# Transformation of the Uranyl Peroxide Studtite, $[(UO_2)(O_2)(H_2O)_2](H_2O)_2$, to Soluble Nanoscale Cage Clusters

*Haylie L. Lobeck [1], Jordan K. Isner [2], and Peter C. Burns* [1, 3]*

[1] Department of Civil and Environmental Engineering and Earth Sciences, University of Notre Dame, Notre Dame, IN 46556

[2] Department of Chemical and Biomolecular Engineering, University of Notre Dame, Notre Dame, IN 46556

[3] Department of Chemistry and Biochemistry, University of Notre Dame, Notre Dame, IN 46556

*To whom correspondence should be addressed. Email: pburns@nd.edu

**Supporting Information**

**Table S1.** Molar ratios of initial reactants and concentration of dissolved uranium in solution normalized to 1 mole initial studtite, average pH of reaction solution, and presence of nanoclusters.

| Reaction | Initial Studtite (mol) | Initial $H_2O_2$ (mol) | OOH- Fraction (mol) | TEAOH (mol) | Studtite dissolved (mol) | Average pH | Clusters |
|---|---|---|---|---|---|---|---|
| pH_H2O_Blank | 1 | 0 | 0 | 0 | 0.00 ± 0.00 | 4.57 ± 0.41 | No |
| pH_H2O_7 | 1 | 0 | 0 | 0.01 ± 0.00 | 0.00 ± 0.00 | 7.00 ± 0.05 | No |
| pH_H2O_8 | 1 | 0 | 0 | 0.01 ± 0.00 | 0.00 ± 0.00 | 8.04 ± 0.07 | No |
| pH_H2O_9 | 1 | 0 | 0 | 0.03 ± 0.00 | 0.00 ± 0.00 | 8.95 ± 0.02 | No |
| pH_H2O_10 | 1 | 0 | 0 | 0.07 ± 0.03 | 0.00 ± 0.00 | 9.98 ± 0.04 | No |
| pH_H2O_11 | 1 | 0 | 0 | 0.11 ± 0.02 | 0.00 ± 0.00 | 10.95 ± 0.01 | No |
| pH_001_Blank | 1 | 0.2 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 4.03 ± 0.18 | No |
| pH_001_7 | 1 | 0.2 | 0.00 ± 0.00 | 0.01 ± 0.00 | 0.00 ± 0.00 | 6.99 ± 0.10 | No |
| pH_001_8 | 1 | 0.2 | 0.00 ± 0.00 | 0.02 ± 0.01 | 0.00 ± 0.00 | 7.97 ± 0.05 | No |
| pH_001_9 | 1 | 0.2 | 0.00 ± 0.00 | 0.04 ± 0.01 | 0.00 ± 0.00 | 8.99 ± 0.04 | No |
| pH_001_10 | 1 | 0.2 | 0.00 ± 0.00 | 0.07 ± 0.03 | 0.00 ± 0.00 | 9.95 ± 0.04 | No |
| pH_001_11 | 1 | 0.2 | 0.04 ± 0.00 | 0.16 ± 0.03 | 0.08 ± 0.03 | 10.94 ± 0.05 | U28 |
| pH_0035_Blank | 1 | 0.7 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 4.03 ± 0.17 | No |
| pH_0035_7 | 1 | 0.7 | 0.00 ± 0.00 | 0.01 ± 0.00 | 0.00 ± 0.00 | 7.00 ± 0.03 | No |
| pH_0035_8 | 1 | 0.7 | 0.00 ± 0.00 | 0.02 ± 0.00 | 0.00 ± 0.00 | 8.10 ± 0.12 | No |
| pH_0035_9 | 1 | 0.7 | 0.00 ± 0.00 | 0.05 ± 0.00 | 0.00 ± 0.00 | 8.98 ± 0.03 | No |
| pH_0035_10 | 1 | 0.7 | 0.02 ± 0.00 | 0.06 ± 0.02 | 0.00 ± 0.00 | 9.98 ± 0.03 | No |
| pH_0035_11 | 1 | 0.7 | 0.13 ± 0.01 | 0.30 ± 0.07 | 0.25 ± 0.02 | 10.89 ± 0.05 | U28 |
| pH_010_Blank | 1 | 2 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 4.03 ± 0.14 | No |
| pH_010_7 | 1 | 2 | 0.00 ± 0.00 | 0.01 ± 0.00 | 0.00 ± 0.00 | 7.03 ± 0.05 | No |
| pH_010_8 | 1 | 2 | 0.00 ± 0.00 | 0.02 ± 0.00 | 0.00 ± 0.00 | 8.00 ± 0.03 | No |
| pH_010_9 | 1 | 2 | 0.00 ± 0.00 | 0.05 ± 0.01 | 0.00 ± 0.00 | 8.96 ± 0.01 | No |
| pH_010_10 | 1 | 2 | 0.04 ± 0.00 | 0.06 ± 0.03 | 0.00 ± 0.00 | 9.97 ± 0.03 | No |
| pH_010_11 | 1 | 2 | 0.39 ± 0.01 | 1.00 ± 0.06 | 0.64 ± 0.01 | 10.92 ± 0.02 | U28 |
| pH_050_Blank | 1 | 10 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 3.90 ± 0.14 | No |
| pH_050_7 | 1 | 10 | 0.00 ± 0.00 | 0.01 ± 0.00 | 0.00 ± 0.00 | 7.01 ± 0.06 | No |
| pH_050_8 | 1 | 10 | 0.00 ± 0.00 | 0.03 ± 0.00 | 0.00 ± 0.00 | 7.97 ± 0.01 | No |
| pH_050_9 | 1 | 10 | 0.02 ± 0.00 | 0.08 ± 0.01 | 0.00 ± 0.00 | 8.93 ± 0.02 | No |
| pH_050_10 | 1 | 10 | 0.22 ± 0.00 | 0.40 ± 0.04 | 0.17 ± 0.07 | 9.96 ± 0.01 | U28 |
| pH_050_11 | 1 | 10 | 2.51 ± 0.37 | 1.83 ± 0.15 | 0.99 ± 0.05 | 11.02 ± 0.06 | U28 |
| pH_100_Blank | 1 | 20 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 3.83 ± 0.19 | No |
| pH_100_7 | 1 | 20 | 0.00 ± 0.00 | 0.01 ± 0.00 | 0.00 ± 0.00 | 7.05 ± 0.04 | No |
| pH_100_8 | 1 | 20 | 0.00 ± 0.00 | 0.03 ± 0.01 | 0.00 ± 0.00 | 7.94 ± 0.02 | No |
| pH_100_9 | 1 | 20 | 0.04 ± 0.00 | 0.08 ± 0.02 | 0.00 ± 0.00 | 8.97 ± 0.05 | No |
| pH_100_10 | 1 | 20 | 0.47 ± 0.06 | 0.78 ± 0.14 | 0.42 ± 0.01 | 9.99 ± 0.05 | U28 |
| pH_100_11 | 1 | 20 | 6.45 ± 0.66 | 2.23 ± 0.25 | 1.02 ± 0.04 | 11.13 ± 0.05 | U28 |

| Reaction | Initial Studtite (mol) | Initial $H_2O_2$ (mol) | OOH- Fraction (mol) | TEAOH (mol) | Studtite dissolved (mol) | Average pH | Clusters |
|---|---|---|---|---|---|---|---|
| moles_H2O_Blank | 1 | 0 | 0 | 0 | 0.00 ± 0.00 | 4.57 ± 0.42 | No |
| moles_H2O_0.02 | 1 | 0 | 0 | 0.02 ± 0.01 | 0.00 ± 0.00 | 8.20 ± 1.02 | No |
| moles_H2O_0.1 | 1 | 0 | 0 | 0.10 ± 0.02 | 0.00 ± 0.00 | 10.56 ± 0.54 | No |
| moles_H2O_0.5 | 1 | 0 | 0 | 0.51 ± 0.00 | 0.05 ± 0.01 | 12.05 ± 0.30 | U24 |
| moles_H2O_1 | 1 | 0 | 0 | 1.02 ± 0.01 | 0.24 ± 0.03 | 12.08 ± 0.24 | U24 |
| moles_H2O_2 | 1 | 0 | 0 | 2.01 ± 0.01 | 0.79 ± 0.04 | 12.69 ± 0.25 | U24 |
| moles_H2O_3 | 1 | 0 | 0 | 3.05 ± 0.02 | 1.04 ± 0.01 | 13.01 ± 0.11 | U24 |
| moles_H2O_4 | 1 | 0 | 0 | 4.07 ± 0.06 | 1.08 ± 0.07 | 13.11 ± 0.14 | U24 |
| moles_H2O_5 | 1 | 0 | 0 | 5.04 ± 0.06 | 1.09 ± 0.03 | 13.13 ± 0.14 | U24 |
| moles_001_0 | 1 | 0.2 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 4.03 ± 0.18 | No |
| moles_001_0.03 | 1 | 0.2 | 0.00 ± 0.00 | 0.03 ± 0.03 | 0.00 ± 0.00 | 8.48 ± 1.16 | No |
| moles_001_0.2 | 1 | 0.2 | 0.04 ± 0.00 | 0.16 ± 0.03 | 0.08 ± 0.03 | 10.95 ± 0.05 | U28 |
| moles_001_0.5 | 1 | 0.2 | 0.20 ± 0.00 | 0.51 ± 0.00 | 0.23 ± 0.01 | 11.69 ± 0.24 | U28 |
| moles_001_1 | 1 | 0.2 | 0.20 ± 0.00 | 1.01 ± 0.00 | 0.43 ± 0.02 | 12.29 ± 0.25 | U28 |
| moles_001_1.5 | 1 | 0.2 | 0.20 ± 0.00 | 1.49 ± 0.02 | 0.64 ± 0.07 | 12.49 ± 0.29 | U28 |
| moles_001_2 | 1 | 0.2 | 0.20 ± 0.00 | 2.03 ± 0.01 | 0.96 ± 0.00 | 12.82 ± 0.23 | U28 |
| moles_001_2.5 | 1 | 0.2 | 0.20 ± 0.00 | 2.53 ± 0.00 | 1.05 ± 0.01 | 13.11 ± 0.07 | U28 |
| moles_0035_0 | 1 | 0.7 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 4.04 ± 0.17 | No |
| moles_0035_0.03 | 1 | 0.7 | 0.00 ± 0.01 | 0.03 ± 0.02 | 0.00 ± 0.00 | 8.52 ± 1.15 | No |
| moles_0035_0.3 | 1 | 0.7 | 0.14 ± 0.01 | 0.32 ± 0.05 | 0.25 ± 0.04 | 10.94 ± 0.03 | U28 |
| moles_0035_0.8 | 1 | 0.7 | 0.39 ± 0.35 | 0.78 ± 0.00 | 0.38 ± 0.04 | 11.26 ± 0.37 | U28 |
| moles_0035_1 | 1 | 0.7 | 0.29 ± 0.05 | 1.00 ± 0.00 | 0.53 ± 0.07 | 11.25 ± 0.06 | U28 |
| moles_0035_1.5 | 1 | 0.7 | 0.70 ± 0.20 | 1.51 ± 0.01 | 0.75 ± 0.12 | 11.64 ± 0.13 | U28 |
| moles_0035_2 | 1 | 0.7 | 0.70 ± 0.00 | 2.03 ± 0.02 | 0.94 ± 0.12 | 12.25 ± 0.51 | U28 |
| moles_0035_2.5 | 1 | 0.7 | 0.70 ± 0.00 | 2.52 ± 0.02 | 1.03 ± 0.07 | 12.43 ± 0.01 | U28 |
| moles_0035_3 | 1 | 0.7 | 0.70 ± 0.00 | 3.04 ± 0.03 | 1.07 ± 0.03 | 12.54 ± 0.21 | U28 |
| moles_010_0 | 1 | 2 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 4.03 ± 0.14 | No |
| moles_010_0.03 | 1 | 2 | 0.01 ± 0.02 | 0.03 ± 0.02 | 0.00 ± 0.00 | 8.36 ± 1.09 | No |
| moles_010_0.2 | 1 | 2 | 0.10 ± 0.06 | 0.20 ± 0.10 | 0.02 ± 0.02 | 10.27 ± 0.29 | U28 |
| moles_010_0.5 | 1 | 2 | 0.31 ± 0.25 | 0.51 ± 0.01 | 0.18 ± 0.12 | 10.71 ± 0.35 | U28 |
| moles_010_1 | 1 | 2 | 0.40 ± 0.01 | 0.97 ± 0.08 | 0.67 ± 0.06 | 10.92 ± 0.01 | U28 |
| moles_010_1.5 | 1 | 2 | 0.56 ± 0.51 | 1.48 ± 0.01 | 0.65 ± 0.08 | 10.95 ± 0.39 | U28 |
| moles_010_2 | 1 | 2 | 0.53 ± 0.11 | 2.02 ± 0.01 | 0.70 ± 0.11 | 11.04 ± 0.10 | U28 |
| moles_010_2.5 | 1 | 2 | 1.27 ± 0.37 | 2.54 ± 0.01 | 1.05 ± 0.03 | 11.41 ± 0.13 | U28 |
| moles_010_3 | 1 | 2 | 2.00 ± 0.00 | 3.03 ± 0.03 | 1.06 ± 0.03 | 11.86 ± 0.12 | U28 |
| moles_050_0 | 1 | 10 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 3.90 ± 0.14 | No |
| moles_050_0.02 | 1 | 10 | 0.00 ± 0.00 | 0.02 ± 0.01 | 0.00 ± 0.00 | 7.49 ± 0.53 | No |
| moles_050_0.1 | 1 | 10 | 0.02 ± 0.00 | 0.08 ± 0.01 | 0.00 ± 0.00 | 8.94 ± 0.02 | No |
| moles_050_0.3 | 1 | 10 | 0.22 ± 0.01 | 0.33 ± 0.07 | 0.13 ± 0.01 | 9.96 ± 0.03 | U28 |
| moles_050_0.8 | 1 | 10 | 0.59 ± 0.41 | 0.78 ± 0.01 | 0.30 ± 0.02 | 10.33 ± 0.29 | U28 |

| Reaction | Initial Studtite (mol) | Initial $H_2O_2$ (mol) | OOH- Fraction (mol) | TEAOH (mol) | Studtite dissolved (mol) | Average pH | Clusters |
|---|---|---|---|---|---|---|---|
| moles_050_1.3 | 1 | 10 | 0.59 ± 0.17 | 1.33 ± 0.13 | 0.52 ± 0.04 | 10.38 ± 0.12 | U28 |
| moles_050_1.8 | 1 | 10 | 2.51 ± 0.37 | 1.78 ± 0.16 | 0.99 ± 0.04 | 11.22 ± 0.40 | U28 |
| moles_050_2.5 | 1 | 10 | 5.22 ± 0.92 | 2.53 ± 0.04 | 1.06 ± 0.02 | 11.34 ± 0.08 | U28 |
| moles_100_0 | 1 | 20 | 0.00 ± 0.00 | 0 | 0.00 ± 0.00 | 3.83 ± 0.19 | No |
| moles_100_0.02 | 1 | 20 | 0.00 ± 0.00 | 0.02 ± 0.01 | 0.00 ± 0.00 | 7.50 ± 0.49 | No |
| moles_100_0.1 | 1 | 20 | 0.04 ± 0.00 | 0.08 ± 0.02 | 0.00 ± 0.00 | 8.97 ± 0.05 | No |
| moles_100_0.5 | 1 | 20 | 0.48 ± 0.31 | 0.51 ± 0.00 | 0.09 ± 0.04 | 9.93 ± 0.31 | U28 |
| moles_100_0.8 | 1 | 20 | 0.46 ± 0.05 | 0.80 ± 0.11 | 0.42 ± 0.01 | 9.98 ± 0.04 | U28 |
| moles_100_1.5 | 1 | 20 | 1.02 ± 0.78 | 1.56 ± 0.13 | 0.62 ± 0.05 | 10.24 ± 0.29 | U28 |
| moles_100_2 | 1 | 20 | 4.42 ± 2.75 | 1.94 ± 0.07 | 0.99 ± 0.01 | 11.23 ± 0.57 | U28 |
| moles_100_2.4 | 1 | 20 | 6.49 ± 0.93 | 2.39 ± 0.04 | 1.04 ± 0.02 | 11.35 ± 0.39 | U28 |

**Figure S1.** Time resolved dissolution of studtite in water and 1.0 M $H_2O_2$ with a TEAOH:U molar ratio of 1.5. Error bars are derived from the stadnard deviation of triplicate reactions. Solution pH was not adjusted in these reactions after the initial addition of TEAOH at time = 0.

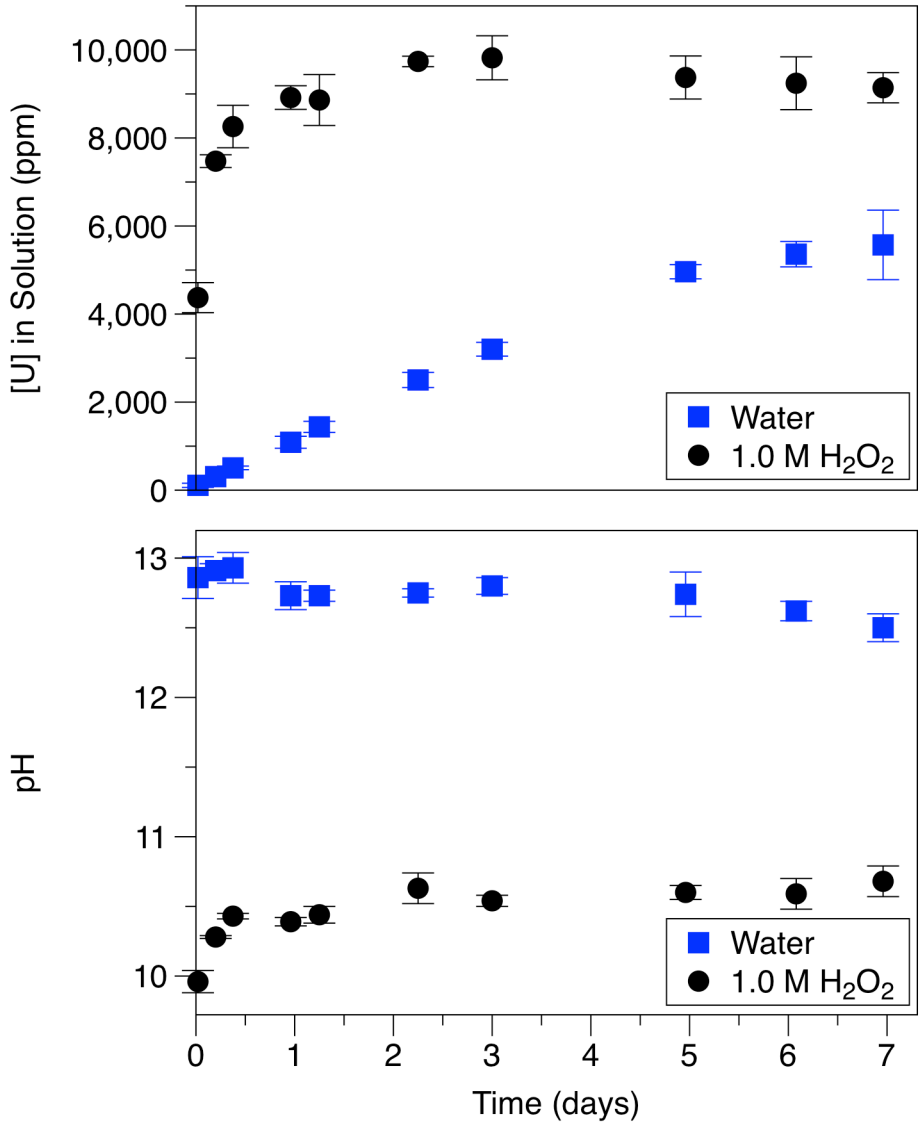

**Figure S2**. PXRD spectra of synthesized studtite phase matched with PDF # 00-016-0206 Studtite, syn. ($UO_4 \bullet 4H_2O$)

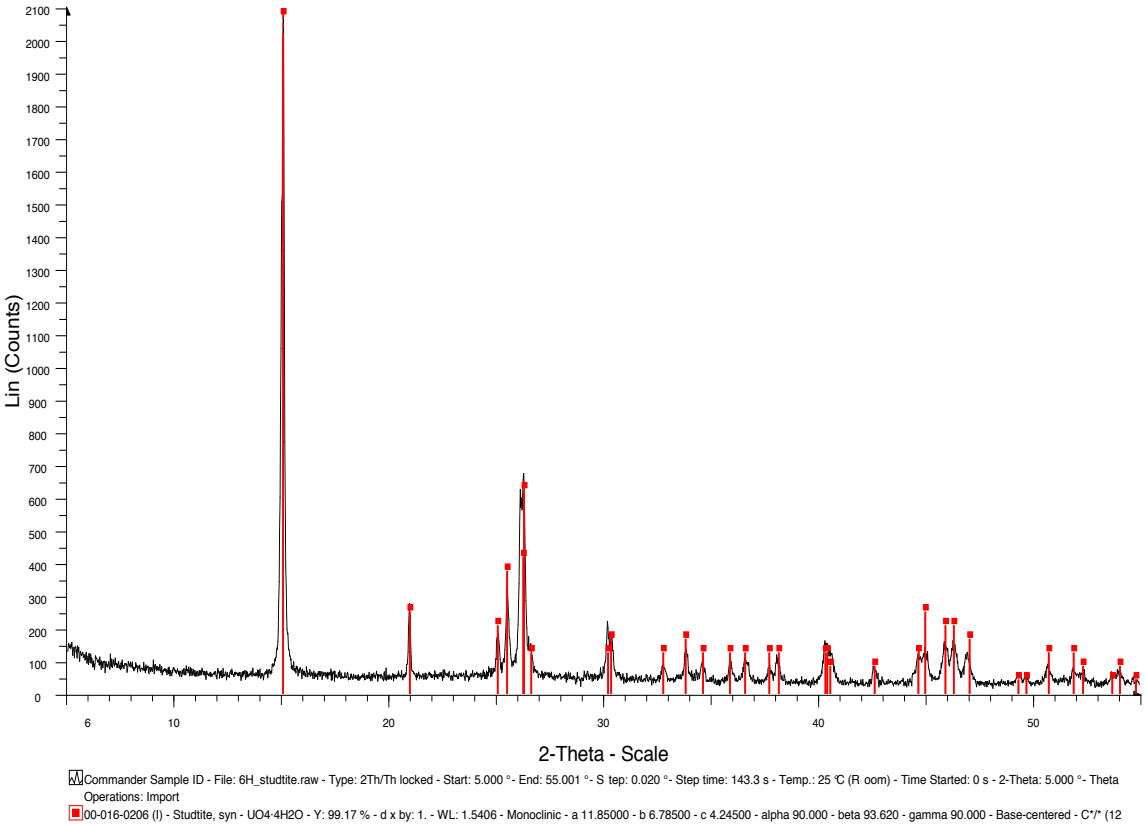

**Figure S3.** TGA analysis of synthetic studtite. Mass loss calculations indicate 4.3 mol $H_2O$ in starting material, confirming the starting material is studtite.

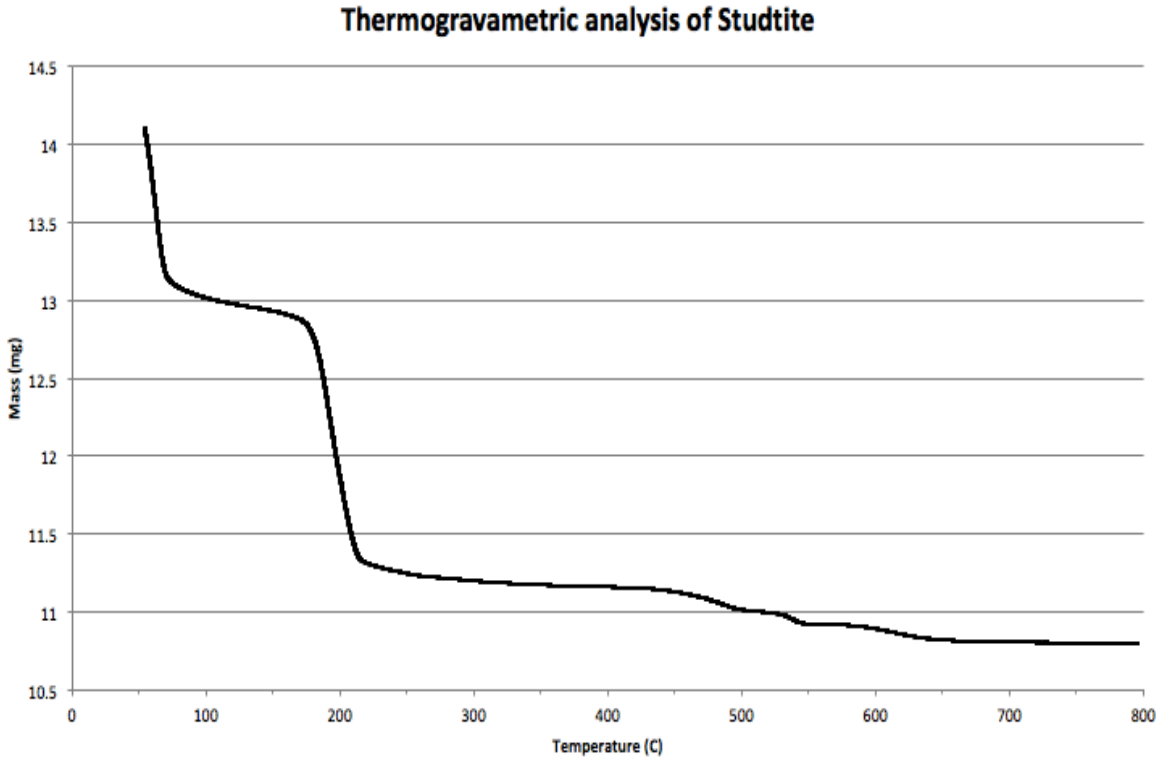

**Table S2.** ICP-OES analysis of uranium content in synthetic studtite compared to estimated uranium content calculated from molecular mass.

| Sample | %w U |
|---|---|
| Calculated from Molecular Mass | 63.63 % |
| Synthesized Studtite | 61.01 ± 1.06 % |

**Figure S4.** PXRD pattern of studtite phase after 7-day reaction with water at varying pH values.

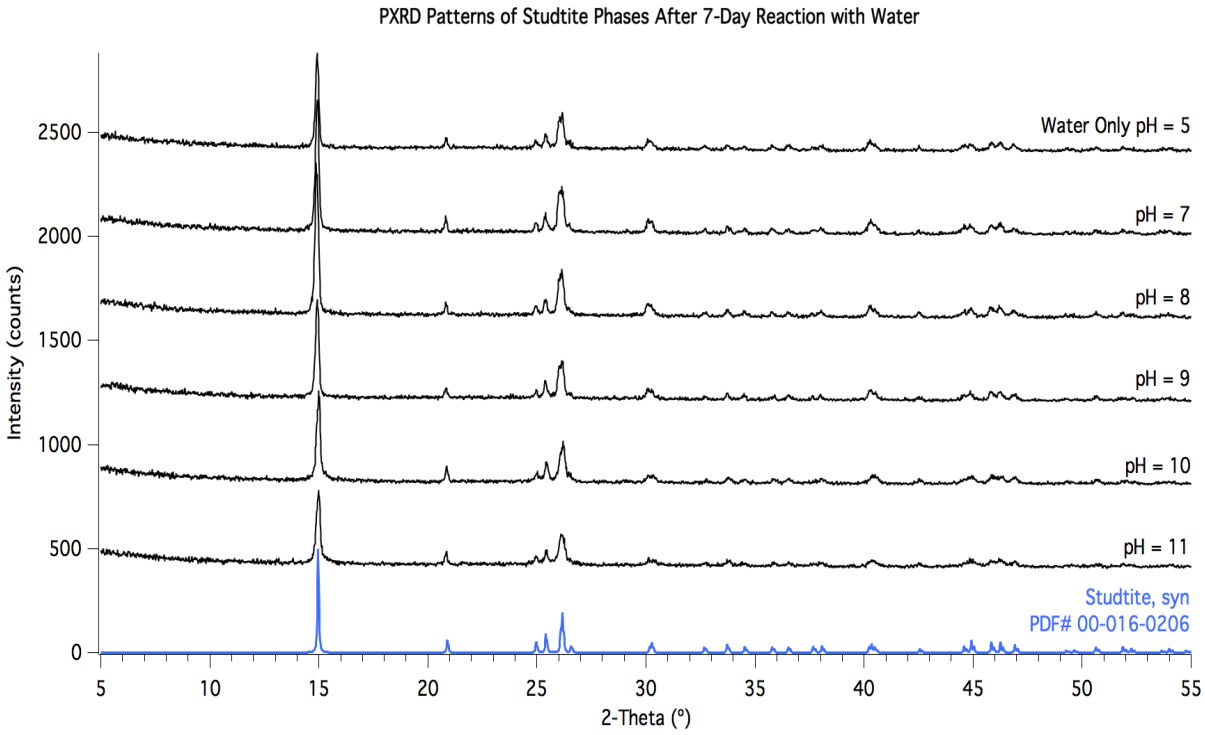

**Figure S5.** PXRD pattern of studtite phase after 7-day reaction with 0.01M $H_2O_2$ at varying pH values.

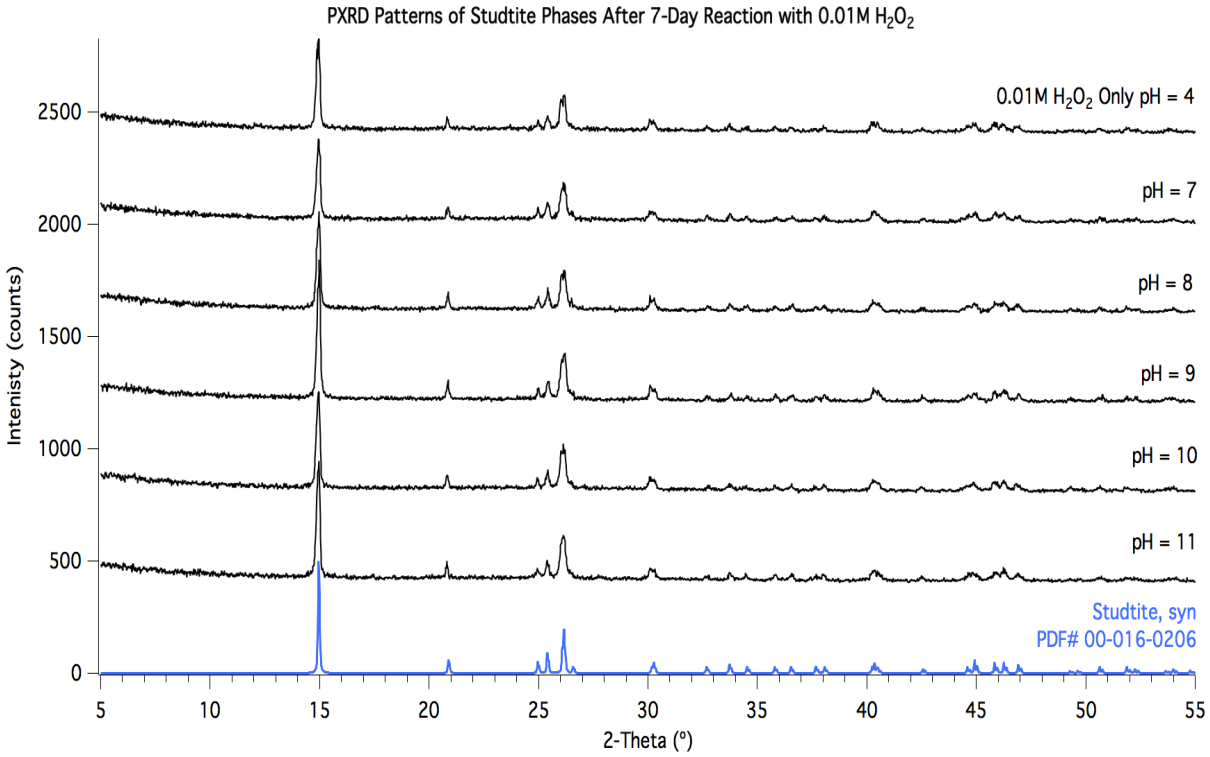

**Figure S6.** PXRD pattern of studtite phase after 7-day reaction with 0.035M $H_2O_2$ at varying pH values.

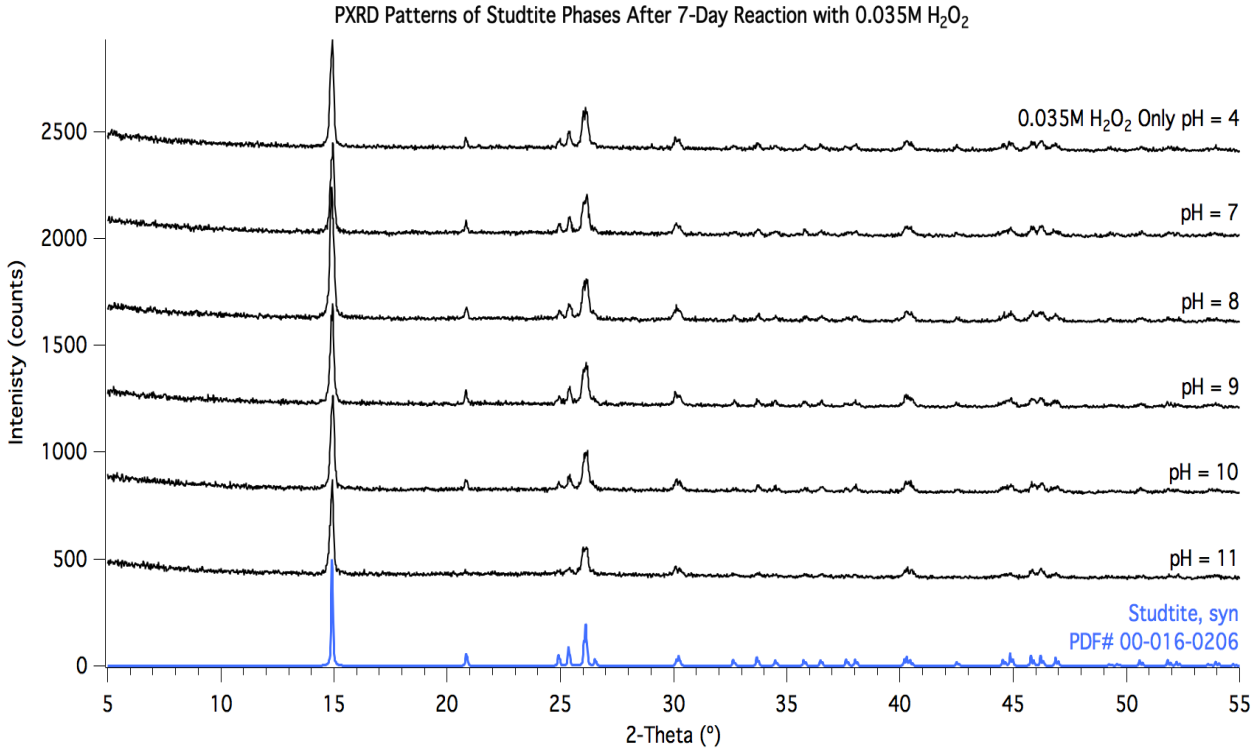

**Figure S7.** PXRD pattern of studtite phase after 7-day reaction with 0.10M $H_2O_2$ at varying pH values.

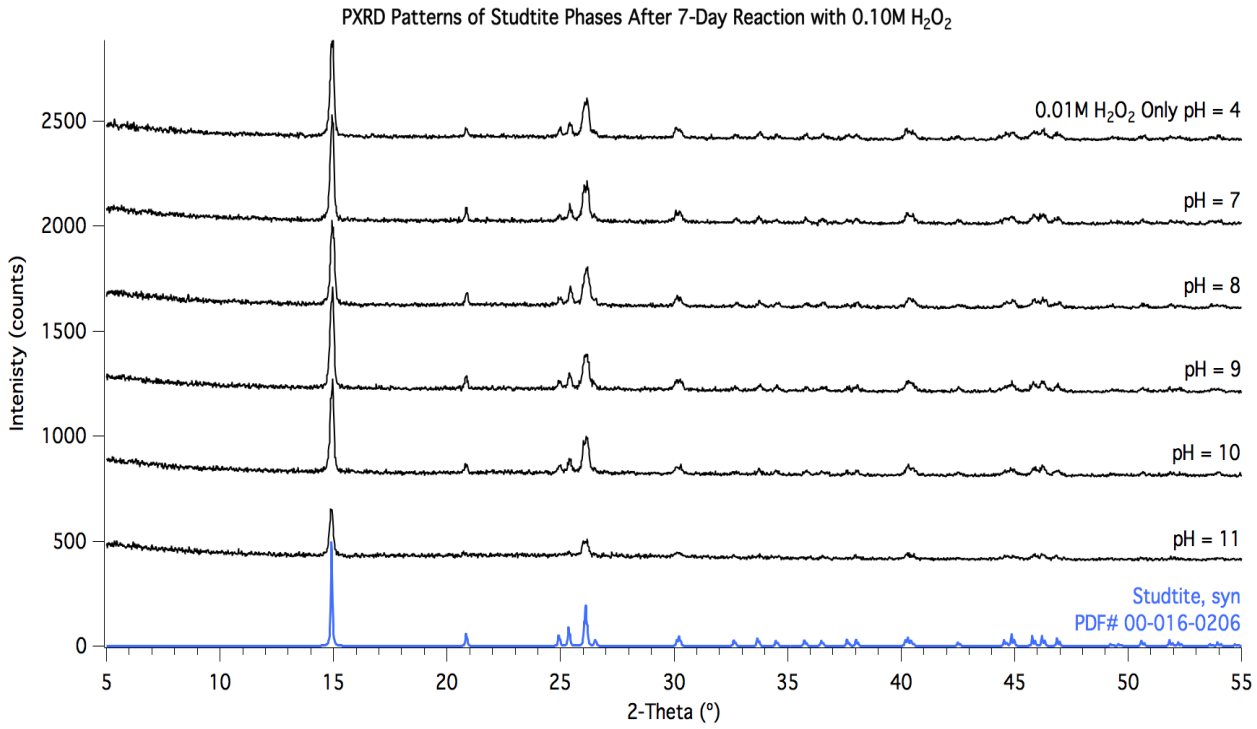

**Figure S8.** PXRD pattern of studtite phase after 7-day reaction with 0.50M $H_2O_2$ at varying pH values.

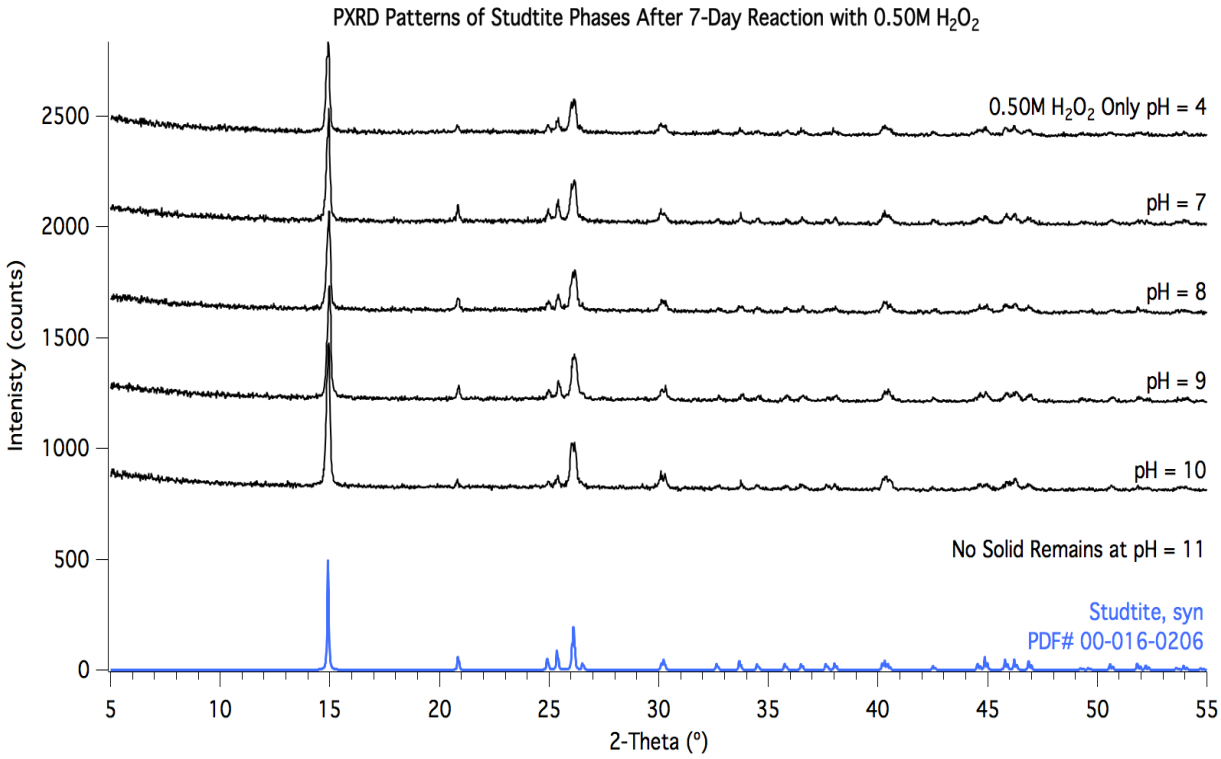

**Figure S9.** Raman spectra of studtite phase after 7-day reaction with water at varying pH values.

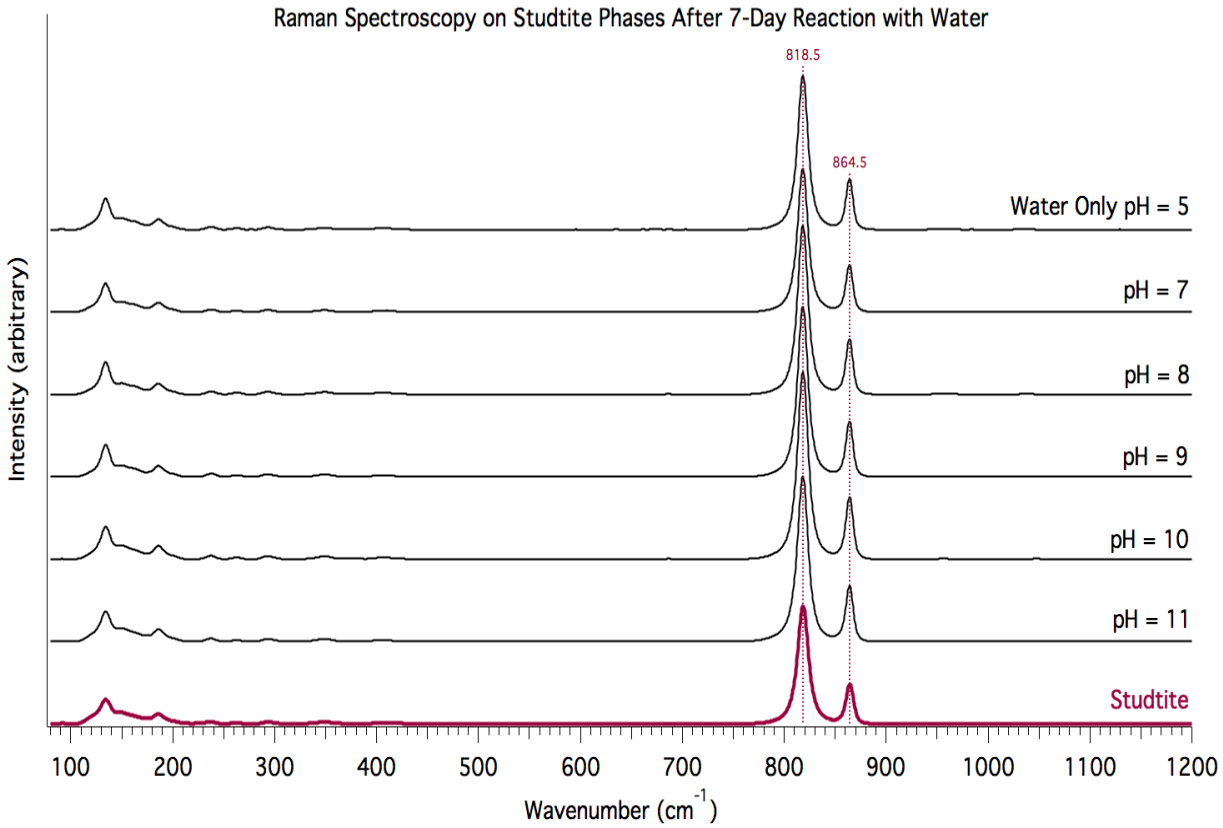

**Figure S10.** Raman spectra of studtite phase after 7-day reaction with 0.01 M $H_2O_2$ at varying pH values.

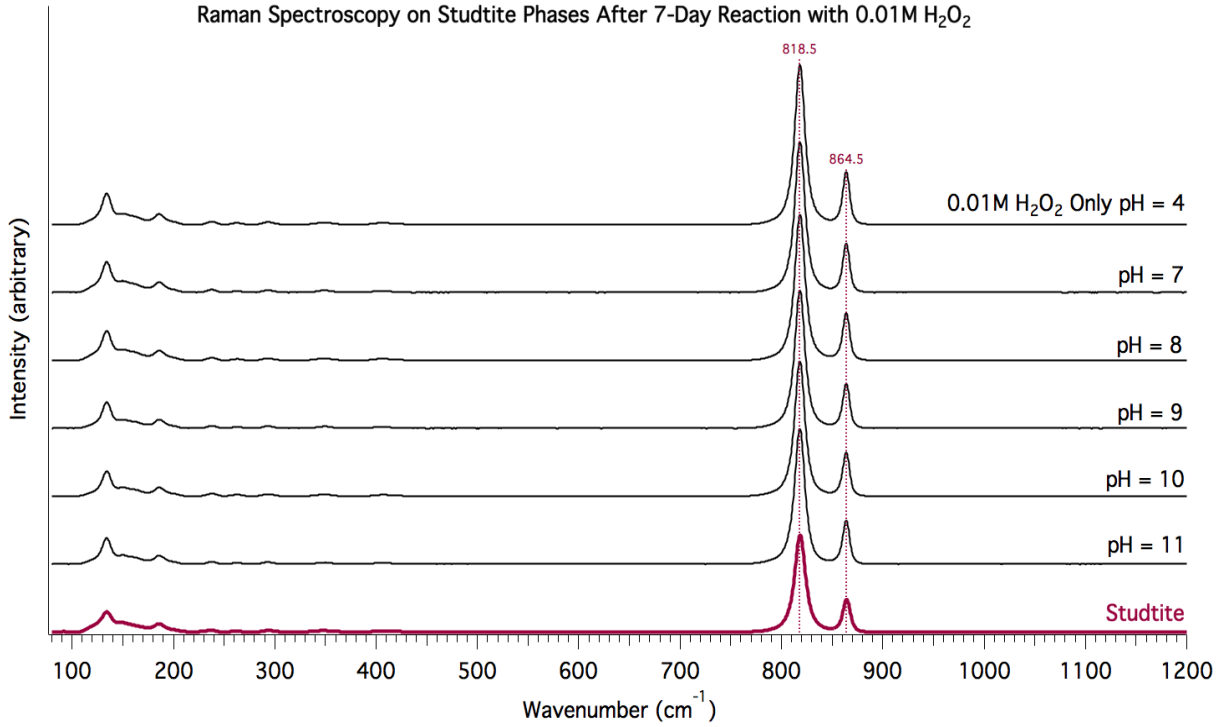

**Figure S11.** Raman spectra of studtite phase after 7-day reaction with 0.035 M $H_2O_2$ at varying pH values.

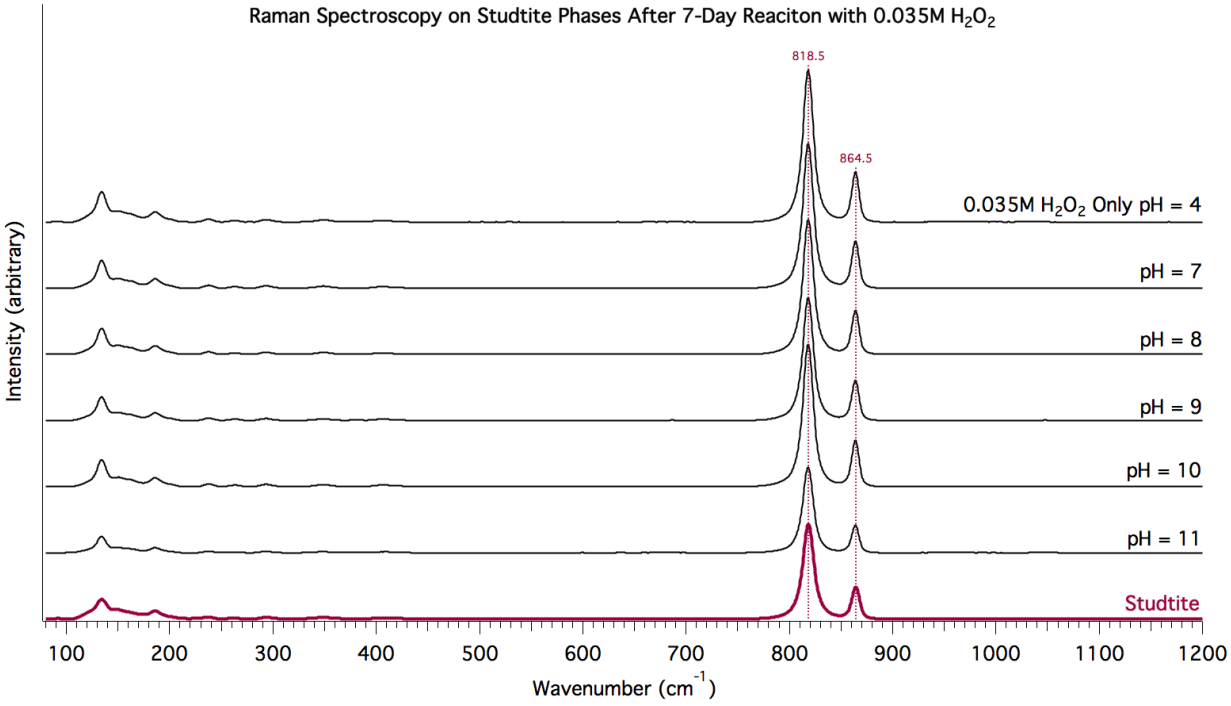

**Figure S12.** Raman spectra of studtite phase after 7-day reaction with 0.10 M $H_2O_2$ at varying pH values.

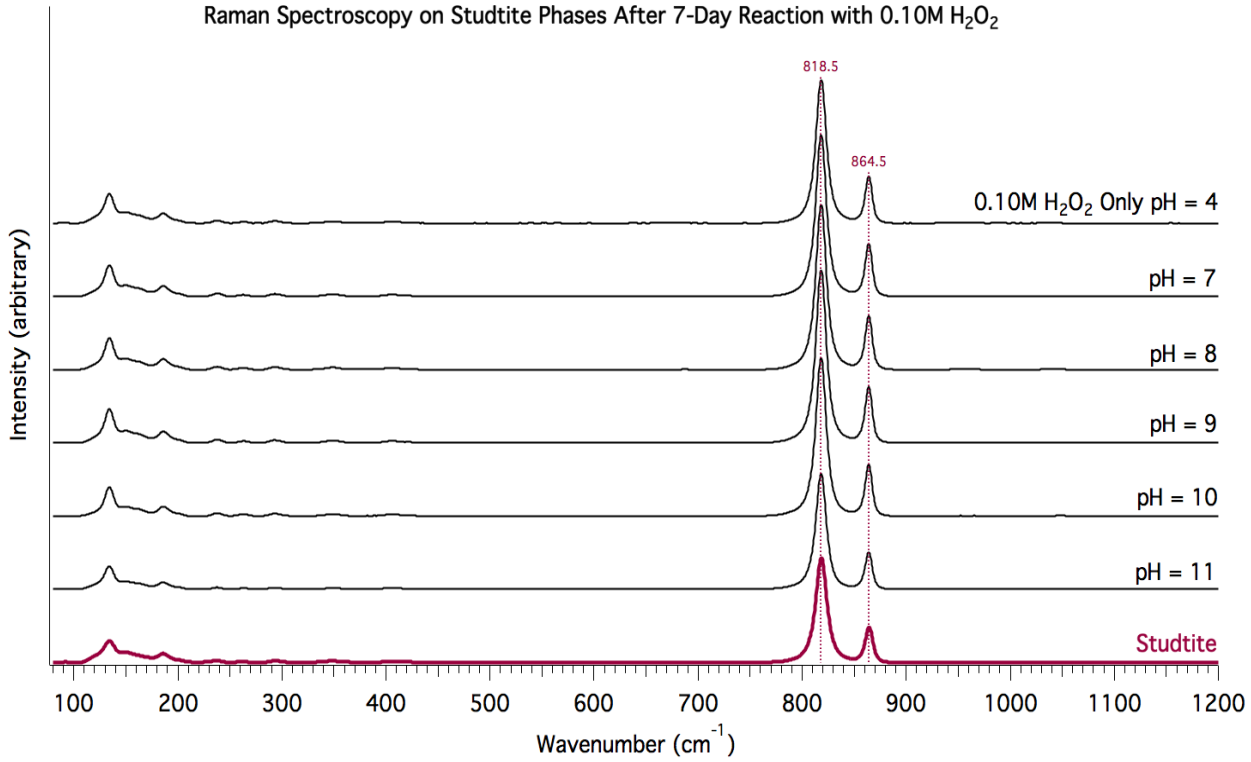

**Figure S13.** Raman spectra of studtite phase after 7-day reaction with 0.50 M $H_2O_2$ at varying pH values.

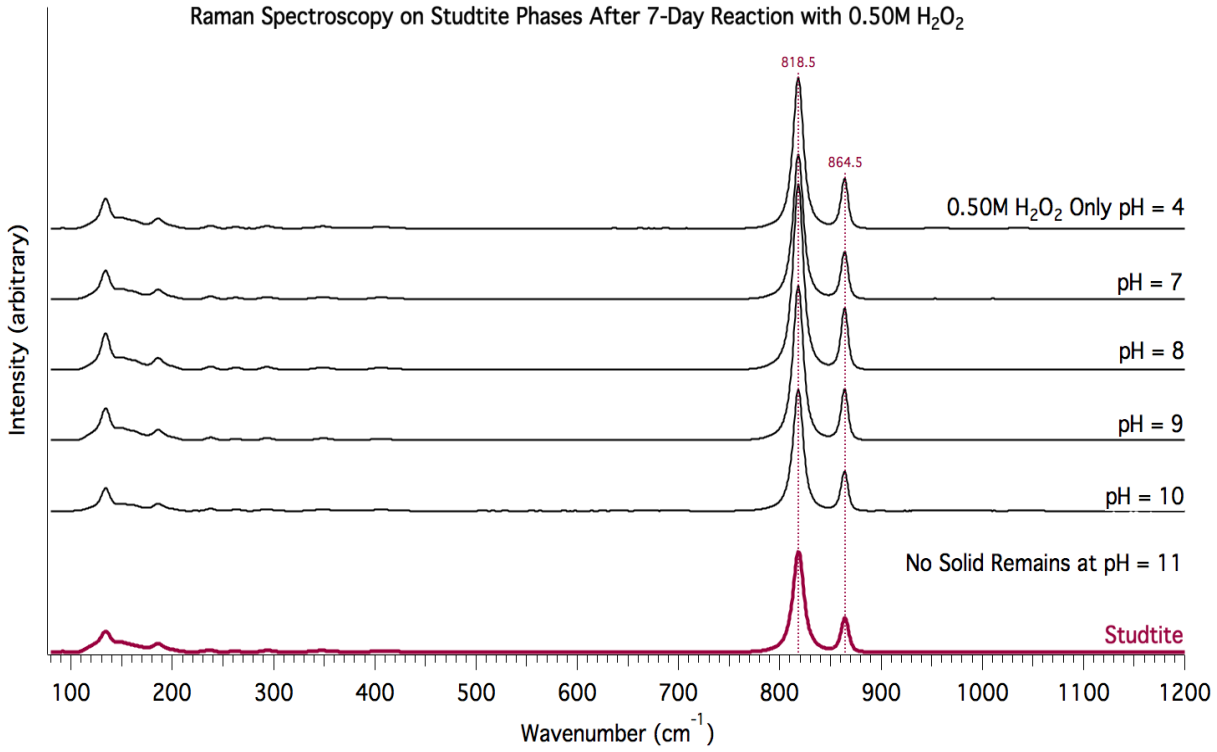

**Figure S14.** Raman spectra of solutions in U:TEAOH molar reactions in water only environments. Lines are marked at 810 and 847 $cm^{-1}$.

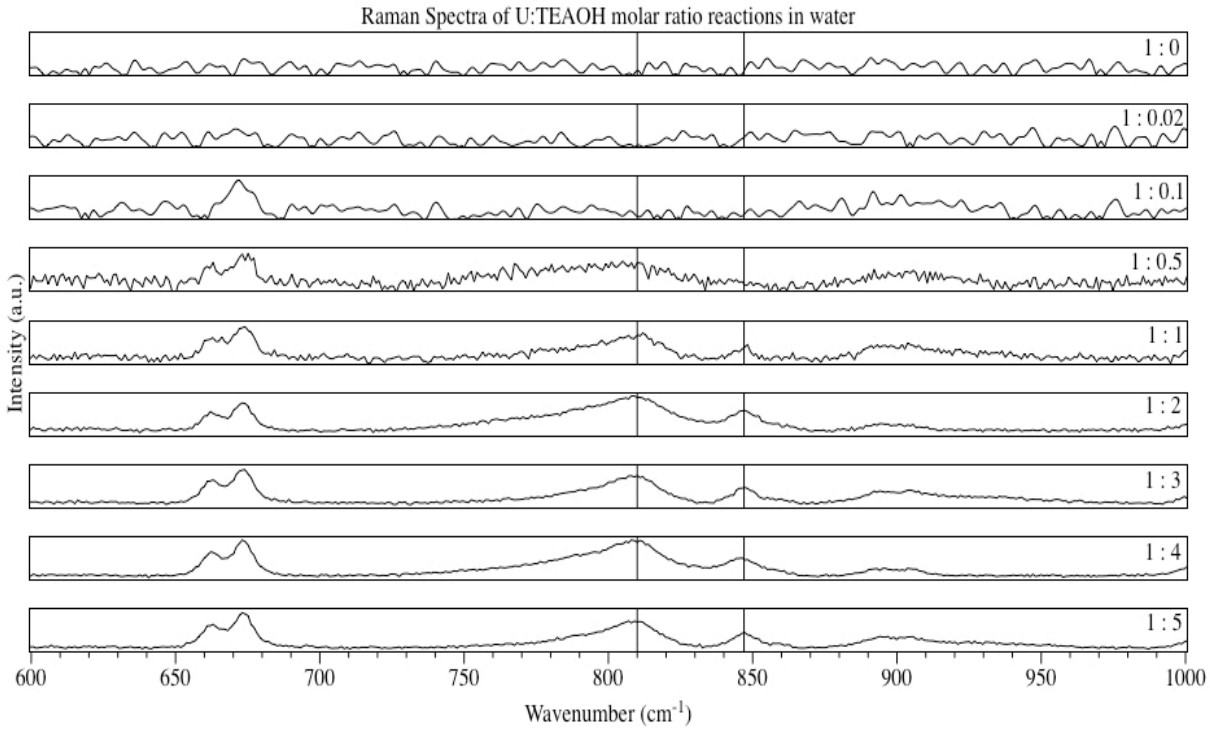

**Figure S15.** ESI-MS spectra of solutions in U:TEAOH molar reactions  water only environments. Red lines represent -5, -4, -3 charge state and a cluster with a $M_{avg}$ = 8.5 kDa.

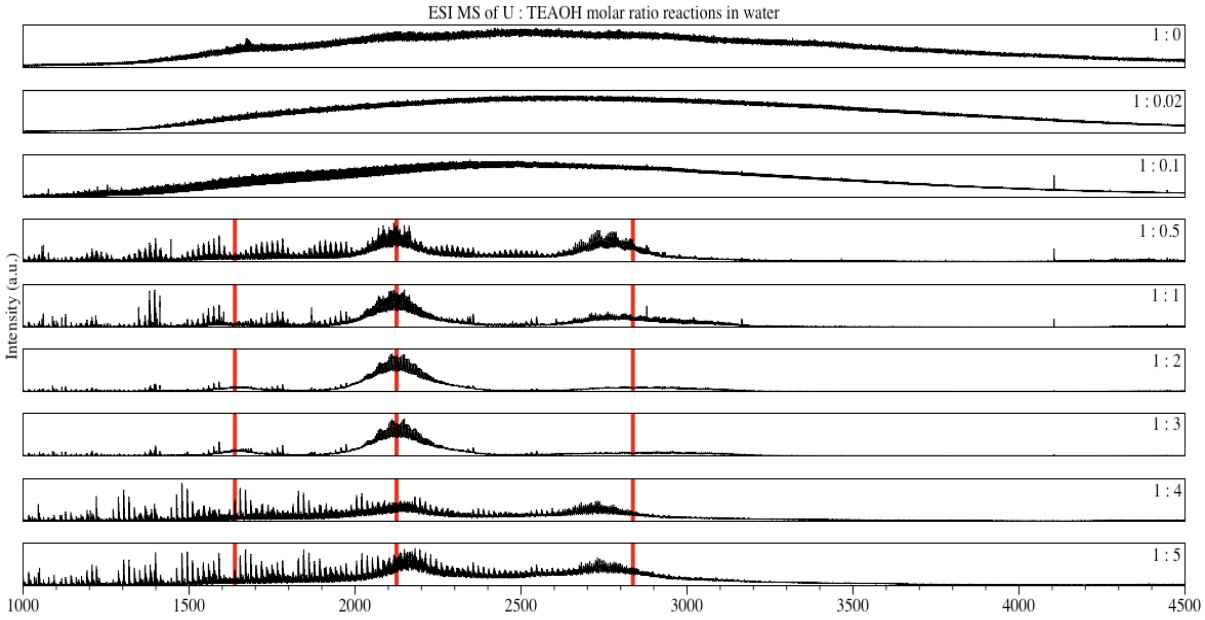

**Table S3.** Uranium concentration (ppm) detected in solution after studtite reacted with peroxide solutions of variable concentration at different pH values after seven days. The uranium concentrations of the control solutions remain below 12 ppm U for all experimental conditions

| [U] (ppm) | | Initial $H_2O_2$ Concentration | | | | | |
|---|---|---|---|---|---|---|---|
| | | **Water Only** | **0.01 M** | **0.035 M** | **0.10 M** | **0.50 M** | **1.00 M** |
| **pH** | **Blank** | 12 ± 8 | < 1 | < 1 | < 1 | < 1 | < 1 |
| | **7** | 4 ± 4 | < 1 | < 1 | < 1 | < 1 | 1 ± 1 |
| | **8** | 4 ± 3 | 1 ± 1 | 2 ± 2 | 1 ± 1 | 3 ± 2 | 5 ± 4 |
| | **9** | 13 ± 8 | 4 ± 3 | 4 ± 3 | 13 ± 2 | 20 ± 10 | 31 ± 9 |
| | **10** | 24 ± 11 | 10 ± 6 | 30 ± 23 | 50 ± 17 | 2065 ± 842 | 4901 ± 108 |
| | **11** | 19 ± 7 | 939 ± 386 | 2877 ± 176 | 7627 ± 202 | 11476 ± 478 | 11721 ± 492 |

**Figure S16.** Raman spectra of final solutions following seven-day reaction of studtite with water at varying pH values.

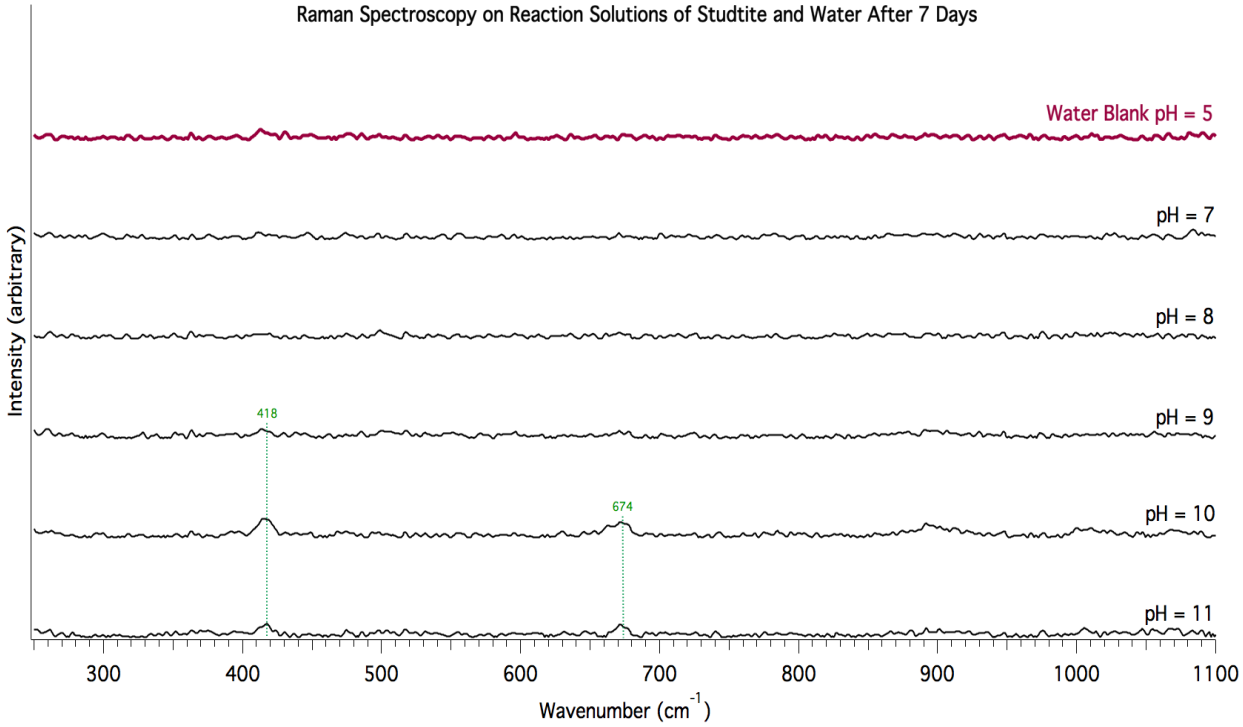

**Figure S17.** Raman spectra of final solutions following seven-day reaction of studtite with 0.01 M peroxide at varying pH values.

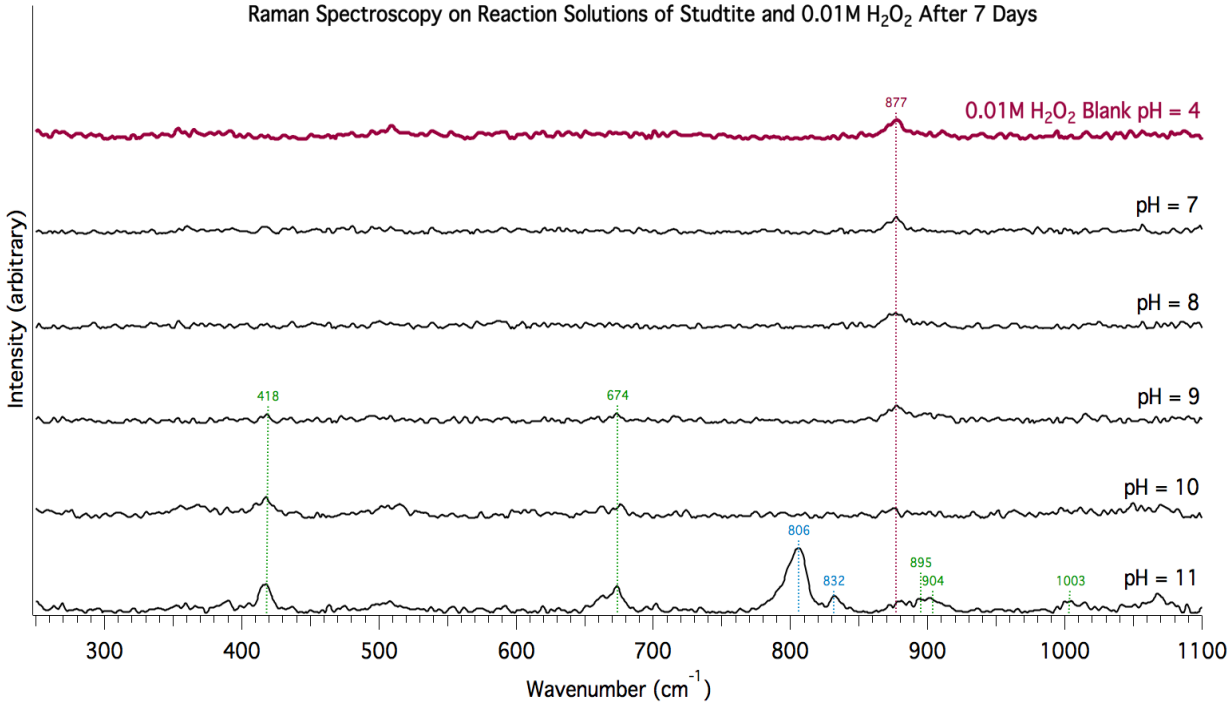

**Figure S18.** Raman spectra of final solutions following seven-day reaction of studtite with 0.035 M peroxide at varying pH values.

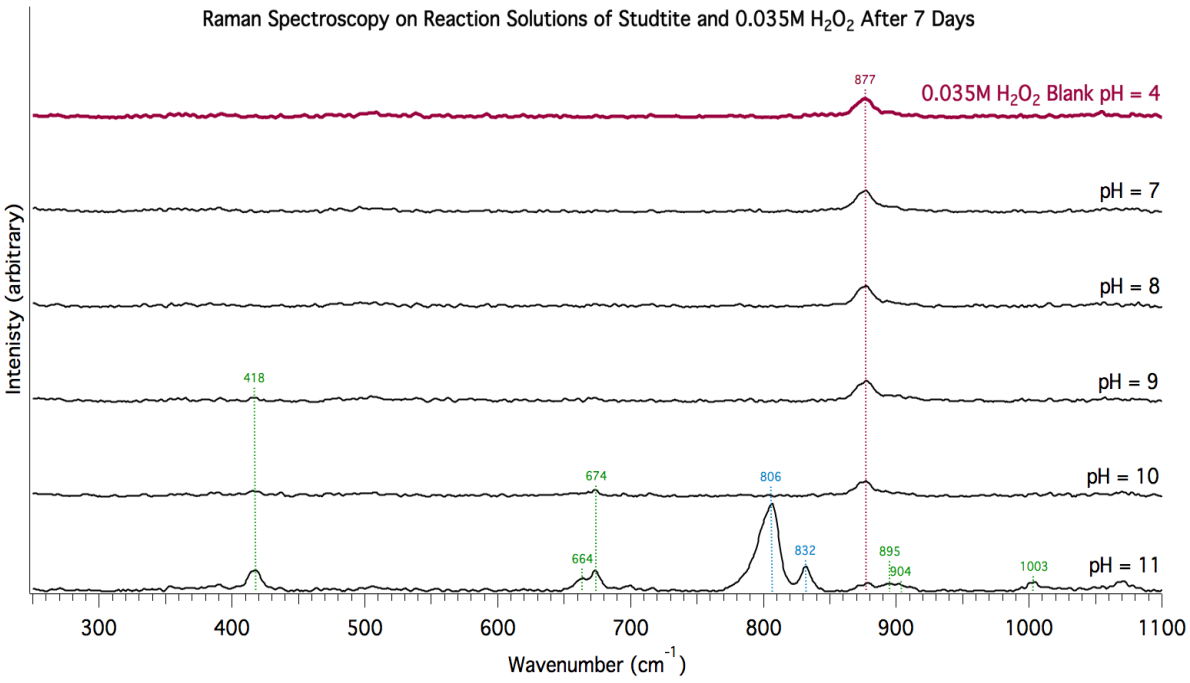

**Figure S19.** Raman spectra of final solutions following seven-day reaction of studtite with 0.10 M peroxide at varying pH values.

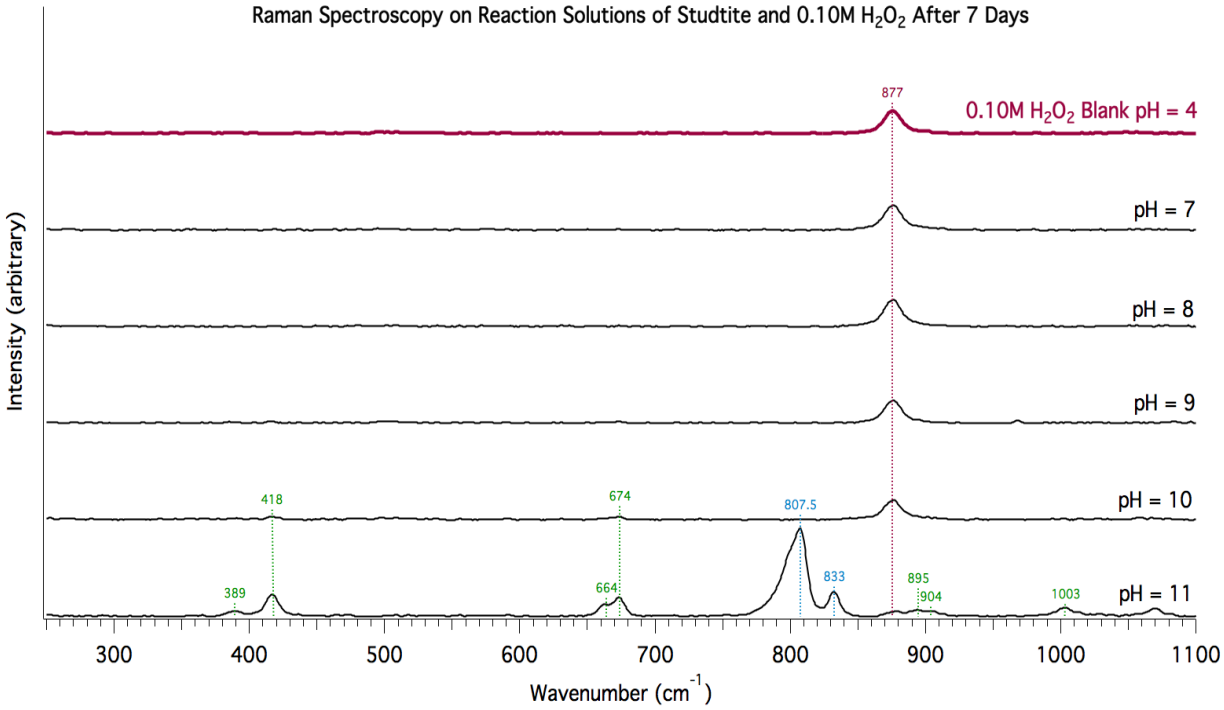

**Figure S20.** Raman spectra of final solutions following seven-day reaction of studtite with 0.50 M peroxide at varying pH values.

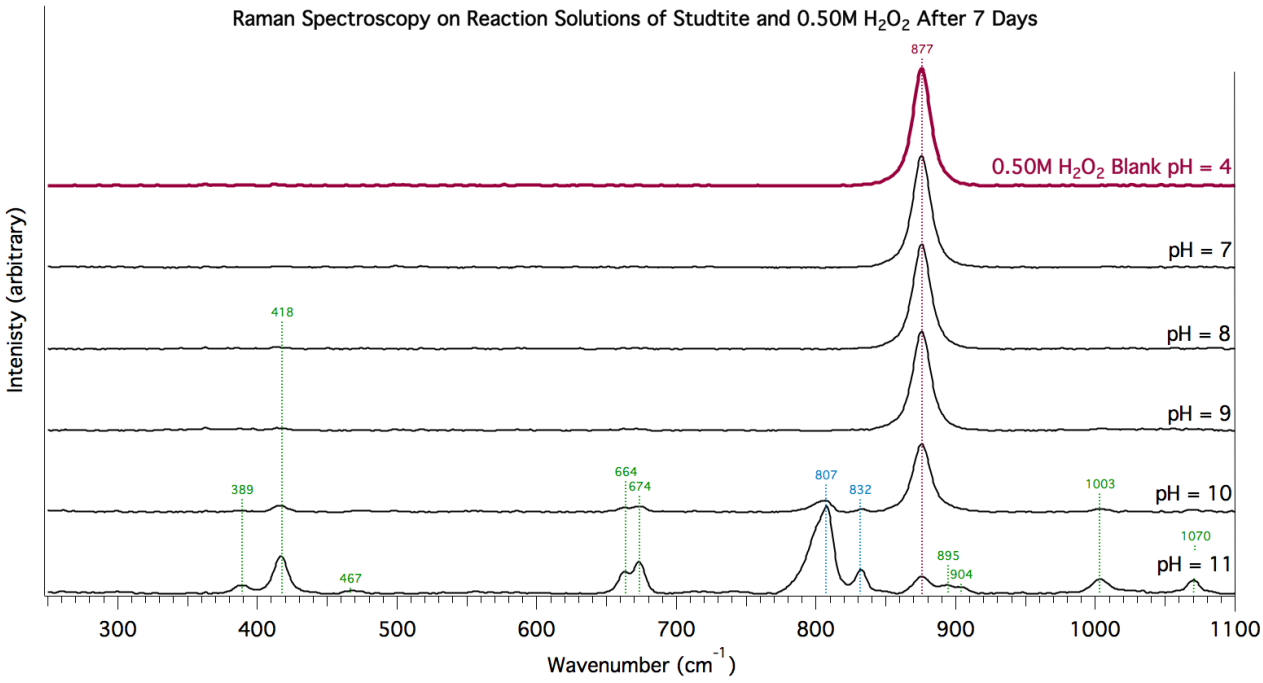

**Figure S21.** ESI-MS spectra of final solutions following seven-day reaction of studtite with water at varying pH values.

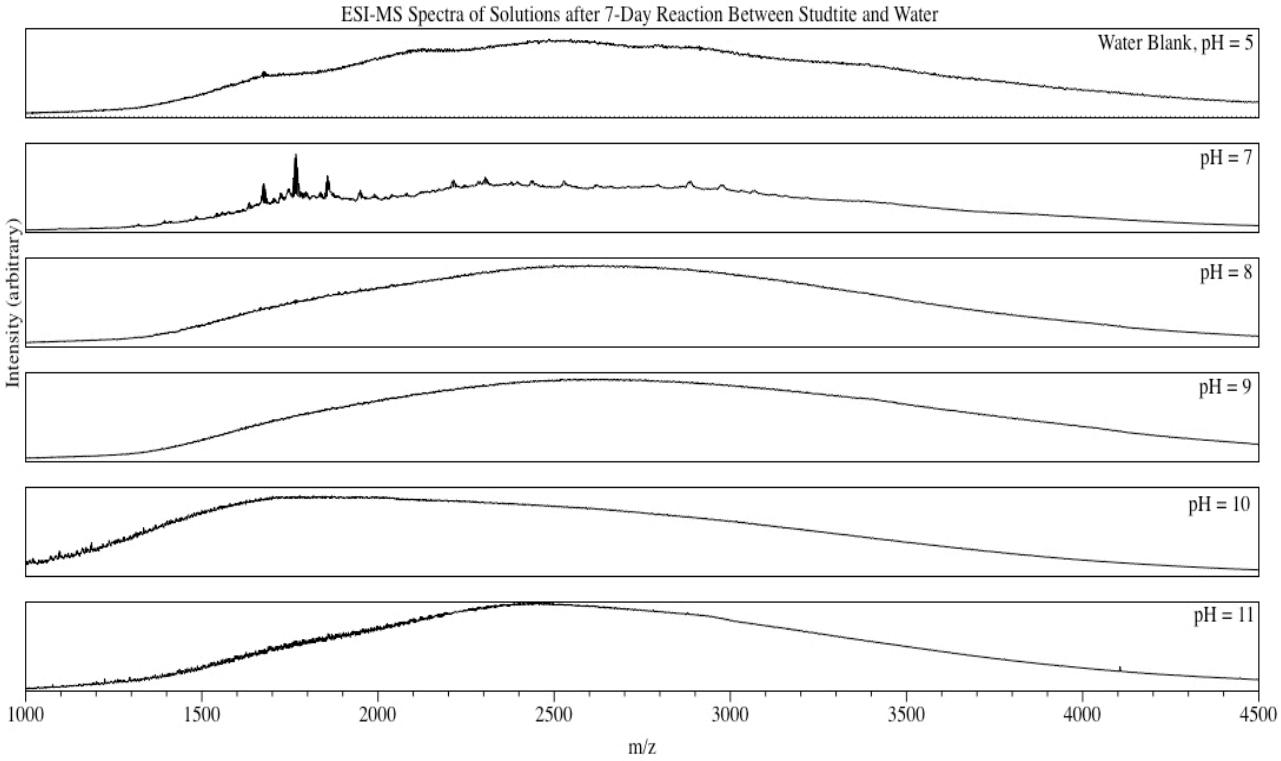

**Figure S22.** ESI-MS spectra of final solutions following seven-day reaction of studtite with 0.01 M peroxide at varying pH values. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

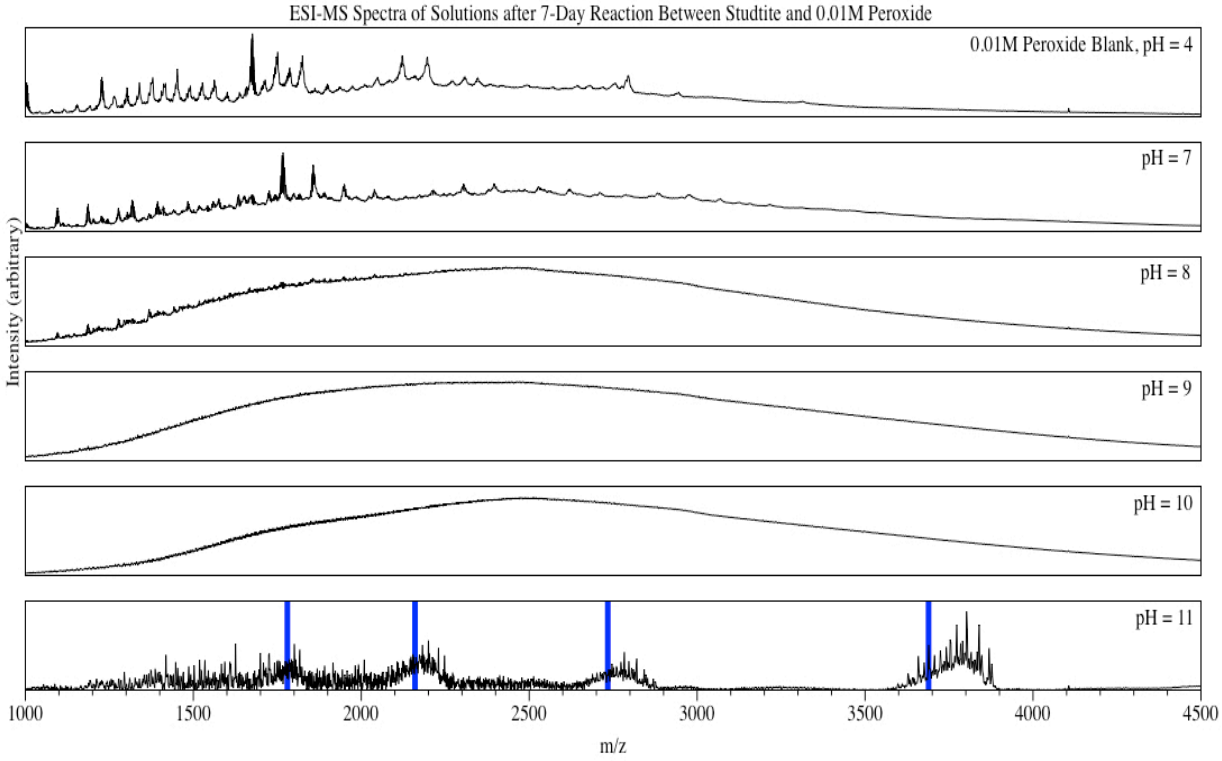

**Figure S23.** ESI-MS spectra of final solutions following seven-day reaction of studtite with 0.035 M peroxide at varying pH values. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

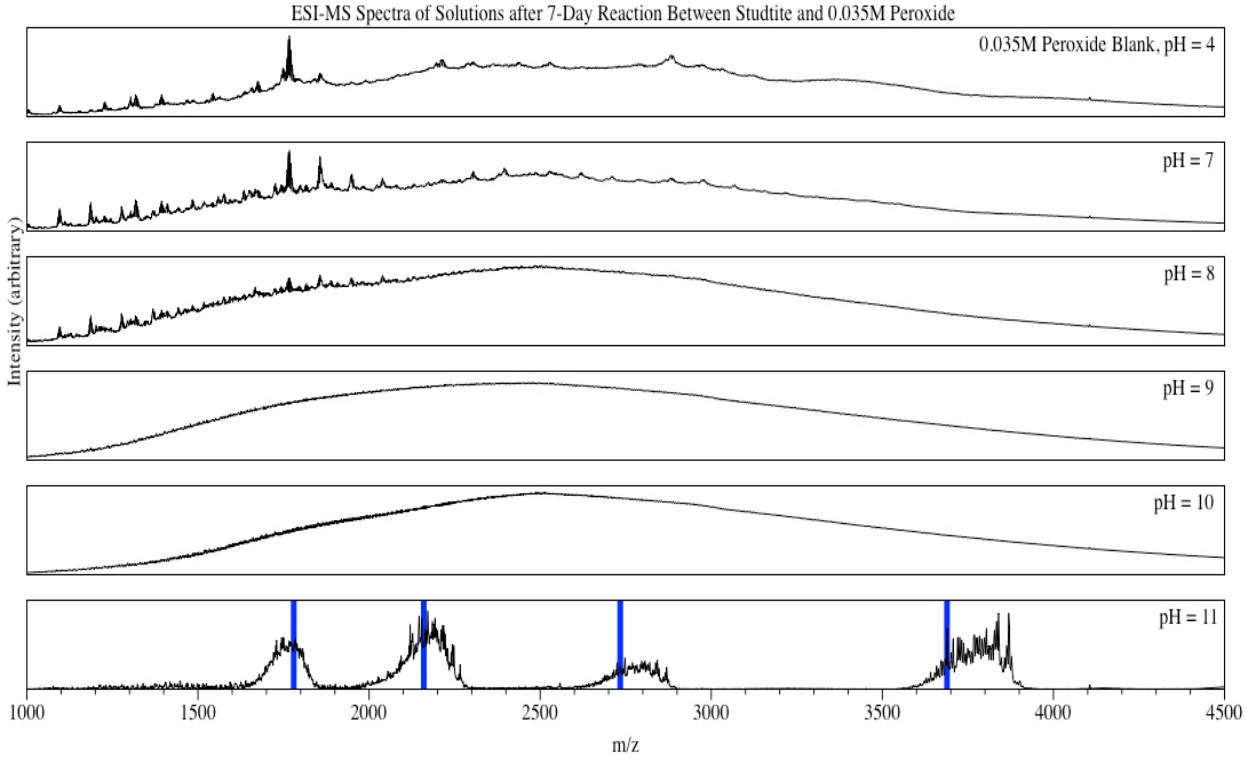

**Figure S24.** ESI-MS spectra of final solutions following seven-day reaction of studtite with 0.10 M peroxide at varying pH values. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

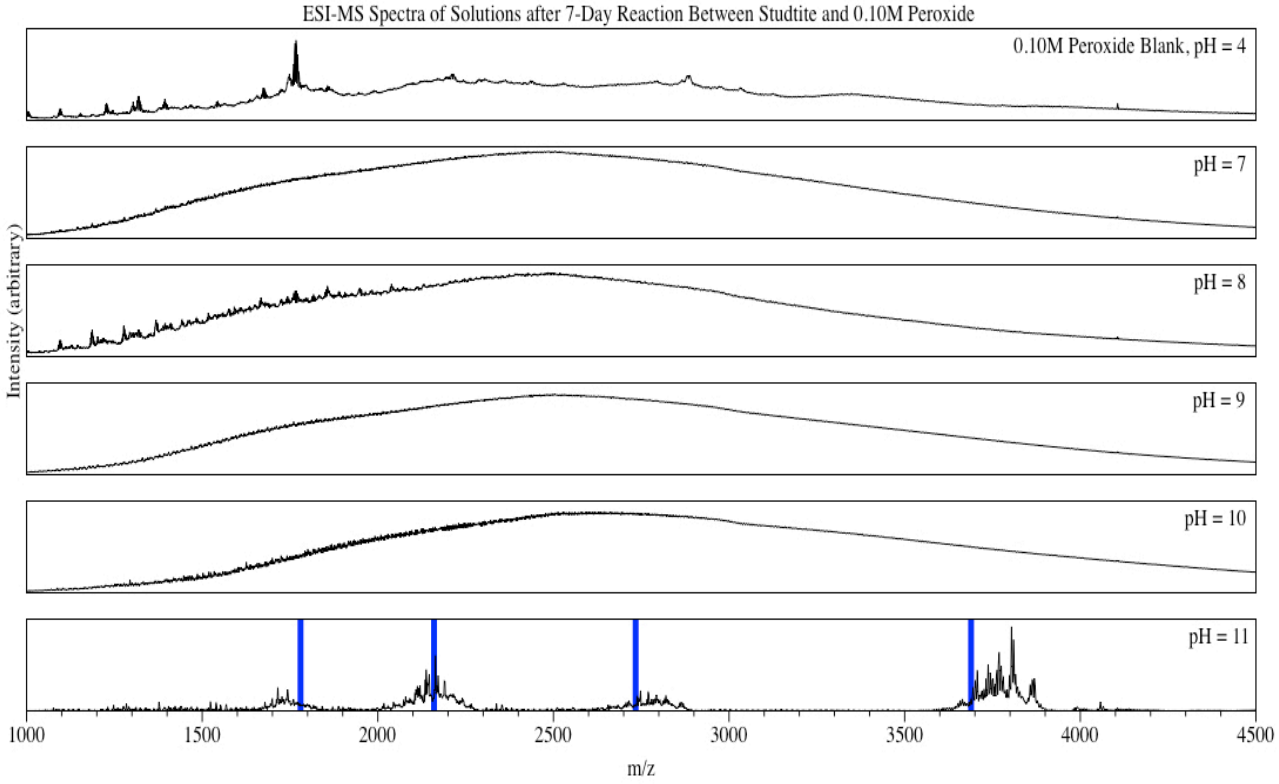

**Figure S25.** ESI-MS spectra of final solutions following seven-day reaction of studtite with 0.50 M peroxide at varying pH values. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

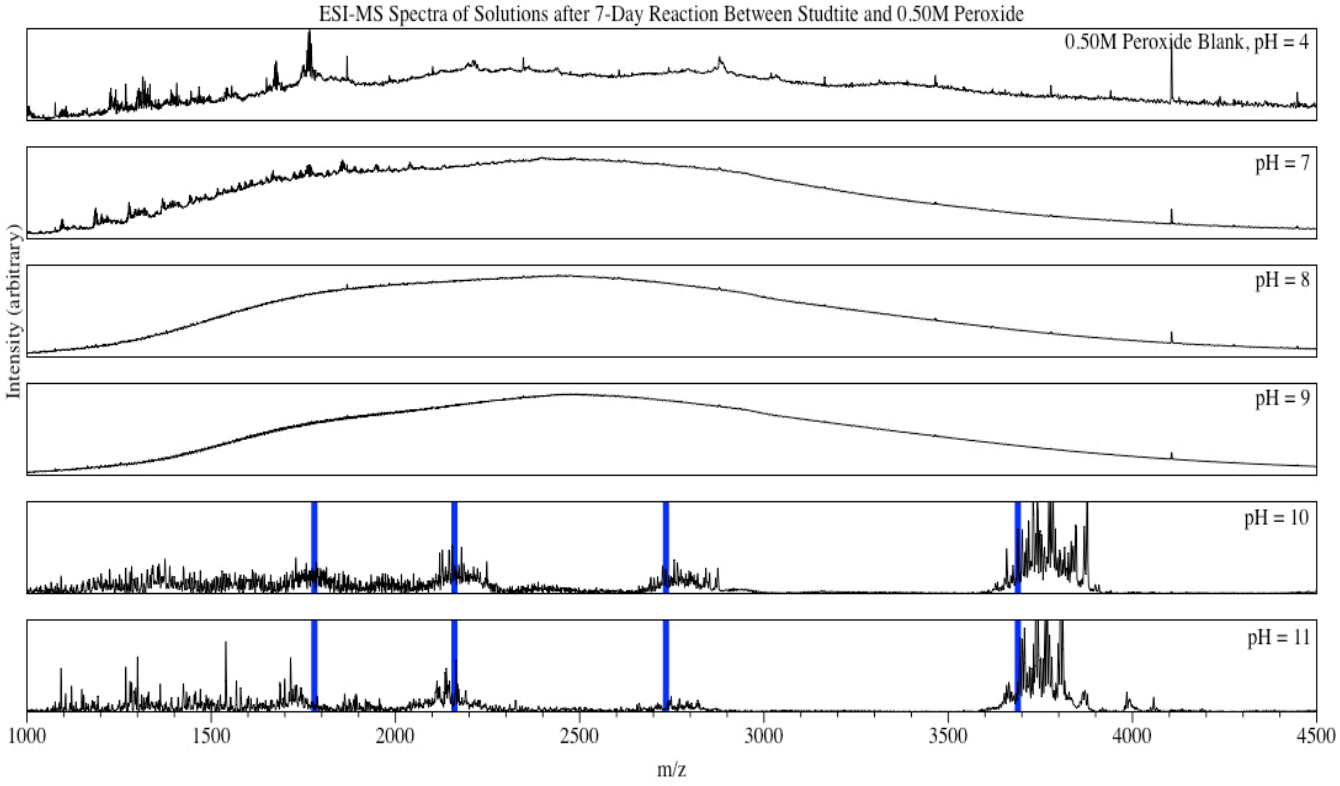

**Figure S26.** Raman spectra of solutions in U:TEAOH molar reactions in 0.01 M peroxide environments. Lines are marked at 808, 832, and 876 $cm^{-1}$.

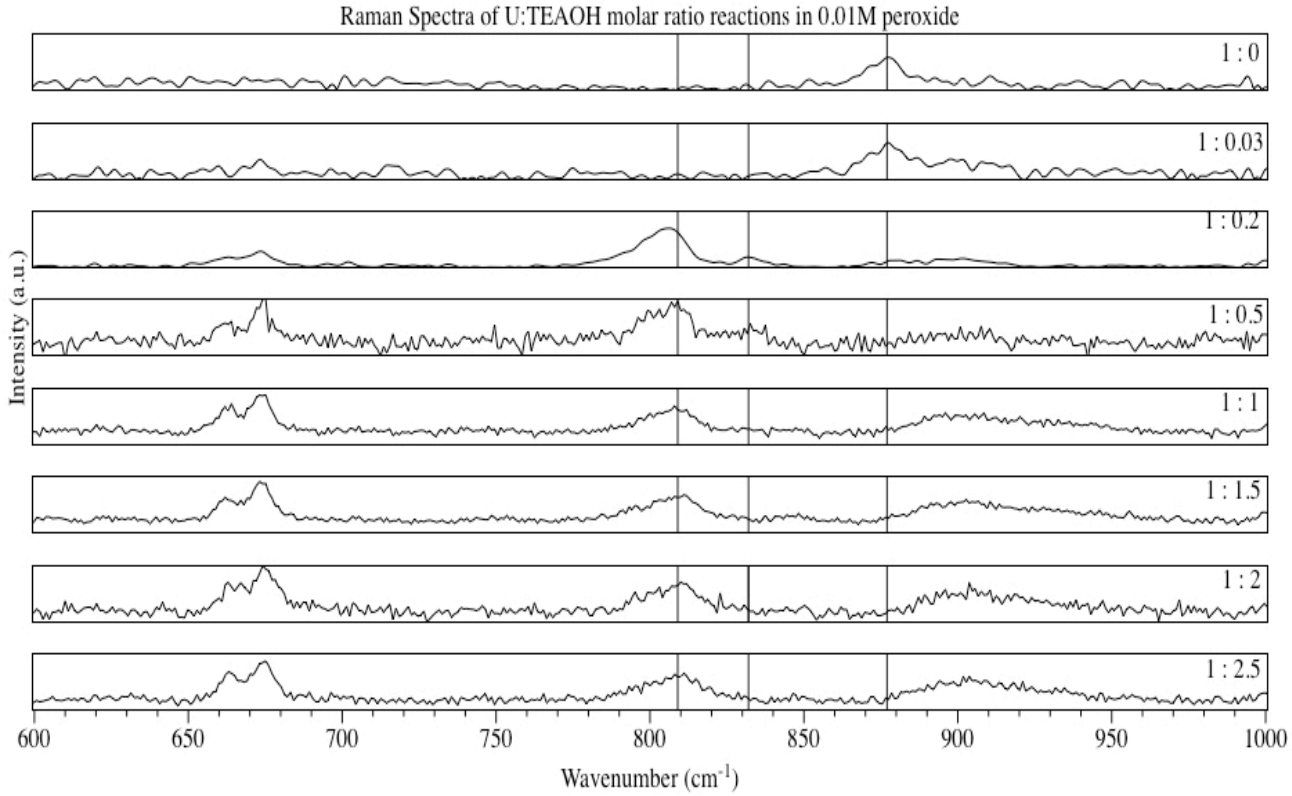

**Figure S27.** Raman spectra of solutions in U:TEAOH molar reactions in 0.035 M peroxide environments. Lines are marked at 808, 832, and 876 $cm^{-1}$.

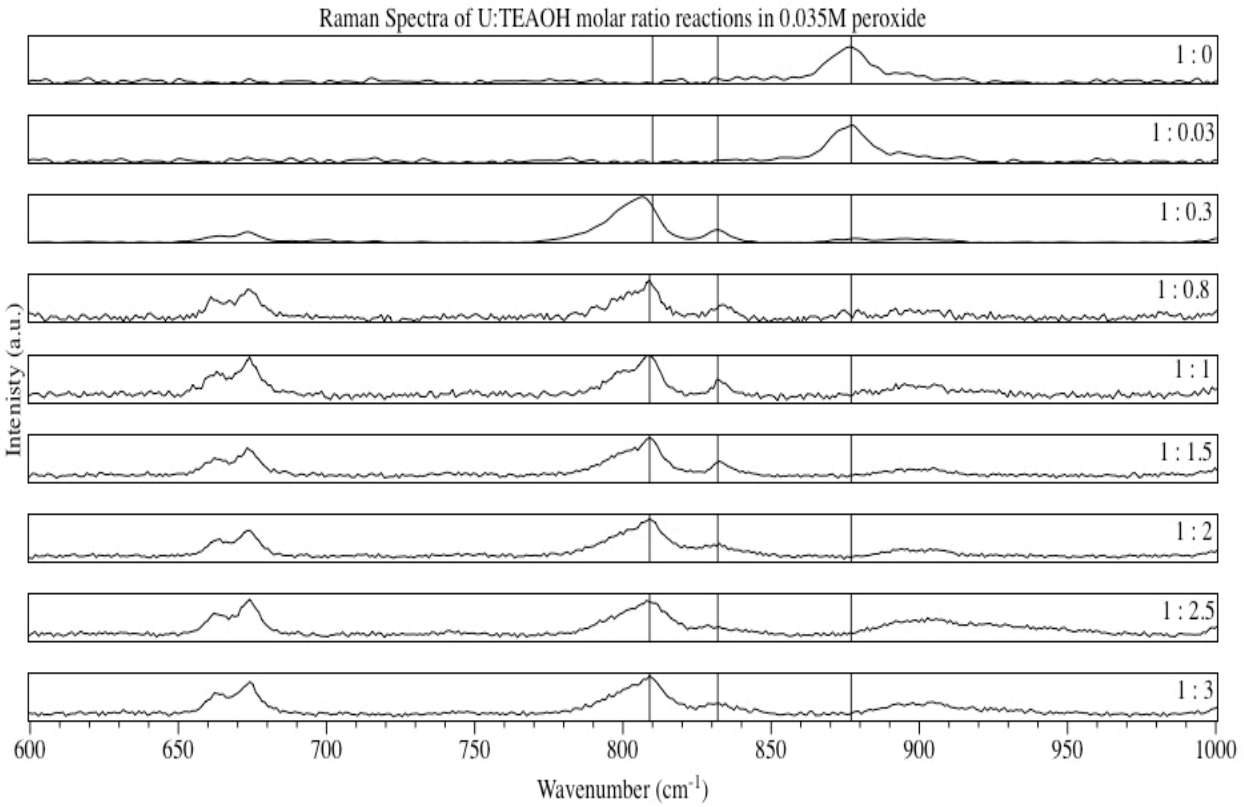

**Figure S28.** Raman spectra of solutions in U:TEAOH molar reactions in 0.10 M peroxide environments. Lines are marked at 808, 832, and 876 $cm^{-1}$.

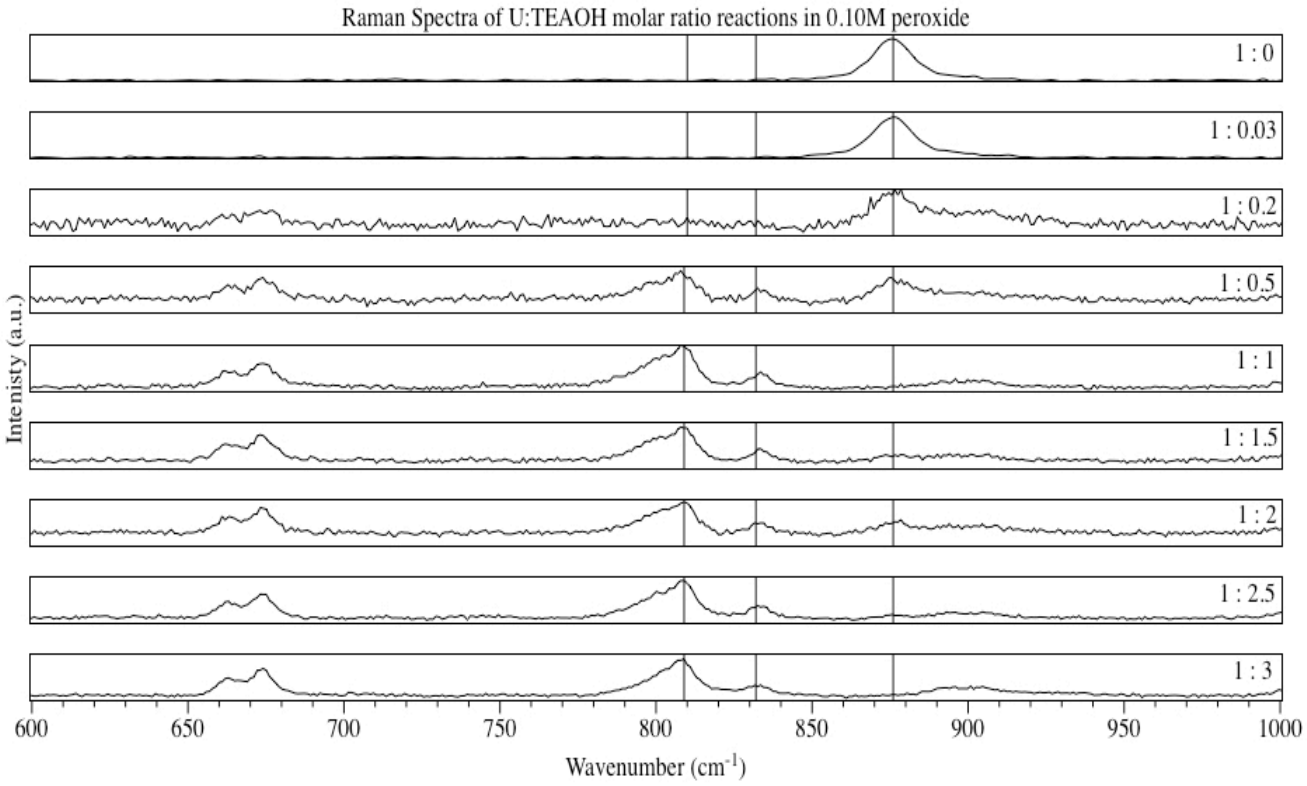

**Figure S29.** Raman spectra of solutions in U:TEAOH molar reactions in 0.50 M peroxide environments. Lines are marked at 808, 832, and 876 $cm^{-1}$.

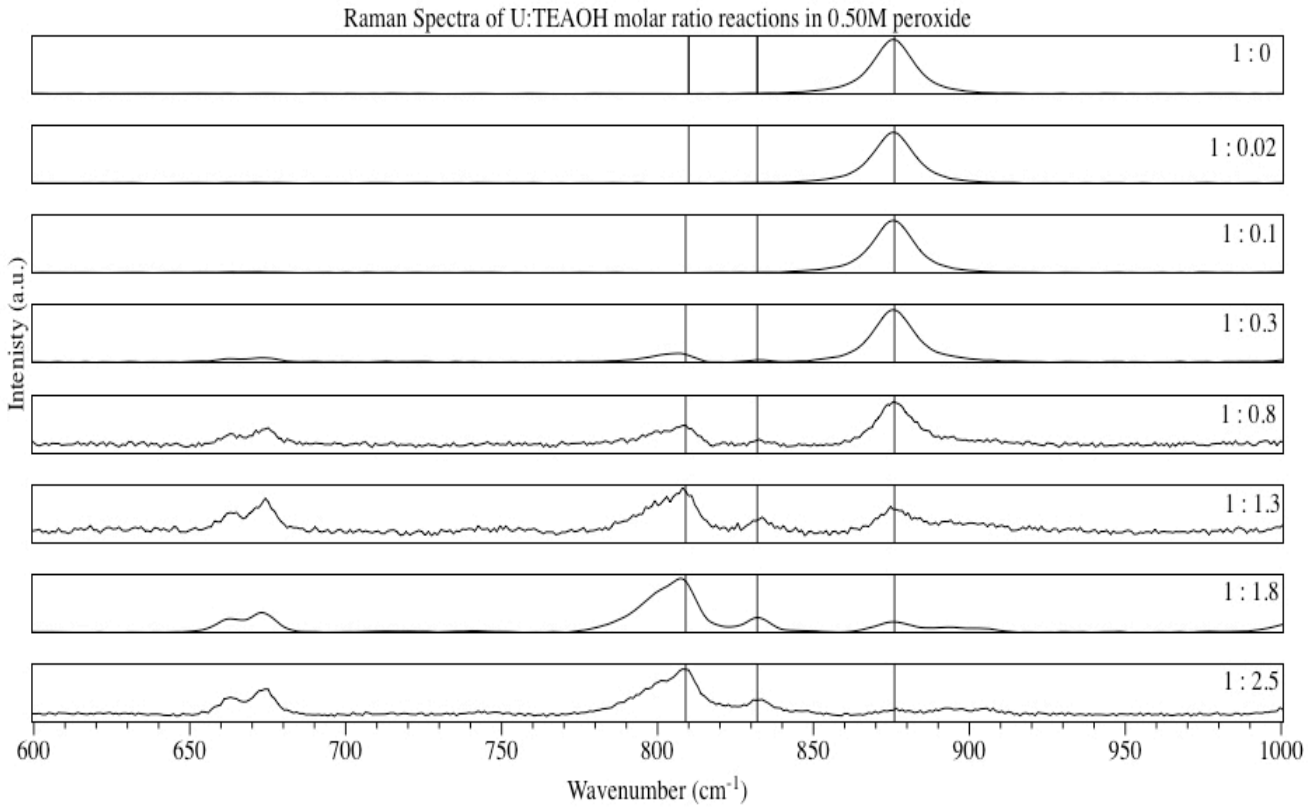

**Figure S30.** Raman spectra of solutions in U:TEAOH molar reactions in 1.00 M peroxide environments. Lines are marked at 808, 832, and 876 $cm^{-1}$.

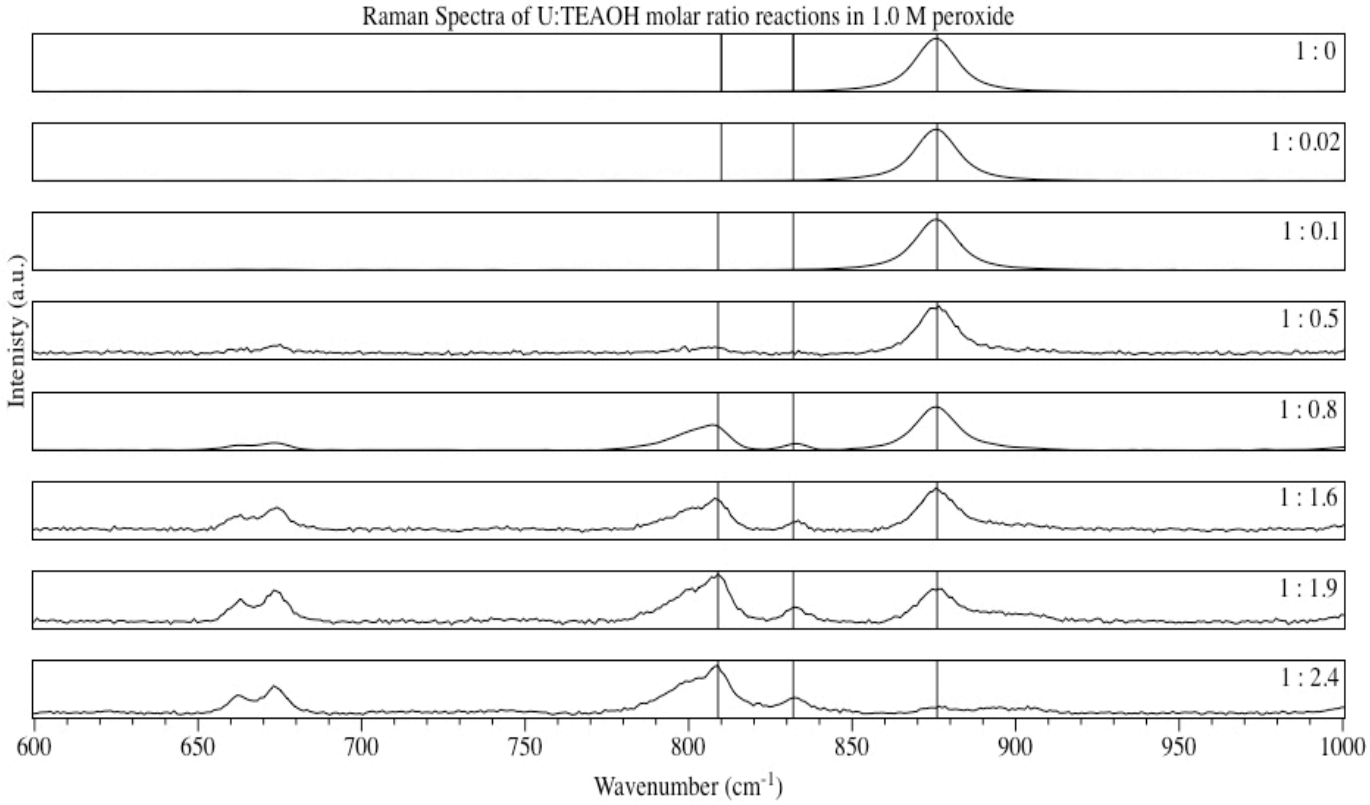

**Figure S31.** ESI-MS spectra of solutions in U:TEAOH molar reactions in 0.01 M peroxide environments. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

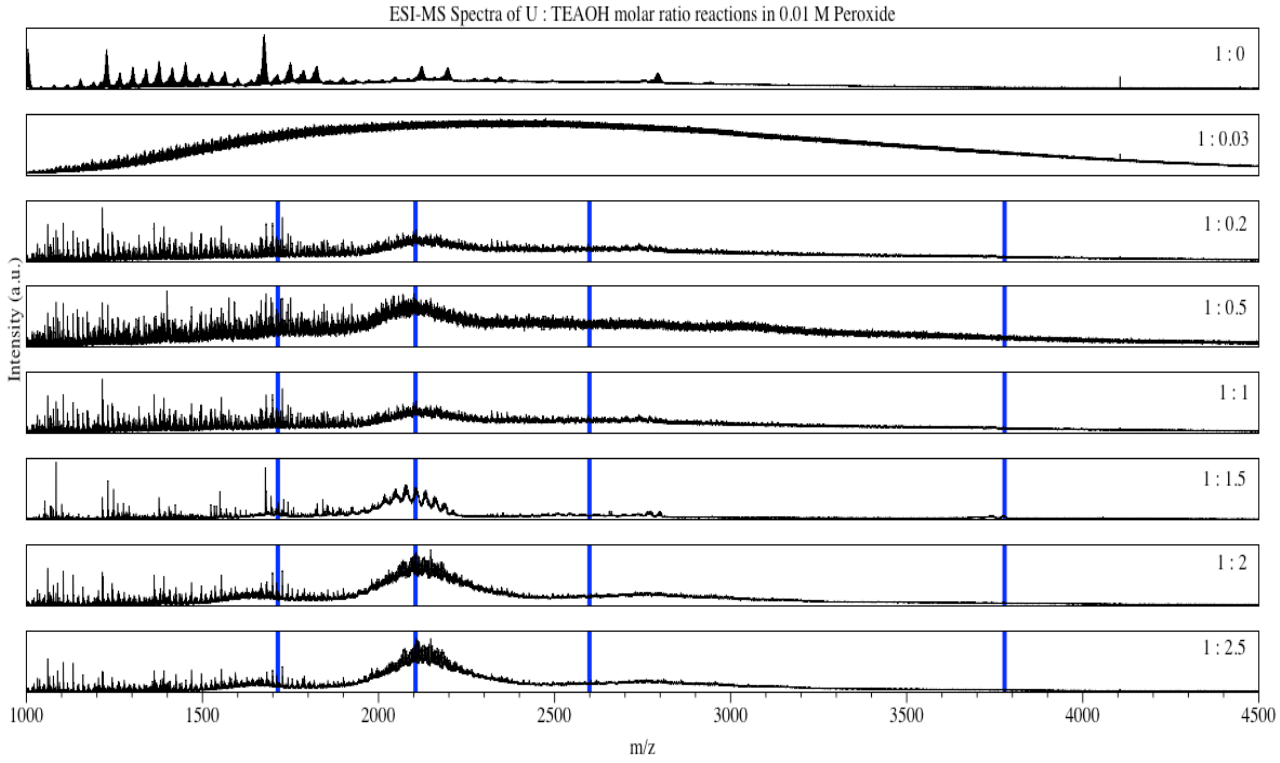

**Figure S32.** ESI-MS spectra of solutions in U:TEAOH molar reactions in 0.035 M peroxide environments. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

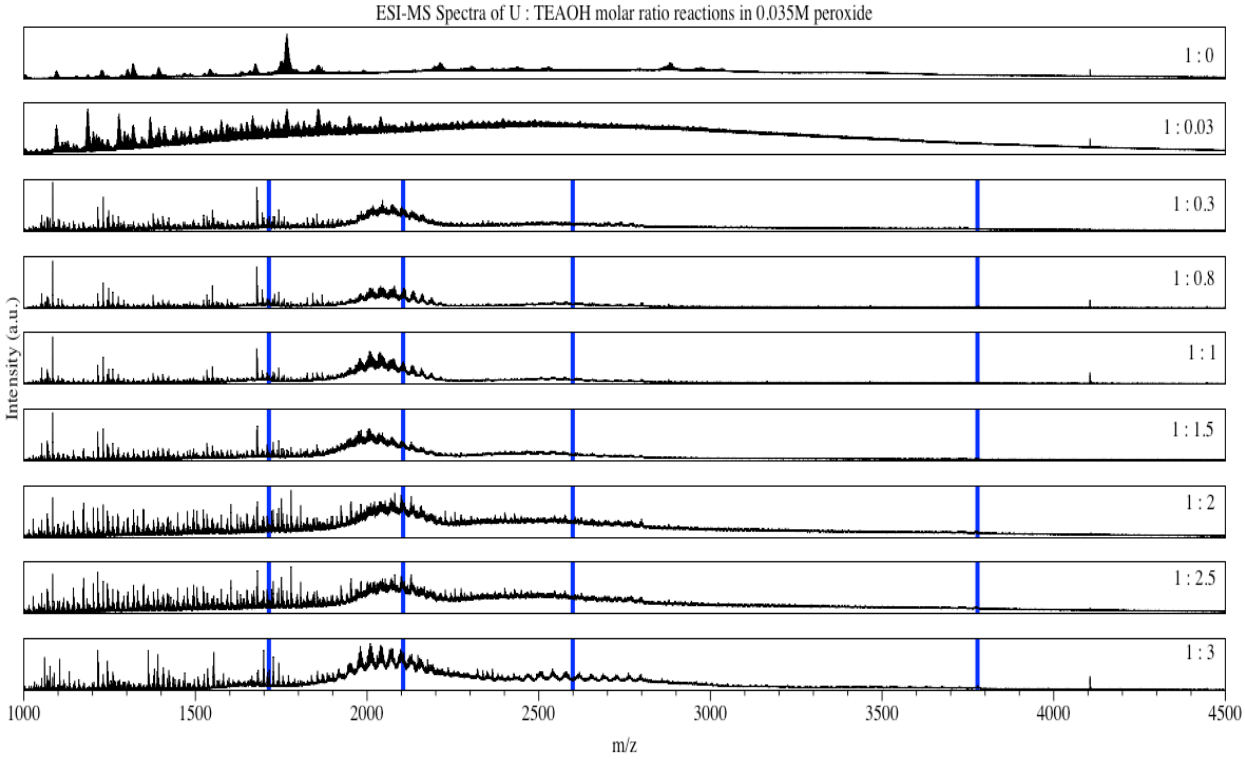

**Figure S33.** ESI-MS spectra of solutions in U:TEAOH molar reactions in 0.10 M peroxide environments. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

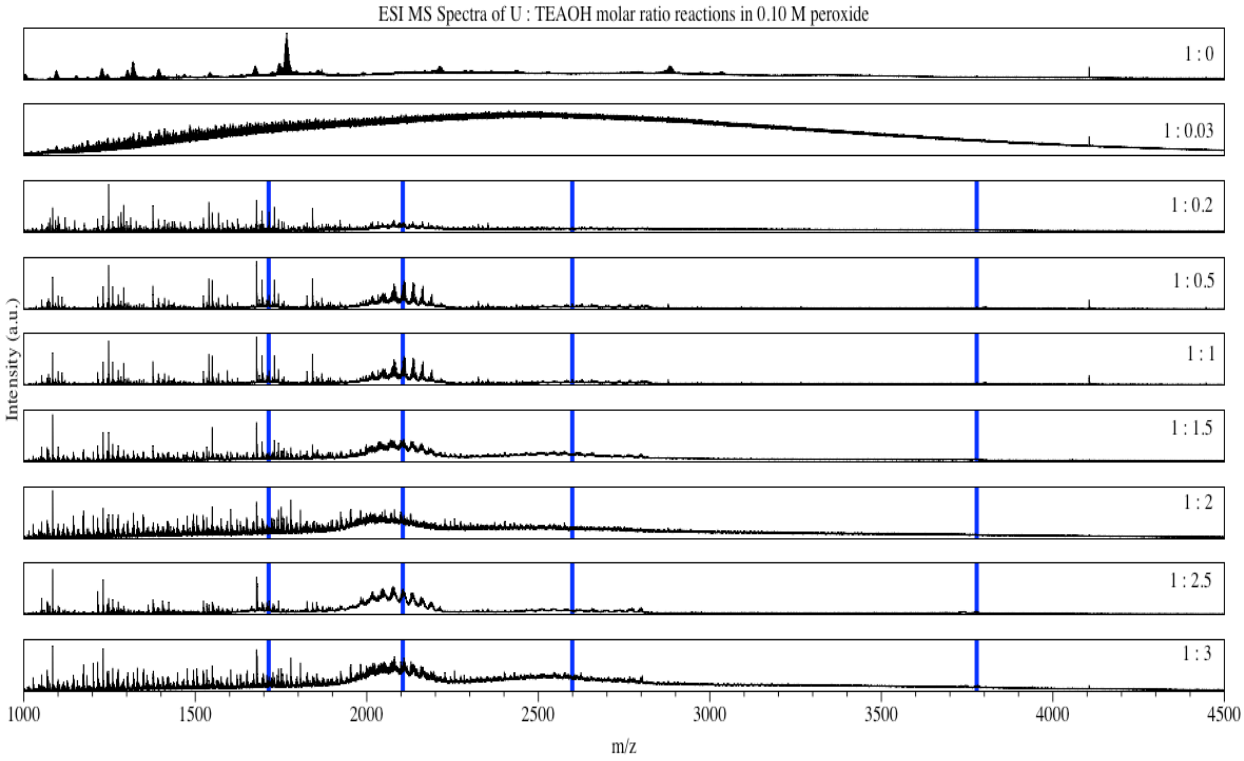

**Figure S34.** ESI-MS spectra of solutions in U:TEAOH molar reactions in 0.50 M peroxide environments. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

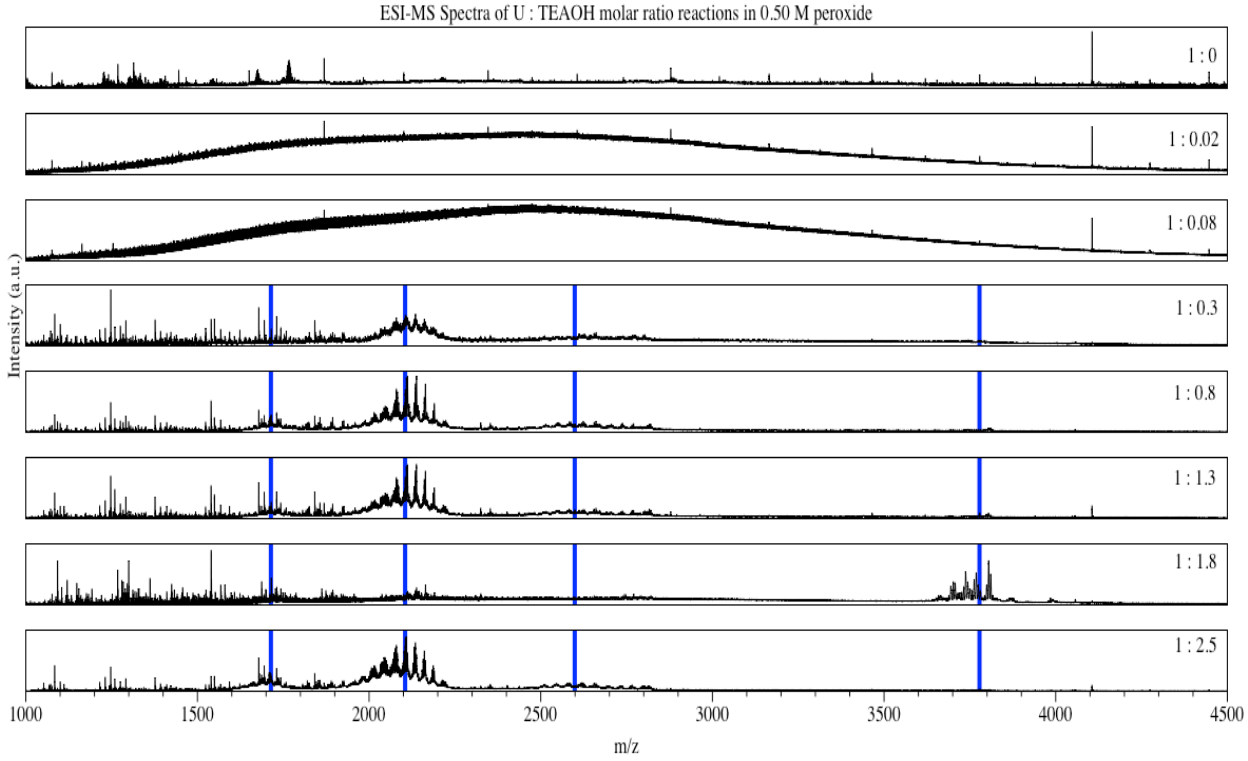

**Figure S35**. ESI-MS spectra of solutions in U:TEAOH molar reactions in 1.0 M peroxide environments. Blue lines are assigned charges of -6, -5, -4, and -3, respectively.

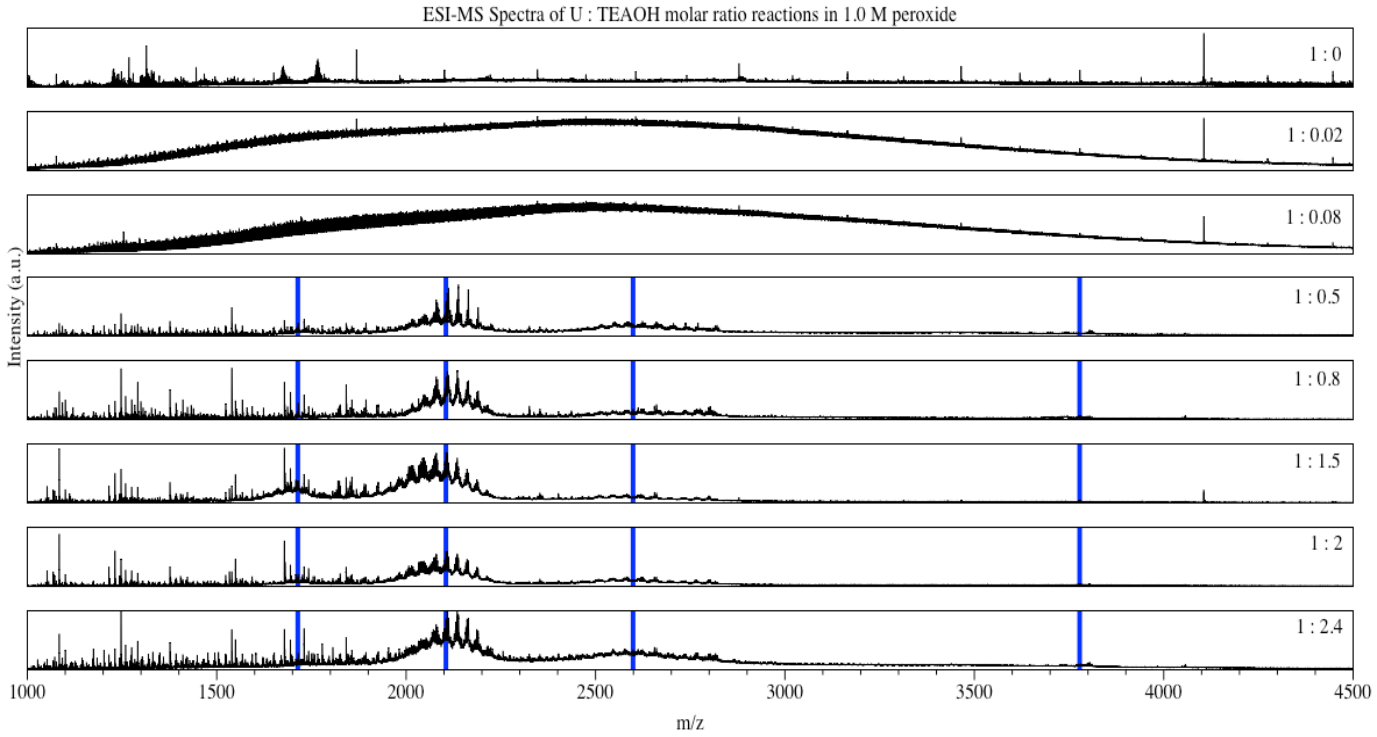

**Figure S36.** Raman spectrum of 40% tetraethylammonium hydroxide (TEAOH).