Thermal stability investigations of magnetron-sputtered Ir-HfO_2 thin films

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Introduction

Thermophotovoltaics (TPV)

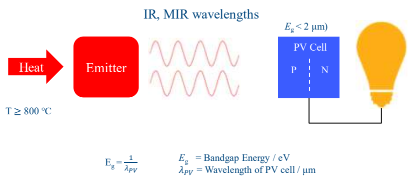

Figure 1: The basic working principle of a TPV system. Heat from a source is absorbed by a emitter to emit tailored radiation in the infrared regime to a photovoltaic cell that generates electricity

- A thermal emitter allows to transfer heat into electricity
- Operating at high *T* is the primary objective

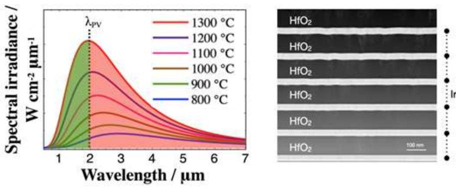

Figure 2: The total efficiency of the TPV system can be improved by using selective emitters which absorbs long-wavelength radiation highlighted in red. On the right is the STEM cross section of an as-prepared multilayered metamaterial selective emitter

- Challenges of TPV application: Thermal stability is required at high temperatures

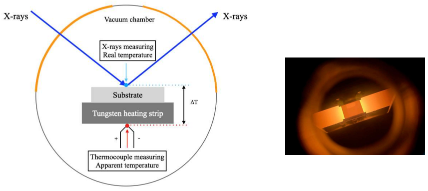

Figure 3: Schematic of in-situ x-ray diffraction chamber and image of a sample annealed at 1000 °C, 2 × 10^{-6} mbar

- Grain size evolution, phase transformation, thermal expansion, chemical reactions and kinetic processes of thin films are investigated

Experimental Results & Discussion

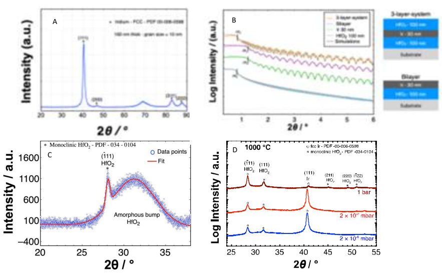

Figure 4: (A&C) XRD diffractograms of as-prepared Ir and HfO_2 layers. (B) XRR plots of various films. (D) XRD diffractograms of annealed 3-layer-systems at 1000 °C and various pressures

- Phase formation of as prepared Ir and HfO_2 films
- Layer thickness control by x-ray reflectometry (XRR)
- Ir is stable at different vacuum pressures
- Oxidation of Ir occurs at atmospheric pressure

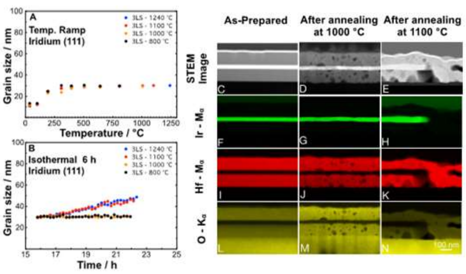

Figure 5 : Grain size Ir as a function of temperature and time during in-situ annealing. Cross sectional STEM-EDX images of 3-layer system before and after annealing

- Grain size of Ir calculated from (111) FCC peak using Scherrer's formula
- Grain size reaches value of 30 nm up to 1000 °C
- At 1100 °C, grains grow up to 50 nm exceeding the layer thickness: loss of interface stability

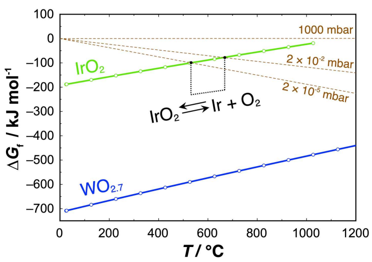

Figure 6: Ellingham diagrams showing Gibbs energy of oxide formation ΔG_f for IrO_2 and $WO_{2.7}$. Equilibrium conditions of chemical reactions estabilished at different pressures.

- Ellingham diagram shows the equilibrium points of the metal oxide, metal, and oxygen
- At low vacuum pressures the equilibrium points are at lower ΔG.

Future Work

- Stabilizing the grain structure in the metallic and dielectric layers of a selective thermal emitter
- Alloying of Ir layers with a second metal
- Doping of the HfO_2 layers

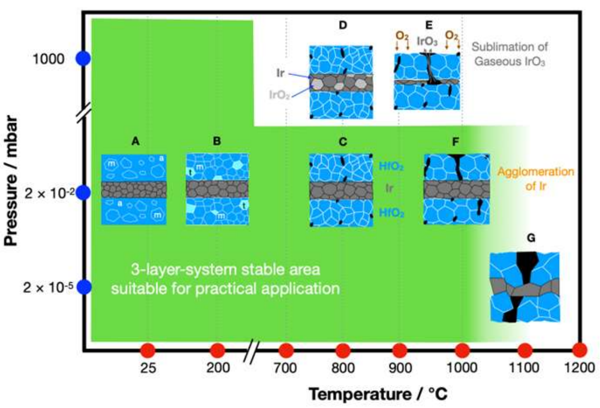

Figure 7: Schematic summary showing the major mechanisms and changes in an Ir 3-layer-system at different temperature and pressure ranges. The green area show the thermally stable area of the 3-layer-system suitable for practical application.

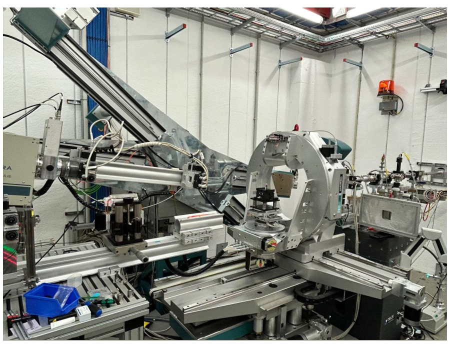

Figure 8: Structural and microstructural characterization of Ir-based thin films using energy-dispersive x-ray diffraction at PETRA III beamline P61A: Hereon's beamline at DESY/Hamburg

Publications

1. Vaidhyanathan Krishnamurthy, G., Chirumamilla, M., Krekeler, T., Ritter, M., Raudsepp, R., Schieda, M., Klassen, T., Pedersen, K., Petrov, A. Y., Eich, M., Störmer, M., Iridium-Based Selective Emitters for Thermophotovoltaic Applications. Adv. Mater. 2023, **35**, 2305922.
2. Krishnamurthy, G.V., Chirumamilla, M., Rout, S.S. et al. Structural degradation of tungsten sandwiched in hafnia layers determined by in-situ XRD up to 1520 °C. Sci Rep **11**, 3330 (2021)
3. Chirumamilla, M., Krishnamurthy, G.V., Knopp, K. et al. Metamaterial emitter for thermophotovoltaics stable up to 1400 °C. Sci Rep **9**, 7241 (2019).
4. Chirumamilla, M., Krishnamurthy, G.V., Rout, S.S. et al. Thermal stability of tungsten based metamaterial emitter under medium vacuum and inert gas conditions. Sci Rep **10**, 3605 (2020)

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