Supporting Information for Publication

Noninvasively imaging pH at the surface of implanted orthopedic devices with X-ray excited luminescence chemical imaging (XELCI).

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S1. Absorption spectra of the dye and sensor film as a function of pH

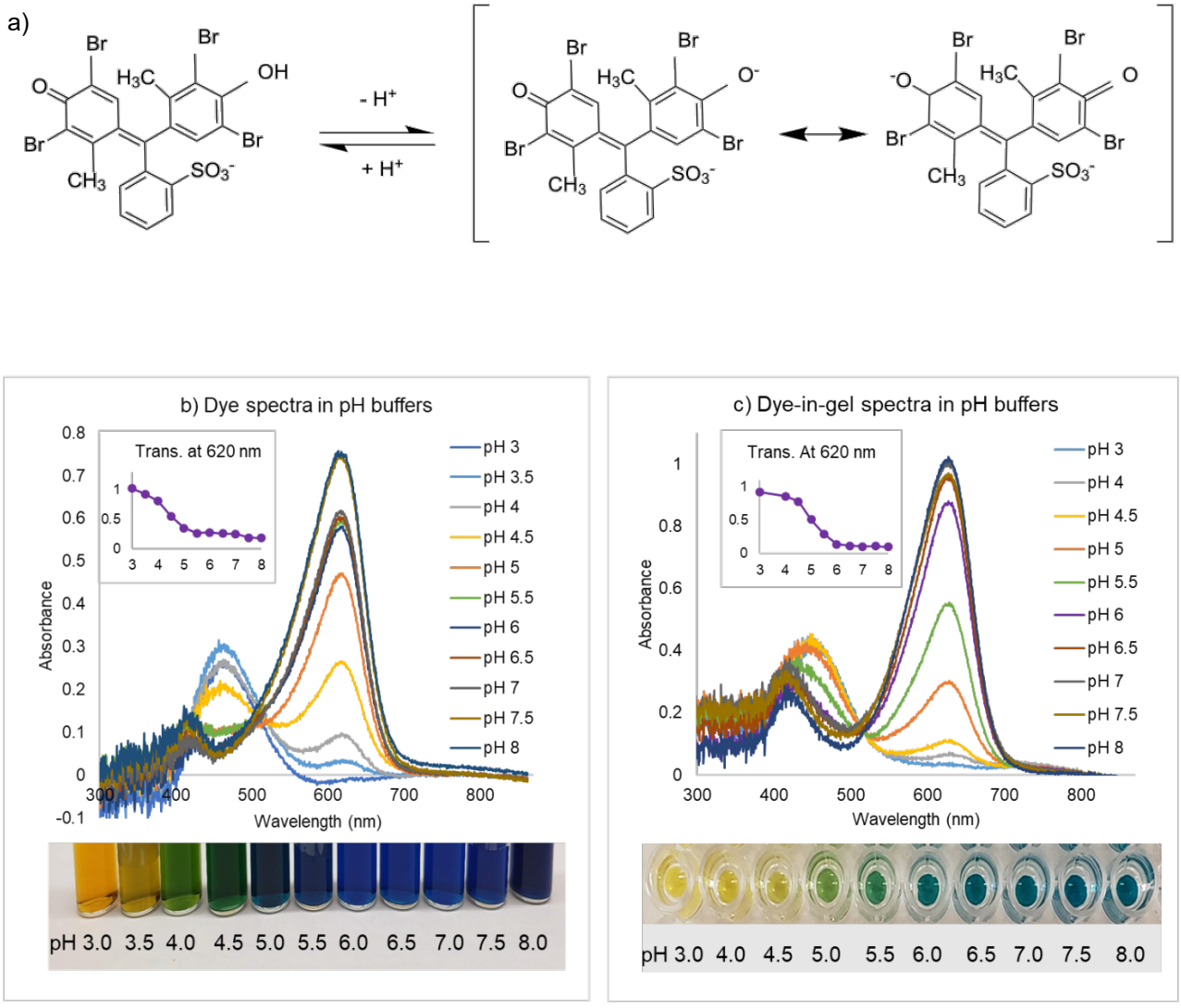

Figure S1. Absorption spectra of bromocresol green (BCG) pH dye as a function of pH. (a) Molecular structure of BCG. (b) Absorption spectra of the pH dye in free form (aqueous solution) taken at different pH (3-8) and photographs showing color change at the respective pH. Inset: Transmittance of BCG dye at 620 nm in different pH. (b) Absorption spectra of the pH dye encapsulated in the hydrogel taken at different pH (3-8) and photographs showing color change at the respective pH. Spectra shown is the average from 3 sample films per pH. Inset: Transmittance of BCG dye-in-gel at 620 nm in different pH.

S2. Ratio vs. pH

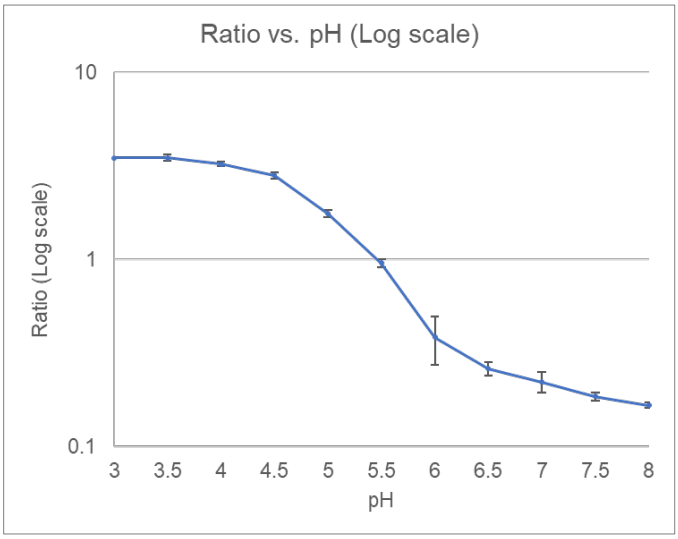

Figure S2. Ratio vs. pH. Attenuation of the scintillator emission signal by the pH dye (bromocresol green) in PEG hydrogel at different pHs. Ratio of 620 and 700 nm intensities plotted on a log scale for each pH with 0.5 pH unit intervals. Error bars represent standard deviation of 3 samples at each pH.

S3. Leaching study

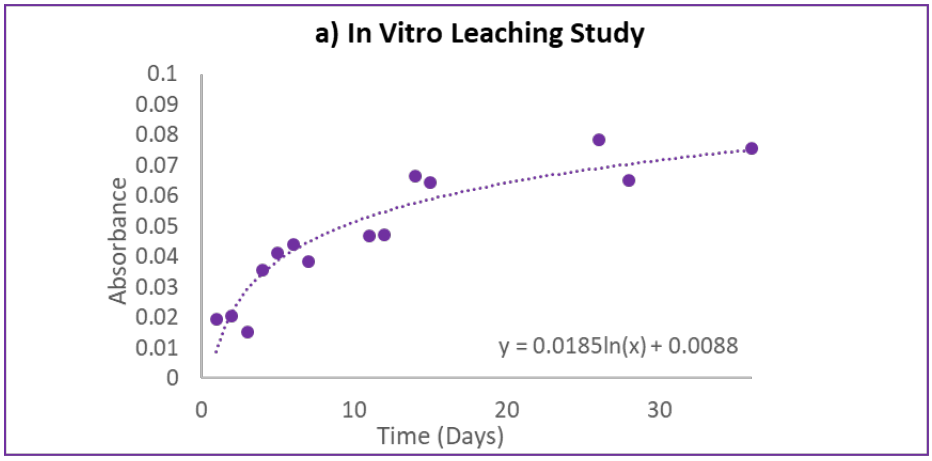

Figure S3. In vitro leaching study: Plot showing accumulative absorbance (at 610 nm) of the bromocresol green dye leaching from a18x18 mm piece of sensor gel kept in 10 ml of phosphate saline buffer (pH 7.4) over a period of 36 days with less than 10% of the total dye leached. Data was fitted to a logarithmic trendline.

S4. Sensor reversibility

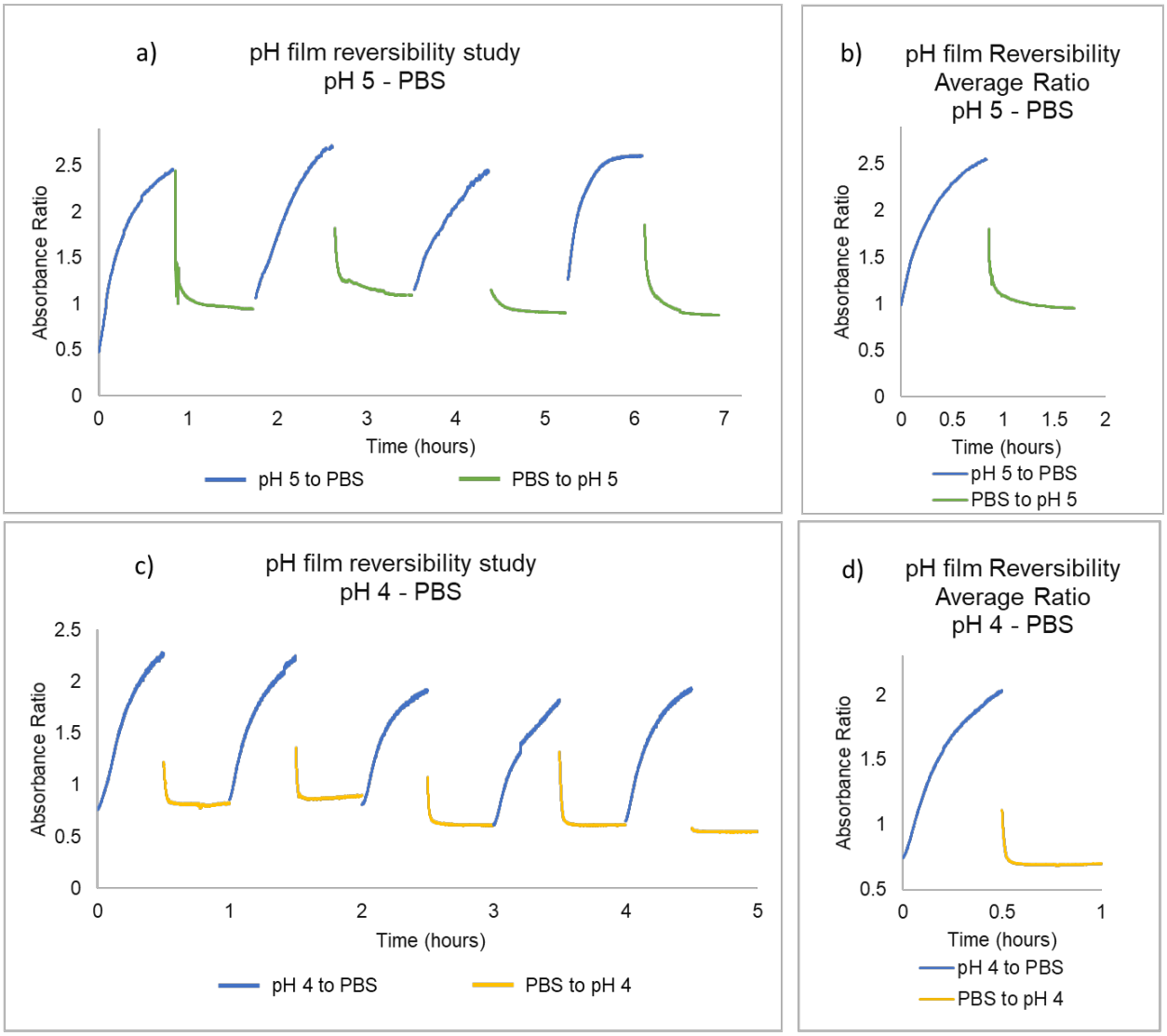

Figure S4. Reversibility study of the pH sensor film (PEG hydrogel with bromocresol green pH dye). (a) Phosphate buffered saline solution (PBS, pH 7.4) was added to the pH film that was initially kept in water and spectra recorded every 1 second for a total of 50 minutes. The film was cycled between PBS and pH 5 buffer and spectra recorded for 50 minutes in each buffer. The film did not reach the initial absorbance as it started in a more acidic medium (leaching of free acid from gel into the unbuffered water) but cycles between green (in pH 5 buffer) and blue (in PBS). (b) Average absorbance ratio of the 4 cycles in (a). (c) pH film was cycled between PBS and pH 4 buffer and spectra recorded every 1 second for 30 minutes in each buffer. (d) Average absorbance ratio of the 5 cycles in (c). Gaps correspond to times when the pH buffers were being changed and spectra acquisition was paused to prevent artefacts during pipetting of buffer solutions.

S5. Sensor images of signal intensities through chicken tissue

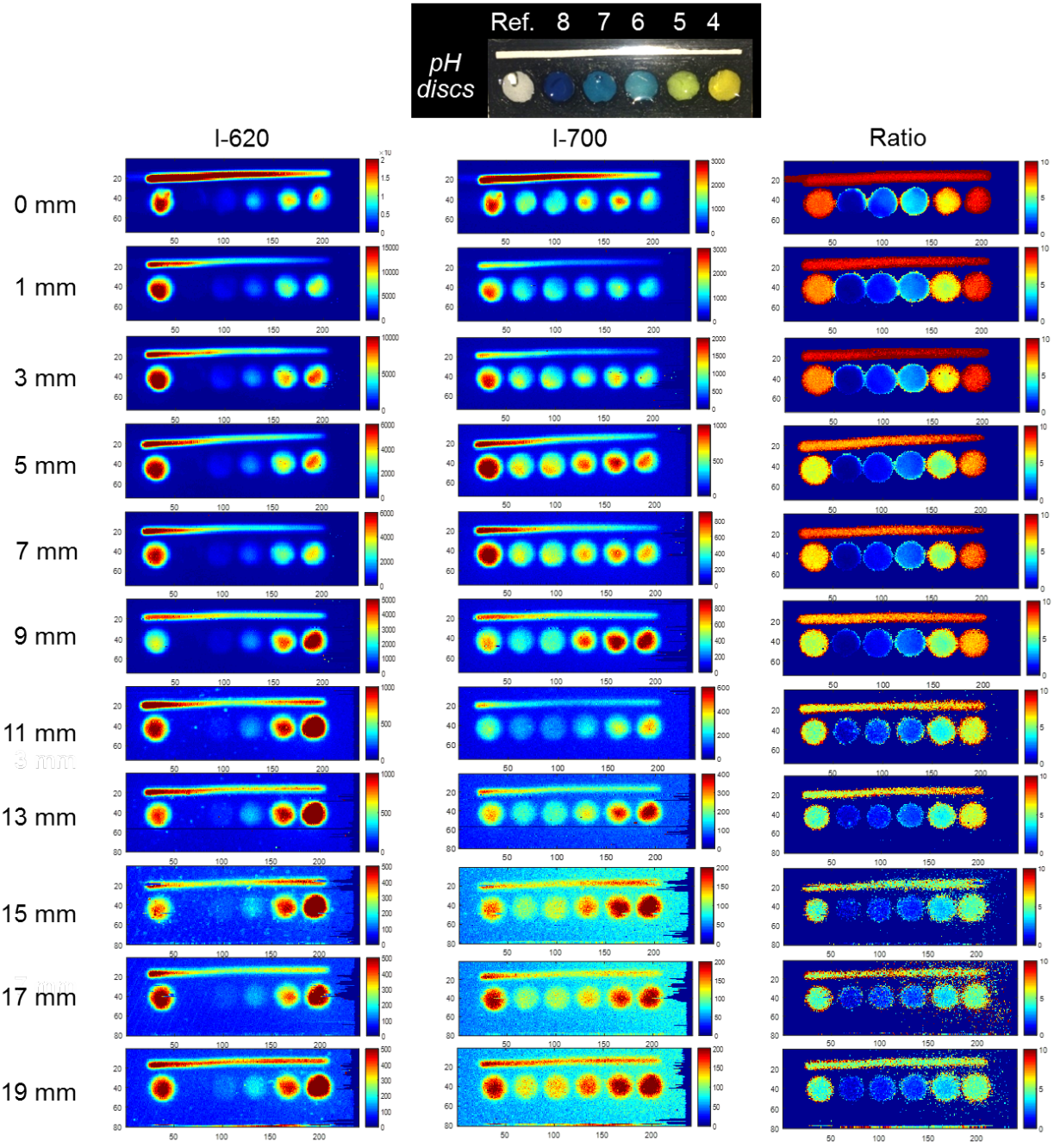

Figure S5. Sensor images of signal intensities through chicken tissue. Photograph showing the pH sensor discs placed in a 3-D printed holder in pH buffers 8, 7, 6, 5 and 4 and a reference disc without any pH coating. The holder was sandwiched between two pieces of chicken tissue and thickness of the top piece was increased from 1 – 19 mm with 2 mm intervals. XELCI images showing the 620 nm, 700 nm and ratio of 620 to 700 nm signal intensities of the pH sensor discs at respective pH obtained without tissue and through 1 – 19 mm of chicken tissue.

S6. Plot of signal to noise ratio versus 620 nm intensity for discs measured through 0-19 mm chicken breast tissue.

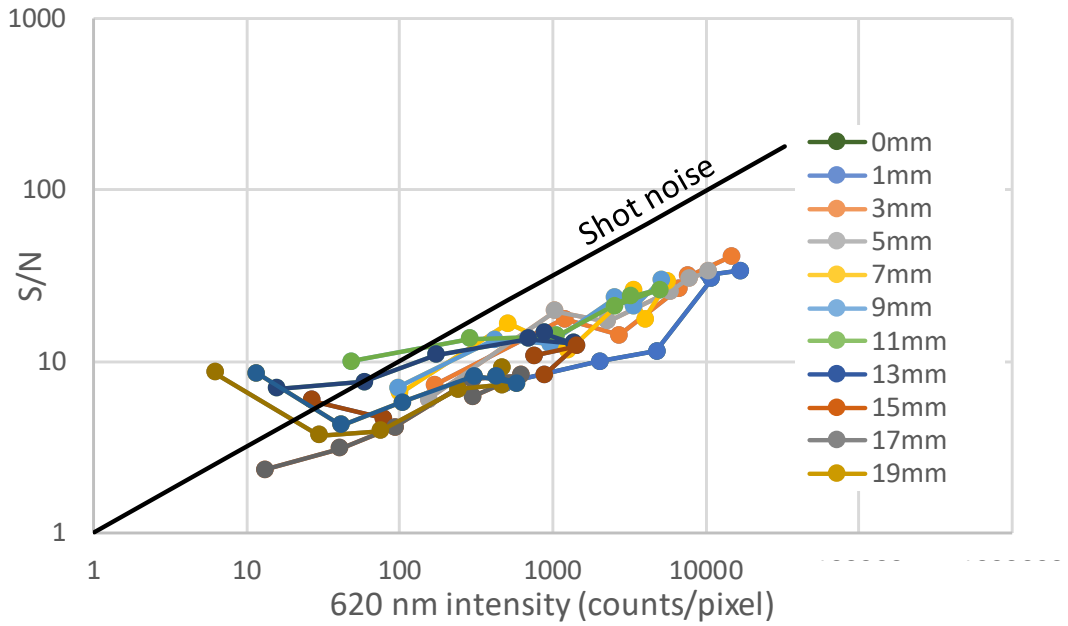

Figure S6. Signal/Noise ratio (Ratio/standard deviation of ratio) as a function of average 620 nm intensity for discs imaged through 0-19 mm of chicken breast tissue.

S7. Plot of signal intensities through human cadaveric tissue

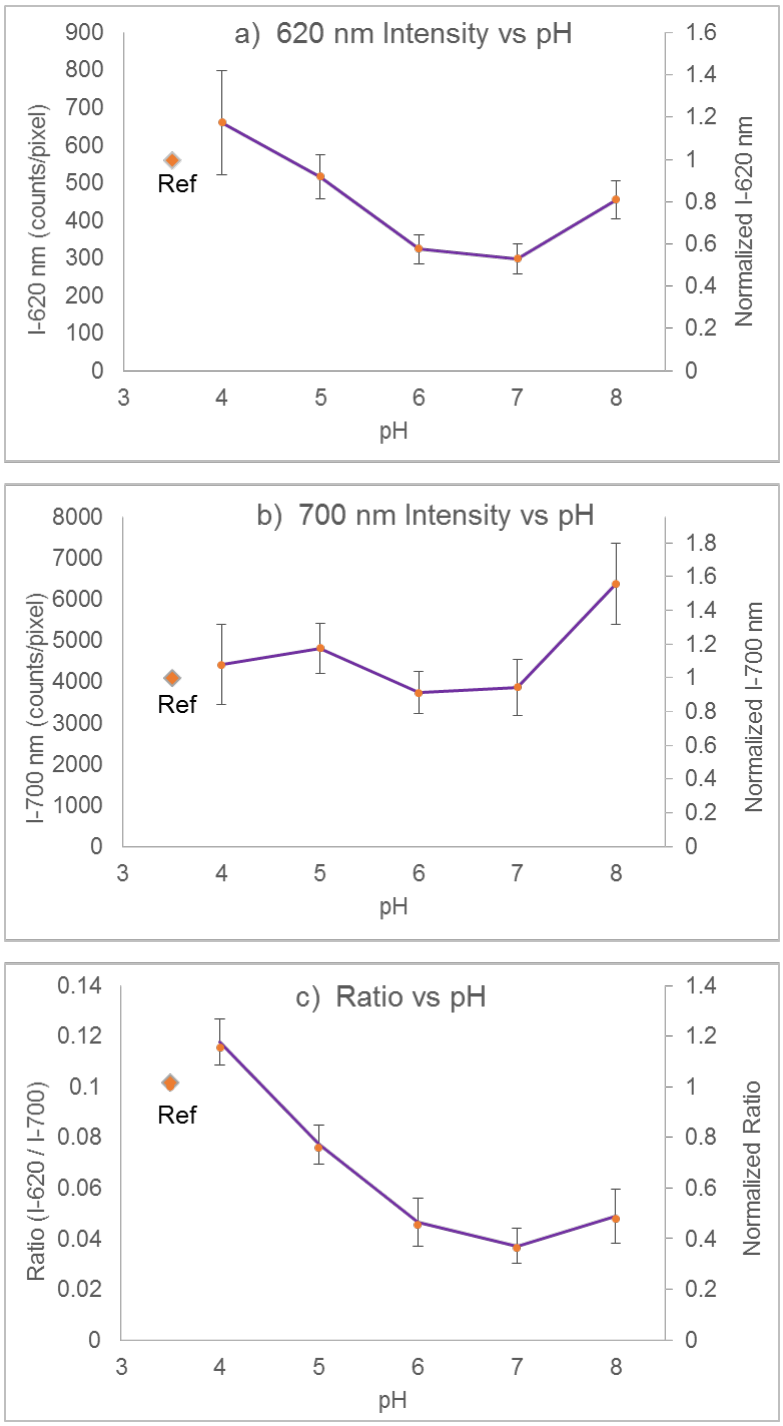

Figure S7. Plot of signal intensities as a function of pH through human cadaveric tissue. (a) 620 nm light intensity at pH 4, 5, 6, 7 and 8 after passing through 1 cm of human cadaveric tissue. (b) 700 nm light intensity at pH 4, 5, 6, 7 and 8 after passing through 1 cm of human cadaveric tissue. (c) Ratio of 620 and 700 nm intensities at pH 4, 5, 6, 7 and 8 after passing through 1 cm of human cadaveric tissue. Note: Right axis scale in each plot is normalized with respective to reference disc. Error bars represent the pixel-to-pixel standard deviation within a disc.