

Supporting Information

Comparative study of high-k Metal Oxide-PVP/PMMA hybrid Gate Dielectrics for low-temperature, all-solution-processed flexible In₂O₃ thin film transistors

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Fig. S1. High resolution XPS spectra of O 1s for d) AlO_x, e) ZrO_x, and f) HfO_x-based hybrid dielectrics, and C 1s for g) AlO_x, h) ZrO_x, and i) HfO_x-based hybrid dielectrics.

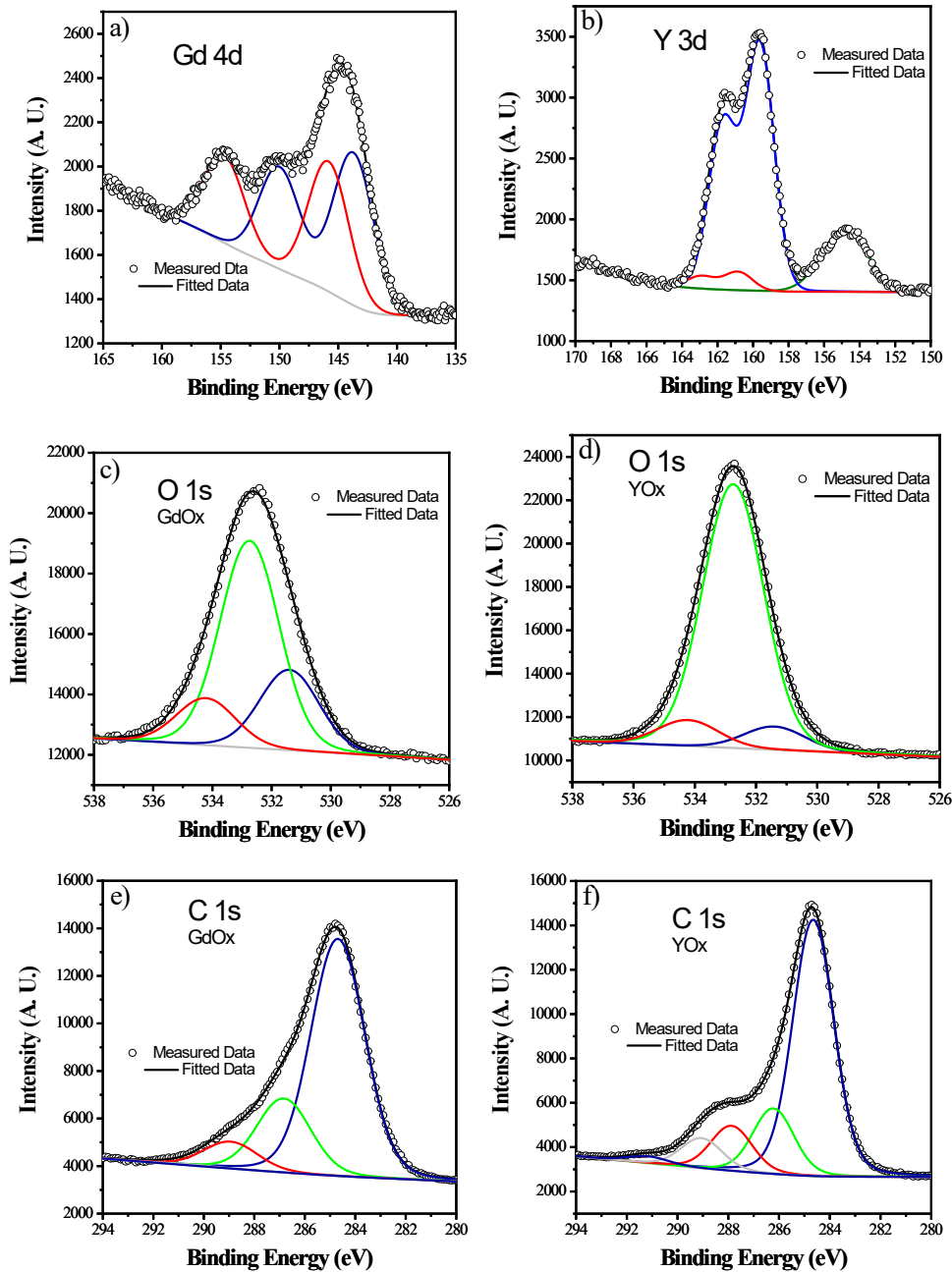


Fig. S2. High resolution XPS spectra of a) Gd 4d, b) Y 3d, O 1s for c) GdOx, and d) YOx-based hybrid dielectrics, and C 1s for e) GdOx, and f) YOx-based hybrid dielectrics.

Table S1 Parameters used for the XPS peak-fitting procedure.

Peak	Fitting parameters			Atomic Percentage (%)					
	Binding Energy (eV)	Gaussian	Lorentzian	PMMA	Gd	Zr	Hf	Y	Al
C ₁	284.68	1.97	0.35	66.42	50.36	48.10	47.40	39.23	40.75
C ₂	286.47	1.97	0.35	9.49	15.67	9.74	11.67	9.99	9.18
C ₃	288.02	1.97	0.35	--	5.72	6.57	5.80	6.91	10.93

C ₄	289.73	1.97	0.35	--	0.22	2.35	3.30	4.60	1.68
C ₅	291.53	1.97	0.35	--	--	0.98	0.65	1.04	0.80

<i>Fitting parameters</i>				<i>Atomic Percentage (%)</i>					
<i>Peak</i>	<i>Binding Energy (eV)</i>	<i>Gaussian</i>	<i>Lorentzian</i>	<i>PMMA</i>	<i>Gd</i>	<i>Zr</i>	<i>Hf</i>	<i>Y</i>	<i>Al</i>
O ₁	531.35	2.18	0.25	3.23	4.35	4.44	3.56	1.59	2.61
O ₂	532.73	2.18	0.25	8.92	11.71	14.39	15.02	18.17	13.30
O ₃	34.25	2.18	0.25	1.60	2.54	1.40	2.69	1.77	0.86

<i>Core</i>	<i>Peak</i>	<i>Branch</i>	<i>Binding Energy (eV)</i>	<i>Spin-orbit Splitting</i>	<i>Gaussian</i>	<i>Lorentzian</i>	<i>Atomic Percentage (%)</i>
Gd 4d	Gd ₁	5/2	143.69	6.29	3.92	0	0.65
		3/2	149.98□		3.92	0	
	Gd ₂	5/2	145.79	8.86	3.92	0	0.56
		5/2	154.65□		3.92	0	
Zr 3d	Zr ₁	5/2	183.92	2.4	1.62	0.38	3.14
		3/2	186.32		1.62	0.38	
	Zr ₂	5/2	185.44	2.4	1.62	0.02	0.24
		3/2	187.84		1.62	0.02	
Hf 4f	Hf ₁	7/2	19.38	1.66	1.89	0.13	0.19
		5/2	21.04		1.89	0.13	
	Hf ₂	7/2	21.93	1.66	1.89	0.13	3.92
		5/2	23.59		1.89	0.13	
Y 3d	Y ₁	5/2	159.59	2.09	1.79	0.047	3.08
		3/2	161.68		1.79	0.047	
	Y ₂	5/2	160.84	2.09	1.79	0.047	0.22
		3/2	162.93□		1.79	0.047	
Al 2p	Al ₁	3/2	75.87	0.44	2.48	0.085	2.37
		1/2	76.31		2.48	0.085	

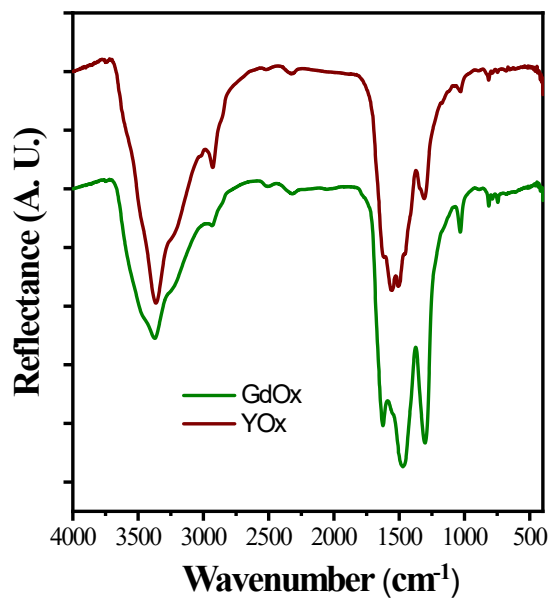


Fig. S3. FTIR spectra of GdOx/PVP-PMMA and YOx/PVP-PMMA hybrid films

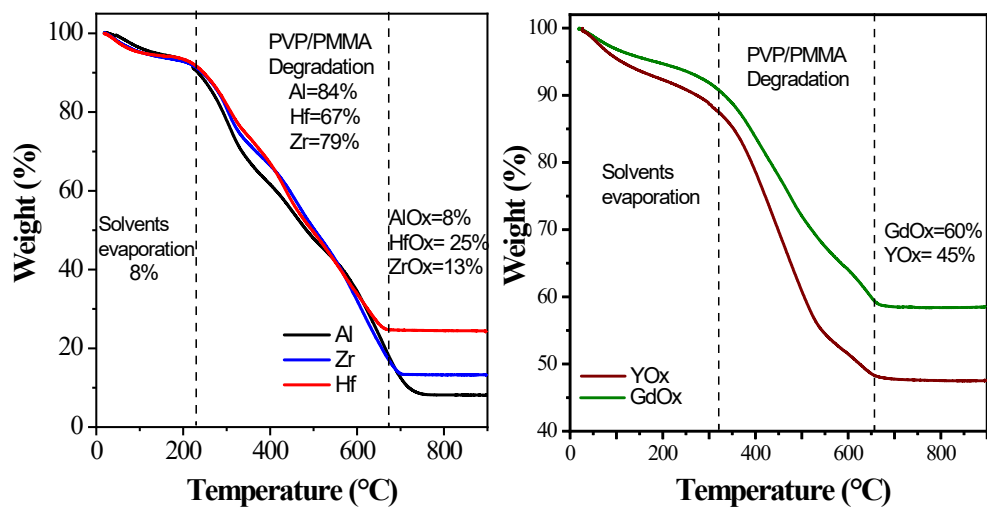


Fig. S4. TGA curves of hybrid dielectrics

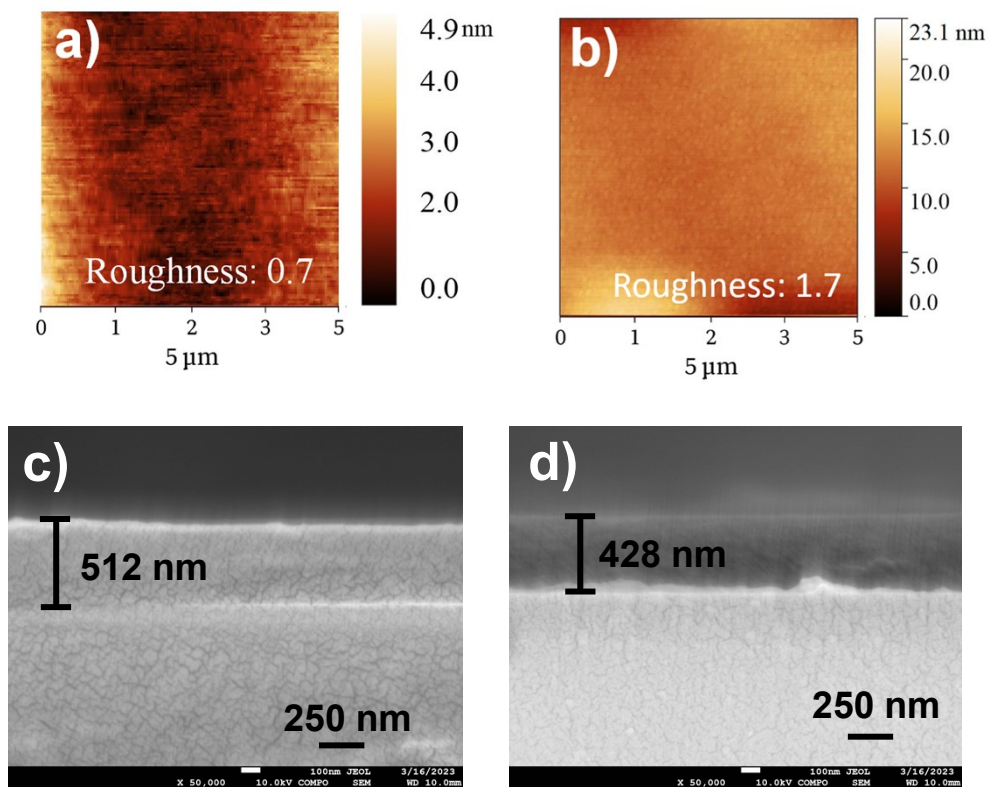


Fig. S5. AFM surface images of a) GdOx-PVP/PMMA, and b) YOx-PVP/PMMA hybrid dielectrics. Cross sectional SEM images of c) GdOx-PVP/PMMA, and d) YOx-PVP/PMMA

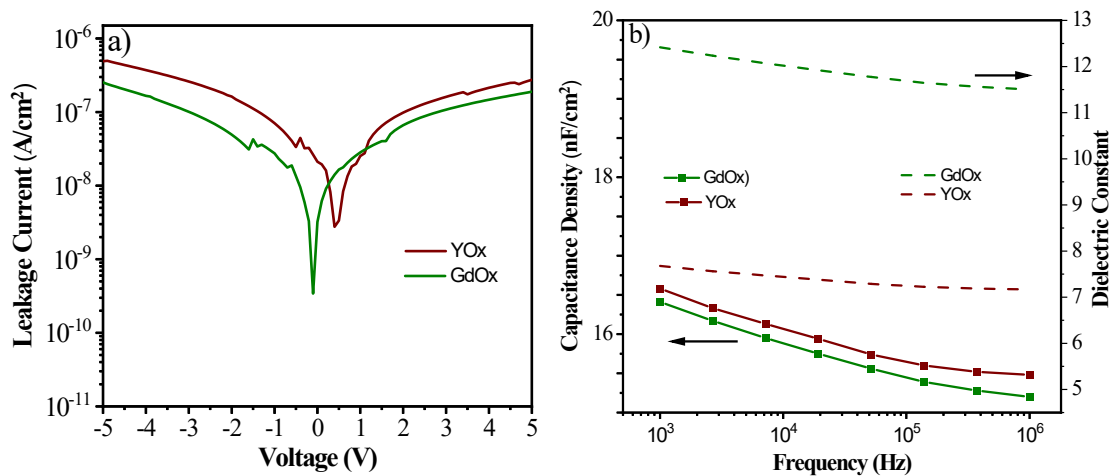


Fig. S6. a) Current density versus voltage and b) capacitance density and dielectric constant versus frequency characteristics of GdOx-PVP/PMMA and YOx-PVP/PMMA hybrid thin films.

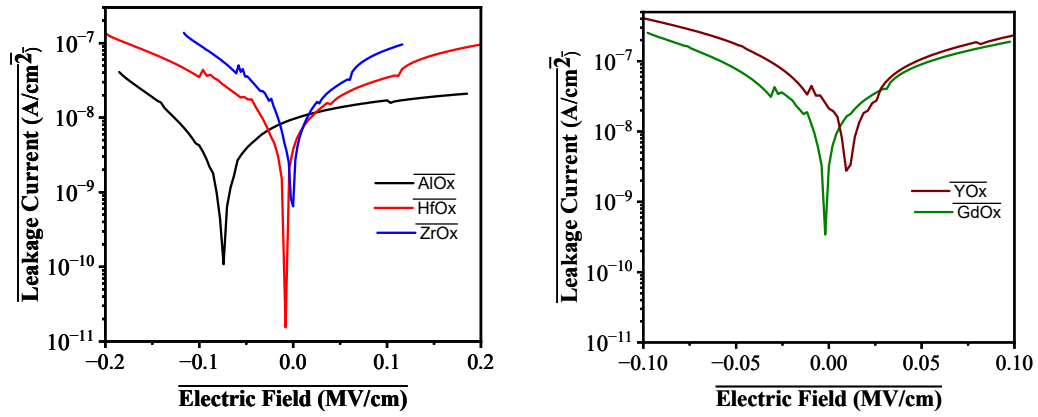


Fig. S7. a) Current density versus electric field of MOx-PVP/PMMA hybrid thin films.

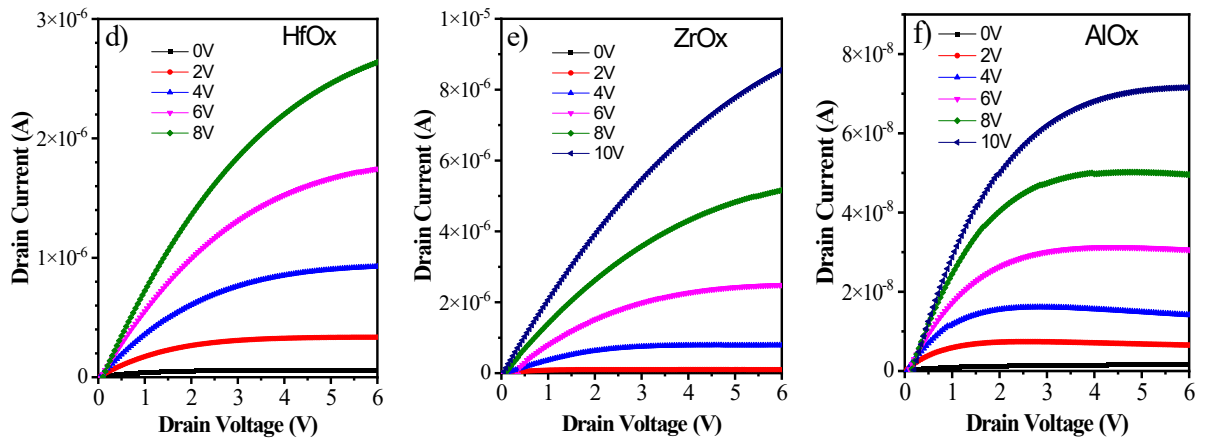


Fig. S8. Output curves of d) HfOx-PVP/PMMA, e) ZrOx-PVP/PMMA, and f) AlOx-PVP/PMMA gate dielectrics.

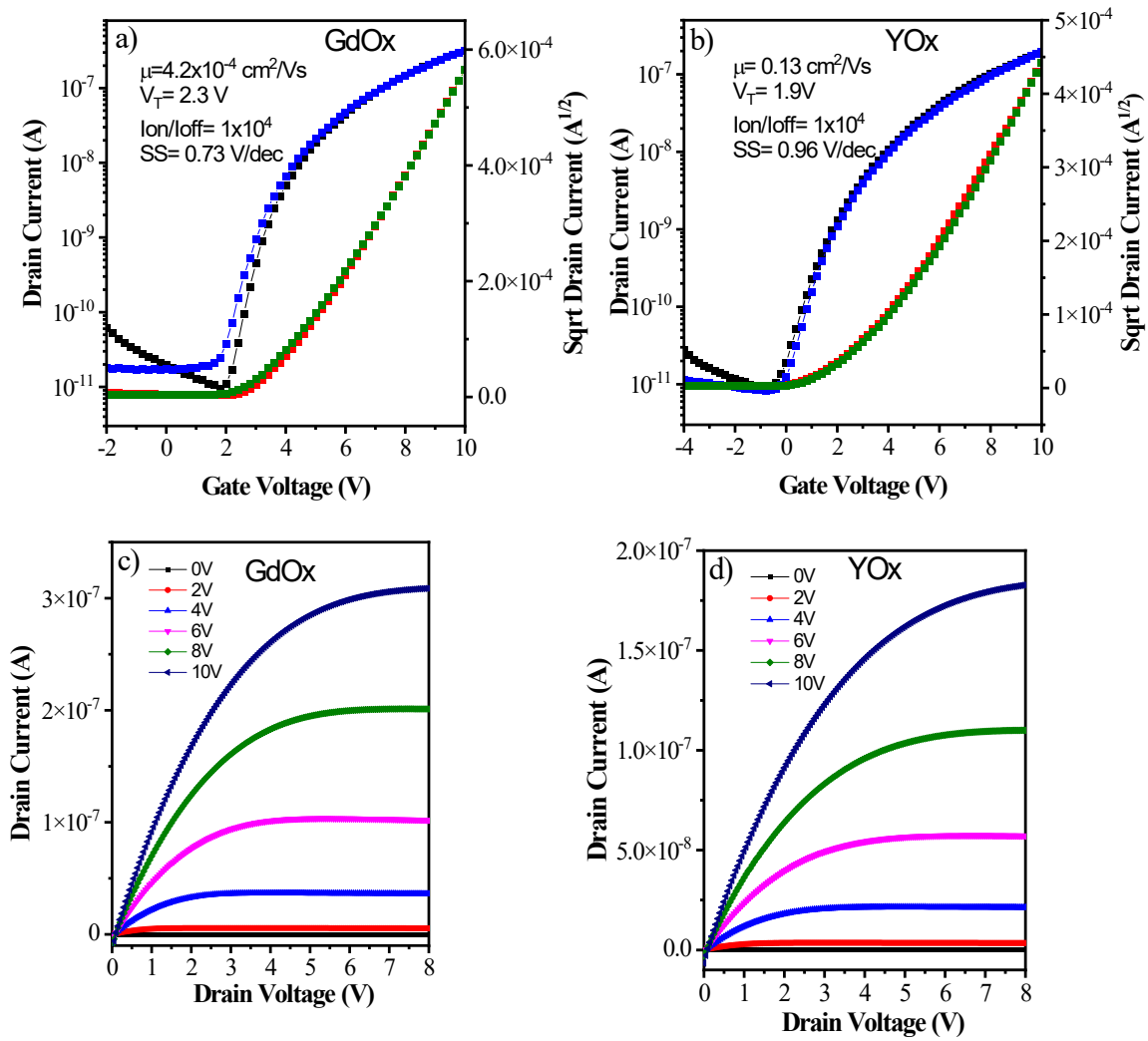


Fig. S9. Transfer curves of the In_2O_3 -based TFTs with a) GdOx-PVP/PMMA, and b) YOx-PVP/PMMA gate dielectrics, and corresponding output curves of c) GdOx-PVP/PMMA, and d) YOx-PVP/PMMA gate dielectrics.

Table S2. Electrical performance parameter obtained for the In_2O_3 -based TFT over rigid substrates.

Sample	Mobility (cm^2/Vs)	Threshold Voltage (V)	Subthreshold swing (V/dec)	$I_{\text{ON}}/I_{\text{OFF}}$
GdOx	4.3×10^{-4}	2.3	0.73	10^4
YOx	0.13	1.9	0.97	10^4

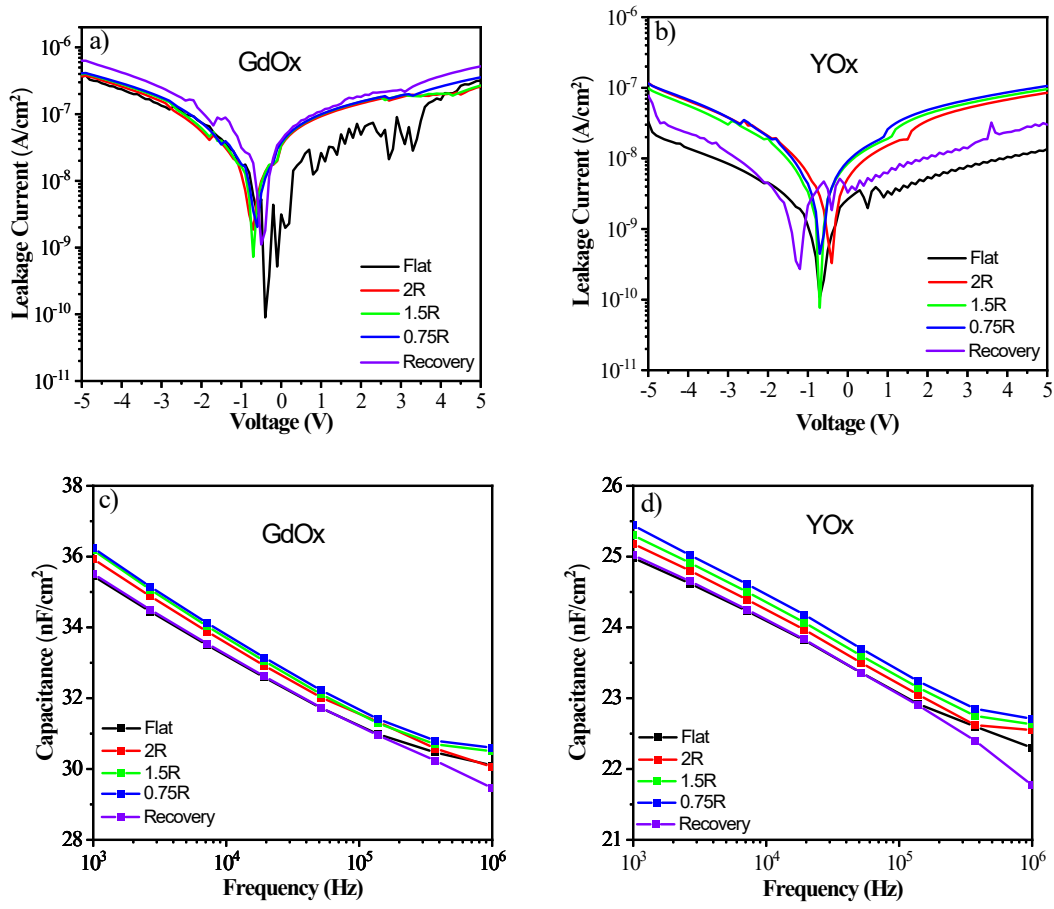


Fig. S10. Current density versus voltage measured at different bending radii on MIM structures fabricated on Al-coated PEN flexible substrate with a) GdOx-PVP/PMMA, and b) YOx-PVP/PMMA. Capacitance density versus frequency measured at different bending radii on the flexible MIM with c) GdOx-PVP/PMMA, and d) YOx-PVP/PMMA hybrid insulators.

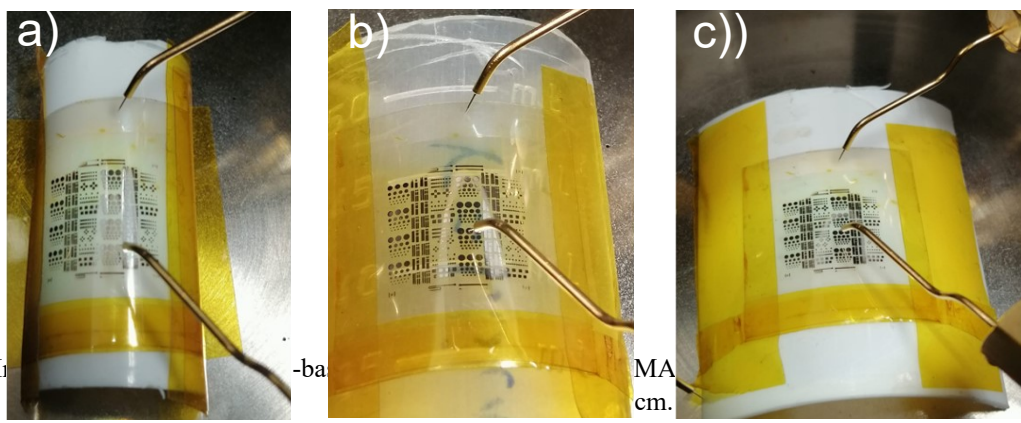


Fig. S11. Images of the MIM structures on a flexible substrate with different bending radii: a) 0.75 cm.

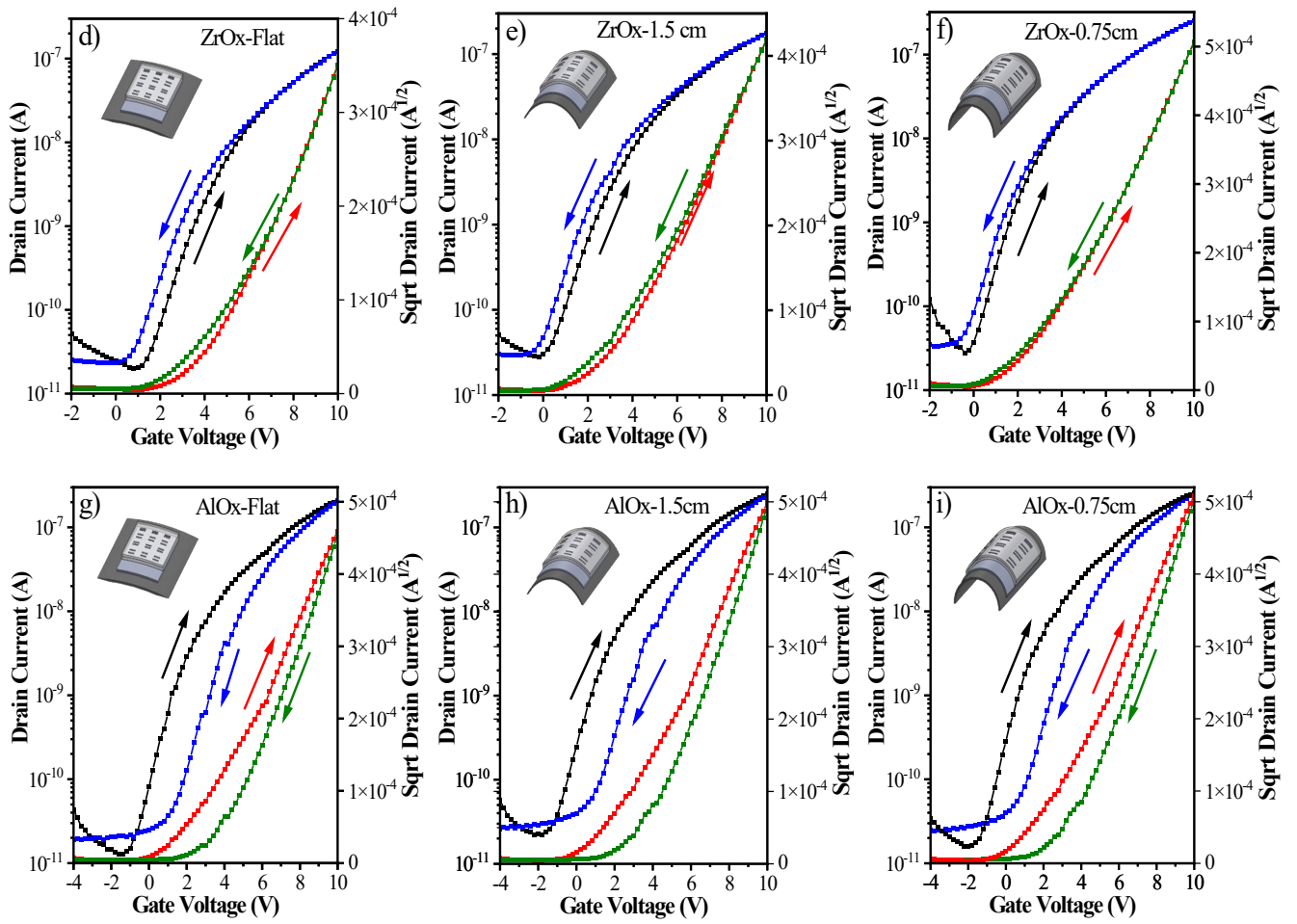


Fig. 12. Transfer curves (left axis) and Sqrt Drain Current versus Gate Voltage (right axis) of the In₂O₃-based TFTs with ZrOx-PVP/PMMA and AlOx-PVP/PMMA gate dielectrics, measured at different bending radii: a) and d) without bending, b), and e) 1.5 cm, and c) and f) 0.75 cm, respectively.

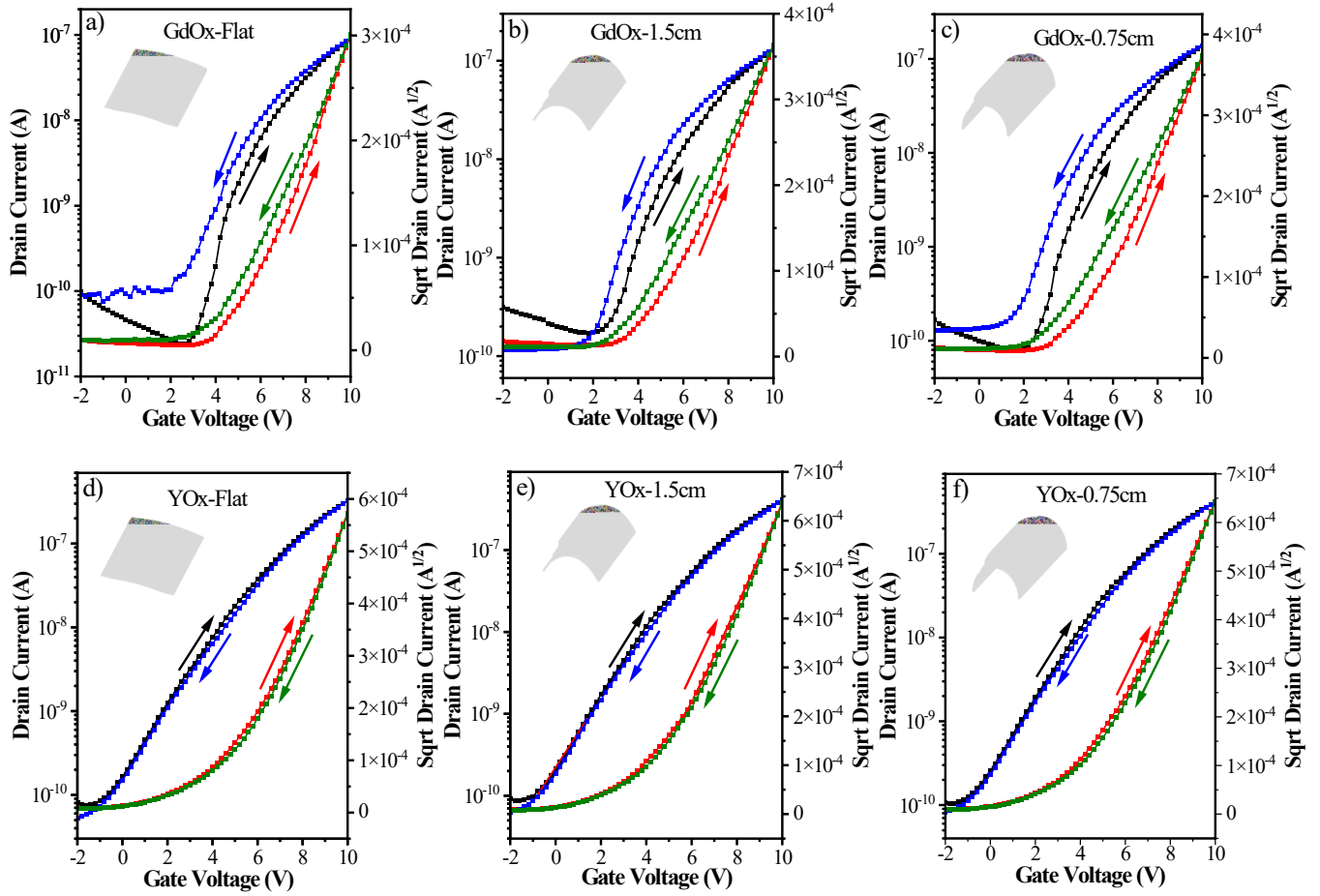


Fig. S13. Transfer curves (left axis) and Sqrt Drain Current versus Gate Voltage (right axis) of the In_2O_3 -based TFTs with GdOx-PVP/PMMA, and YOx-PVP/PMMA gate dielectrics, measured at different bending radii: a), and d) without bending, and b), and e) 1.5 cm, and c), and f) 0.75 cm, respectively.

Table S3. Parameters obtained for the In_2O_3 -based TFT over flexible substrates under bending conditions.

Sample	Bending Radii (cm)	Mobility (cm^2/Vs)	V_T (V)	SS (V/dec)	I_{ON}/I_{OFF}
GdOx-PVP/PMMA	Flat	1.06E-02	4.6	1.01	4.00E+03
	2R	1.18E-02	4.04	1.35	1.92E+03
	1.5R	1.77E-02	3.98	1.54	7.38E+02
	0.75R	1.86E-02	3.98	1.37	1.60E+03
	Recover	2.95E-03	4.6	1.18	2.98E+03
YOx-PVP/PMMA	Flat	6.48E-02	4.1	2.56	4.13E+03
	2R	6.98E-02	3.9	2.50	5.85E+03
	1.5R	7.08E-02	3.9	2.38	4.53E+03
	0.75R	7.05E-02	3.7	2.50	3.90E+03
	Recover	7.06E-03	4.0	2.13	6.60E+03