ITO SPUTTER COATED FILMS FOR TOUCH PANEL APPLICATIONS USING ROTARY SINTERED CERAMIC ITO TARGETS: WHAT CAN BE LEARNED FROM GLASS COATING?

Paul Lippens

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Outline

- Introduction:
  rotary sintered ceramic ITO targets - product, applications and advantages

- Experiences from the glass coating industry relevant for web coating:
  - Instability of RT deposited ITO coatings
  - Influence of hydrogen added to the plasma

- Coating cost implication of rotary ITO targets in web coating

- Conclusions
I. Introduction:
rotary sintered ceramic ITO targets:
product, applications and advantages
Rotary sintered ceramic ITO targets

- Physico-chemical properties of ITO cylinders are identical to ITO-plates:
  - Density: $\geq 7.10 \text{ g/cc}$
  - Chemical purity 4N
- Multiple cylinders are bonded onto Ti-backing tube. Typical backing tube OD = 133 mm.
- ITO-cylinders:
  - ITO-ID: standard = 135 mm
  - ITO-wall thickness, standards today:
    - 6 - 10 mm
    - 12 mm wall thickness in development
  - Segment length: appr. 200 mm
Established applications

- PV:
  - (Flexible) CIGS & TF-Si PV-modules (substrate: stainless steel and polymer foil)
  - New kinds of rigid PV products

- Architectural glass

- Display (substrate: glass):
  - TFT-array (OLED and LCD)
  - Touch Panel on glass
  - Colour Filter (LCD)

- Products were introduced in the market in 2007
- Take-up in web coating only emerging today
Advantages of rotary ITO

- Higher TU: typically 85 % (up to 90 %) vs. 20-35 % for planar.
- Main advantage: avoidance of black nodule formation at high power loads
Nodules when sputtering ITO

- Nodules originate due to re-deposition of sputtered species onto the target.
- Black nodules consist mainly of Indium-sub-oxide ($\text{In}_2\text{O}$): their formation can be understood as an equilibrium reaction [1]:

$$\text{In}_2\text{O}_3 \leftrightarrow \text{In}_2\text{O} + \text{O}_2$$

- The equilibrium is shifted to the right:
  - For high target temperatures (associated with high power levels)
  - For low target density ($< 7.10 \text{ g/cc}$) [2]
  - If residual $\text{SnO}_2$ traces are present (the latter can cause arcing) [3]
  - At accumulations of impurities. [4]
Ceramic sintered rotary ITO

- Black (In$_2$O) nodules are avoided for power loads up to 15 kW/m
- Grey, less harmful, In$_2$O$_3$ nodules are avoided for power loads up to approx. 10 kW/m (DDR = 75 – 110 nm.m/min depending on $T_{sub}$, pressure, magnet array and shielding)

15 kW/m during 36 hours (approx. 500 kWh)
Consequential advantages

• More stable, arc free sputtering process
  ➢ Less particles, higher product yield
• Higher machine up time due to (less target changes/cleanings) :
  ▪ Higher available amount of ITO per target
  ▪ Higher TU and higher CE (less ITO on shields)

➢ Overall: much lower coating costs for rotary ITO!
II. Experiences from the glass coating industry relevant for web coating
Experiments in AT Lab Balzers, FL

UTTU: Umicore Target Testing Unit

Twin planar magnetron    Twin rotary magnetron
Instability of RT deposited ITO coatings

135-150 nm ITO-90/10 coatings (2 passes @1.54 m/min) on glass at RT (10 kW/m; 2.6-2.7E-03 mbar). Exposure to normal atmosphere.
Instability of RT deposited ITO coatings

![Graph showing the thickness and Hall mobility of ITO coatings over time after RT deposition with different oxygen concentrations.]

- **Thickness t [nm]**
- **Hall mobility μ [cm²/(V.s)]**

# days after RT deposition

- **0 % O2, t**
- **1 % O2, t**
- **1.5 % O2, t**
- **2 % O2, t**
- **0 % O2, μ**
- **1 % O2, μ**
- **1.5 % O2**
- **2 % O2, μ**
Instability of RT deposited ITO coatings

Upon exposure to atmosphere at RT:

- Hall resistivity drops with 10-15 % in about 1 month:
  - Carrier concentration increases with same amount
  - Mobility remains quasi constant
- Most probably, intergrain residual amorphous traces are spontaneously crystallized.

- Web coating requires RT deposition or cooling:
  - Fix electro-optical properties by post-annealing at min. some 150°C (e.g. during hot lamination).
  - Or: stack design must take initial instability into account.
Influence of hydrogen in the sputter plasma

- Hydrogen can be added to the sputter gas as either H$_2$O flow or H$_2$ flow
- Hydrogen usage in the sputter gas:
  - Yields smoother coatings (Keran Zhang et al. and E. Nishimura et al.)
  - Improves electrical properties (Zhang and Lippens)
  - Is known to yield better coating uniformities.
- In polymer web coating: there is an inherent water vapour load in the plasma (even when Meissner trap is used).
Influence of hydrogen in the plasma

ITO-90/10 coatings deposited at 10 kW/m (rotary magnetron).

Annealing: 200°C, 30’ in air. Main (222) In$_2$O$_3$-peak

As-deposited coatings: quasi amorphous for ≥ 1.6 % H$_2$ in flow

After annealing: all coatings are fully crystalline (higher d-spacing)
Influence of hydrogen in the plasma

As deposited ITO-90/10 coatings with > 1.6 % H₂ in flow have lower conductivity; H shifts resistivity minimum to higher O₂ flow and lower values.
Influence of hydrogen in the plasma

ITO-97/3 coatings deposited at 10 kW/m (rotary magnetron).
Annealing: 200°C, 30’ in air. Main (222) peak

As-deposited coatings: less pronounced amorphization effect of H
After annealing: all coatings are fully crystalline
Influence of hydrogen in the plasma

ITO-coatings deposited at 10 kW/m (rotary magnetron) on glass.

RT + annealing: 200°C, 30’ in air.

If H is present in the sputter plasma, lowly doped ITO-grades become of greater interest.
III. Coating cost implication of rotary ITO targets in web coating
Example: ITO-coated film for projected capacitive touch panels

Optical coating stack, increases refractive index of PET/HC to appr. 1.95, equal to ITO

\[ R_{ITO} = R_{etch} \text{ – ‘invisible’ ITO} \]

High index film: e.g. \( \text{Nb}_2\text{O}_5, \text{SiN}_x, \text{TiO}_2, \text{ZrO}_2, \ldots \) (selected a.o. to guarantee colour neutrality)
Web coater configurations for ‘invisible’ ITO

• Consider: single or double drum web coaters
• Assume 5 coating bays per drum (either single or twin rotary, single or twin planar)
• All coatings in optical stack are reactively sputtered with metallic targets, twin cathodes and MF-AC
• ITO is deposited with single cathode and DC (rotary or planar)
# Table: Web coater configurations for ‘invisible’ ITO

<table>
<thead>
<tr>
<th></th>
<th>Single drum with sprayed Si</th>
<th>Double drum with sprayed Si</th>
<th>Double drum with cast Si</th>
</tr>
</thead>
<tbody>
<tr>
<td># magnetrons for SiOx</td>
<td>1</td>
<td>2 (1)</td>
<td>2 (1)</td>
</tr>
<tr>
<td>Coating thickness SiOx [nm]</td>
<td>12</td>
<td>12</td>
<td>12</td>
</tr>
<tr>
<td>DDR SiOx needed [nm.m/min]</td>
<td>19.2</td>
<td>19.2 (38.4)</td>
<td>26.4 (52.8)</td>
</tr>
<tr>
<td># magnetrons for high index film</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Coating thickness high n [nm]</td>
<td>7</td>
<td>7</td>
<td></td>
</tr>
<tr>
<td>DDR high n needed [nm.m/min]</td>
<td>11.2</td>
<td>22.4</td>
<td>30.8</td>
</tr>
<tr>
<td># magnetrons for SiO₂</td>
<td>3</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>Coating thickness SiO₂ [nm]</td>
<td>60</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>DDR SiO₂ [nm.m/min]</td>
<td>32</td>
<td>32</td>
<td>44</td>
</tr>
<tr>
<td>Web speed [m/min]</td>
<td>1.6</td>
<td>3.2</td>
<td>4.4</td>
</tr>
<tr>
<td># single magnetrons for ITO</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Coating thickness ITO [nm]</td>
<td>21</td>
<td>21</td>
<td>21</td>
</tr>
<tr>
<td>DDR ITO needed [nm.m/min]</td>
<td>33.6</td>
<td>67.2</td>
<td>92.4</td>
</tr>
<tr>
<td>Power on ITO needed [kW/m]</td>
<td>3.73</td>
<td>7.47</td>
<td>10.27</td>
</tr>
</tbody>
</table>

Determines coater configuration and web speed.
Web coater configurations for ‘invisible’ ITO

- SiOx nucleation/barrier layer:
  - DDR is higher than for SiO₂ (about 10 %)
  - Can be done with single twin rotary cathode on double drum if thickness is reduced. Assume two twin rotary cathodes in this calculation.

- High index material:
  - Nb₂O₅ from stoichiometric rotary ceramic targets (MF-AC, twin cathode):
    DDR = 100 nm.m/min (ref. T. Preussner et al.)
  - TiO₂ from rotary ceramic targets (MF-AC, twin cathode):
    DDR = 53 nm.m/min (ref. T. Preussner et al.)
  - Hence 1 twin cathode is always enough

- Consider also cast-Si rotary targets for SiO₂/SiOₓ deposition so as to exploit max. possible ITO power load to avoid (grey) nodules (i.e. appr. 10 kW/m).
## Parametrization for ITO coating cost comp.

<table>
<thead>
<tr>
<th>Case 1: planar, single drum; sprayed Si</th>
<th>Case 2: rotary, single drum; sprayed Si</th>
<th>Case 3: rotary, double drum; sprayed Si</th>
<th>Case 4: rotary, double drum; cast Si</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>ITO Target Utilization, TU [%]</strong></td>
<td>25.5*</td>
<td>85</td>
<td>85</td>
</tr>
<tr>
<td><strong>Collection efficiency, CE [%]</strong></td>
<td>50</td>
<td>57.5</td>
<td>57.5</td>
</tr>
<tr>
<td><strong>Power on ITO target [kW/m]</strong></td>
<td>4.293</td>
<td>3.733</td>
<td>7.47</td>
</tr>
<tr>
<td><strong>Change time target (2 oper.) [hrs]</strong></td>
<td>1</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td><strong>Wt available per ITO target [kg]</strong></td>
<td>14.38</td>
<td>46.25</td>
<td>46.25</td>
</tr>
<tr>
<td><strong>Total stand-time including maintenance and set up [hrs]</strong></td>
<td>125.19</td>
<td>1644.24</td>
<td>1011.03</td>
</tr>
<tr>
<td><strong>Relative ITO target cost</strong></td>
<td>1</td>
<td>2.5</td>
<td>2.5</td>
</tr>
<tr>
<td><strong>Substrate speed [m/min]</strong></td>
<td>1.60</td>
<td>1.60</td>
<td>3.20</td>
</tr>
<tr>
<td><strong>Product yield [%]</strong></td>
<td>95</td>
<td>98</td>
<td>98</td>
</tr>
<tr>
<td><strong>Dynamic Deposition Rate (DDR) ITO [nm.m/min]</strong></td>
<td>33.60</td>
<td>33.60</td>
<td>67.23</td>
</tr>
</tbody>
</table>

* 30 % minus 15 % additional loss due to nodule grinding
### Parametrization for ITO coating cost comp.

<table>
<thead>
<tr>
<th></th>
<th>Case 1: planar, single drum; sprayed Si</th>
<th>Case 2: rotary, single drum; sprayed Si</th>
<th>Case 3: rotary, double drum; sprayed Si</th>
<th>Case 4: rotary, double drum; cast Si</th>
</tr>
</thead>
<tbody>
<tr>
<td>Investment cost web coater (5 or 10 magnetrons), [M$]</td>
<td>6.25</td>
<td>6.25</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>Effective yielded production [m²/hr]</td>
<td>87.60</td>
<td>92.89</td>
<td>151.06</td>
<td>182.13</td>
</tr>
<tr>
<td>Yielded production for 1 ITO target [# rolls]</td>
<td>5.7</td>
<td>79.38</td>
<td>79.38</td>
<td>79.38</td>
</tr>
</tbody>
</table>

**Other fixed parameters:**

- ITO-target thickness: 9 mm (rotary: length 1600 mm; planar 1500 mm X 125 mm). In-metal price of 500 $/kg used.
- Substrate roll: length 1500 m, usable width 1300 mm (effect. width 1500 mm)
- Electricity cost: 0.094 c$/kWh. Labour rate: 30 $/hr. 350 coating days (in 24h/7day system). Depreciation coater over 5 years.
## Coating cost ITO: comparison

<table>
<thead>
<tr>
<th>Case</th>
<th>Description</th>
<th>Cost (c$/m²)</th>
<th>Materials cost</th>
<th>Labour cost</th>
<th>Energy cost</th>
<th>Machine cost</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>planar, single drum; sprayed Si</td>
<td>2.038</td>
<td>0.398</td>
<td>0.721</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Case 2</td>
<td>rotary, single drum; sprayed Si</td>
<td>1.922</td>
<td>0.371</td>
<td>0.424</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Case 3</td>
<td>rotary, double drum; sprayed Si</td>
<td>1.891</td>
<td>0.247</td>
<td>0.424</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Case 4</td>
<td>rotary, double drum; cast Si</td>
<td>1.569</td>
<td>0.213</td>
<td>0.424</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Cost savings between cases:
- Case 2 vs. Case 1: -13.9%
- Case 3 vs. Case 1: -18.8%
- Case 4 vs. Case 1: -30.1%
Conclusions
CONCLUSIONS

• ITO coating costs are considerably reduced when using rotary magnetrons with sintered ceramic ITO rotary targets due to nodule free sputtering at high power loads combined with much higher target utilization.

• This is also true for polymer web coating. In the specific case of ‘invisible’ ITO for projected capacitive touch panels, coating cost reduces with at least 14% (single drum): using double drum and cast-Si rotary targets, the cost benefit rises even to 30%.

• Quite some of the established know how on room temperature, high power rotary ITO sputtering from the glass industry is helpful for polymer film coating applications (RT instability requiring the need for post-annealing and the influence of hydrogen in the sputter plasma).
Thank you for your attention!

Photograph courtesy of Apple