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Structural and Electrical Properties of Al_2O_3 film grown by Mist Chemical Vapour Deposition

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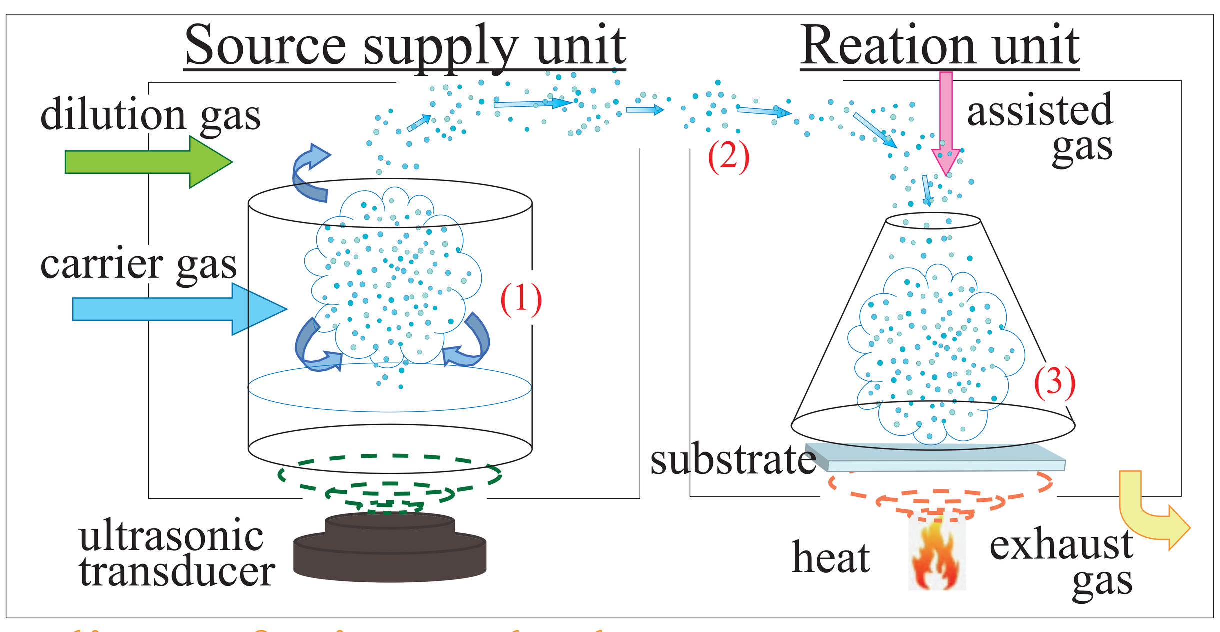
– Mist Chemical Vapour deposition (Mist CVD) –

A technique promising as an atmospheric pressure process!

Schematic view of mist method



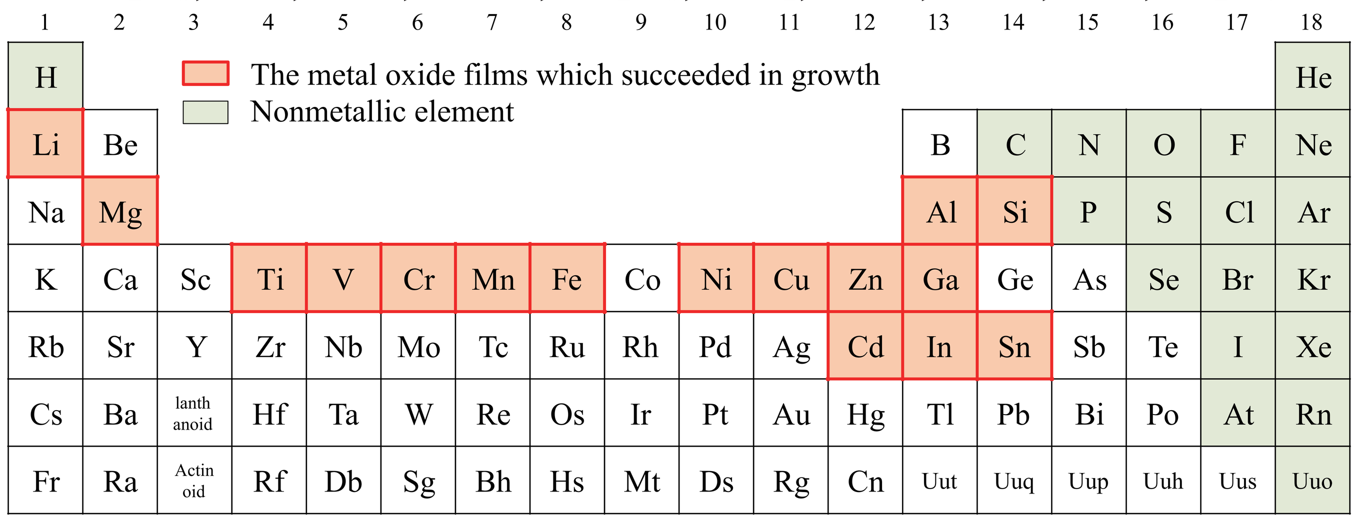
Outlines of mist method

1) The source supply unit
   The material solution is atomized to form mist droplets in the diameter scale of several micrometers with ultrasonic transducer.
2) The transportation unit
   The mist droplets was transferred by carrier gas and dilution gas from the source supply unit to the reaction unit.
3) The reaction unit
   The thin films or particles are formed by the thermal decomposition.

Producible films

Metal oxide & organic films

Al_2O_3, SiO, ZnO, IGZO, Ga_2O_3, TiO_2, AZO, GZO, ITO, Fe_2O_3

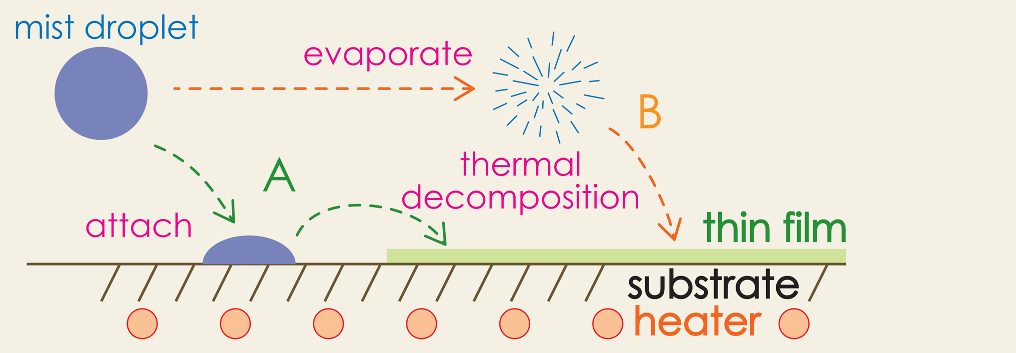


Advantage of mist method

A. "Stable materials" can be treated as "gas".
B. Selection operation of CVD and spray.
C. High experimental efficiency.
D. Less oxide-defect oxide thin film can be grown.

B. Selection operation of CVD & Spray.

A. Mist droplet directly attach to the substrate
B. Mist droplet evaporates before attaching to the substrate.

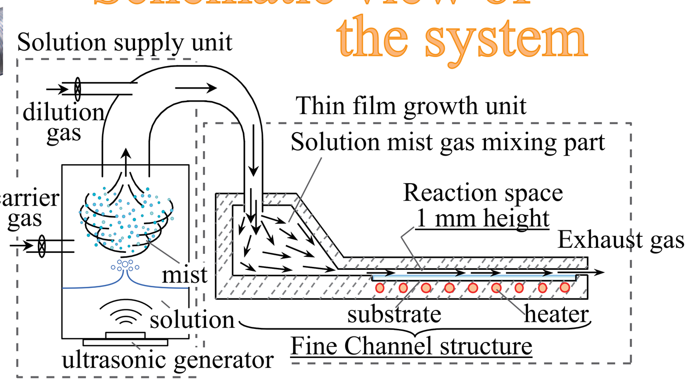


There is a big merit that stacked films can be continuously grown because the source solution does not directly attach to surface of the target film [5].

5) T. Kawaharamura, et al., Convertech, Vol.39 (2011) pp.111 [in Japanese].

Fine channel type Mist CVD system in our lab.

Schematic view of the system



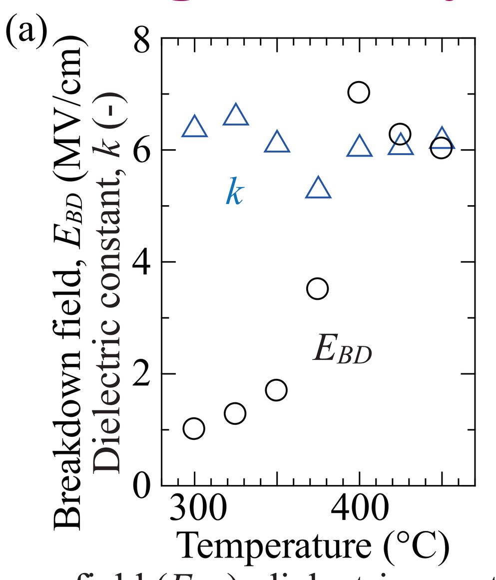
Please refer **"Study on mist CVD and its application to the growth of ZnO thin films"**
T. Kawaharamura,, Ph.D. Thesis, Faculty of Engineering, Kyoto-Univ., 2008 [in Japanese]
http://repository.kulib.kyoto-u.ac.jp/dspace/bitstream/2433/57270/1/26041.pdf

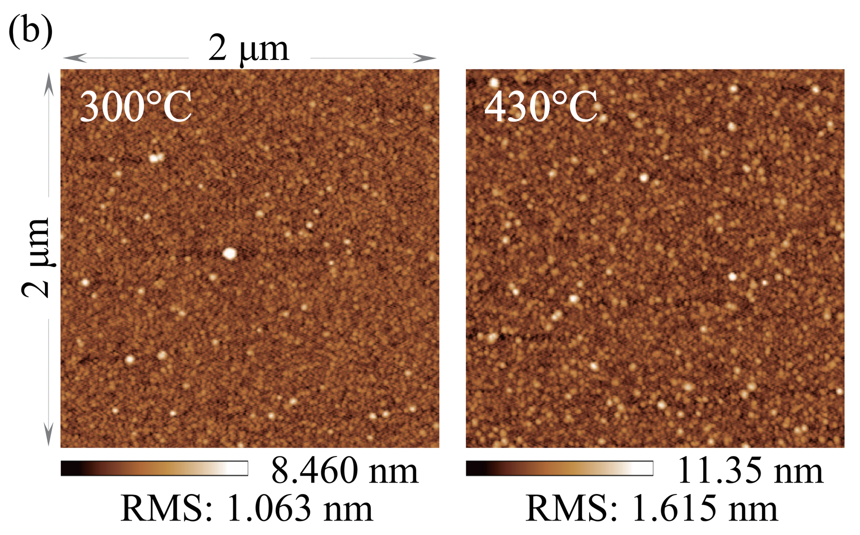
– Properties of AlO_x thin films grown by Mist CVD –

Table 1. Deposition conditions of AlO_x thin films

Solute	Aluminum acetylacetonate ($AlAcac_3$)*2
Solvent (mixing ratio)	Distillated water*3, Methanol*4 (10 : 90)
Solution concentration	0.020 mol/L
Thickness	≈ 50 nm, 200 nm
Substrate temperature	300, 350, 400, 430 °C
Substrate	p+-Si*5
Growth system	φ100 mm ver. FC type mist CVD system*6
Carrier gas / flow rate	Air, 2.5 L/min × 2
Dilution gas / flow rate	Air, 10.0 L/min × 2
Ultrasonic transducer*1	2.4 MHz, 24 V・0.625 A, 6

*1 HONDA ELECTRONICS, HM-2412
*2 Aluminum acetylacetonate, 99%, Sigma-Aldrich
*3 Wako Pure Chemical Industries
*4 Methanol, 99.8%, Wako Pure Chemical Industries
*5 Advantec
*6 Refs. 9 and 16.

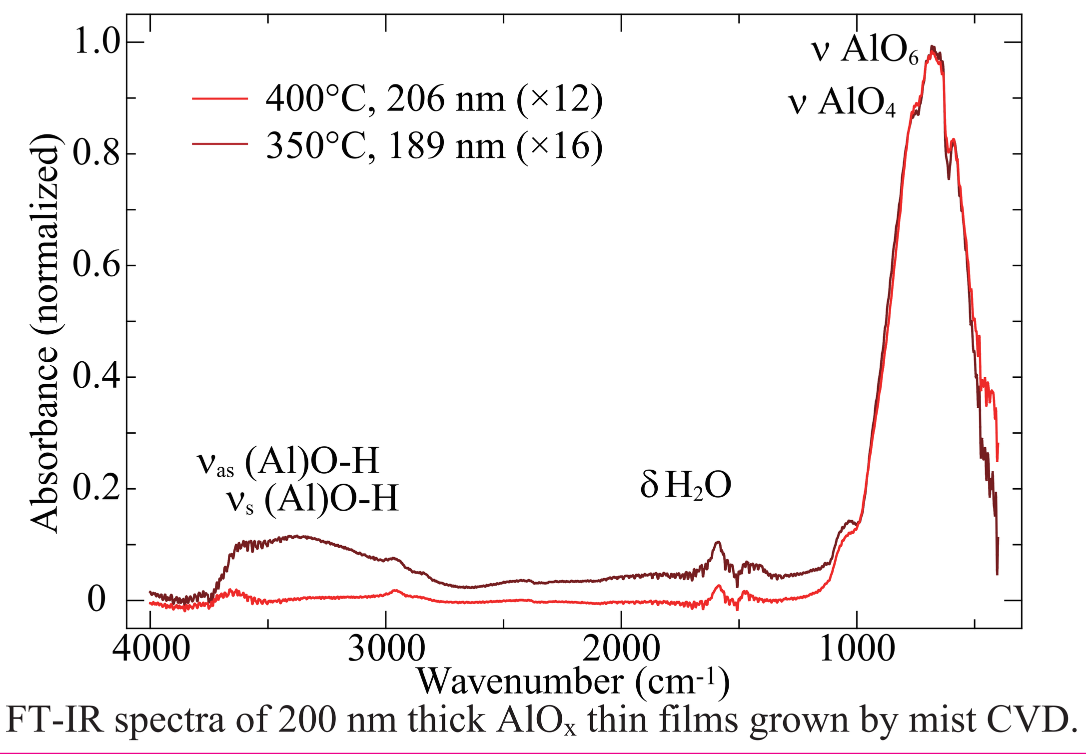




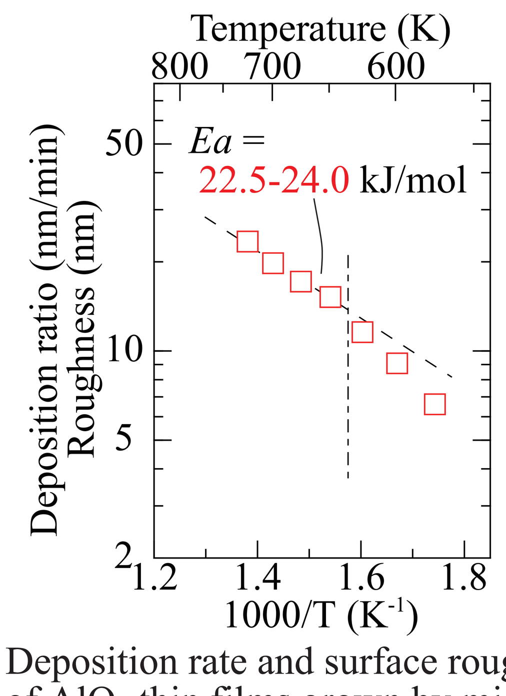
$T > 400$°C
$E_{BD} > 6.0$ MV/cm
$k > 6.1$
$RMS = 1.6$ nm

(a) Breakdown field (E_{BD}), dielectric constant (k), and (b) surface structure of AlO_x thin films grown by mist CVD.

The AlO_x thin films grown over 400°C exhibited the breakdown field and the dielectric constant of 6.0 MV/cm and 6.1, respectively. However, the E_{BD} of AlO_x thin film grown at temperatures below 350°C was dramatically declined.



FT-IR spectra of 200 nm thick AlO_x thin films grown by mist CVD.



Deposition rate and surface roughness of AlO_x thin films grown by mist CVD.

Compared with the AlO_x thin films deposited at various temperature, the difference of the shoulder around 1100 cm^{-1} corresponding to the bending vibration of Al-O-H (ν_s (Al)O-H or ν_{as} (Al)O-H) and the difference of peak around 2350 cm^{-1} corresponding to the stretching vibration of CO_2 adsorbed can be seen.

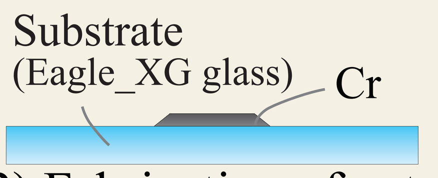
The logarithm of deposition ratio and surface roughness are not directly proportion to the reciprocal of the substrate temperature. The line has changed around 350°C.

It is suggested that a residual as boehmite (γ-AlO(OH)) remains in the AlO_x thin film grown at the temperature below 350°C. It is thought that that is one of the reasons why the E_{BD} dramatically declines.

- Fabrication of Oxide TFT with an IGZO/AlO_x stack by mist CVD -

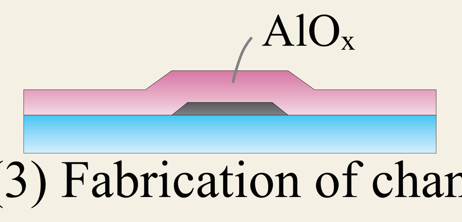
- Fabrication process -

(1) Fabrication of gate electrode



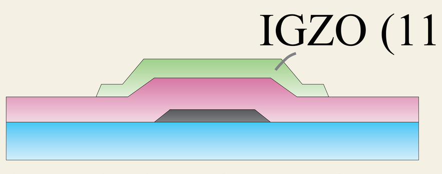
sputtering, 50 nm 150°C
Patterning: Wet etching

(2) Fabrication of gate insulator



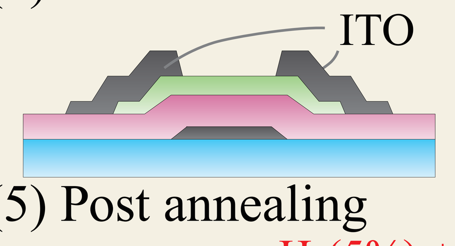
mist CVD (1 atm)
430°C, ≈ 100 nm
Patterning: Wet etching

(3) Fabrication of channel layer



mist CVD (1 atm)
350°C, ≈ 50 nm
Patterning: Wet etching

(4) Fabrication of source & drain electrode

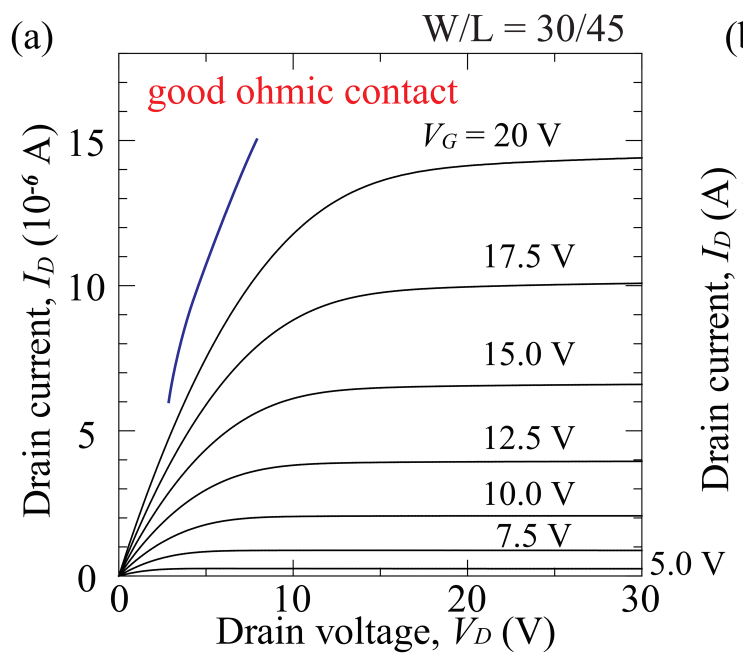


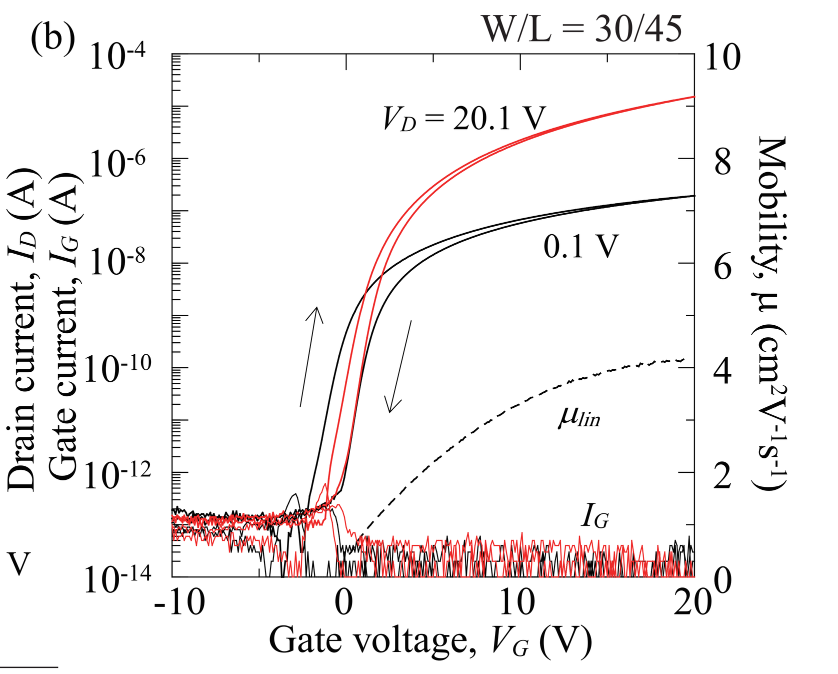
sputtering, 50 nm, RT
Patterning: Wet etching (lift-off)

(5) Post annealing

H_2(5%) + N_2, 350°C, 1 h

- The drive of the first prototype -





Mobility µ ($cm^2V^{-1}s^{-1}$)	linear	:	4.2
	saturation	:	4.0
V_{GS} at I_{DS} = 1 nA (V)		:	0.39
S (V/dec.) @ 10-100 pA		:	0.55
Hysteresis ΔV_H (V)		:	0.68
I_{on}/I_{off} at V_{GS} = 30/-10 V		:	> 10^8
Gate leakage current @ V_G = 20V		:	<10^{-12}

The oxide TFT was driven with low leakage current & good on/off ratio.

S & ΔV_H are worse than previous reports

(a) Output and (b) transfer characteristics of oxide TFT with a IGZO/AlOx stack fabricated by mist CVD.

Conclusion

1) Aluminium oxide (AlO_x) thin film was grown using aluminium acetylacetonate ($AlAcac_3$) as a source solute by mist chemical vapour deposition (mist CVD).
2) The AlO_x thin films grown over 400°C exhibited the breakdown field (E_{BD}) and the dielectric constant (k) of over 6.0 MV/cm and 6.1, respectively.
3) The oxide TFT consisting of channel layer (IGZO) (47 nm) and gate insulator (AlO_x) (116 nm) exhibits the field-effect mobility (μ_{lin}) and on/off ratio of 4.2 cm^2/(V・s) and over 10^8, respectively

Please refer
"Electrical Properties of the Thin-Film Transistor With an Indium–Gallium–Zinc Oxide Channel and an Aluminium Oxide Gate Dielectric Stack Formed by Solution-Based Atmospheric Pressure Deposition"
M. Furuta, T. Kawaharamura, D. Wang, T. Toda, and T. Hirao,
IEEE Electron Device Lett. Vol.33 (2012) pp.851-853.

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