# Deposition of stoichiometric $VO_2$ on ultra-thin glass in roll-to-roll process

Fraunhofer FEP

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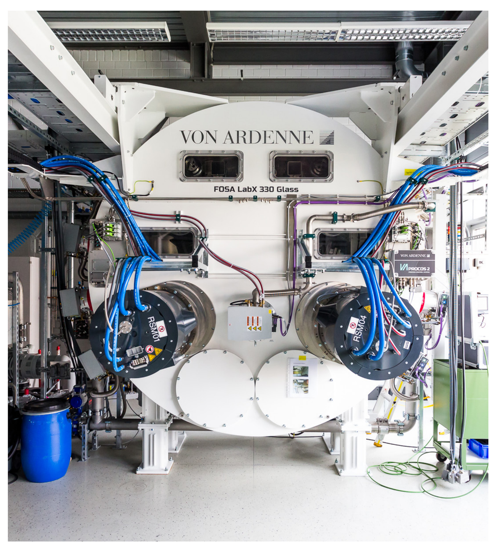
## Introduction

Vanadium dioxide ($VO_2$) exhibits different polymorphic structures, among them monoclinic $VO_2$(B) and $VO_2$(M1), which are widely studied in science and technology development for their application potential: $VO_2$(B) can be used as e.g. electrode materials for batteries and $VO_2$(M1) as thermochromic layer for Insulating Glass Units (IGU). Through reversible, temperature-dependent semiconductor-to-metal transition (at the switching temperature $T_{tr}$) of low-temperature monoclinic $VO_2$(M1) to high-temperature tetragonal $VO_2$ (R); electrical and optical properties are changed. Especially the transmission in infrared (IR) wavelength range can be significantly reduced. Such thermochromic behaviour can lead to a reduction of the energy consumption of buildings, particularly for cooling.

## Methods

- Challenging technology transfer from a laboratory-scale (University of West Bohemia) to roll-2-roll process on FOSA labX 330 glass coater (Fraunhofer FEP, Figure 1)
- Deposition of $ZrO_2$ bottom and top layer from ceramic $ZrO_x$ target with unipolar pulsed direct current magnetron sputtering
- Deposition of thermochromic monoclinic $VO_2$ (M1) with high-power impulse magnetron sputtering (HiPIMS) from V target doped with W (1.5 at%), using a pulsed $O_2$ flow control (Vlcek group, University of West Bohemia - UWB).
- Deposition of non-thermochromic monoclinic $VO_2$ (B) with unipolar pulsed direct current magnetron sputtering from V target doped with W (1.5 at%)
- Substrate: ultra-thin glass (100 µm thickness, 300 mm width)



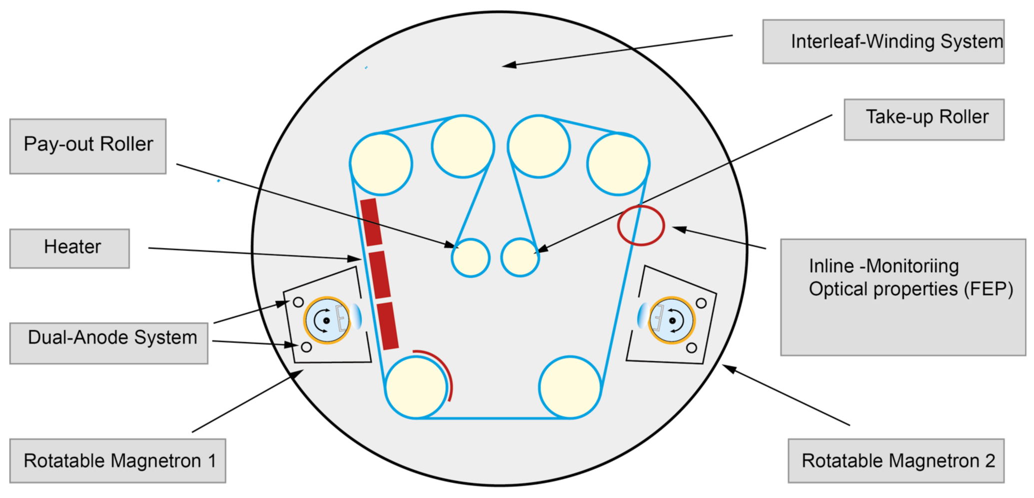
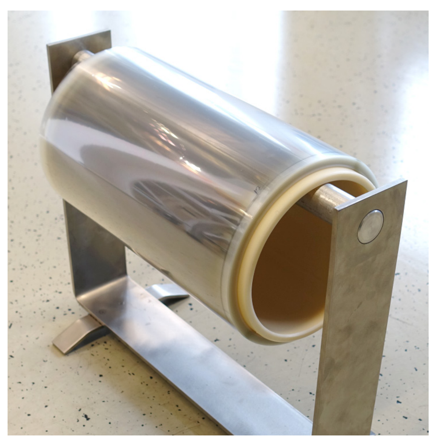


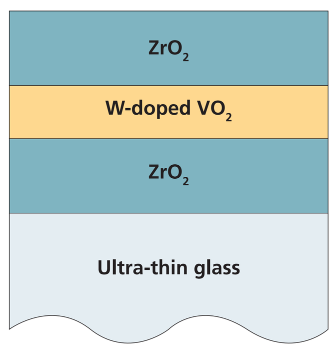

*Figure 1: Scheme of the FOSA labX 330 glass coater.*



| UWB (batch process) | Fraunhofer FEP (roll-to-roll) |
|---|---|
| Planar target, 50 mm diameter | Tube target, 665 mm length |
| Substrate fixed on rotating holder | Moving substrate (dynamic process) |
| V target, W target, W amount, variable, co-sputtering | V target doped with W (fixed amount of W) |
| 2.5 cm × 5 cm sheet substrates | 300 mm × 20 m roll substrates |
| Up to 4 different compounds in one run | 2 different compounds in one run possible |

Three-layer system:

- $ZrO_2$ bottom and top layer → optimal optical properties
- $ZrO_2$ bottom layer → seed layer
- $ZrO_2$ top layer → mechanical and chemical protection
- $VO_2$ → active layer
- Doping of W → decrease in switching temperature




## Results: Crystalline Structure

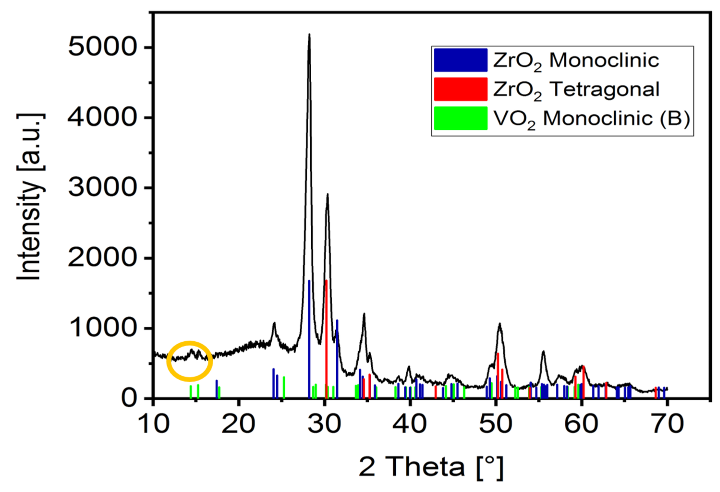
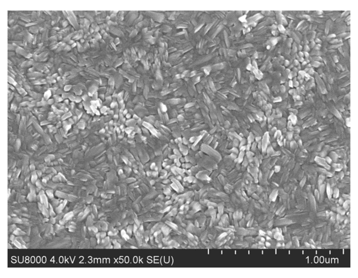



*Figure 2: GIXRD spectrum of $ZrO_2/V_{1-x}W_xO_2$ with the monoclinic $VO_2$(B) on ultra-thin glass.*

- $ZrO_2$ structure: monoclinic and tetragonal → suitable as a seed layer for $VO_2$
- $VO_2$ structure: single phase monoclinic (B), Figure 2, and dominant tetragonal (R) with thermochromic properties, Figure 3



*Crystalline $VO_2$*

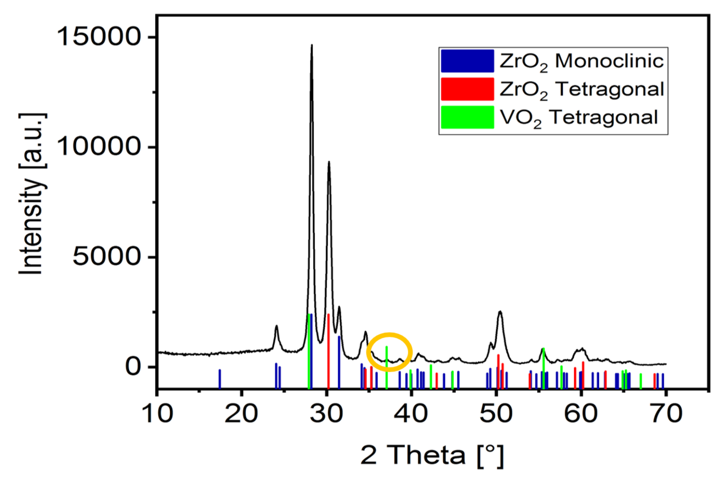



*Figure 3: GIXRD spectrum of $ZrO_2/V_{1-x}W_xO_2/ZrO_2$ with the dominant tetragonal $VO_2$ (R) on ultra-thin glass.*

## Process Parameters

| | $ZrO_2$ | W-doped $VO_2$ |
|---|---|---|
| Power | 6 kW | 6 kW |
| Pulse on | 16 µs | 70 µs |
| Frequency | 50 kHz | 750 Hz |
| Pressure | 0.38 Pa | 0.20 – 0.22 Pa |
| Band speed | 0.15 m/min | 0.10 m/min |
| $O_2$ flow | 8 sccm | 0 – 50 sccm |

## Optical Properties

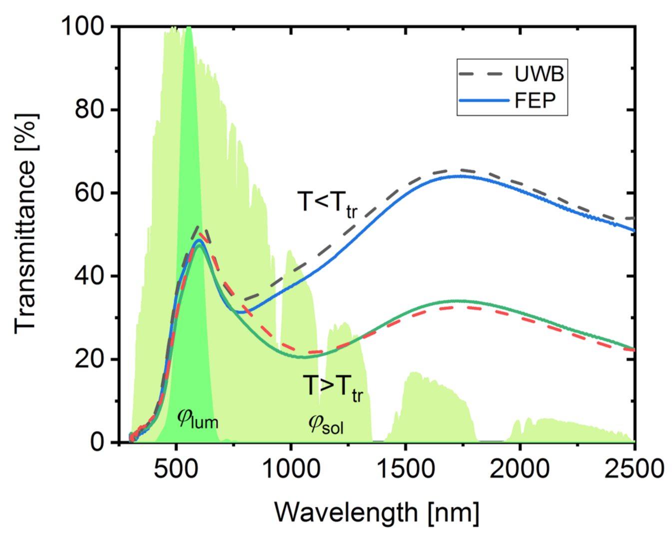



*Figure 4: Spectral transmittance for thermochromic $ZrO_2/V_{1-x}W_xO_2/ZrO_2$ coatings on 0.1 mm ultra-thin glass at a low (blue lines) and high (red lines) temperature. Dashed lines for a sample prepared at the UWB*, solid line at the FEP.*

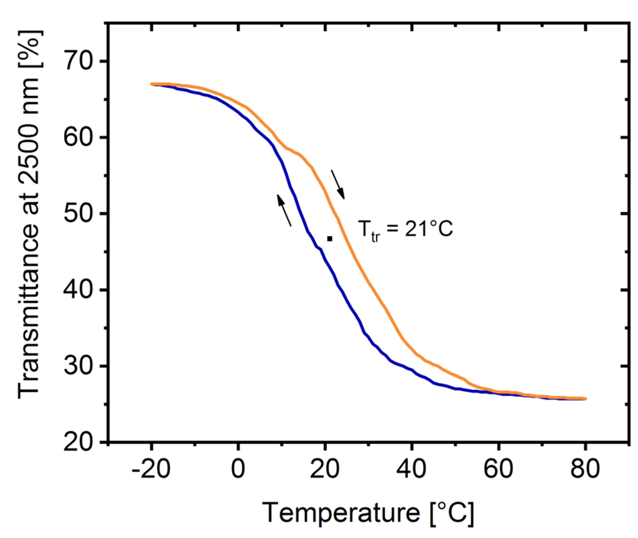



*Figure 5: Temperature dependence of the transmittance at 2500 nm for the $ZrO_2/V_{1-x}W_xO_2/ZrO_2$ film coating on ultra-thin glass deposited in R2R. The switching temperature is presented.*

*Barta et. al., Coatings 2020, 10, 1258; doi:10.3390/coatings10121258

- Significant drop in the transmittance crossing the switching temperature (Figure 4)
- Evaluation of the switching temperature to 21 °C (Figure 5)
- Comparable results to the coatings in laboratory-scale at UWB

Parameter set for deposition of $VO_2$ at UWB

- Power density: 13.5 W/cm$^2$
- Voltage pulse duration: 80 µs
- Frequency: 625 Hz
- Deposition temperature: 330 °C

| | $T_{lum}^{**}$ [%] | $\Delta T_{sol}$ [%] | $T_{tr}$ [°C] |
|---|---|---|---|
| UWB | 44.0 | 9.6 | 22 |
| FEP-R2R | 41.9 | 8.7 | 21 |

**The presented measured values of $T_{lum}$ are underestimated by up to 4 %.

## $O_2$ Flow Control

$p_{ox}(t) \geq (p_{ox})_{crit}$ → $\Phi_{ox}(t) = 0$,

$p_{ox}(t) < (p_{ox})_{crit}$ → $\Phi_{ox}(t) = \Phi_{ox} \neq 0$

- Pulsed $O_2$ inlet system
- $O_2$ inlet orientation towards the substrate
- $O_2$ inlet location in the dense plasma in front of the sputtered target

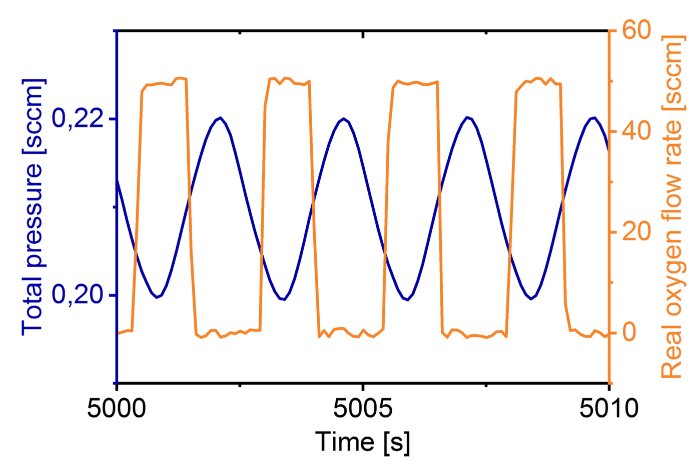



*Figure 6: Time evolution of the total pressure and the corresponding oxygen flow pulses during a HiPIMS deposition of W-doped $VO_2$ layers.*

## Conclusion

The worldwide first thermochromic $VO_2$-based coatings on ultra-thin glass were deposited in roll-to-roll process with HiPIMS. The comparable optical properties between the results from Fraunhofer FEP and UWB show the successful transfer of sputter deposition technique from the laboratory-scale to roll-to-roll coater and a very good upscaling possibility for preparation of termochromic $VO_2$-based coatings in large area production.

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