A New Purification Strategy of 2D $MoS_{2(1-x)}Se_{2x}$ Alloys for Fabrication of Efficient Photodetectors and Photo(electro)catalysts

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Two-dimensional Semiconducting Transition Metal Dichalcogenide (2D-STMD) based semiconductors have emerged as promising materials for future spintronic and optoelectronic applications, including photodetectors and transistors. Transferring monolayer STMDs and their alloys to target substrates is necessary to fabricate devices for such purposes. Unfortunately, current post-transfer methods struggle to completely remove the unwanted contamination residues during wet-transfer processes, adversely affecting material quality and intrinsic properties. The effect of ethanol cleaning on a qualitative and quantitative assessment of adsorbates is attempted to explain based on the atomic-resolution high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) image analysis supporting with density functional theory (DFT) calculations, showcasing ultraclean material structures. We try to estimate the unidentified molecular level adsorbates in the proximity of transition metal (TM) and chalcogenide atomic sites, which are tentatively assigned as OH, H_2O and O2-related adsorbates from HAADF-STEM Gaussian line shape fitting of atomic intensity simulation and corresponding adsorption energy computational values after ethanol treatment of $MoS_{2(1-x)}Se_{2x}$ (MSSE) alloy. Photodetector device measurements revealed a remarkable ~ 90% enhancement in the ultraclean samples' persistent photocurrent and photoresponse values, significantly boosting material photoresponsivity. DFT calculations on adsorption energy and density of electronic states were also conducted to validate our experimental findings.

References

[1] **Ravi K. Biroju*,** Dipak Maity, Viliam Vretenár, Ľubomír Vančo, Rahul Sharma, Mihir Ranjan Sahoo, Jitendra Kumar, Gayathri G, and Tharangattu N. Narayanan, Saroj Kumar Nayak, Materials Today Advances 22 (2024) 100504
[2] Dipak Maity, Rajesh Kumar Yadav, Adi Levi, Rahul Sharma, Emanuel Ber, Eilam Yalon, **Ravi K. Biroju**, Viliam Vretenár and Tharangattu N. Narayanan*, and Doron Naveh* Small Methods, https://onlinelibrary.wiley.com/doi/10.1002/smtd.202401938;
[3] Rahul Sharma, Henry Nameirakpam, David Muradas Belinchón , Prince Sharma , Ulrich Noumbe, Daria Belotcerkovtceva , Elin Berggren , Viliam Vretenár , Ľubomír Vančo , Matúš Maťko , **Ravi K. Biroju**, Soumitra Satapathi , Tomas Edvinsson , Andreas Lindblad and M. Venkata Kamalakar ACS Applied Materials & Interfaces 2024, doi/10.1021/acsami.4c07028
[4] **Ravi K. Biroju**, Deya Das, Rahul Sharma, Shubhadeep Pal, Larionette P. L. Mawlong, Kapil Bhorkar, Abhishek K. Singh, P. K. Giri, Tharangattu N. Narayanan, ACS Energy Lett. 2017, 2, 1355–1361

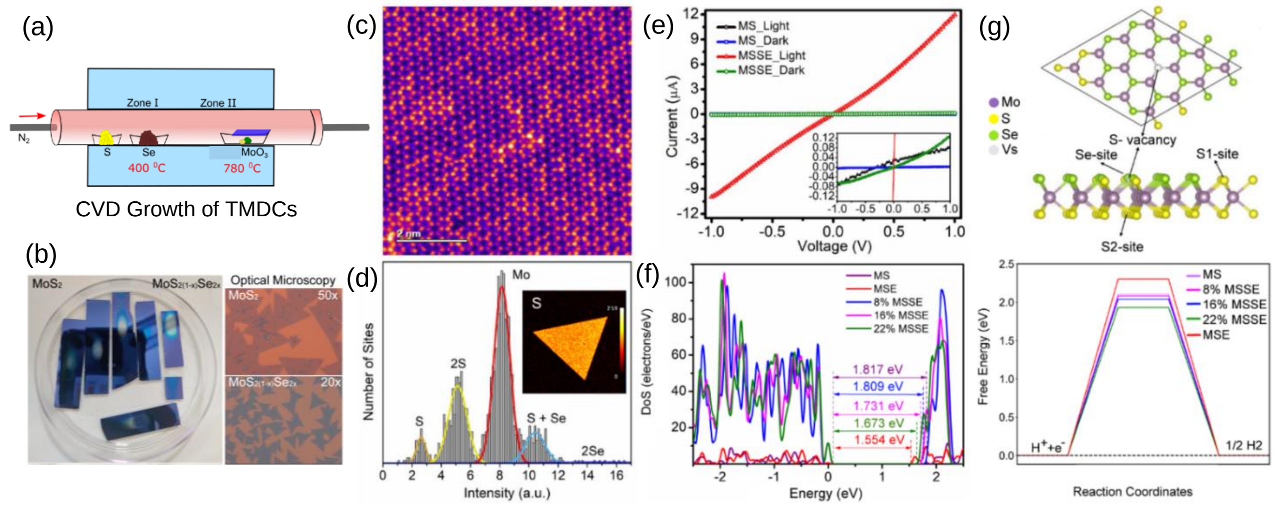

***Figure 1**: (a) The schematic showing a two-zone furnace used as a typical chemical vapour deposition (CVD) for growing engineered TMDCs atomic layers. (b) Digital and optical images of MoS_2 and $MoS_{2(1-x)}Se_{2x}$ atomic layers grown on Si/SiO_2 substrates. (c)Atomic resolution HAADF-STEM image of a (a) $MoS_{2(1-x)}Se_{2x}$ (MSSE) alloy. (d) Atomic site intensity histogram analysis of MSSE showing the distribution of different atomic sites such as Mo, S_2, S+Se and S etc. and inset shows the* AES map of S element in MSSE. *(e) Dark and light I-V characteristics from MSSE alloy using green LED (l_{exc} = 532nm). (f) Electronic density of states in pristine MS, MSE, 8, 16 and 22 Se % MSSE structure and corresponding energy band gaps are shown in the inset. (g) An atomic model showing in-plane and out-of-plane view of 22 % MSSE alloy with single S-vacancy (Vs). Different inequivalent sites for H adsorptions are indicated. (h) Schematic of the transport measurement device setup (with SiO2 as a dielectric material, light blue, on a silicon wafer (blue)). Ti (red)/Au (yellow) metal contacts are made on top of MoS2 monolayer (Mo in green and sulfur in orange).*