

Laser induced ultrafast combustion synthesis of solution-based AlO_x for thin film transistors

Emanuel Carlos^{1,2}, Spilios Dellis², Nikolaos Kalfagiannis², Loukas Koutsokeras³, Demosthenes C. Koutsogeorgis^{2}, Rita Branquinho^{1*}, Rodrigo Martins¹ and Elvira Fortunato^{1*}*

¹CENIMAT/i3N Departamento de Ciência dos Materiais, Faculdade de Ciências e Tecnologia (FCT), Universidade NOVA de Lisboa (UNL), and CEMOP/UNINOVA, 2829-516 Caparica, Portugal.

²School of Science and Technology, Nottingham Trent University, Nottingham, NG11 8NS, UK.

³Cyprus University of Technology, Research Unit for Nanostructured Materials Systems, Kitiou Kyprianou 36, 3041, Lemesos, Cyprus

*E-mail: demosthenes.koutsogeorgis@ntu.ac.uk, ritasba@fct.unl.pt and emf@fct.unl.pt;

The Supplementary Information contains the full data sets related to the production of AlO_x dielectric thin films from solution, by excimer laser annealing (ELA) treatment inducing combustion synthesis, and their application in thin film transistors (TFTs). Table S1 and S2 shows the statistical parameters of dielectric properties obtained from MIS devices. Figure S1 and S2 depicts FTIR spectra for all ELA conditions and for both drying durations, 15 min and 1 min, respectively. Figure S3, S4 and S5-S6 show the capacitance–voltage, capacitance–frequency and breakdown voltage (E) characteristics of Al/p-type Si/(AlO_x)/Al MIS capacitors for both drying durations. Tables S3 and S4 depicts the electrical characteristics summary obtained from IGZO TFTs comprising the sol-gel AlO_x fabricated by all the different ELA treatments, while Figures S7 and S8 show the transfer curves of the same IGZO TFTs devices. Finally, Figure S9 depicts the typical output curves of two test TFT devices and the reference. Table S5 shows the statistical TFT parameters from a previous report using deep ultraviolet (DUV) curing for 30 min¹, alongside those of the optimum devices produced in this work (using 15 min drying cycle followed by ELA). Table S6 depicts relevant low temperature solution-based AlO_x layers processed by different curing methods applied in TFTs reported previously in the literature.

Table S1. Statistical parameters of dielectric properties obtained for the MIS devices dried for 15 min and then treated by ELA. These are all graphically depicted in Figure 3 (a) in the main paper. The reference device (300 °C for 1h) is also shown for comparison.

Fluence (mJ/cm ²)	Number of pulses	Capacitance (μ F/cm ²)	Thickness (nm)	E (MV/cm)	Dielectric constant
125	1	0.08 \pm 0.01	27.9 \pm 0.4	1.94 \pm 0.01	2.62 \pm 0.01
	2	0.12 \pm 0.02	28.3 \pm 0.5	1.82 \pm 0.02	3.8 \pm 0.8
	3	0.20 \pm 0.01	27.7 \pm 0.4	1.93 \pm 0.05	6.2 \pm 0.1
150	1	0.10 \pm 0.01	28.7 \pm 0.3	1.78 \pm 0.01	3.1 \pm 0.4
	2	0.35 \pm 0.02	16.5 \pm 0.3	3.10 \pm 0.02	6.5 \pm 0.4
	3	0.38 \pm 0.01	18.2 \pm 0.4	3.3 \pm 0.3	7.9 \pm 0.1
175	1	0.38 \pm 0.02	22.1 \pm 0.9	2.4 \pm 0.2	9.1 \pm 0.1
	2	0.50 \pm 0.06	15.2 \pm 0.4	4.0 \pm 0.5	8.6 \pm 0.1
	3	0.53 \pm 0.05	13.2 \pm 0.3	3.1 \pm 0.1	8.6 \pm 0.7
200	1	0.51 \pm 0.04	15.2 \pm 0.9	2.72 \pm 0.09	8.8 \pm 0.7
	2	0.54 \pm 0.01	12.7 \pm 0.6	3.5 \pm 0.2	7.8 \pm 0.2
	3	0.55 \pm 0.01	12.8 \pm 0.6	3.61 \pm 0.06	7.9 \pm 0.1
Reference (300 °C, 1h)	0	0.42 \pm 0.02	10.9 \pm 0.1	3.82 \pm 0.03	5.2 \pm 0.2

Table S2. Statistical parameters of dielectric properties obtained for the MIS devices dried for 1 min and then treated by ELA. These are all graphically depicted in Figure 3 (b) in the main paper. The reference device (300 °C for 1h) is also shown for comparison.

Fluence (mJ/cm ²)	Number of pulses	Capacitance (μF/cm ²)	Thickness (nm)	E (MV/cm)	Dielectric constant
200	1	0.69 ± 0.04	47.1 ± 0.9	1.15 ± 0.02	37 ± 2
	2	0.74 ± 0.05	42.3 ± 0.8	0.89 ± 0.03	35 ± 3
	3	0.73 ± 0.04	18.6 ± 1.8	3.0 ± 0.2	15.3 ± 0.9
225	1	0.81 ± 0.04	36.8 ± 0.4	1.5 ± 0.1	33 ± 1
	2	0.72 ± 0.05	14.4 ± 0.7	2.9 ± 0.1	11.7 ± 0.8
	3	0.70 ± 0.04	12.7 ± 0.5	3.5 ± 0.2	9.4 ± 0.1
250	1	0.84 ± 0.03	13.4 ± 0.5	3.1 ± 0.4	12.7 ± 0.5
	2	0.83 ± 0.05	11.4 ± 0.3	3.6 ± 0.3	10.6 ± 0.6
	3	0.84 ± 0.01	11.1 ± 0.4	3.9 ± 0.7	10.2 ± 0.5
300	1	0.79 ± 0.05	14.9 ± 0.7	2.9 ± 0.2	13.2 ± 0.8
	2	0.84 ± 0.02	10.9 ± 0.5	3.5 ± 0.1	10.3 ± 0.2
	3	0.93 ± 0.01	11.0 ± 0.5	3.4 ± 0.3	11 ± 1
Reference (300 °C, 1h)	0	0.42 ± 0.02	10.9 ± 0.1	3.82 ± 0.03	5.2 ± 0.2

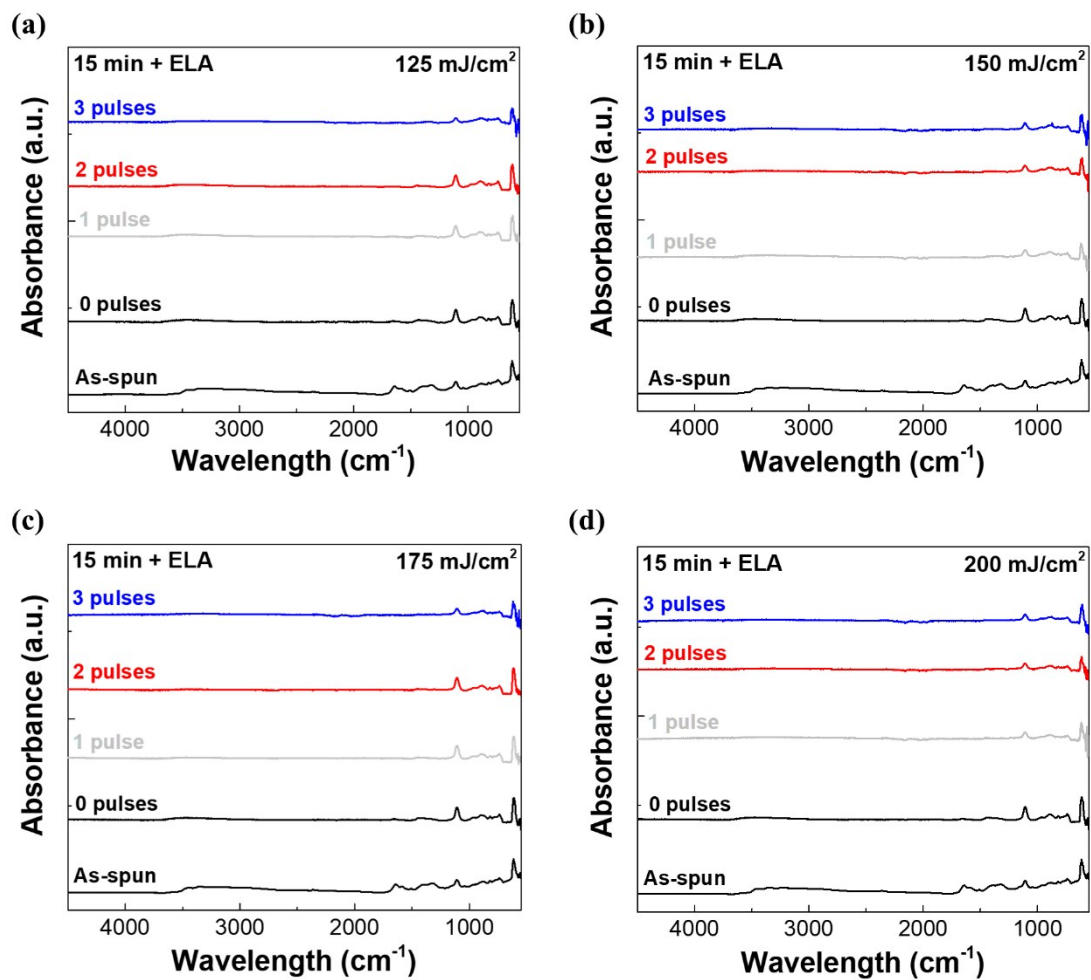


Figure S1. FTIR spectra of AlO_x thin films with 15 min of drying and ELA treatment using different fluences and pulses: (a) 125 mJ/cm^2 ; (b) 150 mJ/cm^2 ; (c) 175 mJ/cm^2 ; (d) 200 mJ/cm^2 .

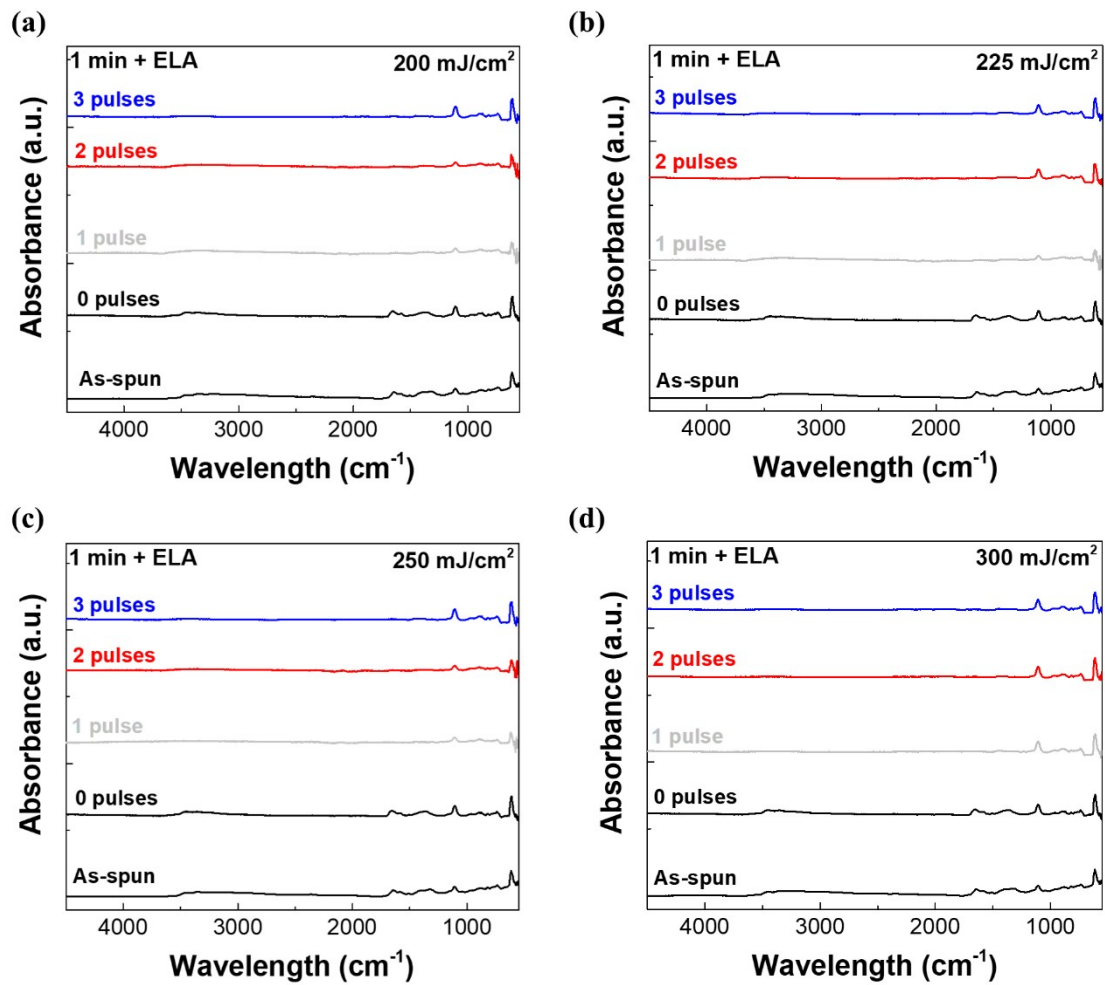


Figure S2. FTIR spectra of AlO_x thin films with 1 min of drying and ELA treatment using different fluences and pulses: (a) 200 mJ/cm^2 ; (b) 225 mJ/cm^2 ; (c) 250 mJ/cm^2 ; (d) 300 mJ/cm^2 .

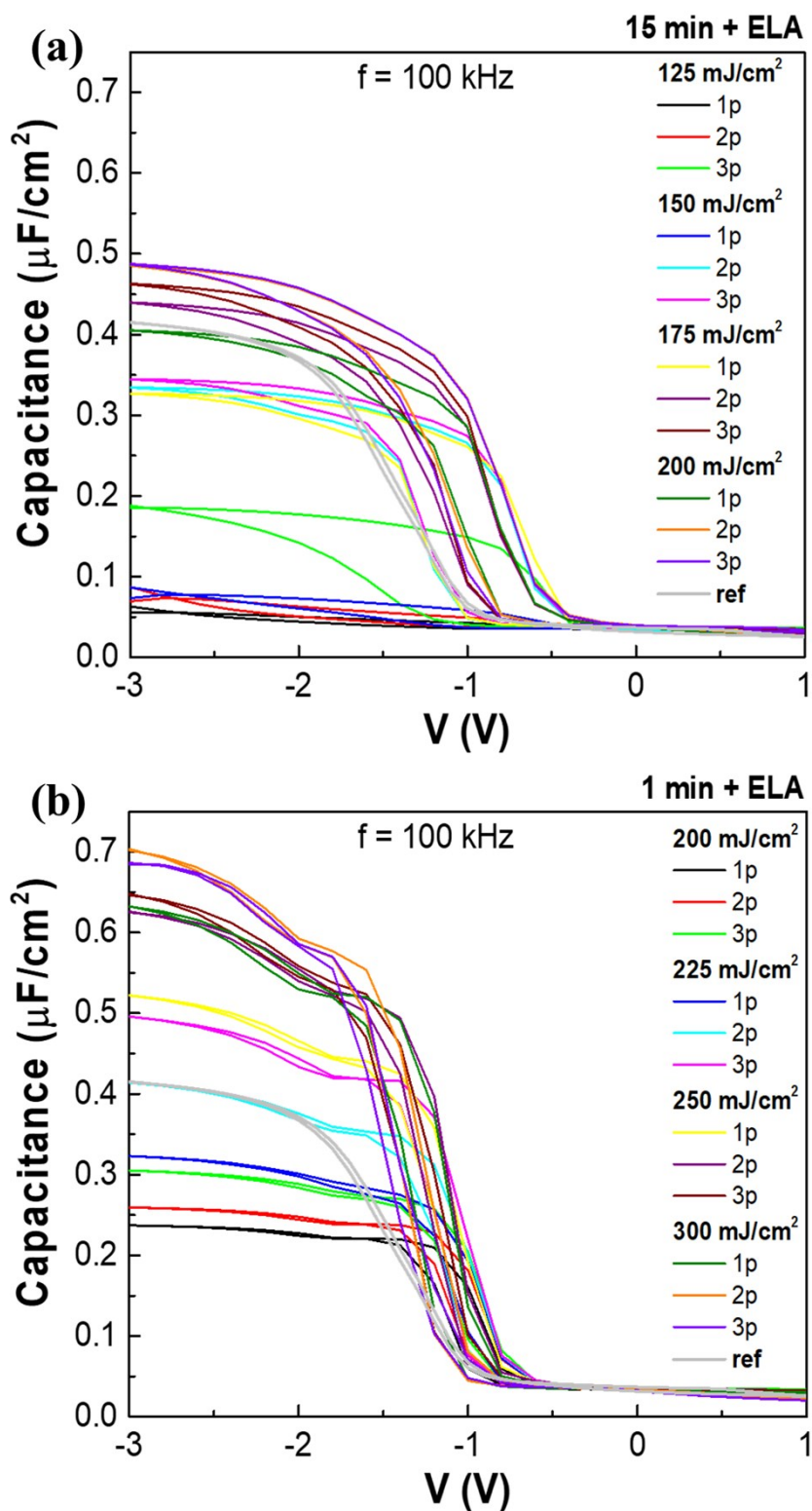


Figure S3. Typical capacitance–voltage characteristics at 100 kHz of solution-based AlO_x MIS capacitors dried at 150 °C for (a) 15 min and (b) 1 min followed by ELA treatment for different fluences and pulses. The reference device (300 °C for 1h) is also shown for comparison.

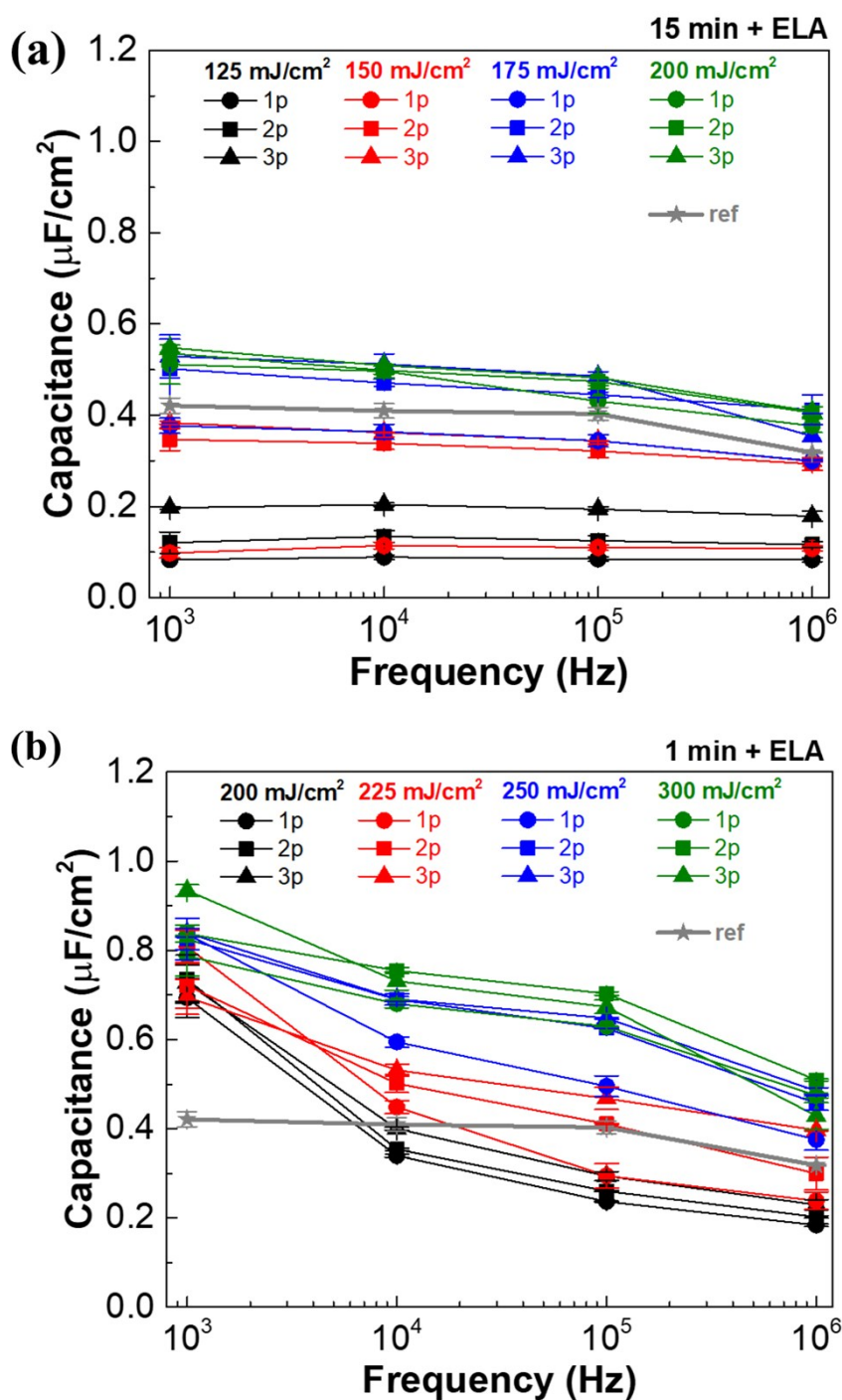


Figure S4. Typical capacitance–frequency characteristics of solution-based AlO_x MIS capacitors dried at 150°C for (a) 15 min and (b) 1 min followed by ELA treatment for different fluences and pulses. The reference device (300°C for 1h) is also shown for comparison.

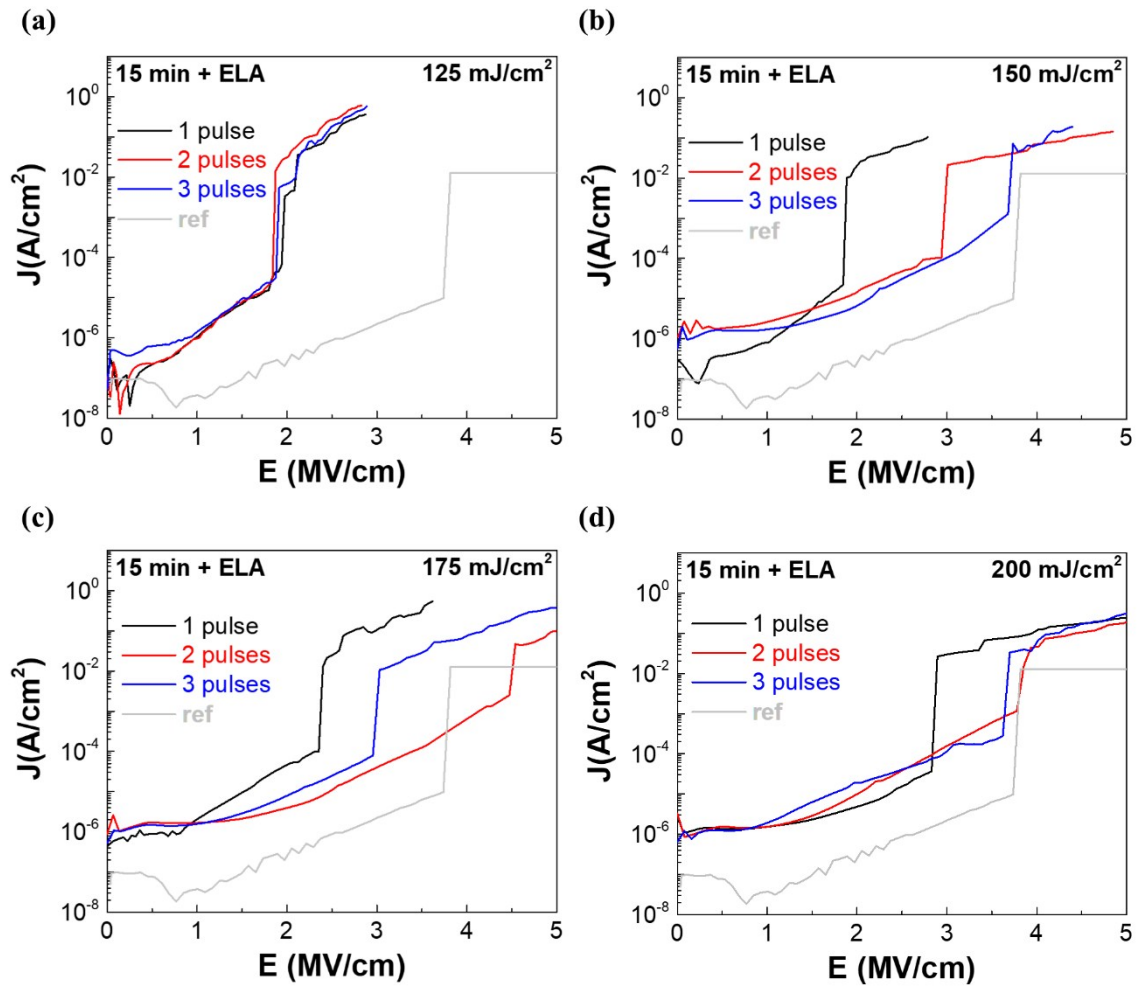


Figure S5. Typical current density (J) and breakdown field (E) characteristics of solution-based AlO_x MIS capacitors with 15 min of drying and ELA treatment using different fluences and pulses: (a) 125 mJ/cm^2 ; (b) 150 mJ/cm^2 ; (c) 175 mJ/cm^2 ; (d) 200 mJ/cm^2 . The reference device ($300 \text{ }^\circ\text{C}$ for 1h) is also shown for comparison.

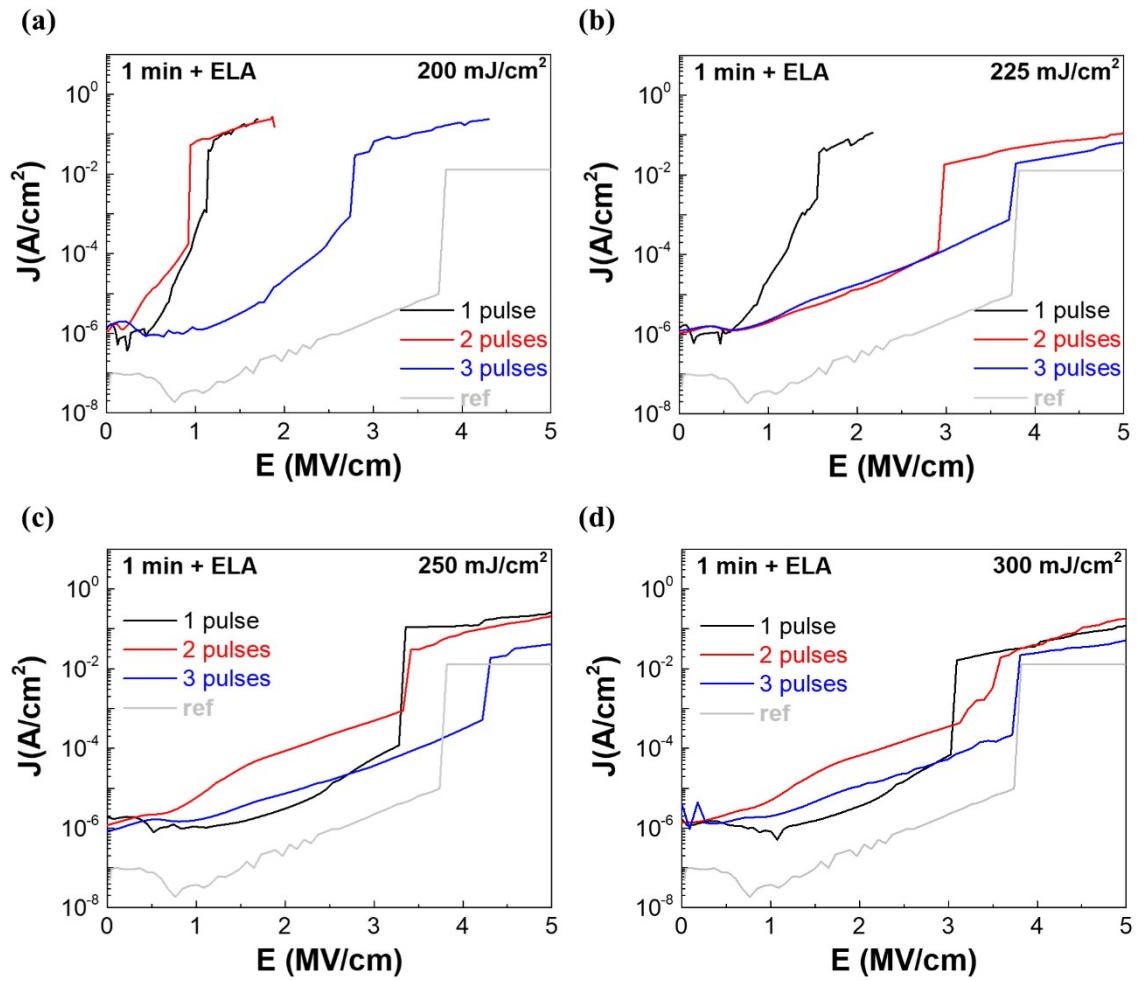


Figure S6. Typical current density (J) and breakdown field (E) characteristics of solution-based AlO_x MIS capacitors with 1 min of drying and ELA treatment using different fluences and pulses: (a) 200 mJ/cm^2 ; (b) 225 mJ/cm^2 ; (c) 250 mJ/cm^2 ; (d) 300 mJ/cm^2 . The reference device ($300 \text{ }^\circ\text{C}$ for 1h) is also shown for comparison.

Table S3. Parameters of TFTs obtained for the devices with 15 min of drying followed by ELA. These are all graphically depicted in Figure 4 (a) in the main paper. The reference device (300 °C for 1h) is also shown for comparison.

Fluence (mJ/cm ²)	Number of pulses	μ_{SAT} (cm ² /V·s)	SS (V/dec)	$I_{ON/OFF}$	V_{ON} (V)	V_{Hyst} (V)
150	2	11.7	0.13	1.5×10^4	-0.27	0.05
	3	17.9	0.14	2.1×10^4	-0.22	0.07
175	1	18.6	0.13	1.0×10^4	0.05	0.08
	2	20.0	0.10	1.6×10^4	- 0.07	0.07
	3	12.8	0.15	9.7×10^3	- 0.22	0.04
200	1	8.0	0.19	3.1×10^3	- 0.36	0.05
	2	14.5	0.21	3.2×10^3	- 0.36	0.04
	3	8.71	0.23	1.4×10^3	- 0.22	0.01
Reference (300 °C, 1h)	0	13.7	0.13	1.1×10^4	- 0.07	0.06

Table S4. Parameters of TFTs obtained for the devices with 1 min of drying followed by ELA. These are all graphically depicted in Figure 4 (b) in the main paper. The reference device (300 °C for 1h) is also shown for comparison.

Fluence (mJ/cm ²)	Number of pulses	μ_{SAT} (cm ² /V·s)	SS (V/dec)	$I_{ON/OFF}$	V_{ON} (V)	V_{Hyst} (V)
225	2	18.4	0.18	1.2×10^4	- 0.36	0.06
	3	22.4	0.12	6.5×10^4	- 0.41	0.09
250	1	5.8	0.12	1.0×10^4	- 0.14	0.04
	2	14.8	0.13	1.7×10^4	- 0.20	0.04
	3	7.6	0.19	7.2×10^3	- 0.04	0.01
300	1	15.5	0.12	2.8×10^4	- 0.30	0.06
	2	14.7	0.12	1.7×10^4	- 0.20	0.02
	3	9.7	0.18	1.0×10^4	- 0.50	0
Reference (300 °C, 1h)	0	13.7	0.13	1.1×10^4	- 0.07	0.06

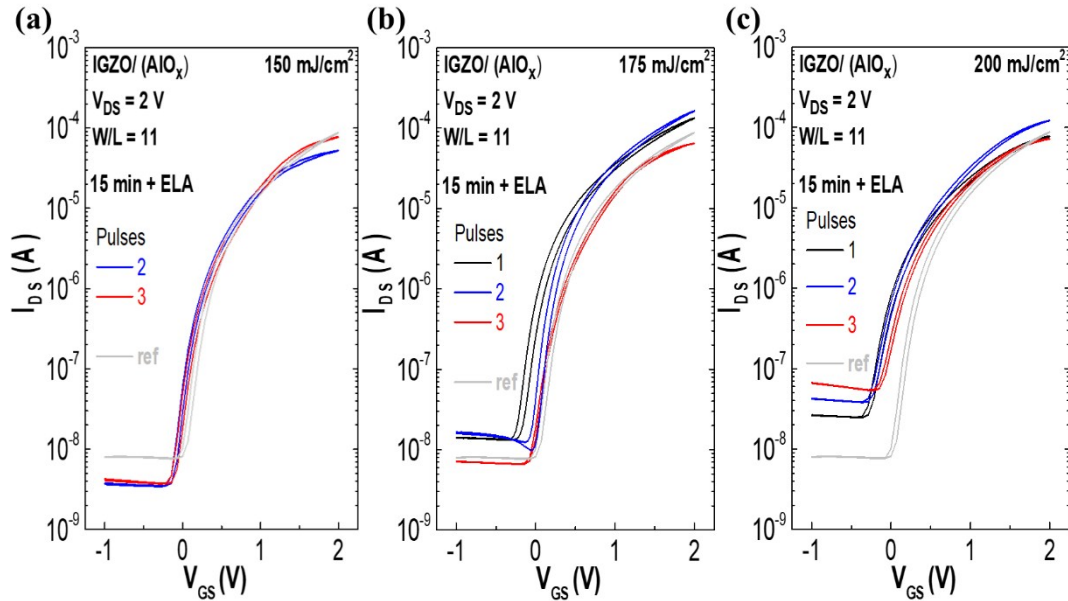


Figure S7. Transfer curves of IGZO TFTs with 15 min of drying followed by ELA treatment using different fluences and pulses: (a) 150 mJ/cm^2 ; (b) 175 mJ/cm^2 ; (c) 200 mJ/cm^2 . The reference device (300 °C for 1h) is also shown for comparison.

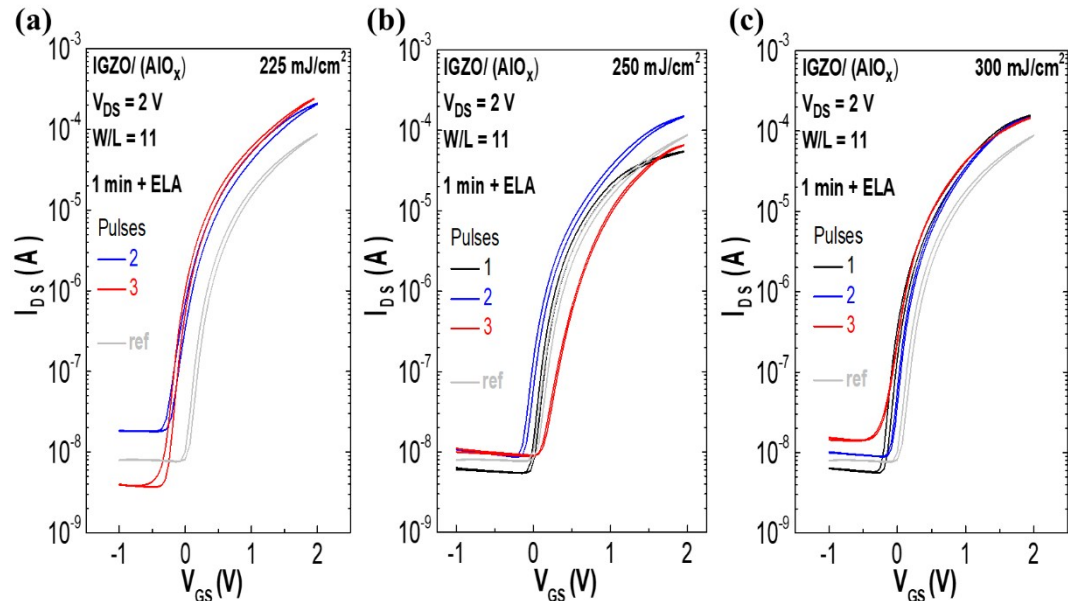


Figure S8. Transfer curves of IGZO TFTs with 1 min of drying followed by ELA treatment using different fluences and pulses: (a) 225 mJ/cm^2 ; (b) 250 mJ/cm^2 ; (c) 300 mJ/cm^2 . The reference device (300 °C for 1h) is also shown for comparison.

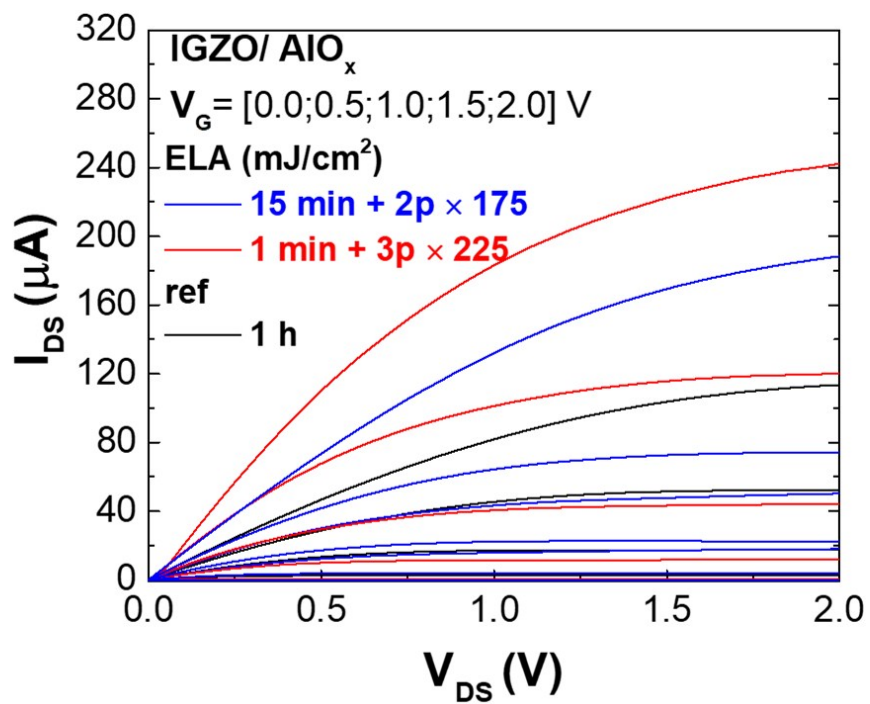


Figure S9. Typical output curves of IGZO/ AlO_x TFTs.

Table S5. Statistics of TFT parameters in a previous report¹ using deep ultraviolet (DUV) for 30 min and this work using the 15 min drying followed by ELA. These are all graphically depicted in Figure 5 (b) in the main paper.

Condition	V_{ON} (V)	μ_{SAT} ($\text{cm}^2/\text{V}\cdot\text{s}$)	SS (V/dec)
150 °C + DUV (30 min)	-0.02 ± 0.06	17.3 ± 0.9	0.09 ± 0.01
150 °C + ELA (15 min)	-0.015 ± 0.027	20.4 ± 0.9	0.103 ± 0.001

Table S6. Literature of relevant low temperature (≤ 150 °C) solution-based AlO_x layers processed by different curing methods applied in TFTs.

Year	Irradiation source (wavelength)	Irradiation time (pre-annealing time)	T (°C)	μ_{SAT} ($\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$)	Selectivity
2015 ²	Deep-ultraviolet lamp (254 nm)	30 min (-)	150	8.33	No
2016 ³	Far ultraviolet lamp (160 nm)	30 min (-)	150	14.10	No
2017 ¹	Deep-ultraviolet lamp (254 nm)	30 min (-)	150	17.3	No
2017 ⁴	Deep-ultraviolet lamp (254 nm)	120 min (-)	60	5.9	No
2019 ⁵	Deep-ultraviolet lamp (254 nm)	120 min (-)	150	~ 6	No
Present study	Excimer laser (248 nm)	2 s (15 min)	150	20.0	Yes
		3 s (1 min)		22.4	Yes

References

- 1 E. Carlos, R. Branquinho, A. Kiazadeh, J. Martins, P. Barquinha, R. Martins and E. Fortunato, *ACS Appl. Mater. Interfaces*, 2017, **9**, 40428–40437.
- 2 S. K. Park, K.-H. K. T. K. H. K.-T. Kim, J.-W. W. Jo, S. Sung, K.-H. K. T. K. H. K.-T. Kim, W.-J. J. Lee, J. Kim, H. J. Kim, G.-R. R. Yi, Y.-H. H. Kim, M.-H. H. Yoon and S. K. Park, *Adv. Funct. Mater.*, 2015, **25**, 2807–2815.
- 3 E. Carlos, R. Branquinho, A. Kiazadeh, P. Barquinha, R. Martins and E. Fortunato, *ACS Appl. Mater. Interfaces*, 2016, **8**, 31100–31108.
- 4 J.-W. Jo, Y.-H. Kim, J. Park, J. S. Heo, S. Hwang, W.-J. Lee, M.-H. Yoon, M.-G. Kim and S. K. Park, *ACS Appl. Mater. Interfaces*, 2017, **9**, 35114–35124.
- 5 B.-S. Yu, J.-Y. Jeon, B.-C. Kang, W. Lee, Y.-H. Kim and T.-J. Ha, *Sci. Rep.*, 2019, **9**, 8416.