Supporting information for:

An Electrochemical Technique for Controlled Dissolution of Zirconium Based Components of Light Water Reactors

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Figure S1. Instrumentation of the tube furnace assembly for loading the liner with specified amounts of H/D.



Figure S2. Dissolution kinetics of H-loaded liner (1:0.5 mol Zr:H) measured by liner thickness changes with time at a constant applied potential (225 mV vs Ag.AgCl) in 1 M aqueous KCl: (Top panels) Cross-sectional images of liner upon progressive dissolution, (bottom panel) Dissolution kinetics of the liner measured by liner thickness changes with time.



Figure S3. Comparison of experimental XRD patterns with those obtained from literature¹: (top, brown trace) Product obtained from electrodissolution of pristine Zr-4 liner, (second from the top, black trace) Product obtained from electrodissolution of H-loaded Zr-4 liner (1: 1 mol Zr:H), (third from the top, yellow trace) Product obtained from electrodissolution of D-loaded Zr-4 liner (1: 1 mol Zr:D), (fourth from the top, green trace) tetragonal ZrO₂ from literature (#00-017-0923),²⁻³ (third from the bottom, blue trace) rhombohedral ZrO₂ from literature (#00-037-0031),⁴ (second from the bottom, red trace) orthorhombic ZrO₂ from literature (#00-037-0031),⁶



Figure S4. Representative secondary electron SEM images of Zr-4 liners prior to electrodissolution at different magnifications: (top row) unloaded lines, (middle row) H-loaded liners 1:1 mol Zr:H, (bottom row) D-loaded liners 1:1 mol Zr:D

References:

1. Kabekkodu, S., *Powder Diffraction File Inorganic and Organic Data Book*. International Center for Diffraction Data: Newtown Square, PA, USA, 2015; Vol. 60.

2. Howard, C. J.; Hill, R. J.; Reichert, B. E., Structures of the Zro2 Polymorphs at Room-Temperature by High-Resolution Neutron Powder Diffraction. *Acta Crystallogr B* **1988**, *44*, 116-120.

3. Teufer, G., Crystal Structure of Tetragonal Zro2. Acta Crystallogr 1962, 15 (Nov), 1187-+.

4. Hasegawa, H., Rhombohedral Phase Produced in Abraded Surfaces of Partially Stabilized Zirconia (Psz). *J Mater Sci Lett* **1983**, *2* (3), 91-93.

5. Lityagina, L. M.; Kabalkina, S. S.; Pashkina, T. A.; Khozyainov, A. I., Polymorphism of Zro2 at High-Pressure. *Fiz Tverd Tela*+ **1978**, *20* (11), 3475-3477.

6. Garvie, R. C., Phase Analysis in Zirconia Systems. J Am Ceram Soc 1972, 55 (6), 303-&.