6 sccm to 20 sccm.