## **Supporting Information**

Interface engineering for stable chemical structure of oxidized-black phosphorus *via* self-reduction in  $AlO_x$  atomic layer deposition

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**Fig. S1** Atomic force microscopy characteristics of oxidized black phosphorus (BP) as a function of air exposure time. Time-dependence AFM images of the exfoliated BP surface from 4 h to 48 h under ambient conditions.



**Fig. S2** X-ray photoelectron spectroscopy characteristics of oxidized BP as a function of air exposure time. Time-dependent XPS of (a) raw, and (b) fitted spectra of P 2p core level of oxidized BP as a function of air exposure time (4 h to 48 h); (c) The peak area ratio of  $P_xO_y$  from the fitted XPS spectra (b).



**Fig. S3** Comparison of chemical states of  $ALD-AlO_x$  and  $HfO_x$  on oxidized BP at surface exposure times. The fitted XPS spectra of the P 2*p* core level prior to (oxidized BP) and following (a) 30 and (b) 50 deposition cycles as a function of air exposure time 26 h and 72 h.



Fig. S4 The cumulative distribution of change in the oxidized phosphorus species ( $P_xO_y$ ) measurements for statistical analysis in  $AlO_x$  grown on oxidized BP (24 h) under the same thickness (30 cycles).



Fig. S5 The stoichiometry and chemical state of the  $P_xO_y$  before and after ALD process. After primary vacuum and N<sub>2</sub> purging process after primary vacuum in ALD, the binding energy and peak ratio of phosphorus species ( $P_xO_y$ ) was not changed, compared with that of air exposure for 24 h.



**Fig. S6** Comparison of cumulative electrical measurement of  $Al_2O_3/AlO_x/oxidized BP$  (24 h) FET device at different exposure time (after 50, 60 and 70 days). The (a)  $I_d$ - $V_{gs}$  characteristics for the carrier transport data and other values (b), and (c) of extracted mobility,  $D_{it}$ ,  $V_{th}$ , and SS are well consistent with those of initial measurement of FET device.



**Fig. S7** Back gating electrical characteristics of the ALD-Al<sub>2</sub>O<sub>3</sub>/AlO<sub>x</sub> and HfO<sub>2</sub>/HfO<sub>x</sub> on oxidized BP (surface exposed for 24 h)/SiO<sub>2</sub>-300 nm/highly doped Si for field-effect transistors (FETs). The (a) and (b) I<sub>d</sub>-V<sub>gs</sub> transfer curves were obtained by back gating FET device (gate voltage (V<sub>g</sub>) ranges of  $\pm 60$  V at a fixed drain voltage (V<sub>sd</sub>) of 0.1 V) and (c) gate leakage current and (d) transconductance.