



## Article

# Hafnium Oxide Nanostructured Thin Films: Electrophoretic Deposition Process and DUV Photolithography Patterning.

Vanessa Proust<sup>1,2,3\*</sup>, Quentin Kirscher<sup>4,5</sup>, Thi Kim Ngan Nguyen<sup>3,6</sup>, Lisa Obringer<sup>4,5</sup>, Kento Ishii<sup>2</sup>, Ludivine Rault<sup>7</sup>, Valérie Demange<sup>7</sup>, David Berthebaud<sup>3</sup>, Naoki Ohashi<sup>2,3</sup>, Tetsuo Uchikoshi<sup>2,3</sup>, Dominique Berling<sup>4,5</sup>, Olivier Soppera<sup>4,5,\*</sup>, Fabien Grasset<sup>3,7\*</sup>

<sup>1</sup> CEA, DES, ISEC, DMRC, Université de Montpellier, F-30200 Marcoule, France

<sup>2</sup> Research Center for Functional Materials, National Institute for Materials Science (NIMS), Tsukuba 305-0044, Japan; ishii.kento@nims.go.jp (K.I.); ohashi.naoki@nims.go.jp (N.O.); uchikoshi.tetsuo@nims.go.jp (T.U.)

<sup>3</sup> CNRS-Saint Gobain-NIMS, IRL 3629, Laboratory for Innovative Key Materials and Structures (LINK), NIMS, Tsukuba 305-0044, Japan; nguyen.thikimngan@nims.go.jp (T.K.N.N.); david.berthebaud@cnrs.fr (D.B.)

<sup>4</sup> Université de Haute-Alsace, CNRS, IS2M UMR 7361, F-68100 Mulhouse, France; quentin.kirscher@uha.fr (Q.K.); lisa.obringer@uha.fr (L.O.); dominique.berling@uha.fr (D.B.)

<sup>5</sup> Université de Strasbourg, F-67081 Strasbourg, France

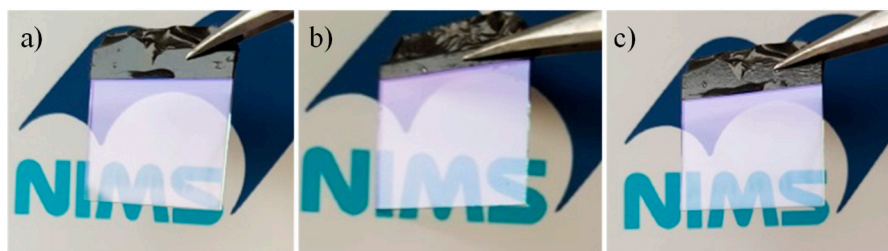
<sup>6</sup> International Center for Young Scientists, ICYS-Sengen, Global Networking Division, NIMS, Tsukuba 305-0047, Japan

<sup>7</sup> Univ Rennes, CNRS, ISCR UMR 6226, ScanMAT UAR 2025, F-35000 Rennes, France; ludivine.rault@univ-rennes1.fr (L.R.); valerie.demange@univ-rennes1.fr (V.D.)

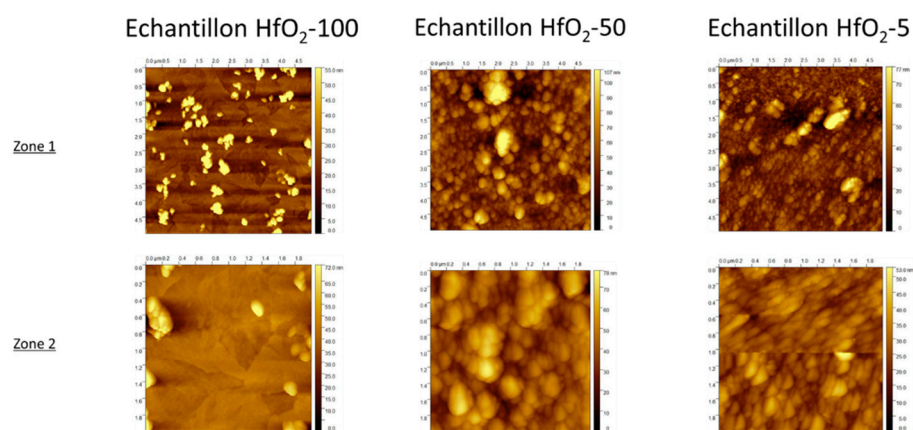
\* Correspondence: vanessa.proust@cea.fr (V.P.); olivier.soppera@uha.fr (O.S.); fabien.grasset@univ-rennes1.fr (F.G.)

	<b>Zone 1</b> <b>5 x 5 <math>\mu\text{m}^2</math></b> <b>(<math>\pm 0.5</math> nm)</b>	<b>Zone 2</b> <b>2 x 2 <math>\mu\text{m}^2</math></b> <b>(<math>\pm 0.5</math> nm)</b>	<b>Average roughness</b> <b>(<math>\pm 0.5</math> nm)</b>
<b>HfO<sub>2</sub>-100</b>	11.1 nm	6.6 nm	9.0 nm
<b>HfO<sub>2</sub>-50</b>	16.8 nm	10.3 nm	14.0 nm
<b>HfO<sub>2</sub>-5</b>	10.6 nm	5.3 nm	9.0 nm

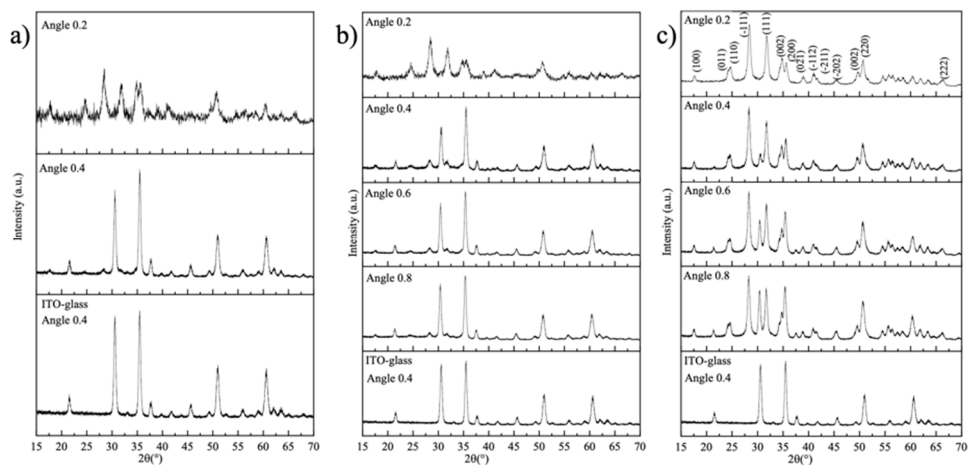
**Table S1:** Values of roughness for the thin films prepared by EPD.



**Figure S1.** (a–c) Digital photographs of coatings prepared by EPD (5 V, 0.7 mA/cm<sup>2</sup>, 90 s) on ITO glass substrates using suspensions of (a) 5 nm, (b) 50 nm, and (c) 100 nm-sized HfO<sub>2</sub>-NPs dispersed in an equimolar mixture of ethanol and acetylacetone at pH 2.



**Figure S2.** A2M images of the films prepared by EPD.



**Figure S3.** Grazing incidence X-ray diffraction patterns of electrophoretic coatings of (a) 5 nm, (b) 50 nm, and (c) 100 nm sized particles at incidence angles of 0.2, 0.4, 0.6 and 0.8° from the surface.