

## Supporting Information

# Ta<sub>2</sub>O<sub>5</sub>/SiO<sub>2</sub> Multicomponent Dielectrics for Amorphous Oxide TFTs

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### Compositional and structural analysis of the multicomponent dielectrics by Rutherford backscattering spectrometry (RBS), spectroscopic ellipsometry (SE), X-ray diffraction (XRD) and Atomic Force Microscopy (AFM).

**Table S1.** Molar concentrations and film density obtained by RBS analysis of Ta<sub>2</sub>O<sub>5</sub>, SiO<sub>2</sub> and Ar in the thin films deposited using different powers in the Ta<sub>2</sub>O<sub>5</sub> and SiO<sub>2</sub> targets. Thickness of the films extracted by SE from [53].

Ta <sub>2</sub> O <sub>5</sub> power (W)	Density (10 <sup>15</sup> atm/cm <sup>2</sup> )	SiO <sub>2</sub> power (W)	Ta <sub>2</sub> O <sub>5</sub> (mol.%)	SiO <sub>2</sub> (mol.%)	Ar (at.%)	Norm. Ta <sub>2</sub> O <sub>5</sub> (mol.%)	Norm. SiO <sub>2</sub> (mol.%)	Thickness (nm)
50	1668	150	37.5	59.1	3.4	39	61	243
75	1495	150	57.6	38.6	3.8	60	40	206
100	1796	150	65.7	30.0	4.3	69	31	245
125	1712	150	70.6	25.0	4.5	74	26	233
150	1923	150	71.7	23.9	4.4	75	25	257
100	1855	-	94.5	-	5.5	100	0	232

While the Ar content is relatively similar across the film compositions presented in Table S1, a small tendency is still noticeable with the increase of the Ta<sub>2</sub>O<sub>5</sub> content. The total atomic fraction of Ar incorporated in the material,  $x_{Ar}$ , can be described as the sum of the fractions incorporated in both the Ta<sub>2</sub>O<sub>5</sub> material and the SiO<sub>2</sub> material, as shown in Equation (1),

$$x_{Ar} = x_{Ar@Ta_2O_5} x_{Ta_2O_5} + x_{Ar@SiO_2} x_{SiO_2} \quad (1)$$

where  $x_{Ta_2O_5}$  and  $x_{SiO_2}$  are the non-normalized molar fractions of Ta<sub>2</sub>O<sub>5</sub> and SiO<sub>2</sub>, respectively, and  $x_{Ar@Ta_2O_5}$  and  $x_{Ar@SiO_2}$  are the fraction of Ar incorporated in each of these materials, respectively. Knowing that the fractions of each material add to 1, as shown in Equation (2), Equation (1) can then be rewritten as Equation (3), describing the Ar content as a function of the Ta<sub>2</sub>O<sub>5</sub> content, as presented in Figure S1. Through linear fitting of this data, the fractions of Ar incorporation in each material,

$x_{Ar@SiO_2}$  and  $x_{Ar@Ta_2O_5}$ , are calculated as 1.9 % and 5.7 %, respectively, proving that  $Ta_2O_5$  is much more prone for Ar incorporation than  $SiO_2$ .

$$1 = x_{Ar} + x_{Ta_2O_5} + x_{SiO_2} \quad (2)$$

$$x_{Ar} = \left( \frac{x_{Ar@Ta_2O_5} - x_{Ar@SiO_2}}{1 + x_{Ar@SiO_2}} \right) x_{Ta_2O_5} + \frac{x_{Ar@SiO_2}}{1 + x_{Ar@SiO_2}} \quad (3)$$

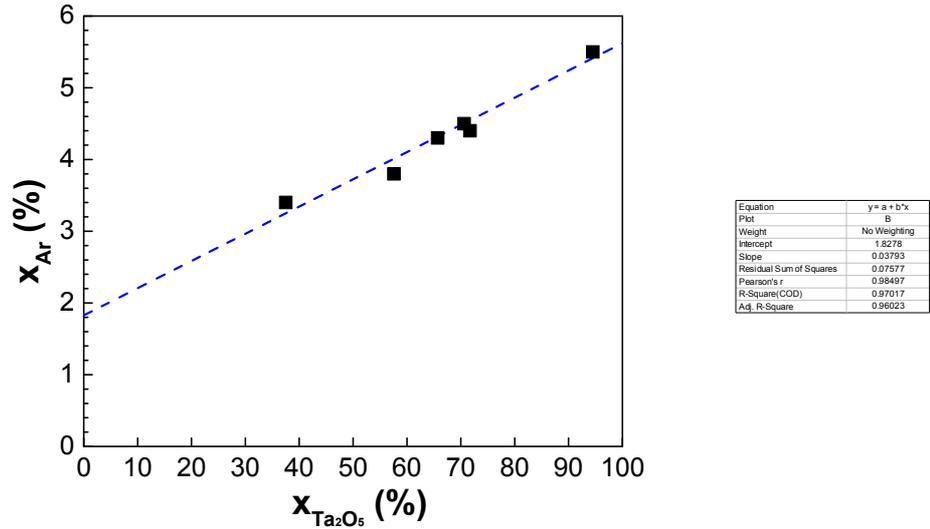


Figure S1. Ar concentration as a function of the  $Ta_2O_5$  concentration.

### Capacitance-Voltage characteristics of MIS structures.

C-V curves measured from the MIS structures. The thicknesses (as described in Table S1) and MIS areas are also included in the figure.

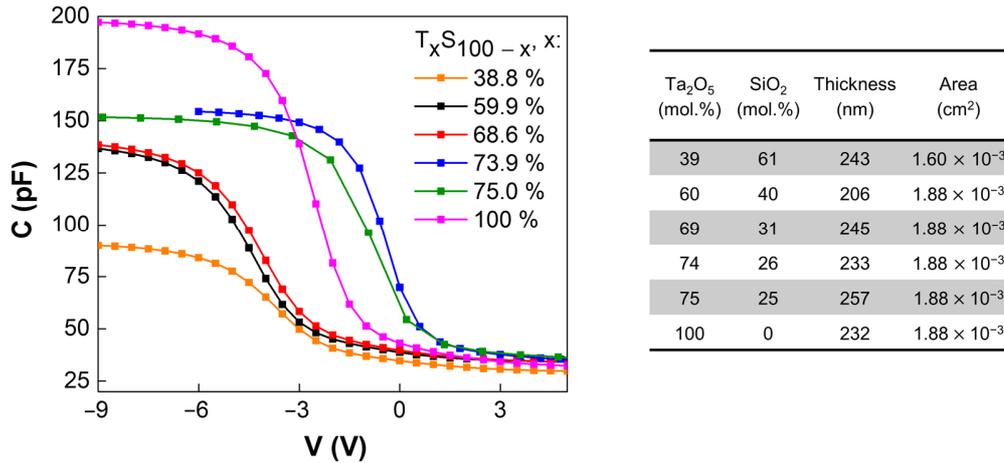
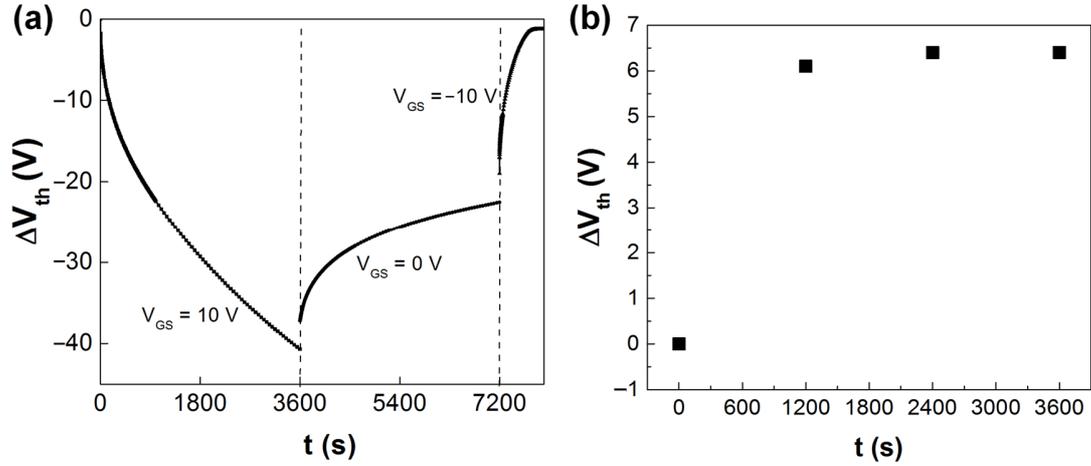


Figure S2. C-V curves of the MIS employing the  $T_xS_{100-x}$  dielectric layers. The composition and geometrical properties of the MIS are presented in the table.

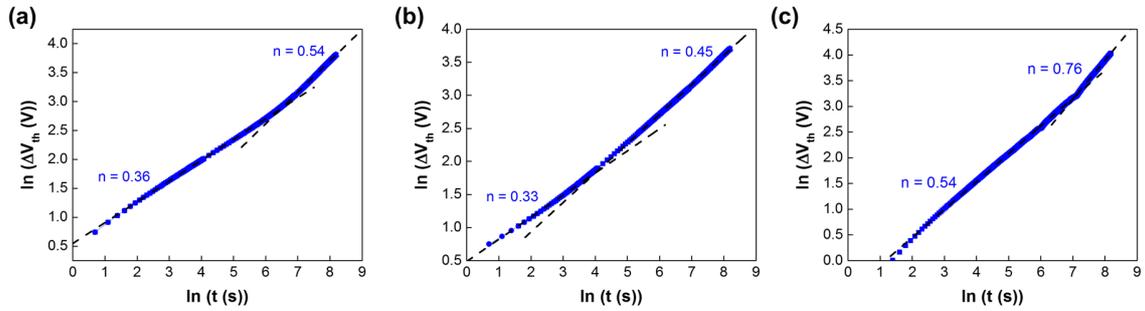
**Anomalous threshold voltage shift ( $\Delta V_{th}$ ) during PGBS, recovery and NGBS.**



**Figure S3.** (a)  $V_{th}$  shift measured during gate biasing with  $V_{GS} = 10$  V,  $V_{GS} = 0$  V and  $V_{GS} = -10$  V, sequentially, for the  $T_{69}S_{31}$  composition. (b)  $V_{th}$  shift measured during negative gate biasing with  $V_{GS} = -10$  V for an as-fabricated device with the  $T_{39}S_{61}$  composition.

While the  $\Delta V_{th}$  shift (observed only during the first minutes) during NGBS in as-fabricated devices is much lower than during PGBS, the channel is depleted during NGBS and thus only charges in the overlap areas between the gate and the source and drain electrodes are expected to respond to the biasing, making a direct comparison of these shift magnitudes unfeasible.

**Power law time dependency of  $\Delta V_{th}$  during positive gate bias stress**



**Figure S4.**  $\Delta V_{th}$  during positive gate bias stress ( $V_{GS} = 10$  V) for the (a)  $T_{60}S_{40}$ , (b)  $T_{69}S_{31}$  and (c)  $T_{69}S_{31}/SiO_2$  compositions, showing power law time dependencies.