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Supporting information

High Mobility Nanometer-Thin Crystalline In-Sm-O Thin-Film Transistors via Aqueous Solution Processing

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1. Detailed analysis of the positive shift of V_{th}, the decrease of I_{off} and SS after Sm incorporation.

The V_{th} change can be explained by follow equation:[1]

$$n_e = \frac{C_{OX}(V_{GS} - V_{th})}{q}$$

Where n_e is the electron concentration, C_{OX} is the capacitance per unit area of SiO_2, and q is the electronic charge. From the formula above, the decline of n_e will lead to the positive shift of V_{th}. Sm doping could effectively suppress oxygen vacancies that are a source of n_e, and ultimately leads to the positive shift of V_{th}.

The change of SS could be explained by follow equation: [2]

$$SS = \left(\frac{q \cdot N_t \cdot}{C_{OX}} + 1\right)\frac{k_B T}{q\log(e)}$$

Where N_t is the trap density, q and $k_B T$ are the elementary electron charge and thermal energy. So the SS value of the device is related to the total trap density (N_t). The incorporation of Sm could suppress the oxygen vacancy-related defects and hence reduce the trap density, leading to the improvement of SS value.

The value of the off-state current (I_{off}) can be calculated by the following expression:[3]

$$I_{off} = \frac{\sigma t w V_{DS}}{L}$$

Where σ represents the conductivity of the In-Sm-O film, t denotes the thickness of In_2O_3, W and L are the channel width and length, respectively. The incorporation of Sm could suppress oxygen vacancies and reduce the conductivity of the semiconducting film, resulting in the decline of I_{off}.

1. X. Zhang, B. Wang, W. Huang, Y. Chen, G. Wang, L. Zeng, W. Zhu, M. J. Bedzyk, W. Zhang, J. E. Medvedeva, A. Facchetti and T. J. Marks, *J Am Chem Soc*, 2018, **140**, 12501-12510.
2. A. Abliz, L. Xu, D. Wan, H. Duan, J. Wang, C. Wang, S. Luo and C. Liu, *Applied Surface*

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3. L. C. Liu, J. S. Chen and J. S. Jeng, *Appl. Phys. Lett.*, 2014, **105**, 023509.

2. XRR patterns of In-Sm-O films with different Sm contents.

We use X-ray reflectivity (XRR) characterization to further confirm the thicknesses of In-Sm-O thin films, as shown in Fig. S1 and summarized in Table S1. The In-Sm-O films with 0%, 2%, 5%, 10% Sm contents are 4.72 nm, 4.88 nm, 5.40 nm, 5.84 nm, respectively. The thickness of 5% Sm doped In-Sm-O film is estimated to be 4-5 nm by cross-sectional TEM measurement. Therefore, the XRR results are in good agreement with the cross-sectional TEM measurement, confirming the correct thickness of In-Sm-O.

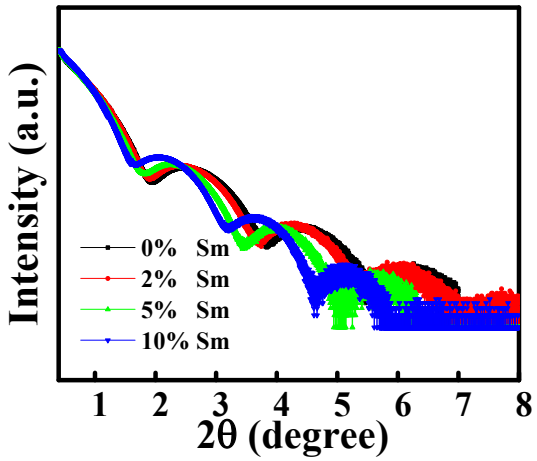

Fig. S1. XRR patterns of In-Sm-O films with different Sm contents.

Table S1 Thicknesses of In-Sm-O films with different Sm concentrations.

Sm concentrations	0%	2%	5%	10%
thicknesses (nm)	4.72	4.88	5.40	5.83

3. Analysis of EDS element distribution lines and mapping of elements

The energy-dispersive X-ray spectroscopy (EDS) element distribution lines are provided in Fig. S2. The energy-dispersive X-ray spectroscopy mapping of elements

are show in Fig. S3. The TEM and EDX results indicate the formation of continuous multilayer architectures with negligible interpenetration. Similar results are reported previously for In-B-O or In-Sm-O channel layers (J Am Chem Soc, 2018, 140, 12501-12510; Nat Mater, 2019, 18, 1091-1097). We also use X-ray reflectivity (XRR) characterization to further confirm the thicknesses of In-Sm-O thin films, as shown in Fig. S1 and summarized in Table S1. The In-Sm-O films with 0%, 2%, 5%, 10% Sm contents are 4.72 nm, 4.88 nm, 5.40 nm, 5.84 nm, respectively. The thickness of 5% Sm doped In-Sm-O film is estimated to be 4-5 nm by cross-sectional TEM measurement. Therefore, the XRR results are in good agreement with the cross-sectional TEM measurement, confirming that the thickness of In-Sm-O is ~5 nm.

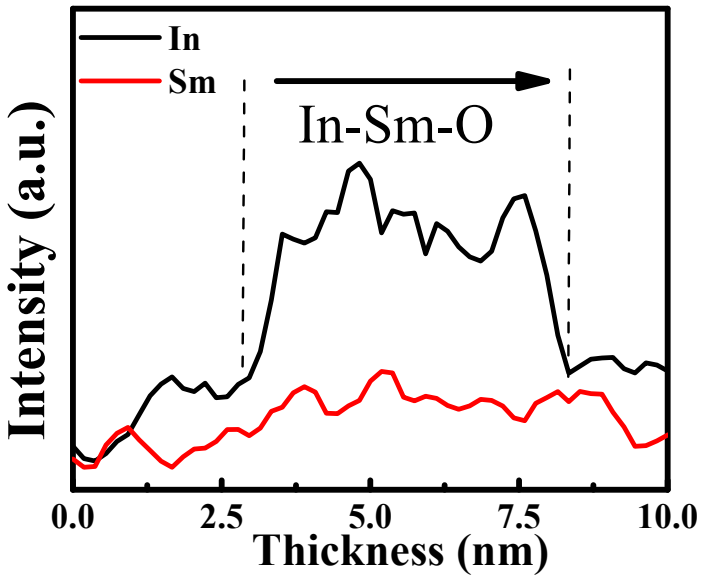

Fig. S2 EDS element distribution lines of the In-Sm-O thin film with 5% Sm concentrations.

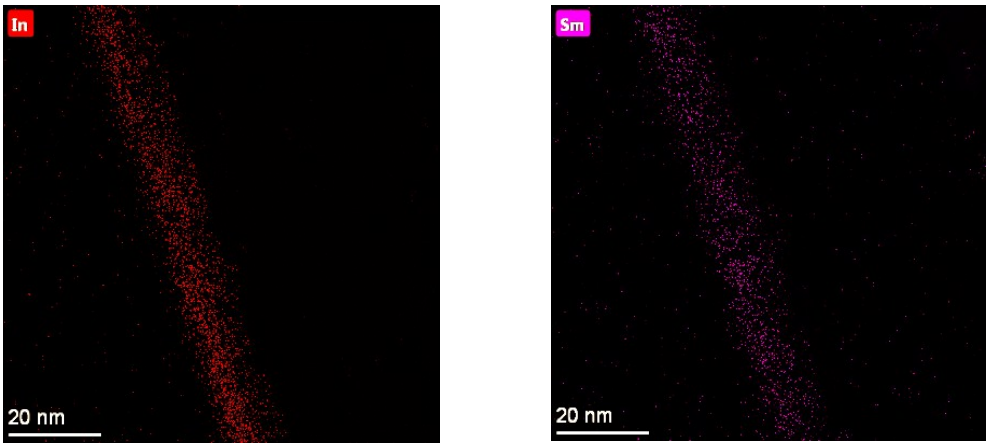

Fig. S3 Energy dispersive X-ray spectroscopy mapping of elements In and Sm.

4. Leakage current of the In-Sm-O TFTs

The leakage current of the SiO_2 results are shown in Fig. S4. The relatively high leakage currents are due to the un-patterned channel layers.[1, 2] However, the leakage current is still 5 orders lower than the drain current, which could effectively avoid mobility overestimation

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2. W. Xu, H. Wang, F. Xie, J. Chen, H. Cao and J. B. Xu, *ACS Appl Mater Interfaces*, 2015, **7**, 5803-5810.

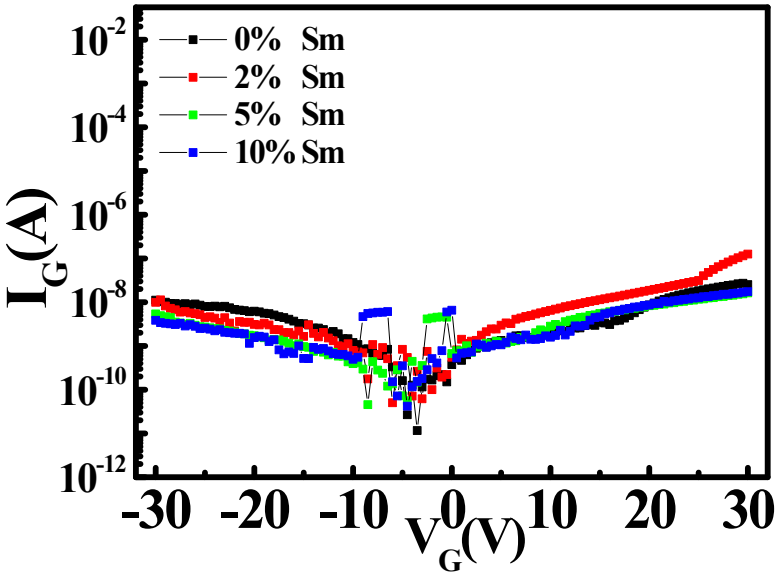

Fig. S4 Leakage current of the In-Sm-O TFTs with different Sm concentrations (V_{DS}

= 20 V)

5. Transfer curves in linear regime of the In-Sm-O TFTs.

The transfer curves in linear regime (V_{DS} = 0.5 V) for In-Sm-O TFTs is shown in Fig. S5, with linear mobility summarized in Table S2. The linear mobility is in good agreement with the saturation mobility.

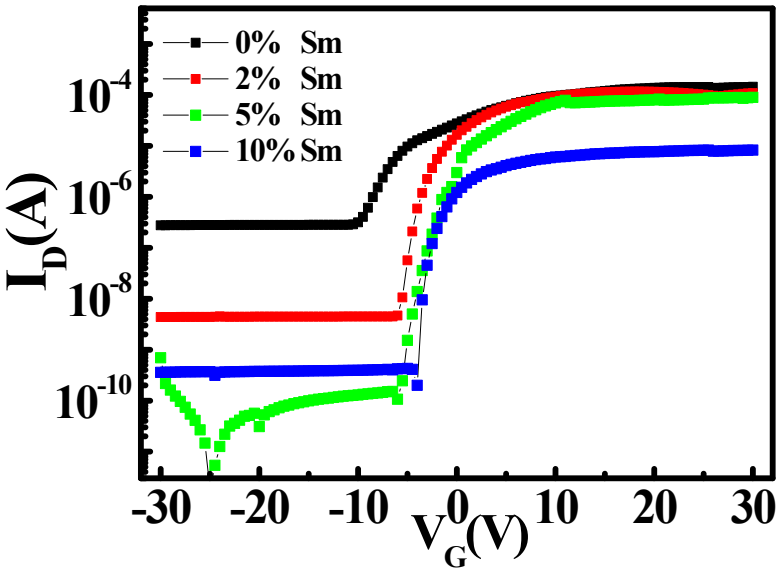

Fig. S5 Transfer curves for In-Sm-O TFTs with various Sm concentrations (V_{DS} = 0.5 V).

Table S2 linear mobility (μ) of In-Sm-O TFTs with different Sm concentrations (V_{DS} = 0.5 V).

Sm concentrations	0%	2%	5%	10%
mobility ($cm^2v^{-1}s^{-1}$)	26.02	25.81	24.64	3.34

6. In-Sm-O TFTs annealed at 250 °C

We have tried the lower temperature annealing (250 °C) for In-Sm-O TFTs, with electrical characteristics shown in Fig. S6 and summarized in Table S3. The 250 °C-annealed devices exhibit relatively poor electrical performance than that of the 350 °C-annealed counterparts. The enhanced performance for the 350 °C-annealed samples is attributed to the improvement of metal-oxygen framework, crystallinity,

and thin film density.[1] Therefore, the annealing temperature of In-Sm-O is fixed at 350 °C.

1. Y. Hwan Hwang, J.-S. Seo, J. Moon Yun, H. Park, S. Yang, S.-H. Ko Park and B.-S. Bae, *NPG Asia Materials*, 2013, **5**, e45-e45.

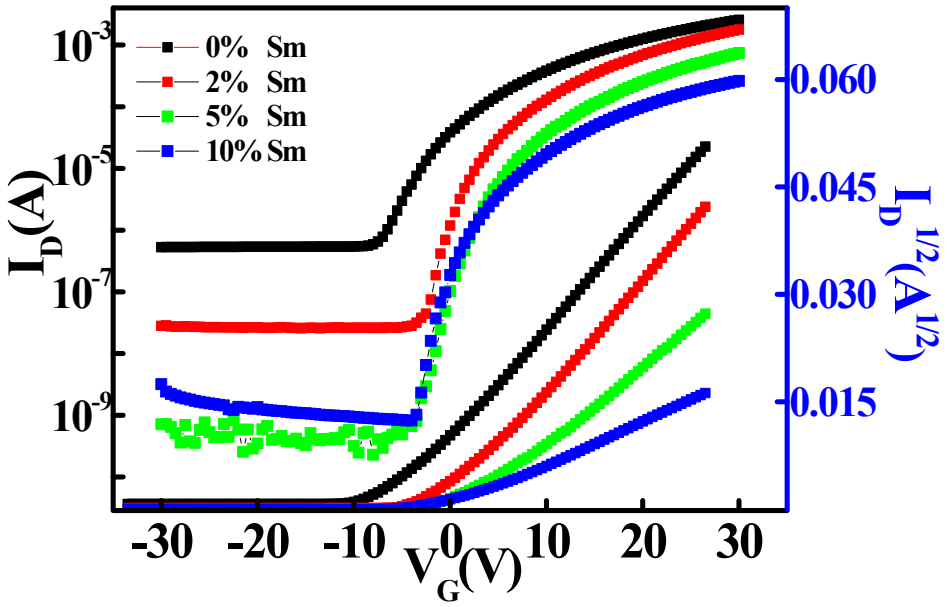

Fig. S6 Transfer curves for In-Sm-O TFTs annealed at 250 °C with different Sm concentrations (V_{DS} = 20 V).

Table S3 Summary of the electrical characteristics of In-Sm-O TFTs annealed at 250 °C with different Sm concentrations.

Sm concentrations	μ ($cm^2V^{-1}s^{-1}$)	I_{on}/I_{off}	SS (V/dec)	V_{th} (V)
0%	10.01	4.84×10^3	3.15	-7.53
2%	9.26	6.95×10^3	1.34	-1.06
5%	5.47	3.24×10^5	1.29	3.14
10%	1.60	1.89×10^5	1.20	3.31

7. Analysis of the electrical characteristics of In-Sm-O TFTs with different thicknesses.

The thickness of thin film is very important for solution-based oxide TFTs. The

thickness of In-Sm-O reported in this study is 4-5 nm. The transfer curves for 5% Sm doped In-Sm-O TFTs with different thicknesses are shown in Fig. S7, with electrical parameters summarized in Table S4. In-Sm-O TFTs with thickness of ~5 nm demonstrate the best performance. It should be noted that the solution processing is somewhat different from that of vacuum-based processing. For spin-coating processing, large amounts of gases are evolved during post-annealing, compromising film density and roughness for one-step thick-film growth processing (PNAS, 2015, 112, 3217-3222). So thicker film is significantly more porous and rougher, leading to poor device performance. Therefore, solution-based oxide thin film must be sufficient thin to maintain smooth, continuity and densification without porosity. However, if the channel layer is too thin (~2-3 nm), interface scattering would deteriorate the device performance.

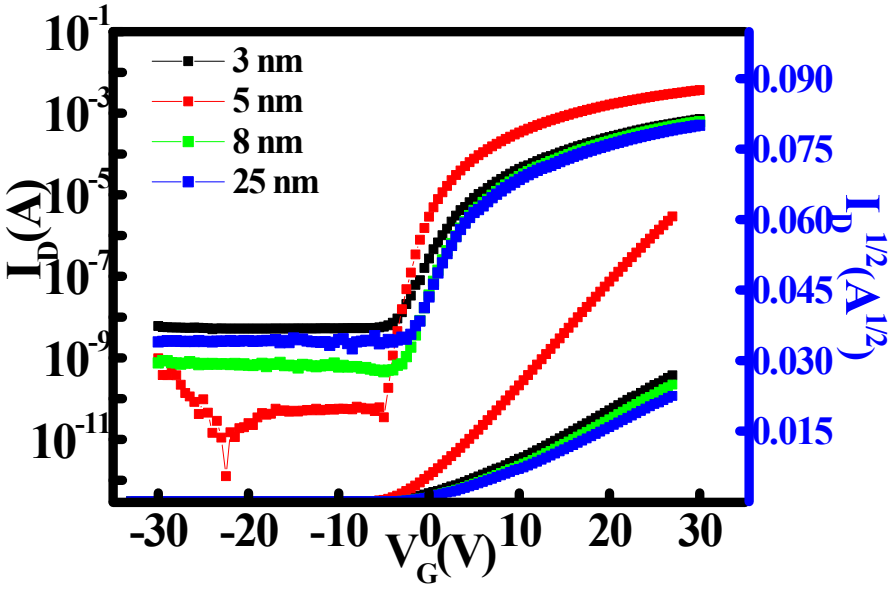

Fig. S7 Transfer curves for 5% Sm In-Sm-O TFTs with different thickness (V_{DS} = 20 V).

Table S4 Summary of the electrical characteristics of In-Sm-O TFTs with different thicknesses.

thickness(nm)	μ ($cm^2V^{-1}s^{-1}$)	I_{on}/I_{off}	SS (V/dec)	V_{th} (V)
4	5.11	1.44×10^5	1.55	2.43
5	21.51	1.09×10^8	0.66	2.14
8	4.93	3.24×10^6	1.50	2.51

25	3.80	1.41×10^6	1.69	2.63

8. Off state current variation after bias stress.

The value of the off-state current (I_{off}) can be calculated by the following expression:[1]

$$I_{off} = \frac{\sigma t w V_{DS}}{L}$$

where σ is the conductivity of the In-Sm-O film, t denotes the thickness of In-Sm-O, W and L are the channel width and length, respectively.

Under positive bias stress, electrons could be trapped at the active layer/dielectric interface, causing the reduction of carrier concentration in In-Sm-O active layer.[2] Besides, the electric field can also induce the desorption of O_2 on the back channel, leading to the reduction of carrier concentration in In-Sm-O channel.[3] Both factors reduce the electron concentration and conductivity in the active layer, resulting in the reduction of off current with bias stress time. Similar phenomena could be found in our previous report for In-Ni-O TFTs.[4]

1. L. C. Liu, J. S. Chen and J. S. Jeng, *Appl. Phys. Lett.*, 2014, **105**, 023509.
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