Supporting Information

Low temperature sol-gel processed AlO_x gate dielectric buffer layer for improved performance in pentacene based OFETs

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Experimental

Preparation of AlO_x

96 % aluminum tri-secbutoxide which is the major precursor material for this synthesis was purchased from J&K Chemicals and was used as received without further purification. The preparation was conducted under dry nitrogen atmosphere due to the alkoxide's sensitivity to atmospheric moisture. Ethanolic sol of AIO_x was prepared by a low temperature sol-gel process. 2 ml of aluminum tri-secbutoxide was measured into 40 ml ethanol in a 100 ml 2-neck schlenk flask fitted with a condenser. The reaction temperature was steadily increased and the mixture gently aggitated until a clear and colourless solution was obtained. 0.5 ml of acetic acid was then added to stabilize the solution. The reaction was allowed to reflux at the boiling point of ethanol (80 °C) for 1 hour. The clear and colourless sol obtained was transferred into a 100 ml sample bottle and was allowed to age for 48 hours to form a polymeric gel.

To enable us determine the minimum annealing temperature required for the formation of a pure AIO_x thin film, the gradual decomposition of the organic components in the precursor sol was monitored using FT-IR. The polymeric gel obtained as product after ageing was spin coated on pre-fabricated KBr (Potassium bromide) pellets at 4000 rpm for 40 sec to obtain films with thickness of 20 nm. The films were annealed at 50 °C, 100 °C and 150 °C each for 20 min. A separate film was also prepared using the same deposition conditions but without annealing.

Device fabrication

Vacuum and dry nitrogen conditions were used as appropriate in the fabrication and testing of our devices. Before devices were fabricated, a contact layer of 10 nm titanium/100 nm gold was deposited on the back (rough side) of the thermal oxide wafer [200 nm SiO₂ on Si(100)] by electron-beam (e-beam) evaporation. To achieve this, the polished side was first protected with a photoresist. The wafer was subsequently cut into convenient sizes and the photoresist carefully stripped off with UV/ozone cleaning.

To characterize the dielectric properties of SiO_2 single layer and the SiO_2/AIO_x bilayer such as capacitance and leakage current, we fabricated simple capacitors using these materials. For the SiO_2 single layer, the prepared thermal oxide wafer having 200 nm thick SiO_2 layer was used directly while for the SiO_2/AIO_x bilayer, a 20 nm AIO_x buffer layer was deposited on 200 nm SiO_2 layer of the prepared thermal oxide wafer by spin coating following the aforementioned deposition conditions and then annealed at 150 °C for 20 min. A 70 nm thick top gold electrode was then deposited via thermal evaporation using a thermal evaporation system (Kurt J. Lesker) attached to the glove box. This gives simple MIM device structures of $Si/SiO_2/Au$ and $Si/SiO_2/AlO_x/Au$ respectively. A shadow mask was used to obtain a plate area of 0.09 cm².

Pentacene based OFET was fabricated using the bilayer gate dielectric system. A bottom gate top contact device structure was adopted for this fabrication. For the bilayer gate dielectric, the 20 nm AlO_x layer was deposited on the 200 nm SiO₂ and annealed at 150 °C for 20 min as already discussed. After the dielectric layer had been deposited, 40 nm of the organic active layer (pentacene) was deposited by vacuum deposition using a Kurt J. Lesker thermal evaporator at $3x10^{-4}$ ps chamber pressure and 0.1 Å/s growth rate. Au top electrodes (70 nm) which serves as the source and drain were then deposited at a growth rate of 1 Å/s without breaking the vacuum by using a shadow mask, giving the active layer a channel length of 70 µm and a channel width of 1500 µm. Fig. S1 shows the schematic diagram of the fabricated OFET. A reference pentacene-based OFET was also fabricated using the single SiO₂ layer as gate dielectric following the same device fabrication procedure.



Fig. S1. Device structure of the pentacene based OFET with AlO_x as gate dielectric buffer layer.

Characterization

The gradual decomposition of the organic components in the sol-gel processed AlO_x as a result of the low temperature annealing was monitored using Bruker Vertex 70 FTIR instrument in the 600 – 4000 nm wavenumber

range. Temperature dependent phase transition analysis was carried out by an X ray defraction technique using PANalytical designed Empyrean X ray diffractometer with Cu K α radition $\lambda = 0.154056$ nm at 40 mA and 40kV. The AlO_x thin film thickness was determined by AFM technique using Veeco atomic force microscope which was also used in the tapping mode to characterise the material's surface morphology. Chemical composition of the material was studied by using an Ultra DLD Shimadzu X-ray photoelectron spectrometer (XPS) and Casa XPS program (Casa Software Ltd, UK). C-F charateristics were obtained for the frequency dependent capacitance of the single layer and bilayer gate dielectric materials using Wayne Kerr 6500B precision impedance analyzer. The gate dimensions of OFETs fabricated were defined by using a shadow mask and the resulting channel length *L* and width *W* were mearsured under high vacuum using the length measurement software on FEI designed Quanta 200 FEG scanning electron microscope (SEM). The temperature dependent electronic behavior of the OFETs such as output characteristics were studied by using a high vacuum probe Lakeshore probe station connected to Keithley 4200 semiconductor parameter analyzer. Contact angle measurements were conducted on the on the surfaces of SiO₂/AlO_x bilayer and SiO₂ single layer gate dielectrics by using a goniometer (G 10, Krüss) equipped with an image capture software (Drop Shape Analysis 1.0 software).

Table S1. Peak parameters of Al 2p and O 1s core levels and the quantified Al:O ratio in AlO_x thin film annealed at 150 °C.

Name	Position	FWHM	Area	At %
Al 2p	74.6	1.72	18543.3	37.7
O 1s	531.2	2.67	64901.7	62.3



Fig. S2. XRD pattern of the AlO_x thin film annealed at 150 °C confirms the amorphous nature of the material.



Fig. S3. Temperature dependent transfer curves of OFET (a) without AlO_x buffer layer and (b) with AlO_x buffer layer. The transfer curves are logarithmic plots of $-I_{DS}$ vs V_{GS} and a plot of $(-I_{DS})^{1/2}$ vs V_{GS} taken at $V_{DS} = -30$ V.



Fig. S4. Images of droplets of different test liquids with contact angle values. (a) water on SiO_2/AIO_x bilayer, (b) water on SiO_2 single layer, (c) diiodomathane on SiO_2/AIO_x bilayer and (d) diiodomathane on SiO_2 single layer.