Supporting Information

CVD Synthesis of $Mo_{(1-x)}W_xS_2$ and $MoS_{2(1-x)}Se_{2x}$ Alloy Monolayers

Aimed at Tuning the Bandgap of Molybdenum Disulfide

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Fig. S1 (a) The dark-field TEM image of a ML- $Mo_{(1-x)}W_xS_2$ nanosheet supported on holey carbon grid. The folds and black dots were produced during the transfer onto the grid. (b) HRTEM image taken from (a) and its SAED pattern (c) which demonstrates it is a single crystal with hexagonal structure.



Fig. S2 (a)-(b) Raman and PL spectrum of the representative sample ML- $Mo_{(1-x)}W_xS_2$ respectively. (c) EDS characterization of ML- $Mo_{(1-x)}W_xS_2$, TEM image of a nanosheet transferred to a grid and the corresponding 2D elemental mapping for the three detected elements: Mo, S, and W, respectively.



Fig. S3 (a) The dark-field TEM image of a ML-MoS_{2(1-x)}Se_{2x} nanosheet supported on holey carbon grid. The folds were produced during the transfer onto the grid. (b) HRTEM image taken from (a), the lattice fringe measured from it is 0.28 nm, corresponding to the (100) lattice plane. (c) The corresponding SAED pattern which showing only one set of six-fold symmetry diffraction spots, demonstrates it is a single crystal with hexagonal structure.



Fig. S4 (a)-(b) Raman and PL spectrum of the representative sample ML-MoS_{2(1-x)}Se_{2x} respectively. (c) EDS characterization of ML-MoS_{2(1-x)}Se_{2x}, TEM image of a nanosheet transferred to a grid and the corresponding 2D elemental mapping for the three detected elements: Mo, S, and Se, respectively. The data shows that the Se mole fraction [x, Se/(Se+S)] of ~0.2, indicating the composition of the nanosheet as $MoS_{2(0.8)}Se_{2(0.2)}$.



Fig. S5 (a) Raman spectrum of the ML-Mo_(1-x)W_xS₂ and ML-MoS_{<math>2(1-x)}Se_{2x} nanosheets excited with a 532 nm argon ion laser (Fig.3 (a) in paper). Part of the spectra indicated by an orange rectangle in (a) is shown in larger scale in (b).</sub>



Fig. S6 Raman peak intensity mapping at the MoS₂-like E for ML-Mo_(1-x) W_xS_2 (a) and MoS₂-like A_{1g} for ML-MoS_{2(1-x)}Se_{2x} (b) respectively.