## **Supporting information**

## Morphological Evolution of Mo<sub>x</sub>W<sub>1-x</sub>S<sub>2</sub>/MoS<sub>2</sub> Heterostructures Modulated with Growth Dynamic Process by One-step Chemical Vapor Deposition

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Table S1 Precursor quantity with different Mo:W

| Mo:W | $Na_2MoO_4•2H_2O(mg)$ | $Na_2WO_4 \bullet 2H_2O(mg)$ |
|------|-----------------------|------------------------------|
| 10:1 | 33.0                  | 4.5                          |
| 2:1  | 24.2                  | 16.5                         |
| 1:2  | 12.1                  | 33.0                         |
| 1:10 | 3.3                   | 45.0                         |



Fig. S1 (a) OM image, (b) Raman spectrum, and (c) PL spectrum of  $MoS_2$  grown by  $Na_2MoO_4$  solution

The Raman peak positions of  $MoS_2$  are 386.8 cm<sup>-1</sup> and 407.1 cm<sup>-1</sup>, respectively, the relative distance of the two peaks is 20.3 cm<sup>-1</sup>(see Fig.S1(b)). The result is in good agreement with the monolayer  $MoS_2$  grown based on VLS.<sup>1</sup> In addition, according to Fig.S1(c), the main PL

peak of  $MoS_2$  is 673 nm, corresponding to direct band gap of 1.84 eV.



Fig. S2 (a) OM image, (b) Raman spectrum, and (c) PL spectrum of  $WS_2$  grown by  $Na_2WO_4$  solution

The in-plane vibration peak  $(E_{2g}^1)$  of WS<sub>2</sub> is 351 cm<sup>-1</sup>, while the out-ofplane vibration peak  $(A_{1g})$  is 416 cm<sup>-1</sup> in Fig.S2(b), and the frequency difference between the two vibration peaks is shown to be ~65 cm<sup>-1</sup>. This result indicates that it is monolayer of WS<sub>2</sub>.<sup>2</sup> In Fig. S2(c), the PL peak of WS<sub>2</sub> is 630 nm and its band gap is 1.97 eV.



Fig. S3 AFM morphology and thickness of the heterostructure at Mo:W of (a) 2:1 and (b) 1:2

## References

S1. Chen Long, Lingyu Zang, Luhua Chen, Jinchao Wu, Chengming Jiang and Jinhui Song, *CRYSTENGCOMM*, 2021, **23**, 5337-5344.

S2. Hang Liu, Guopeng Qi, Caisheng Tang, Maolin Chen, Yang Chen, Zhiwen Shu, Haiyan Xiang, Yuanyuan Jin, Shanshan Wang, Huimin Li, Miray Ouzounian, Travis Shihao Hu, Huigao Duan, Shisheng Li, Zheng Han and Song Liu, *ACS APPL MATER INTER*, 2020, **12**, 13174-13181.