Supporting Information

Effect of Large Work Function Modulation of MoS\textsubscript{2} by Controllable Chlorine Doping Using a Remote Plasma

Ki Hyun Kim,\textsuperscript{a} Ki Seok Kim,\textsuperscript{a,e} You Jin Ji,\textsuperscript{a} Inyong Moon,\textsuperscript{b} Keun Heo,\textsuperscript{c} Dong-Ho Kang,\textsuperscript{c} Kyong Nam Kim,\textsuperscript{d} Won Jong Yoo,\textsuperscript{b} Jin-Hong Park,\textsuperscript{c} and Geun Young Yeom\textsuperscript{a,b,*}

\textsuperscript{a}School of Advanced Materials Science and Engineering, Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do 16419, Republic of Korea.
\textsuperscript{b}SKKU Advanced Institute of Nano Technology (SAINT), Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do 16419, Republic of Korea.
\textsuperscript{c}School of Electronic and Electrical Engineering, Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do 16419, Republic of Korea.
\textsuperscript{d}School of Advanced Materials Science and Engineering, Daejeon University, Yongun-dong, Dong-gu, Daejeon 34520, Korea.
\textsuperscript{e}Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, MA, USA.

*Corresponding Author: gyyeom@skku.edu
**Figure S1.** Schematic drawing of a 13.56 MHz remote ICP system. Double mesh-grid was installed at the center of the chamber to extract reactive radicals only. Almost all of ions were filtered at the grounded mesh grid.
Figure S2. AFM profiles of (a) before and (b) after chlorine doping for 60 s. The thickness of ~2.3 nm is well accord with Raman spectroscopic data in Figure 2a. Any thickness change of MoS$_2$ flake was not observed after the chlorine radical treatment.
Figure S3. (a) EDS mapping image of MoS$_2$ in TEM after the chlorine radical doping for 30s. (b) High resolution TEM image of MoS$_2$. The inset shows a fast Fourier transform (FFT) image as a moiré pattern that appears for the crystalline multilayer-MoS$_2$[1]. (c) The mapping images of each element (Mo, S, and Cl) was illustrated separately. Surface distribution of Cl atoms on MoS$_2$ after the treatment was observed using an energy dispersive spectroscopy (EDS). As shown in (a), signals of Mo, S and Cl were observed on a surface of MoS$_2$ after treatment for 30 s. We observed no significant disorder of MoS$_2$ lattice after the chlorine radical doping, which was confirmed through high resolution TEM image in (b). Elemental images of Mo, S and Cl in (a) were separately illustrated in (c).
Figure S4. Photoresponsivity of pristine and chlorine doped MoS$_2$ FETs measured at $\lambda = 520$ nm while varying gate voltage from -30 to 30 V. The photoresponsivity of MoS$_2$ FETs was measured with gate bias (from $V_{gs} = -30$ to $+30$ V) at $\lambda = 520$ nm. The photoresponsivity of pristine MoS$_2$ FET showed the saturation point at $V_{gs} = 0$ V. For the chlorine doped MoS$_2$ FET, however, gradual increase of photoresponsivity was observed until $V_{gs} = 30$ V.
Figure S5. XPS analysis of trilayer MoS$_2$ grown by Chemical Vapor Deposition (CVD) process with various S/Mo ratio from 2.02 to 1.83. (a) The binding energy of two Mo 3d peaks for chlorine doped CVD MoS$_2$ having S/Mo ratio from 2.02 to 1.83. The MoS$_2$ having S/Mo ratio close to stoichiometric number of 2 showed relatively small blue-shift of Mo peaks because little substitutional doping of Cl atoms was occurred. However, those having lower S/Mo ratio under 2 have more probability for Cl atoms to bind on S vacancy sites resulting in more noticeable blue-shift of Mo peaks. (b) Narrow scan XPS data of Mo 3d measured on CVD MoS$_2$ having S/Mo ratio of 1.83 [the blue box in Figure (a)] before and after chlorine radical treatment for 30 s.

Reference