

**Figure 1**. **NiS<sub>x</sub> ALD characteristics:** growth rate and resistivity versus a) NiCl<sub>2</sub>(TMPDA), and b) H<sub>2</sub>S pulse length, and c) deposition temperature. The somewhat atypical saturation characteristics are due to changes in preferred crystalline orientation. Thicknesses were measured by energy-dispersive X-ray spectrometry. The resistivity error bars represent variation in square resistance over the 5×5 cm<sup>2</sup> substrate. Unless otherwise noted, 750 cycles with 2 s NiCl<sub>2</sub>(TMPDA) and 4 s H<sub>2</sub>S pulses separated by 2 s N<sub>2</sub> purges at 165 °C were applied.



**Figure 2. OER catalyst characterization**: a) Scanning electron microscopy images and b) X-ray photoelectron spectra (S 2p region) of approximately 5 nm NiS<sub>x</sub> films deposited on fluorine-doped tin dioxide before and after use as an OER catalyst for approximately 2 hours. SEM suggests the film becomes less dense and more porous after OER. Approximate S/Ni ratios as well as expected positions of S<sup>2-</sup> and SO<sub>4</sub><sup>2-</sup> components are indicated in b) to illustrate the loss of sulfur during OER (no major Ni loss was observed).