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Electronic Supplementary Information

Dual Function Metal Oxide Interlayer as Oxygen-Defects Inhibitor and Gate-Leakage Suppressor for Hysteresis-free, Solution-processed Top-gated IGZO TFT

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Figure S1. Output curves of the standard TFTs prepared at the different IGZO annealing times: (a) 10, (b) 20, (c) 30, (d) 60, and (e) 90 min

Table S1. Representative ($V_{GS} = 40$ V) gate leakage currents of the standard TFTs prepared at the different IGZO annealing times.

| Active Layer-Annealing Duration | $I_{GS}(A)$ $(V_{GS}=40V)$ |
|---------------------------------|----------------------------|
| IGZO-10 | 1.8x10 ⁻⁶ |
| IGZO-20 | 5x10 ⁻⁶ |
| IGZO-30 | 1.2x10 ⁻⁵ |
| IGZO-60 | 2.1x10 ⁻⁵ |
| IGZO-90 | 4.7x10 ⁻⁴ |

Table S2. Relative intensities of the individual peaks in the O 1s XPS spectra of the IGZO films prepared at the different annealing times (Figure 3) and the respective relative weights of the metal hydroxides and oxygen vacancies with respect to M-O.

| Active Layer | М-О | M-O _{vac} | М-ОН | O _I (M-OH/M-O) | $O_{II} (M-O_{vac}/M-O)$ |
|-----------------|------|--------------------|------|---------------------------|--------------------------|
| IGZO-10 | 48.1 | 21.9 | 30.0 | 0.62 | 0.45 |
| IGZO-20 | 51.4 | 26.6 | 22 | 0.43 | 0.52 |
| IGZO-30 | 53.5 | 28.3 | 18.2 | 0.34 | 0.53 |
| IGZO-60 | 55.1 | 30.4 | 14.5 | 0.26 | 0.55 |
| IGZO-90 | 56.8 | 32.2 | 11 | 0.19 | 0.57 |



Figure S2. Output curves of the standard (a) and Mg-ion-engineered (b-e) IGZO TFTs.



Figure S3. Output curves of the standard (a) and Al-ion-engineered (b-e) IGZO TFTs.

| Active Layer | Mobility (cm²/Vs) | I _{on/off} 10 ⁿ | SS (V/dec) | V _{th} (V) | D _{it} (cm ⁻² eV ⁻¹) |
|--------------|----------------------|--|---------------|------------------------|---|
| | 4 34 | 4 72 | 8 17 | 3.4 | |
| IGZO-60 | (+0.39) | (+0.93) | (+4.64) | (+1,7) | $6.27 \ge 10^{12}$ |
| | (± 0.39) | (± 0.93) | () | (± 1.7) | |
| Mg-5% | 1.27 | 5.19 | 3.52 | 4.67 | $2.70 \ge 10^{12}$ |
| 8 | (± 0.41) | (± 0.47) | (± 1.7) | (±2.7) | |
| Mg 109/ | 0.48 | 5.45 | 3.04 | 17.70 | 2.24×1012 |
| lvig-1070 | (± 0.08) | (± 0.27) | (±0.57) | (± 1.0) | 2.24 X 10 ⁻² |
| | 0.37 | 6.11 | 2.22 | 18.10 | 1 (5 1012 |
| Mg-15% | (± 0.07) | (± 0.26) | (± 0.32) | (± 3.1) | $1.65 \ge 10^{12}$ |
| | 0.13 | 5 73 | 2.16 | 19.80 | |
| Mg-20% | (+0.01) | (+0.14) | (+0.26) | (+1,0) | $1.61 \ge 10^{12}$ |
| | (± 0.01) | (±0.14) | (± 0.20) | (± 1.0) | |
| Al-5% | 5.54 | 4.02 | 4.12 | 3.82 | $3.16 \ge 10^{12}$ |
| | (± 0.39) | (± 0.80) | (± 1.7) | (±1.9) | |
| A1_100/ | 2.51 | 5.58 | 4.08 | 9.1 | 3.04×1012 |
| AI-10 /0 | (± 0.40) | (± 0.40) | (±1.3) | (±3.5) | 5.04 X 10 |
| | 2.13 | 5.83 | 3.04 | 11.20 | 2 20 1012 |
| AI-15% | (± 0.59) | (± 0.33) | (± 0.78) | (± 3.8) | 2.28×10^{12} |
| | 17 | 5 99 | 2.57 | 14 3 | |
| Al-20% | (± 0.15) | (± 0.10) | (± 0.53) | $(\pm 1, 1)$ | $1.92 \ge 10^{12}$ |
| | (± 0.13) | (± 0.19) | (± 0.55) | (+1.1) | |

 Table S3. Electrical parameters of the pristine and Mg and Al ion engineered IGZO

TFTs.



Figure S4. O 1s XPS spectra of the standard (a,b) and Mg (a) and Al (b) doped IGZO films. The spectra are decomposed in several individual peaks (see text for details).

Table S4. Relative intensities of the individual peaks in the O 1s XPS spectra of thestandard, and Mg/Al doped IGZO films and the respective relative weights of themetal hydroxides and oxygen vacancies with respect to M-O.

| Active layer | М-О | M-O _{vac} | M-OH | O _I (M-OH/M-O) | O_{II} (M- O_{vac} /M-O) |
|--------------|------|--------------------|------|---------------------------|------------------------------|
| IGZO - 60 | 54.8 | 31.9 | 13.3 | 0.24 | 0.58 |
| Mg - 10% | 59.5 | 27.0 | 13.5 | 0.23 | 0.45 |
| Mg - 20% | 67.1 | 20.6 | 12.3 | 0.18 | 0.30 |
| Al - 10% | 57.3 | 29.9 | 12.8 | 0.22 | 0.52 |
| Al - 20% | 60.5 | 26.8 | 12.7 | 0.20 | 0.44 |



Figure S5. (a-c) Results of the PBS measurements for the standard (a) and Mg (b) and Al (c) engineered IGZO TFTs as well as (d) the respective threshold voltage shifts as functions of bias stress time.

Table S5. Thicknesses of the MgO (M) and AlO_x (A) interlayers at the different

| Interlayer-Annealing Duration | Thickness (nm) |
|-------------------------------|----------------|
| M-30 | 6.5 |
| M-60 | 6 |
| M-90 | 5.5 |
| M-120 | 5 |
| A-30 | 6 |
| A-60 | 5.5 |
| A-90 | 5 |
| A-120 | 4.5 |

annealing times (30-120 min).



Figure S6. Output characteristics of the MgO-engineered IGZO TFTs fabricated with annealing of the MgO interlayer for 30 min (a), 60 min (b), 90 min (c) and 120 min (d).



Figure S7. Output characteristics of the AlO_x -engineered IGZO TFTs fabricated with annealing of the AlOx interlayer for 30 min (a), 60 min (b), 90 min (c) and 120 min (d).



Figure S8. Gate leakage current of the MgO (a) and AlOx (b) engineered IGZO TFTs as a function of V_{GS} .

| Interlayer-Annealing Duration | I _{GS} (A) (V _{GS} =40V) |
|-------------------------------|---|
| M-30 | 2x10 ⁻⁵ |
| M-60 | 1.5x10 ⁻⁵ |
| M-90 | 1.9x10 ⁻⁶ |
| M-120 | 1.9x10 ⁻⁶ |
| A-30 | 7.5x10 ⁻⁶ |
| A-60 | 9.4x10 ⁻⁶ |
| A-90 | 8.4x10 ⁻⁶ |
| A-120 | 4.6x10 ⁻⁶ |

Table S6. Representative ($V_{GS} = 40$ V) gate leakage currents of the MgO (a) and AlOx(b) engineered IGZO TFTs.



Figure S9. O 1s XPS spectra of MgO (a) and AlOx (b) films. The spectra are decomposed in several individual peaks (see text for details).

 Table S7. Relative intensities of the individual peaks in the O 1s XPS spectra of MgO

 and AlOx films (Figure S9) as well as respective relative weights of metal hydroxides

 with respect to M-O.

| Interlayer | М-О | M-OH | O _I (M-OH/M-O) |
|-------------|------|------|---------------------------|
| M-30 | 43.9 | 56.1 | 1.23 |
| M-60 | 60.7 | 39.3 | 0.64 |
| M-90 | 62.3 | 37.7 | 0.61 |
| A-30 | 67.1 | 32.9 | 0.49 |
| A-60 | 68.6 | 31.4 | 0.46 |
| A-90 | 68.9 | 31.1 | 0.45 |



Figure S10. Results of the NBS measurements for the standard (a), MgO (b) and AlOx (c) engineered IGZO TFTs.



Figure S11. TEM EDS mapping of (a) IGZO, (b) MgO/IGZO, and (c) AlOx/IGZO.



Figure S12. XPS depth profiles of (d) MgO/IGZO, (e) AlOx/IGZO

| Active Layer- Annealing Duration | Leakage current density at 10V (A/cm ²) |
|-------------------------------------|---|
| PMMA | 1.1 x 10 ⁻⁴ |
| M-30 | 4.8 x 10 ⁻⁷ |
| M-60 | 4.1 x 10 ⁻⁹ |
| M-90 | 3.3 x 10 ⁻¹⁰ |
| M-120 | 2.9 x 10 ⁻¹⁰ |
| A-30 | 5.7 x 10 ⁻¹⁰ |
| A-60 | 5.7 x 10 ⁻¹⁰ |
| A-90 | 3.4 x 10 ⁻¹⁰ |
| A-120 | 3.4 x 10 ⁻¹⁰ |

 Table S8. Leakage current of the PMMA, PMMA/MgO and PMMA/AlOx films