Supplementary Information

For VB XPS spectra comparison 10 nm thermal-ALD  $Ta_2O_5$  films were also grown via the  $Ta(OC_2H_5)_5/H_2O$  process, at the same substrate temperature on the same substrates. VB XPS spectra of active H\* assisted ALD  $TaO_x$  and thermal-ALD  $Ta_2O_5$  films are Figure1(Supplementary Information).

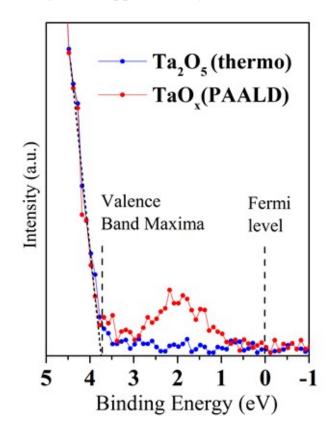


Figure 1 Valence band XPS spectra measured from 10 nm PAALD TaOx ( $t_{pl} = 20$ sec, R = 7% and  $t_{pr} = 0.5$  sec and 10 nm thermal ALD Ta<sub>2</sub>O<sub>5</sub> films.

It can be seen that thermal ALD  $Ta_2O_5$  film revealed no electronic states at the energies higher than VBM. In other words, there no defect states in the band gap in the stoichiometric thermal-ALD  $Ta_2O_5$  film. In contrast, a peak at ~ 1.8 eV higher than VBM is clearly distinguishable at the VB spectrum of PAALD TaOx film.