ELECTRICAL AND OPTICAL PROPERTIES OF HIGH DEPOSITION RATE SPUTTERED ZINC OXIDE AND TITANIA BASED TCOs

Fraunhofer FEP

M. JUNGHÄHNEL, U. HARTUNG, B. HEIMKE, T. KOPTE, T. PREUßNER

FRAUNHOFER-INSTITUT FÜR ELEKTRONENSTRAHL- UND PLASMATECHNIK FEP, DRESDEN

ABSTRACT

We present the results of our investigations of zinc oxide and titania based TCO thin film deposition on large area substrates. Undoped and doped zinc oxide (i-ZnO, ZnO:Al, ZnO:Ga) as well as doped titania (TiO_2:Nb) thin films were produced on large area substrates of (300 × 300) mm² by direct current sputtering with high deposition rates using either single planar or tubular targets. The films were deposited dynamically onto glass using a vertical pilot scale in-line sputtering plant. This setup allows a reproducible preparation of the TCO films with excellent homogenous properties under close to production conditions.

SPUTTERING PROCESS

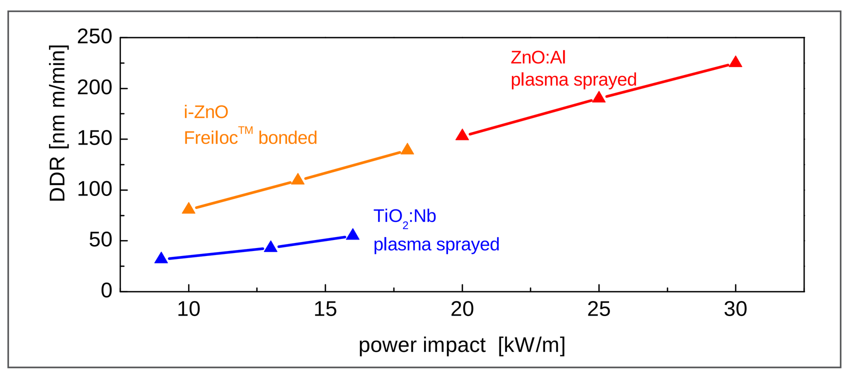
Magnetron system	• Rotatable magnetron system (Soleras Advanced Coatings) • Planar magnetron system (Leybold system)
Target length	• Tubular targets - 780 mm • Planar targets - 750 mm
Target material	• i-ZnO, tubular target • ZnO:Al, Al 2 wt.-%, tubular • ZnO:Ga, Ga 3 wt.-%, planar • TiO_2:Nb, Nb 4 wt.-%, tubular
Powering	• DC sputtering • Pulse DC sputtering (PMS) by using UBS-C2 (Fraunhofer FEP equipment) The UBS-C2 converts DC power into square wave current pulses and ensures minimum energy input in case of an arc even at high discharge power through intelligent arc handling.
Process temperature	Room temperature
Annealing temperature	400°C … 450°C
Annealing atmosphere	High vacuum (< 5 × 10^{-5} mbar)
Substrate size	300 × 300 mm²
Substrates	Low iron float glass, Borofloat® glass

TiO_2:Nb tubular target in a rotatable magnetron arrangement.

Ultrafast switching unit UBS-C2 (Fraunhofer FEP)

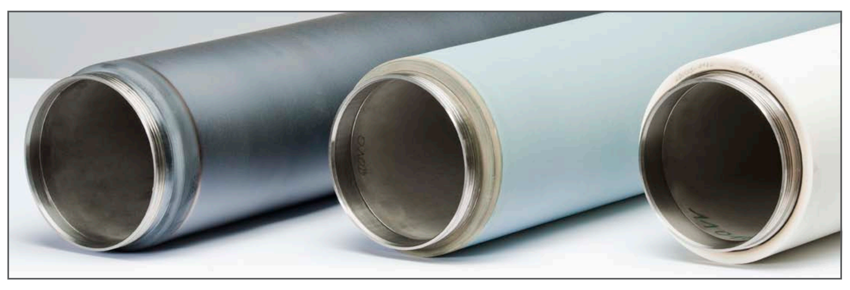
Rotatable magnetron systems show some benefits compared to planar magnetron systems:

- Higher deposition rates because of higher power density
- Higher stability of process conditions, especially in the reactive mode,
- Reduced particle generation during the process,
- Higher utilization of material



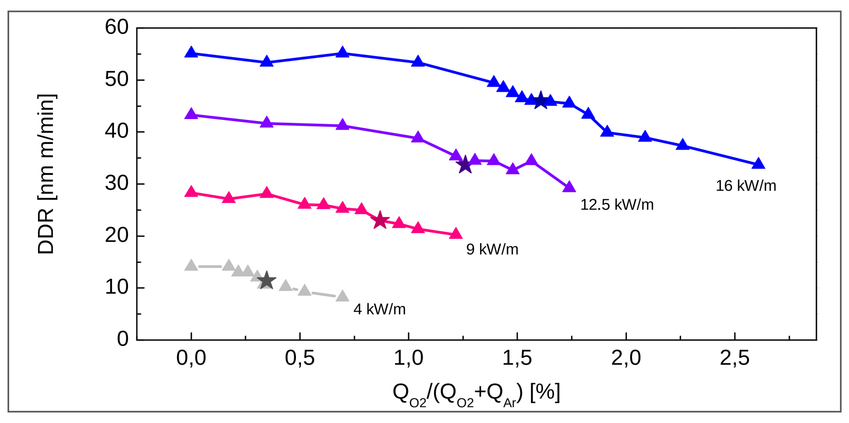
Dynamic deposition rates DDR in dependence on power impact for several tubular targets.

In-line sputter plant ILA 900.

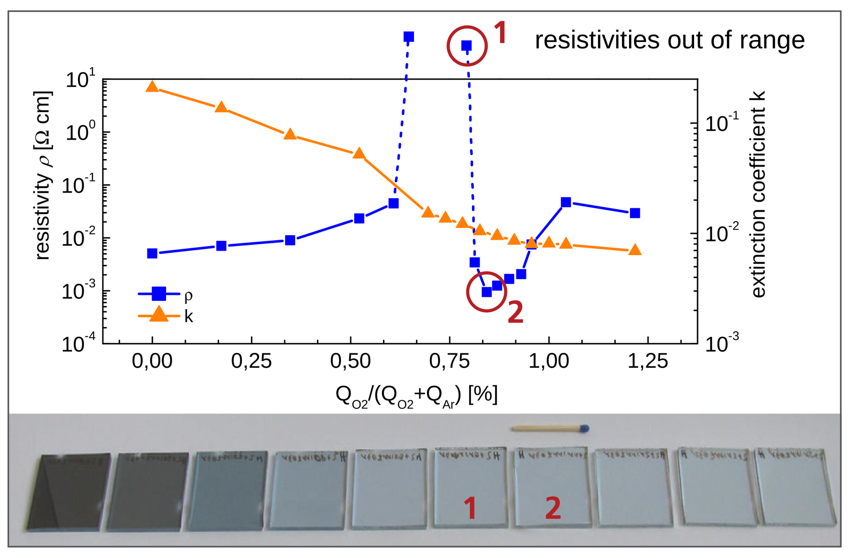


Tubular targets of 780 mm length: TiO2:Nb, ZnO:Al, i-ZnO.

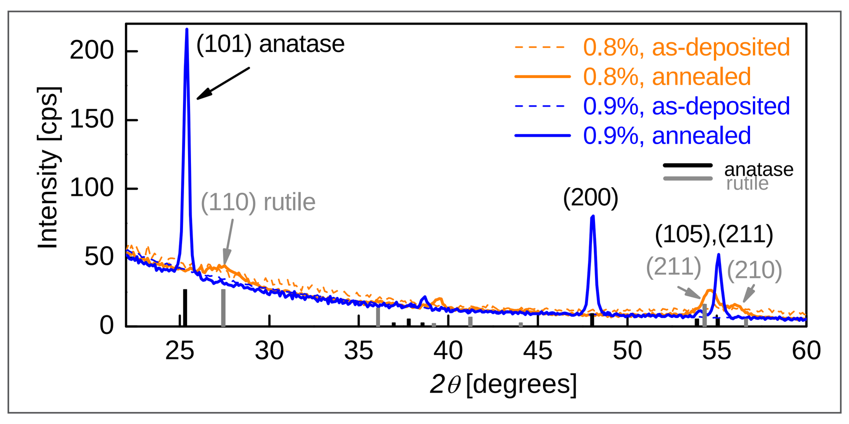
TITANIA BASED TCOs



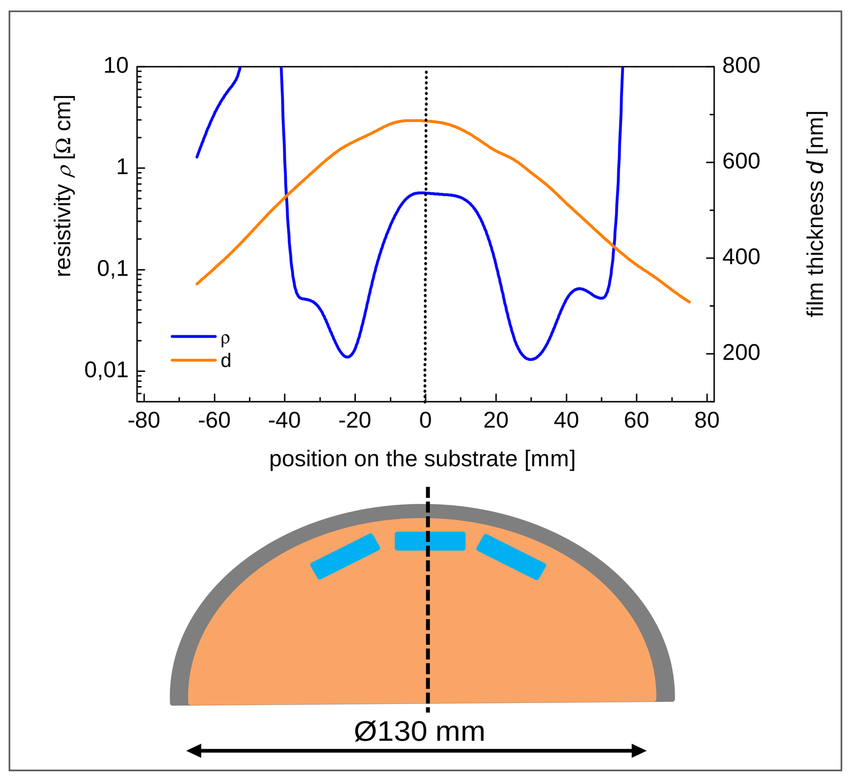
Influence of additional oxygen during the sputtering process on the dynamic deposition rate DDR for TiO_2:Nb films deposited from the TiO_x:Nb target at different DC power impacts.



Influence of additional oxygen during the sputtering process on the resistivity and extinction coefficient for TiO_2:Nb films deposited at a DC power impact of 9 kW/m.



XRD diagram of DC sputtered TiO_2:Nb films with used 0.8% and 0.9% additional oxygen to the total gas during sputtering process in as-deposited state and after annealing.

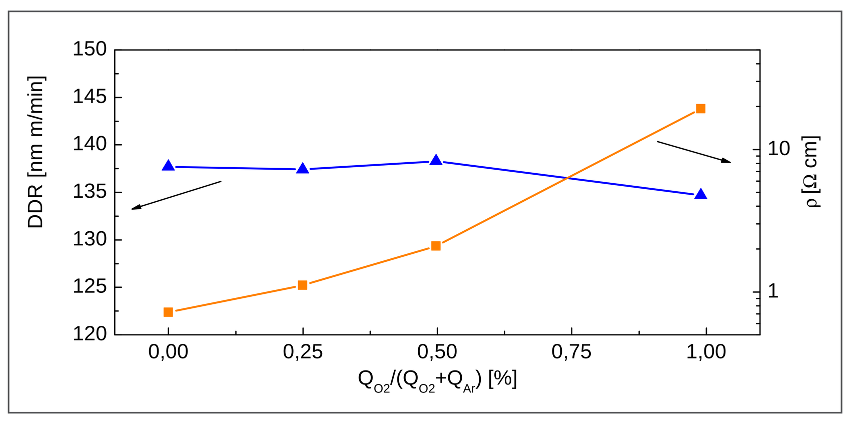


Resistivity and film thickness distribution for static deposition of DC sputtered TiO_2:Nb from plasma sprayed tubular TiO_2:Nb target on glass (after annealing at 450°C in vacuum). Inhomogeneity of the film properties due to different energetic flux in the plasma and substrate sheath.

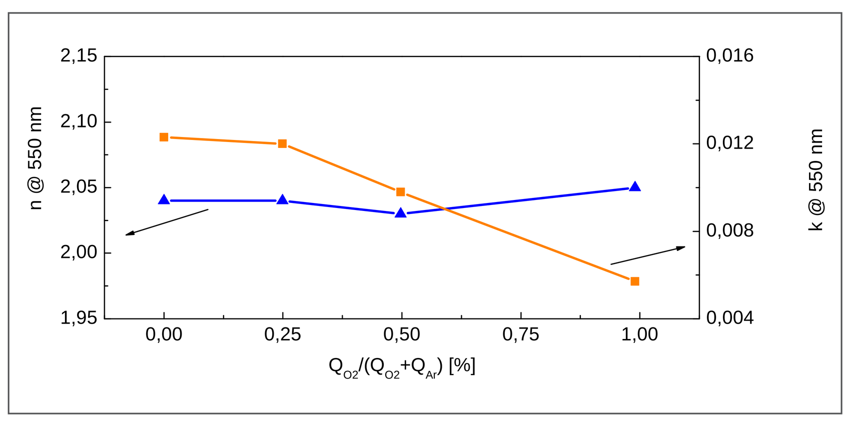
- Deposition at room temperature, thermal annealing at 450°C in vacuum.
- DC sputtering with maximum power impact of 16 kW/m, DDR_{max} ~ 55 nm·m/min, ρ_{min} ~ 1.34 × 10^{-3} Ωcm.
- Adjustment of the stoichiometry by variation of the volume flow rate $Q_{O2}/(Q_{O2} + Q_{Ar})$.
- Dynamic deposition rates comparable with undoped TiO_2.
- Film thickness (100 … 500) nm on Borofloat® glass.
- The as-deposited films are X-ray amorphous.
- After annealing rutile films show high resistivity, while anatase films have lowest resistivity.
- We observed at 150 nm thin films the lowest resistivity of 8.5 × 10^{-4} Ωcm, deposited at DC power impact of 9 kW/m.

ZINC OXIDE BASED TCOs

INTRINSIC ZINC OXIDE – i-ZnO

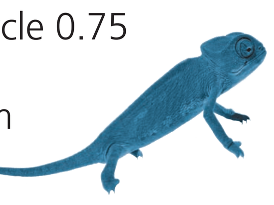


Behavior of dynamic deposition rate and resistivity in dependence on the additional oxygen during the sputtering process.

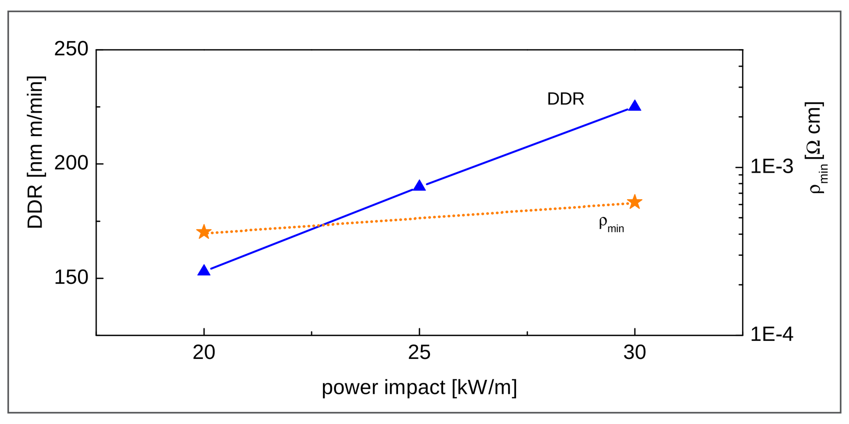


Behavior of refractive index and extinction coefficient for 550 nm wavelength in dependence on the additional oxygen during the sputtering process.

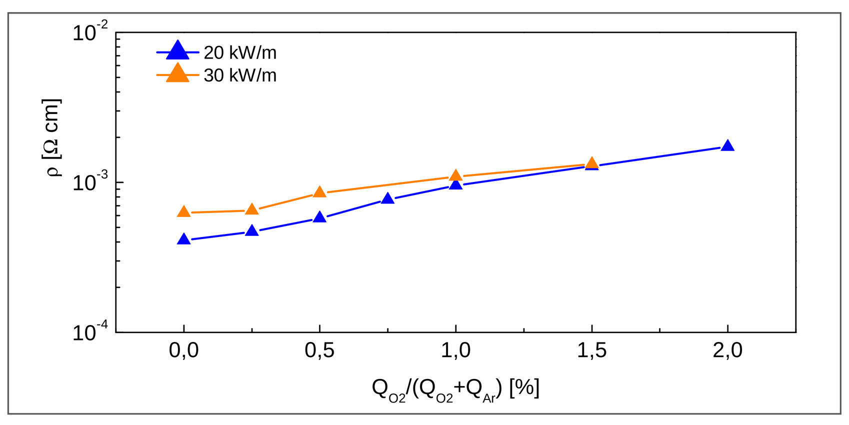
- Deposition at room temperature, no thermal annealing
- Unipolar pulse mode with duty cycle 0.75 (f_p = 50 kHz, t_{on} = 15 μs)
- Maximum power impact 18 kW/m
- Film thickness circa 450 nm
- Deposition on low iron float glass



Al DOPED ZINC OXIDE – ZnO:Al



Behavior of dynamic deposition rate and reachable minimum resistivity in dependence on the power impact on the tubular target.



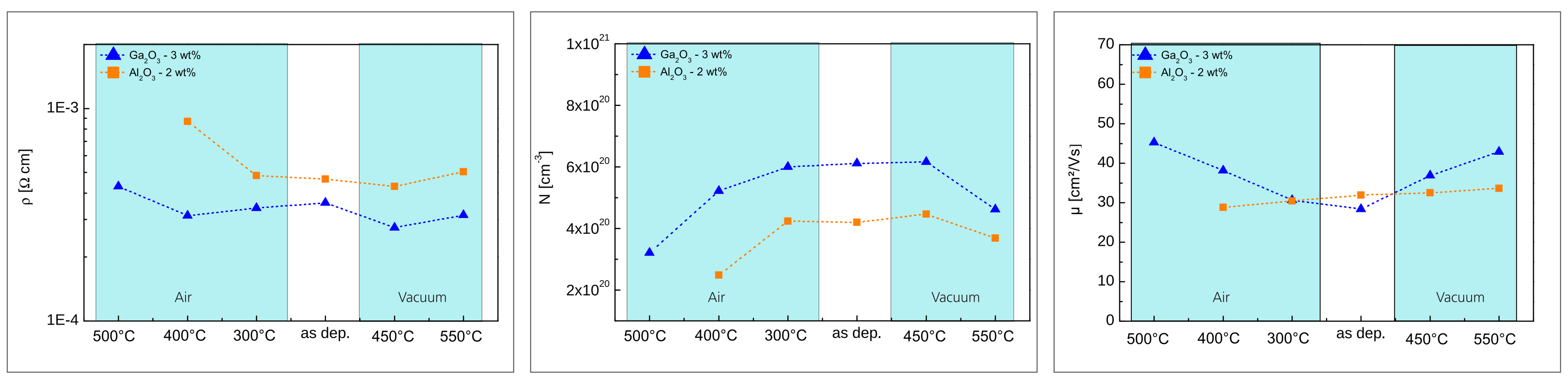
Behavior of resistivity in dependence on the additional oxygen during the sputtering process for different power impacts on the target.

- Deposition at room temperature, thermal annealing at 400°C
- DC sputtering
- Maximum power impact 30 kW/m
- Film thickness circa 1000 nm, R_{sq} ~ (4 … 7) Ω
- Deposition on low iron float glass

PROPERTIES OF TCOs AT FRAUNHOFER FEP

	ITO	i-ZnO	ZnO:Al		ZnO:Ga	TiO_2:Nb		
Dopand	90/10	-	2%	2%	3%	2%	4%	6%
Target	planar	tubular	planar	tubular	planar	planar	tubular	planar
DDR [nm·m/min]	40	135	100	> 225	100	(8…12)	46.5	(8…12)
As deposited								
$T_{deposition}$ [°C]	350	RT	350	RT	350	RT	RT	RT
ρ [Ωcm]	1.6 × 10^{-4}	1	3.9 × 10^{-4}	(6…9) × 10^{-4}	3.6 × 10^{-4}	No	No	No
k [x 10^{-3}]	5	10	1.4	5.5	2.1			
After annealing in vacuum								
$T_{annealing}$ [°C]	500		450	400	450	550	550	550
ρ [Ωcm]	1.3 × 10^{-4}		3.9 × 10^{-4}	3.6 × 10^{-4}	2.7 × 10^{-4}	2 × 10^{-3}	1.3 × 10^{-3}	9.8 × 10^{-4}
k [x 10^{-3}]	6.5		2	2.4	2.1	9.4	15.4	20

ELECTRICAL PROPERTIES OF DOPED ZINC OXIDE



Overview of resistivity, carrier concentration and mobility of ZnO doped with Ga_2O_3 and Al_2O_3 after several thermal annealing methods.

SUMMARY

For the cost effective deposition of zinc oxide based TCOs sputtering from oxidic tubular targets in rotatable magnetron arrangements are established. Through a suitable choice of the process parameters i-ZnO can also be deposited in DC mode with high deposition rates under stable process conditions. For ZnO:Al we reached a dynamic deposition rate of higher than 225 nm·m/min at power impacts of more than 30 kW/m. Nb-doped titania films have been deposited in DC mode with high deposition rates using an oxidic tubular target under pilot scale conditions.

The most important effect on thin film properties can be attributed to additional oxygen flow during sputter deposition. There is a very narrow partial oxygen pressure window for getting anatase-dominated films with lowest resistivity lower than 10^{-3} Ωcm. The cost effective deposition of TiO_2:Nb on large areas opens a wide range of new possible applications. A 150 nm thick film shows a resistivity of 8.5 × 10^{-4} Ωcm after annealing. The resistivity of the films strongly depends on the doping concentration of Nb.

CONTACT

FRAUNHOFER-INSTITUT
FÜR ELEKTRONENSTRAHL- UND PLASMATECHNIK FEP
DR. MANUELA JUNGHÄHNEL
WINTERBERGSTRASSE 28
01277 DRESDEN, GERMANY

PHONE +49 351 2586-128
FAX +49 351 2586-55-128

MANUELA.JUNGHAEHNEL@FEP.FRAUNHOFER.DE
WWW.FEP.FRAUNHOFER.DE

ACKNOWLEDGEMENT

We would like to thank especially Dr. Mykola Vinnichenko (Helmholtz Zentrum Dresden-Rossendorf) for the intensive discussions and the collaboration regarding the investigations of the structure of the TiO_2:Nb films. Furthermore, we would like to thank Dr. Grit Hüttl (GfE Fremat GmbH) and Dr. Wilmert De Bosscher (Soleras Advanced Coatings) for providing the targets and the helpful discussions.

PARTS OF THIS WORK WAS FUNDED BY THE FREE STATE OF SAXONY: 14274/2473