

S.IIa. Modeling Polycondensation in Methanolic Tetramethoxysilane Precursor Solution

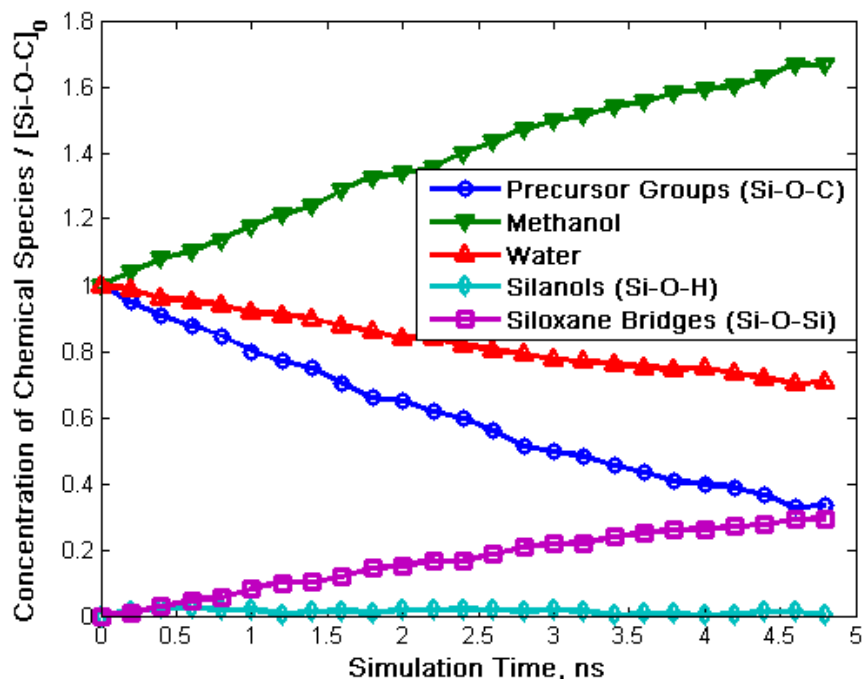


Figure S1: Concentration of chemical groups in system. The initial composition prior to reaction is a 1:4:4 molar ratio of TetraMS, methanol, and water. Concentrations are normalized by the initial number of alkoxy groups in the simulation.

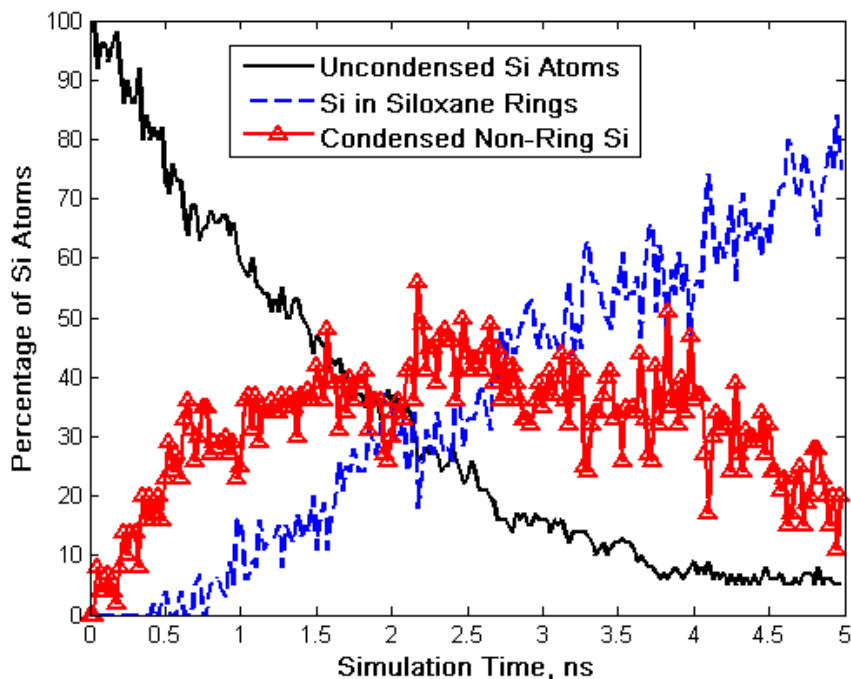


Figure S2: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:4:4 molar ratio of TetraMS, methanol, and water.

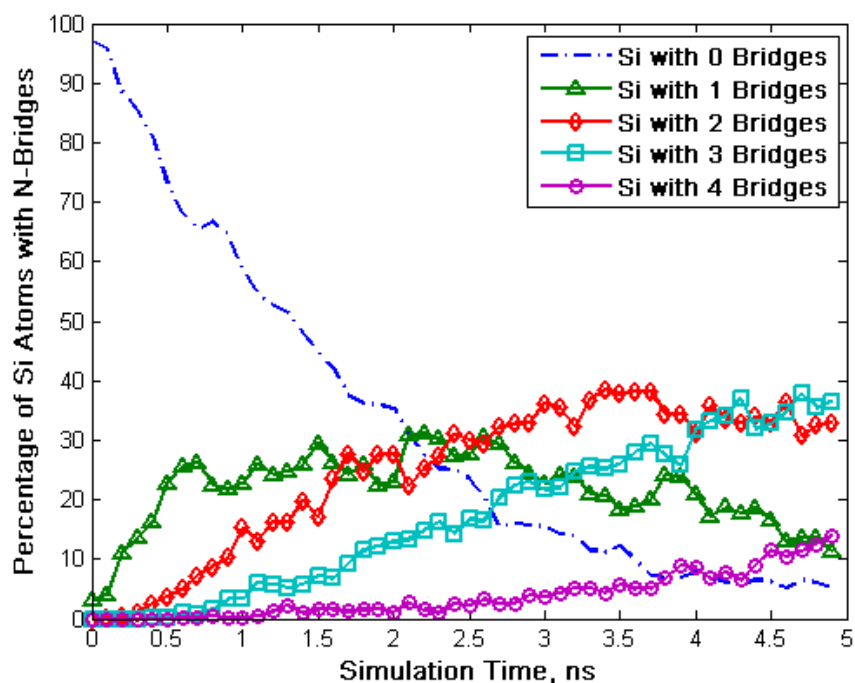


Figure S3: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:4:4 molar ratio of TetraMS, methanol, and water.

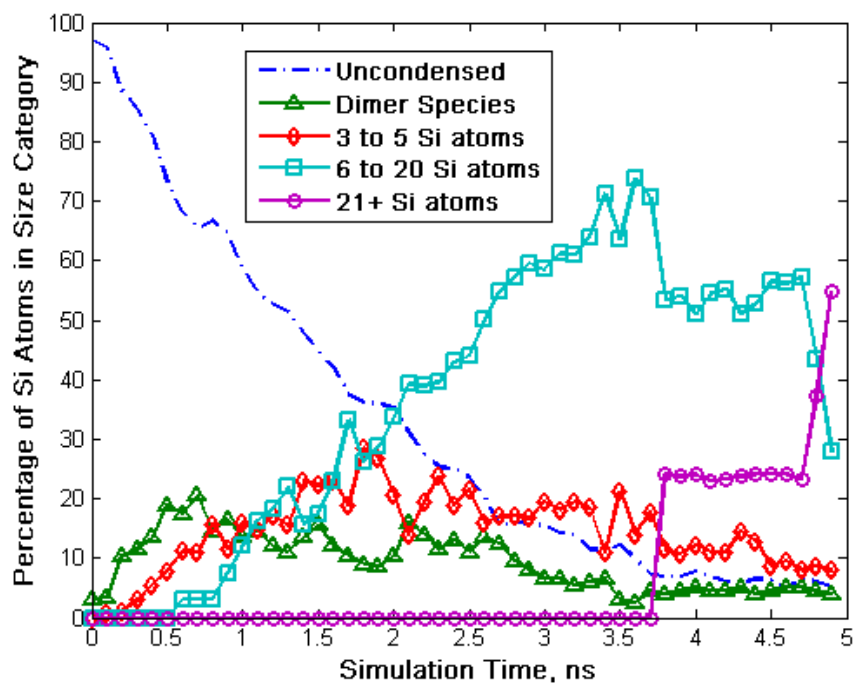


Figure S4: Siloxane cluster size distribution for a solution with an initial composition of a 1:4:4 molar ratio of TetraMS, methanol, and water.

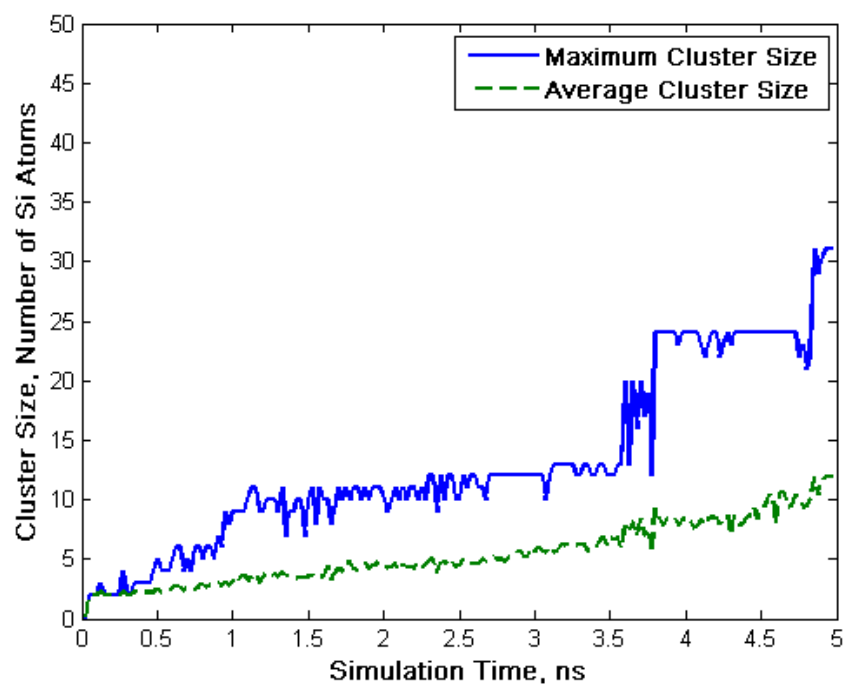


Figure S5: Maximum and average siloxane cluster size for a system containing one hundred tetramethoxysilane molecules. The initial composition of the solution is a 1:4:4 molar ratio of TetraMS, methanol, and water.

S.IIb. Effects of Alkoxysilane Chemical Structure: Silyl Headgroups and Alkoxy Ligands

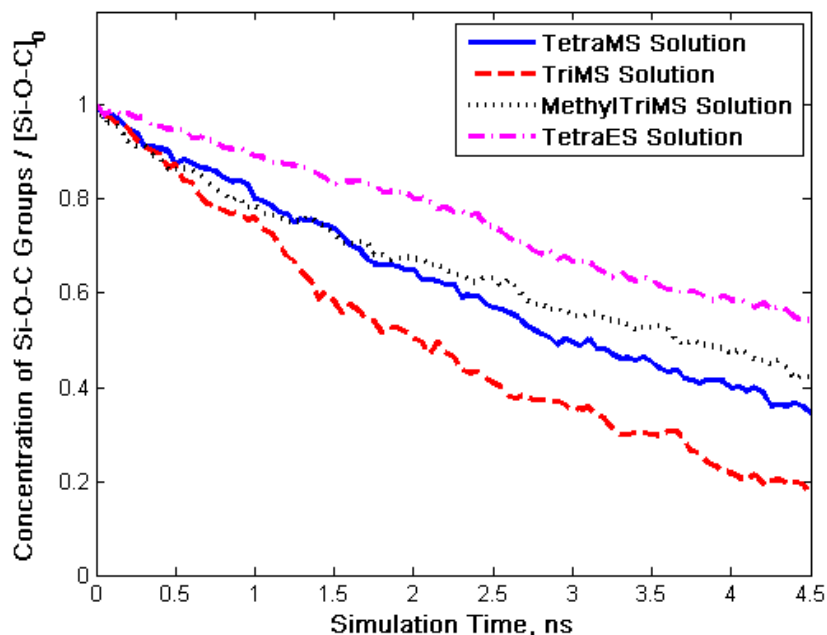


Figure S6: Normalized concentration of alkoxy groups (Si-O-C) in system. Four precursor molecules, TetraMS, TriMS, MethylTriMS and TetraES, with the same initial molar ratio of 1:4:4 precursor:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

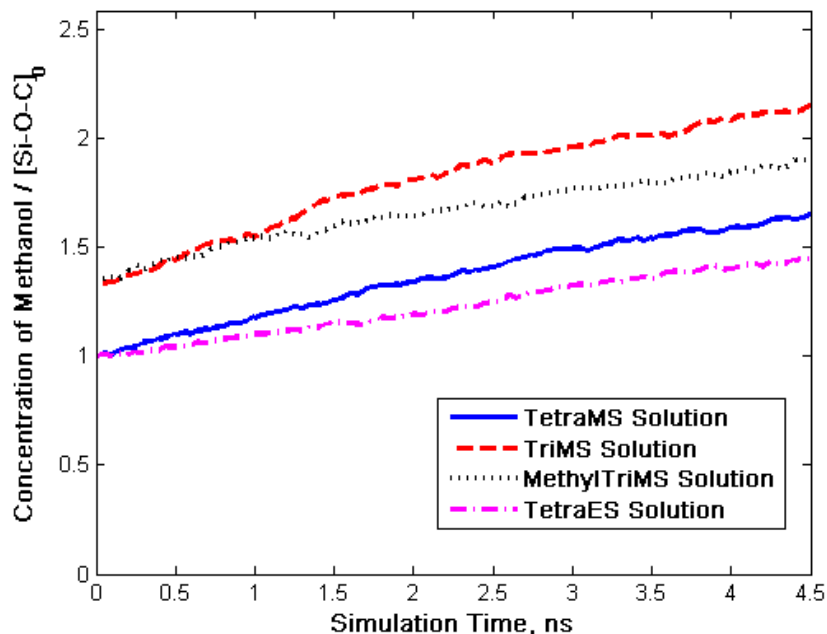


Figure S7: Normalized concentration of methanol in system. Four precursor molecules, TetraMS, TriMS, MethylTriMS and TetraES, with the same initial molar ratio of 1:4:4 precursor:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

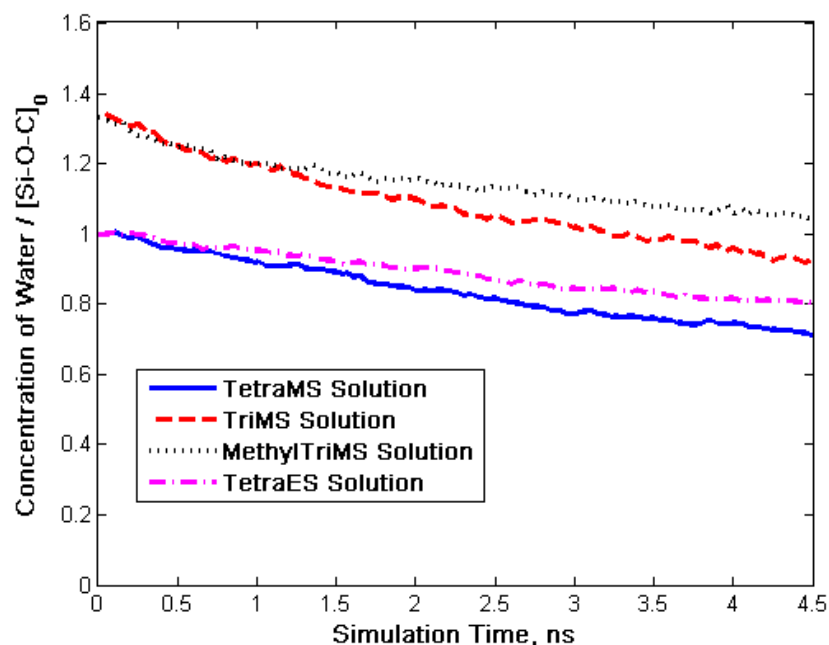


Figure S8: Normalized concentration of water in system. Four precursor molecules, TetraMS, TriMS, MethylTriMS and TetraES, with the same initial molar ratio of 1:4:4 precursor:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy silane groups in each simulation.

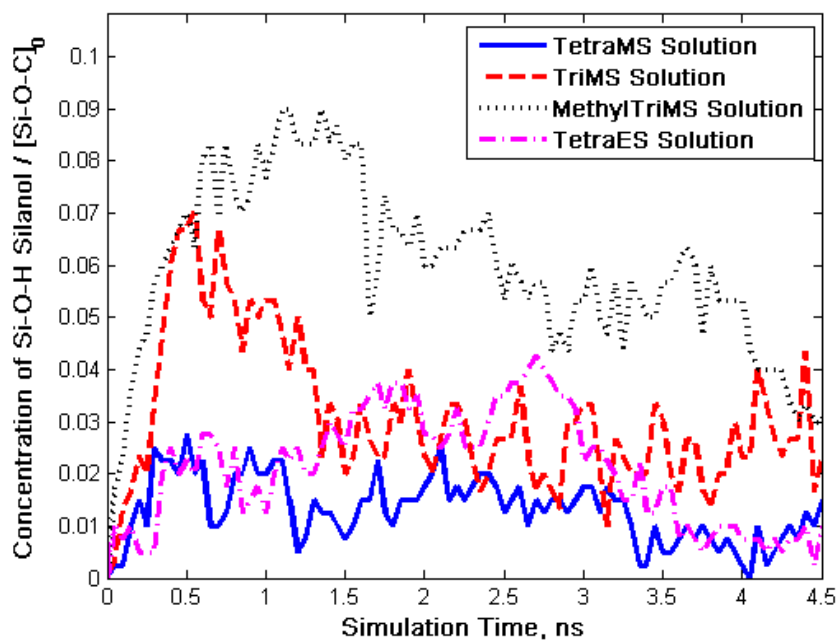


Figure S9: Normalized concentration of silanol (Si-O-H) in system. Four precursor molecules, TetraMS, TriMS, MethylTriMS and TetraES, with the same initial molar ratio of 1:4:4 precursor:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy silane groups in each simulation.

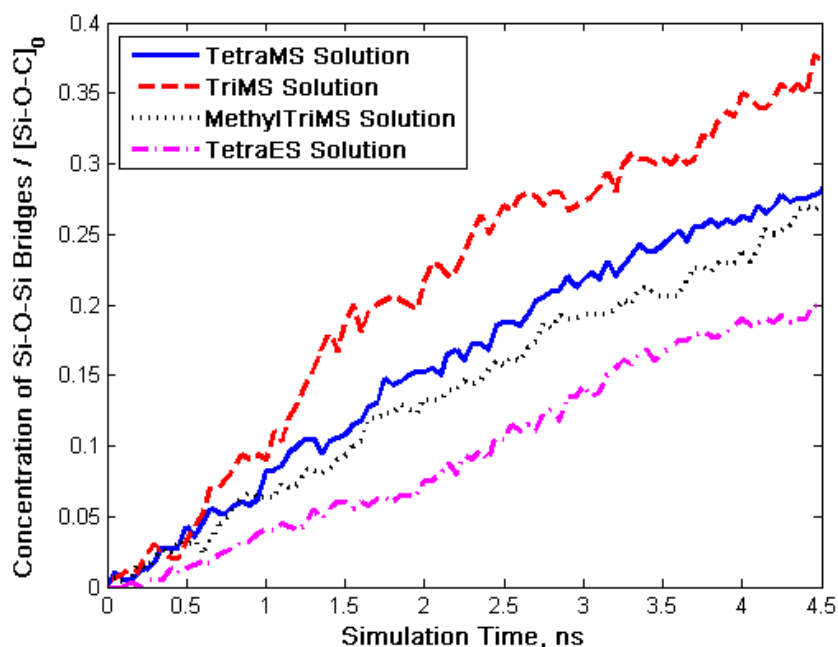


Figure S10: Normalized concentration of siloxane bridges (Si-O-Si) in system. Four precursor molecules, TetraMS, TriMS, MethylTriMS and TetraES, with the same initial molar ratio of 1:4:4 precursor:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy silane groups in each simulation.

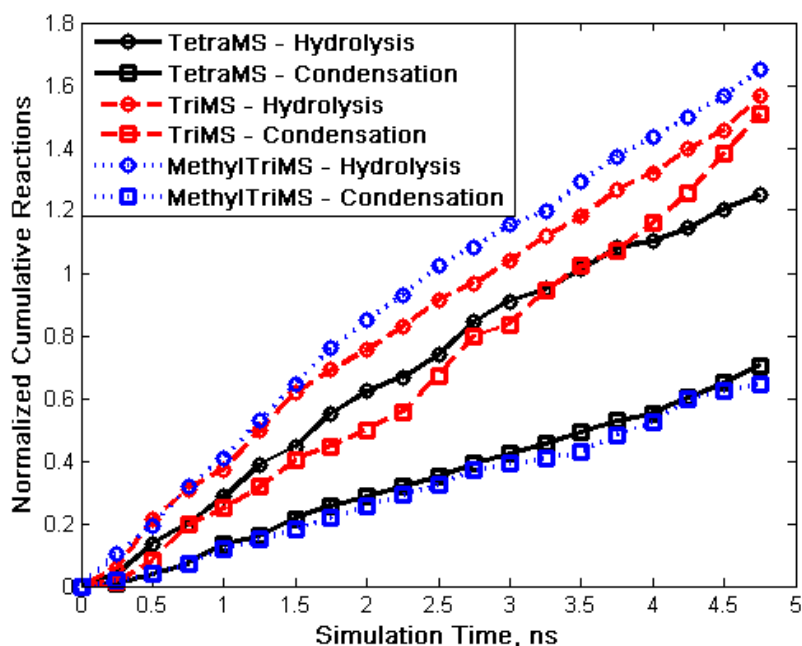


Figure S11: Normalized cumulative hydrolysis and condensation. Three precursor molecules, TetraMS, TriMS, and MethylTriMS, with the same initial molar ratio of 1:4:4 precursor:methanol:water were modeled. Number of reactions are normalized by the initial number of alkoxy silane groups in each simulation.

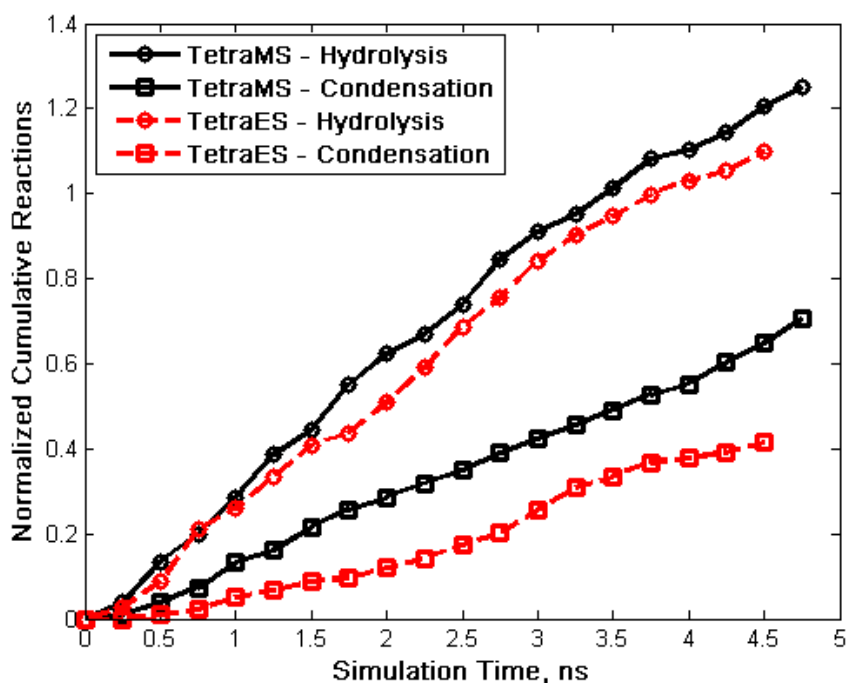


Figure S12: Normalized cumulative hydrolysis and condensation. Two precursor molecules, TetraMS, and TetraES, with the same initial molar ratio of 1:4:4 precursor:methanol:water were modeled. Number of reactions are normalized by the initial number of alkoxysilane groups in each simulation.

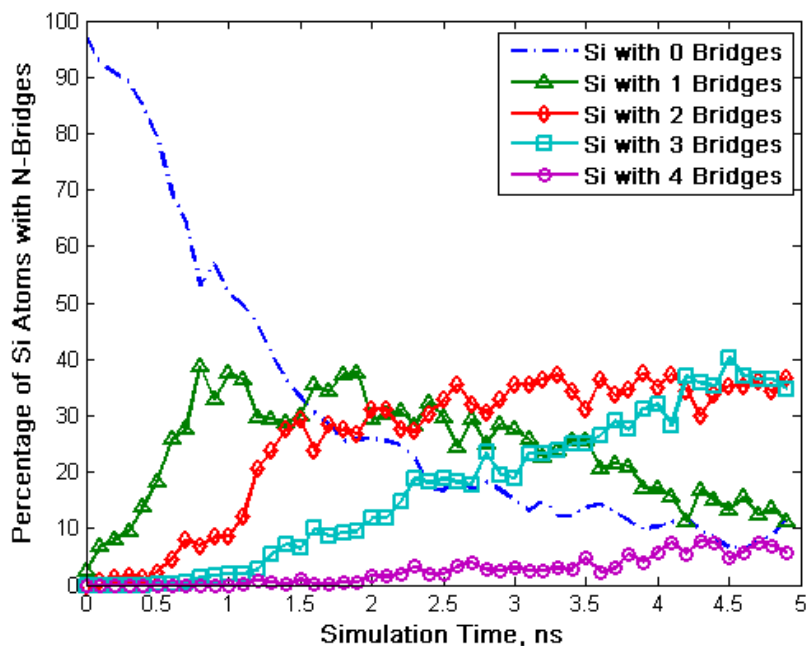


Figure S13: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:4:4 molar ratio of TriMS, methanol, and water.

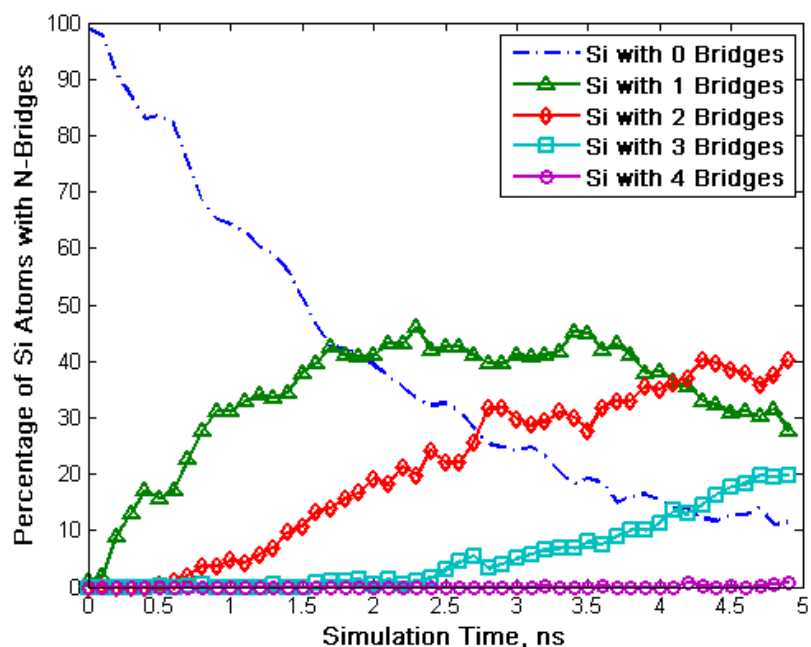


Figure S14: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:4:4 molar ratio of MethylTriMS, methanol, and water.

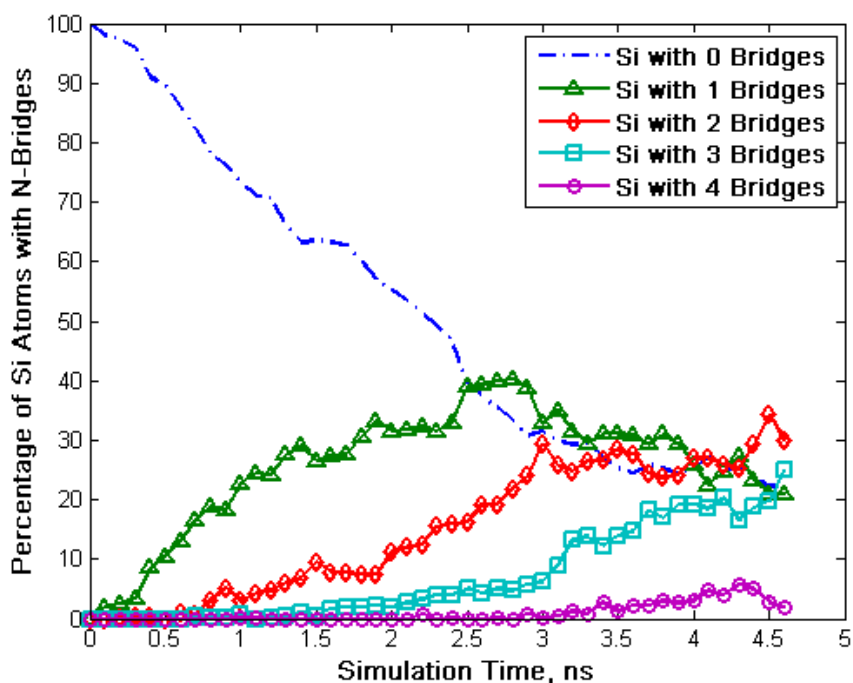


Figure S15: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:4:4 molar ratio of TetraES, methanol, and water.

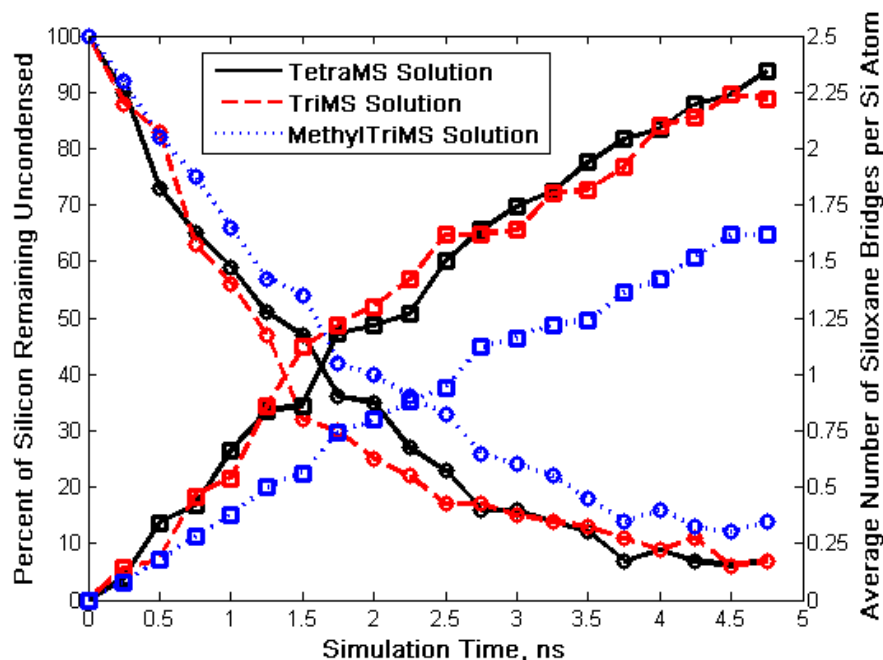


Figure S16: Percent of silicon atoms remaining uncondensed compared with the average number of siloxane Si-O-Si bridges per silicon atom. Three systems with an initial composition of a 1:4:4 molar ratio of precursor, methanol, and water. The three precursor molecules compared are TetraMS, TriMS, and MethylTriMS.

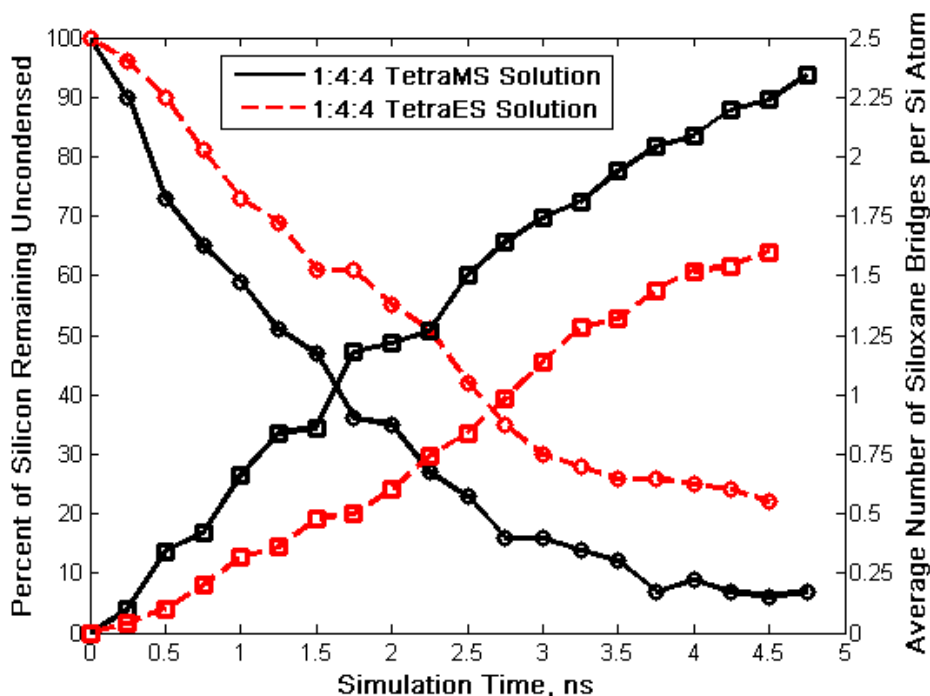


Figure S17: Percent of silicon atoms remaining uncondensed compared with the average number of siloxane Si-O-Si bridges per silicon atom. Two systems with an initial composition of a 1:4:4 molar ratio of precursor, methanol, and water. The two precursor molecules compared are TetraMS, and TetraES.

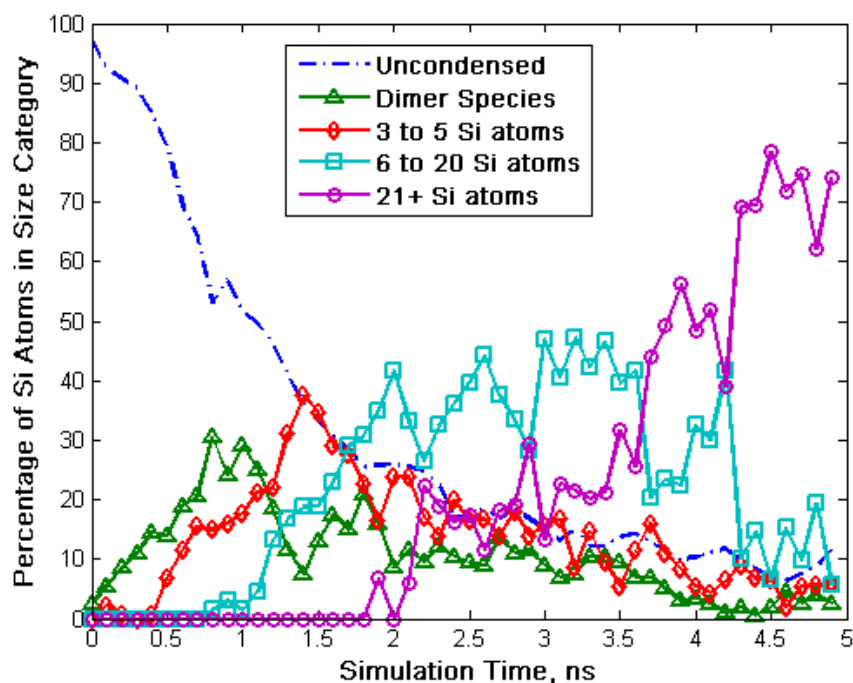


Figure S18: Siloxane cluster size distribution for a solution with an initial composition of a 1:4:4 molar ratio of TriMS, methanol, and water.

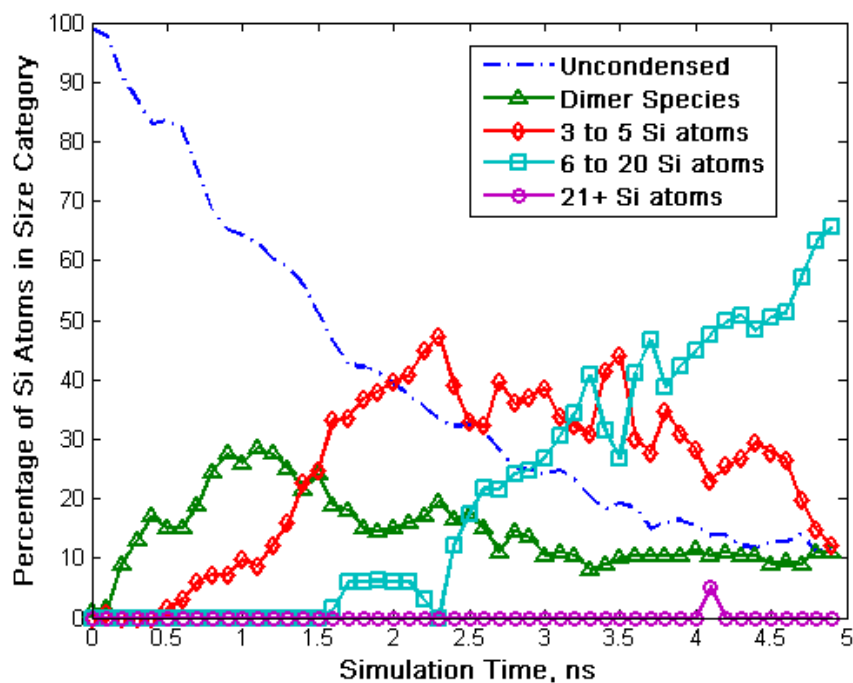


Figure S19: Siloxane cluster size distribution for a solution with an initial composition of a 1:4:4 molar ratio of MethylTriMS, methanol, and water.

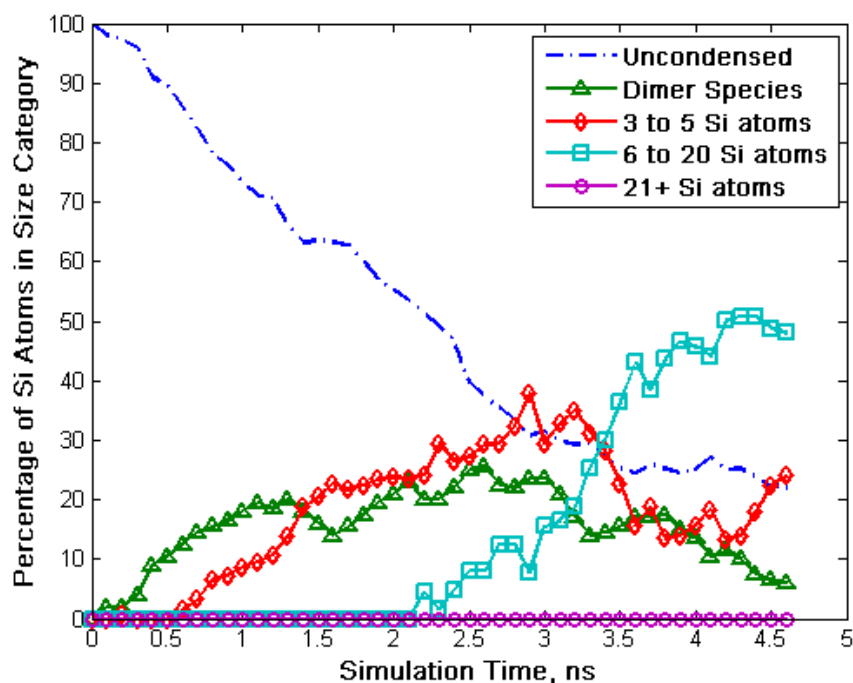


Figure S20: Siloxane cluster size distribution for a solution with an initial composition of a 1:4:4 molar ratio of TetraES, methanol, and water.

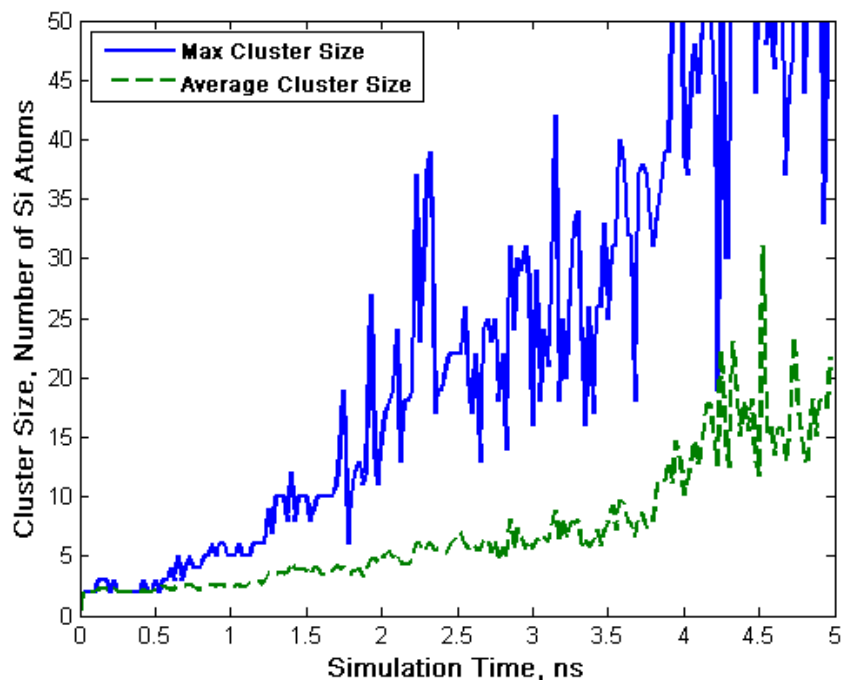


Figure S21: Maximum and average siloxane cluster size for a system containing one hundred trimethoxysilane molecules. The initial composition of the solution is a 1:4:4 molar ratio of TriMS, methanol, and water.

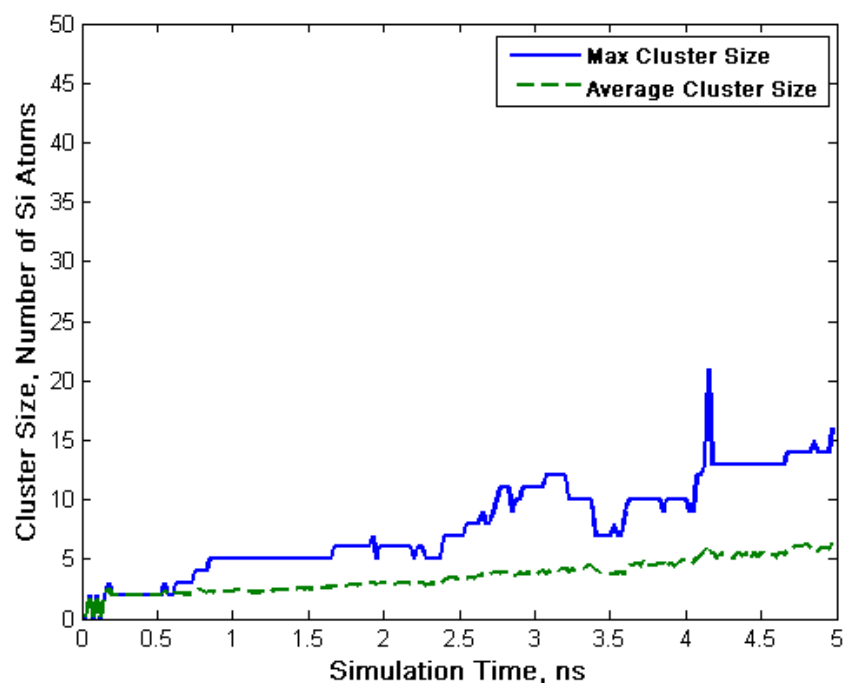


Figure S22: Maximum and average siloxane cluster size for a system containing one hundred methyltrimethoxysilane molecules. The initial composition of the solution is a 1:4:4 molar ratio of MethylTriMS, methanol, and water.

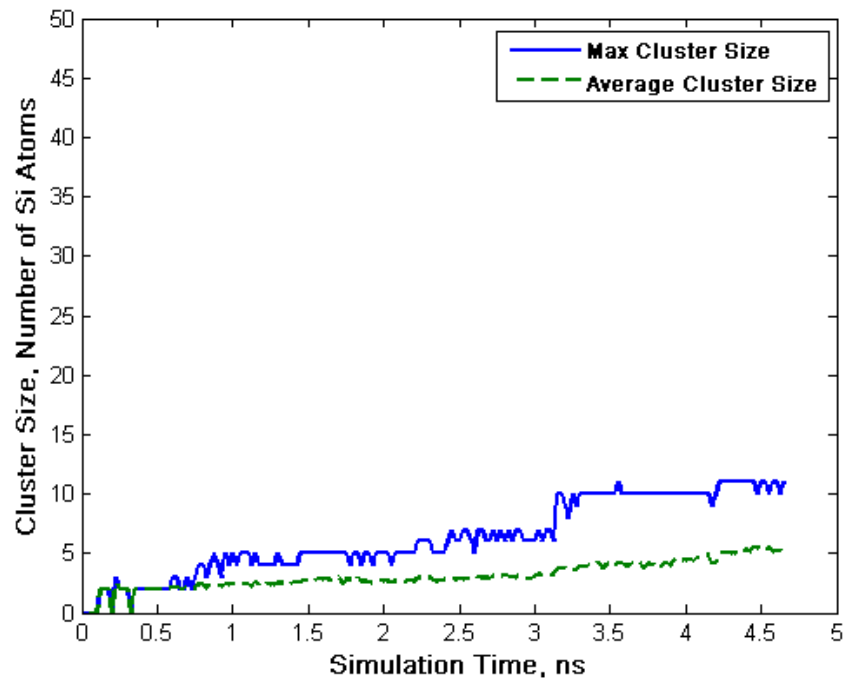


Figure S23: Maximum and average siloxane cluster size for a system containing one hundred tetraethoxysilane molecules. The initial composition of the solution is a 1:4:4 molar ratio of TetraES, methanol, and water.

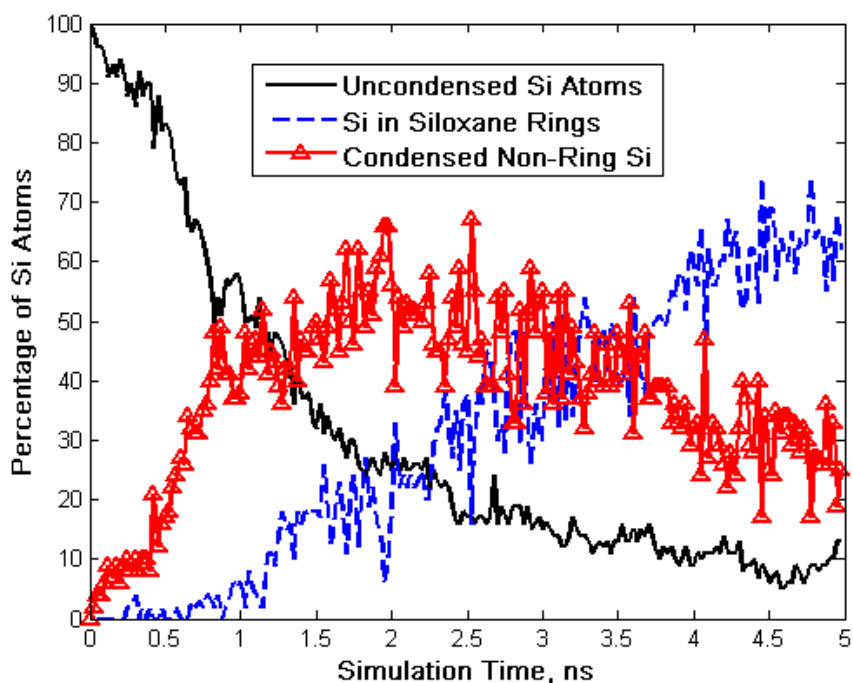


Figure S24: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:4:4 molar ratio of TriMS, methanol, and water.

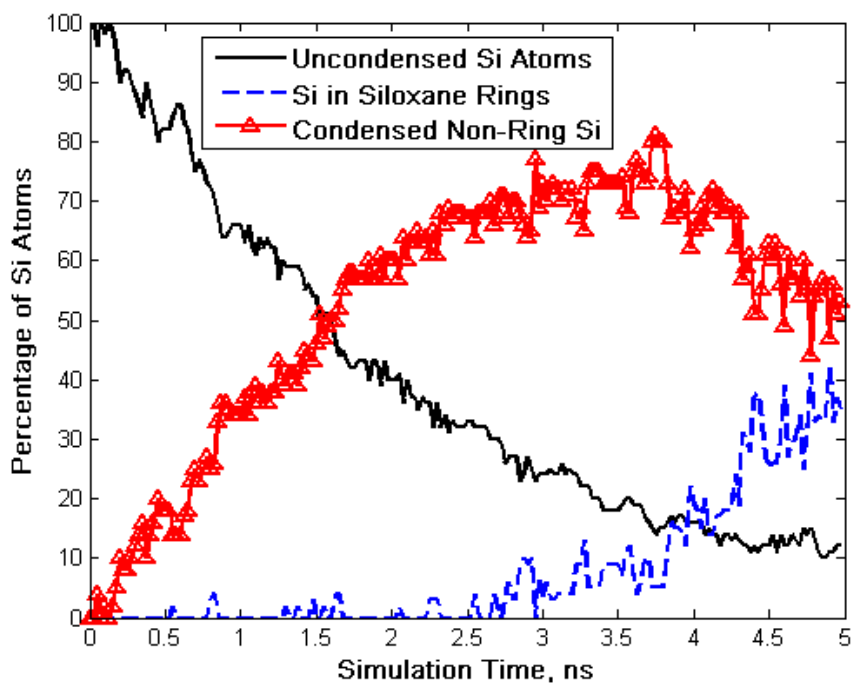


Figure S25: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:4:4 molar ratio of MethylTriMS, methanol, and water.

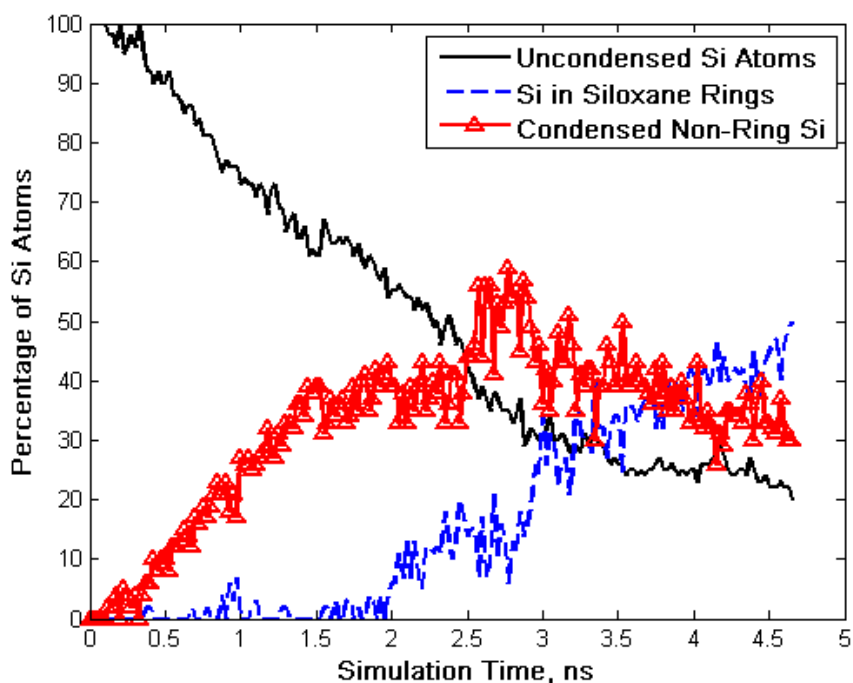


Figure S26: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:4:4 molar ratio of TetraES, methanol, and water.

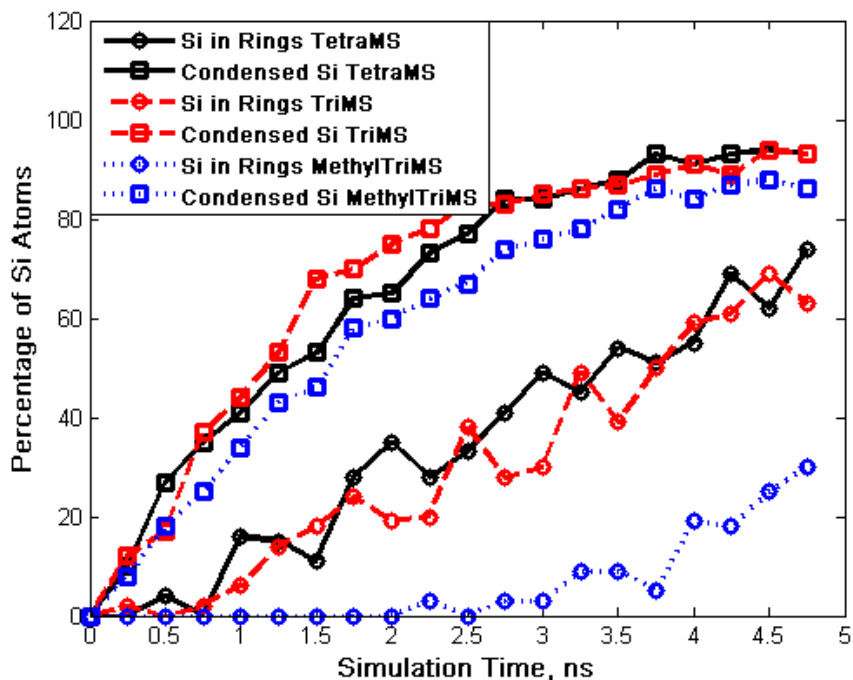


Figure S27: Percent of silicon atoms condensed compared with the percent in siloxane rings. Three systems with an initial composition of a 1:4:4 molar ratio of precursor, methanol, and water. The three precursor molecules compared are TetraMS, TriMS, and MethylTriMS.

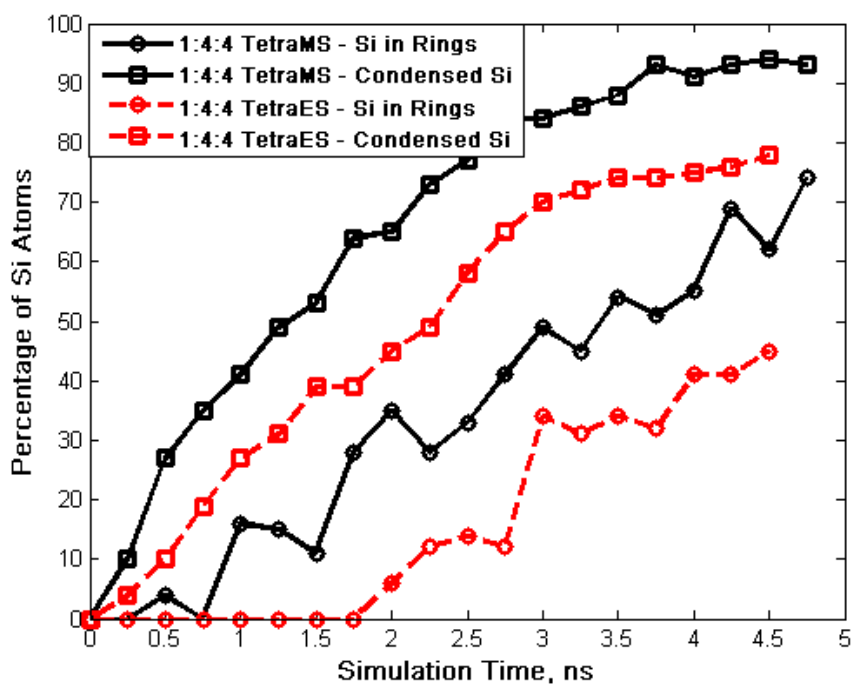


Figure S28: Percent of silicon atoms condensed compared with the percent in siloxane rings. Two systems with an initial composition of a 1:4:4 molar ratio of precursor, methanol, and water. The two precursor molecules compared are TetraMS and TetraES

IIc. Effects of Alkoxysilane, Water, and Alcohol Composition in Solution

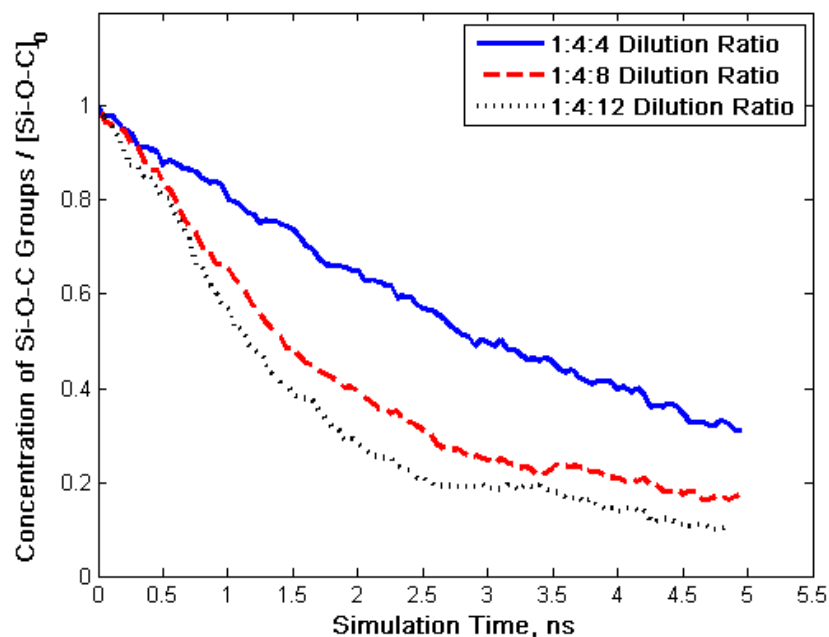


Figure S29: Concentration of alkoxysilanes (Si-O-C) in system. Molar ratios of 1:4:4, 1:4:8, and 1:4:12 tetramethoxysilane:methanol:water modeled. Concentration normalized by initial number of Si-O-C groups.

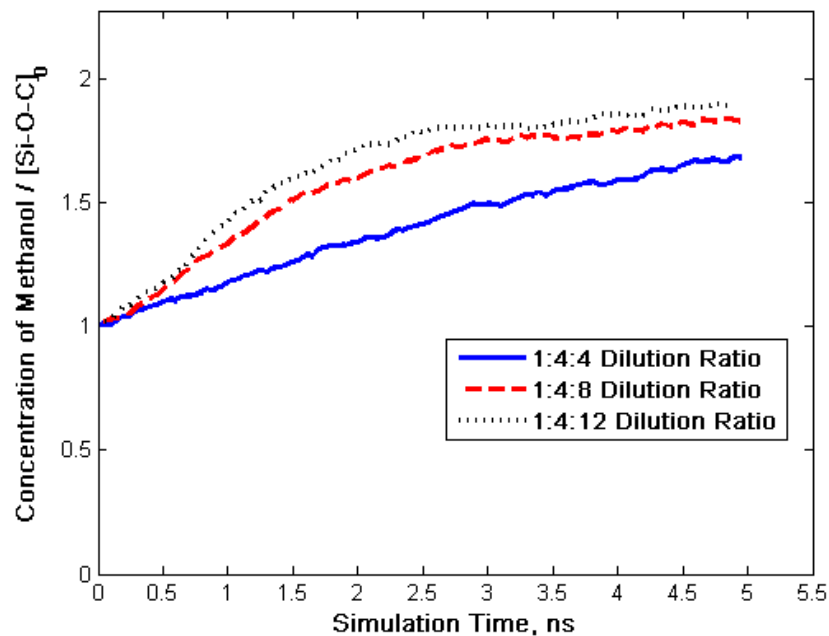


Figure S30: Concentration of methanol in system. Molar ratios of 1:4:4, 1:4:8, and 1:4:12 tetramethoxysilane:methanol:water modeled. Concentration normalized by initial number of Si-O-C groups.

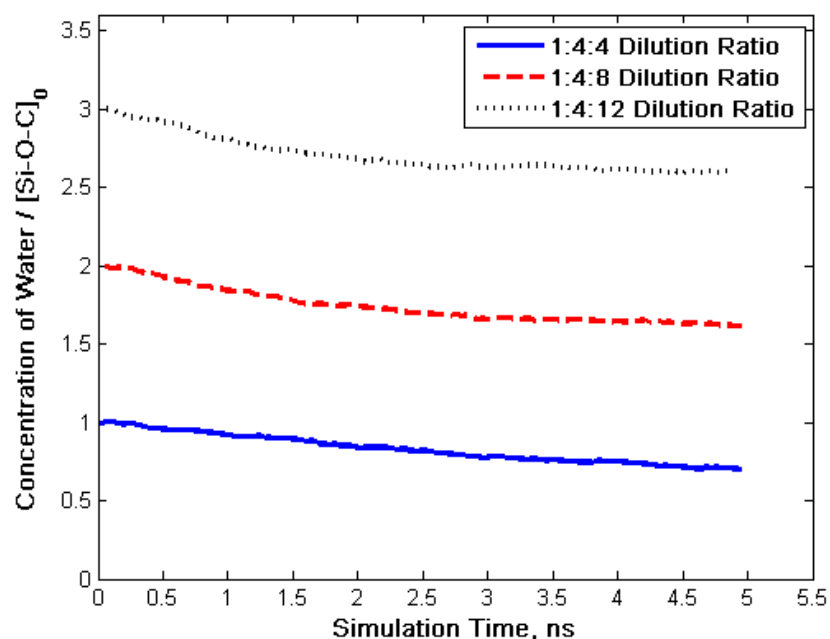


Figure S31: Normalized concentration of water in system. Three molar ratios of 1:4:4, 1:4:8, and 1:4:12 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy silane groups in each simulation.

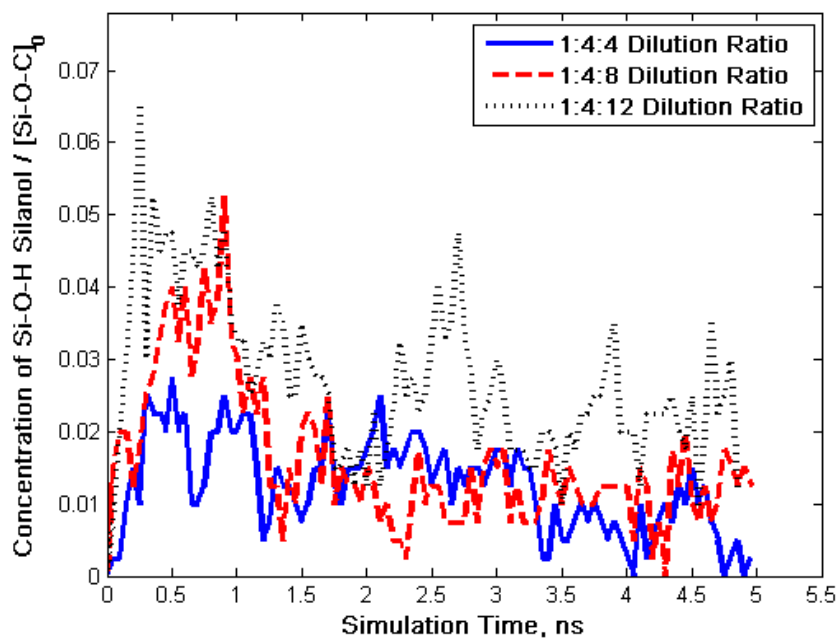


Figure S32: Normalized concentration of silanol (Si-O-H) in system. Three molar ratios of 1:4:4, 1:4:8, and 1:4:12 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy silane groups in each simulation.

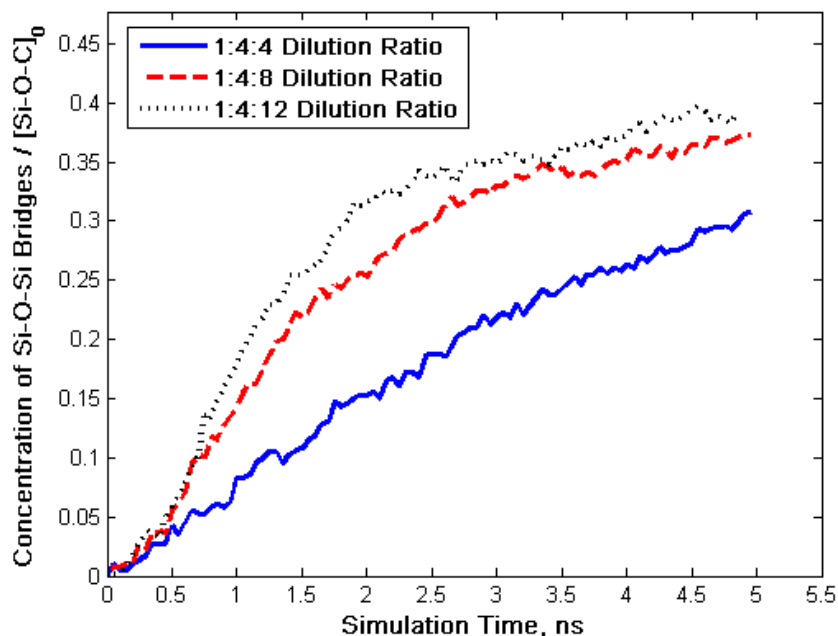


Figure S33: Normalized concentration of siloxane bridges (Si-O-Si) in system. Three molar ratios of 1:4:4, 1:4:8, and 1:4:12 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy silane groups in each simulation.

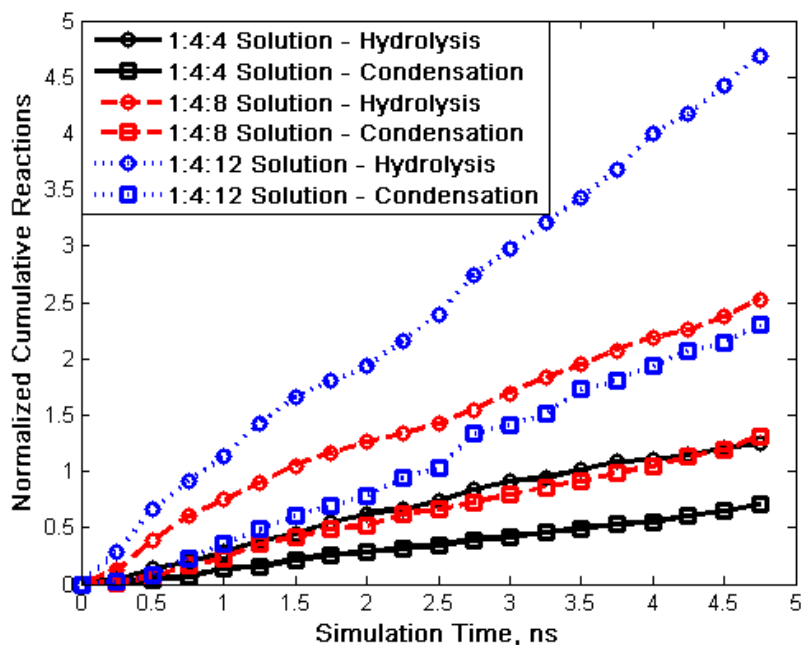


Figure S34: Normalized cumulative hydrolysis and condensation reactions. Three molar ratios of 1:4:4, 1:4:8, and 1:4:12 tetramethoxysilane:methanol:water were modeled. Number of reactions are normalized by initial number of alkoxy silane groups in the simulation.

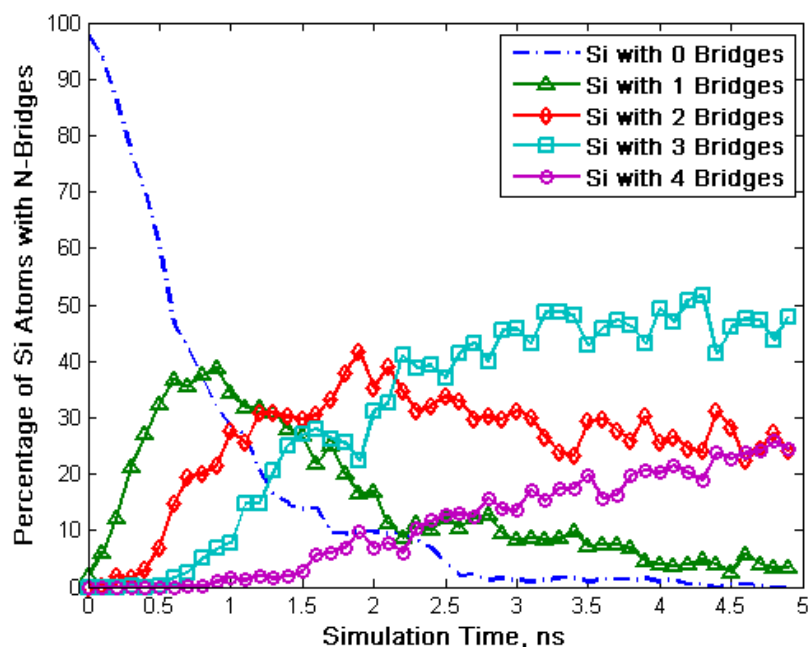


Figure S35: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:4:8 molar ratio of TetraMS, methanol, and water.

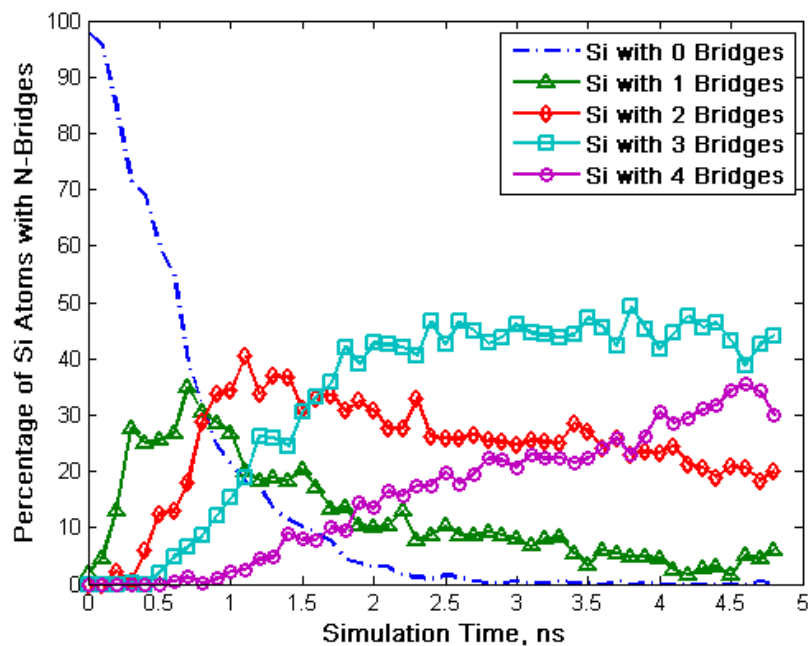


Figure S36: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:4:12 molar ratio of TetraMS, methanol, and water.

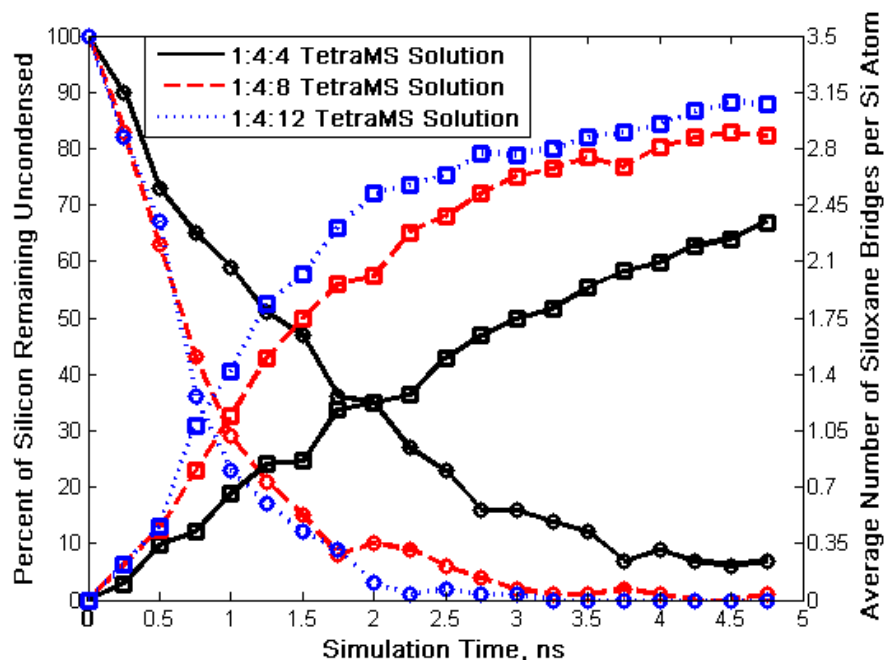


Figure S37: Percent of silicon atoms remaining uncondensed compared with the average number of siloxane Si-O-Si bridges per silicon atom. Three systems are modeled with different ratios of TetraMS:methanol:water. The three systems compared here have different water concentrations at 1:4:4, 1:4:8, and 1:4:12.

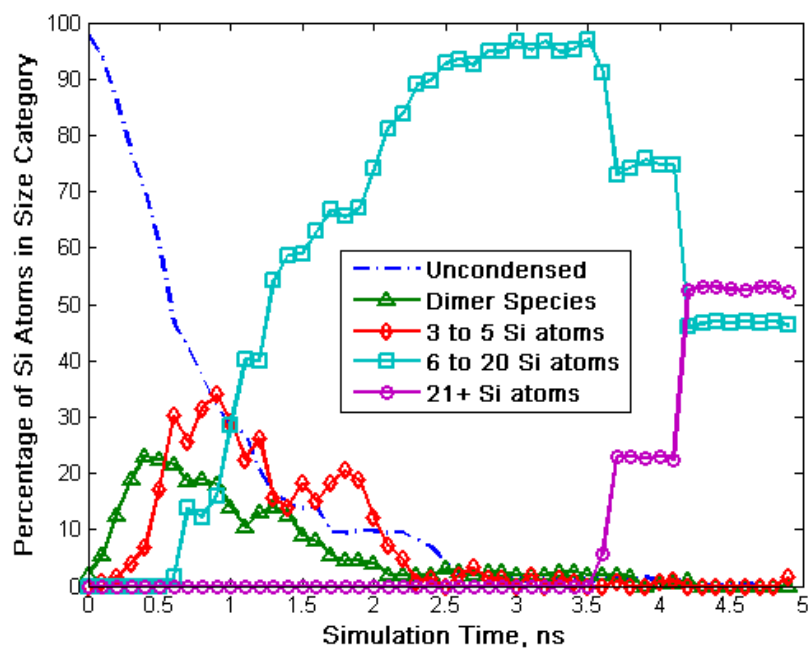


Figure S38: Siloxane cluster size distribution for a solution with an initial composition of a 1:4:8 molar ratio of TetraMS, methanol, and water.

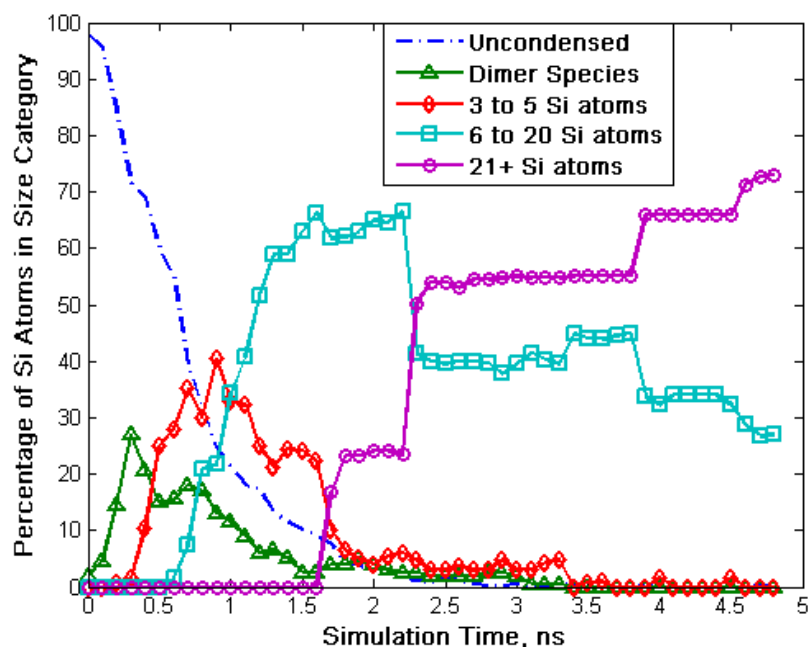


Figure S39: Siloxane cluster size distribution for a solution with an initial composition of a 1:4:12 molar ratio of TetraMS, methanol, and water.

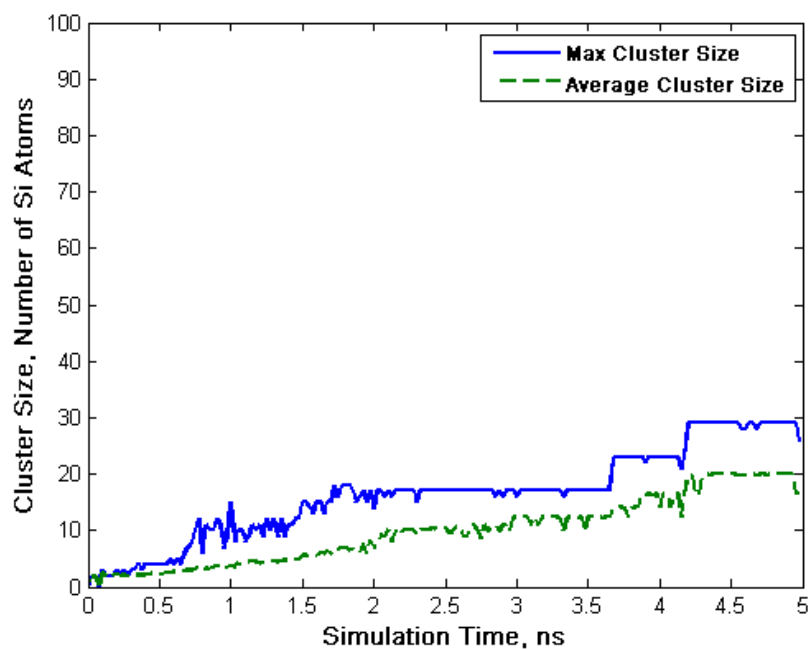


Figure S40: Maximum and average siloxane cluster size for a system containing one hundred tetramethoxysilane molecules. The initial composition of the solution is a 1:4:8 molar ratio of TetraMS, methanol, and water.

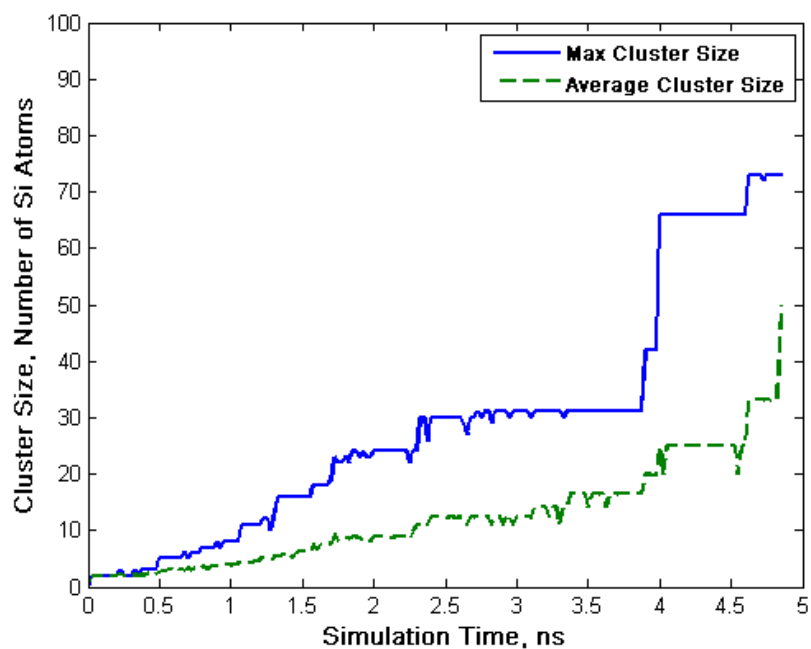


Figure S41: Maximum and average siloxane cluster size for a system containing one hundred tetramethoxysilane molecules. The initial composition of the solution is a 1:4:12 molar ratio of TetraMS, methanol, and water.

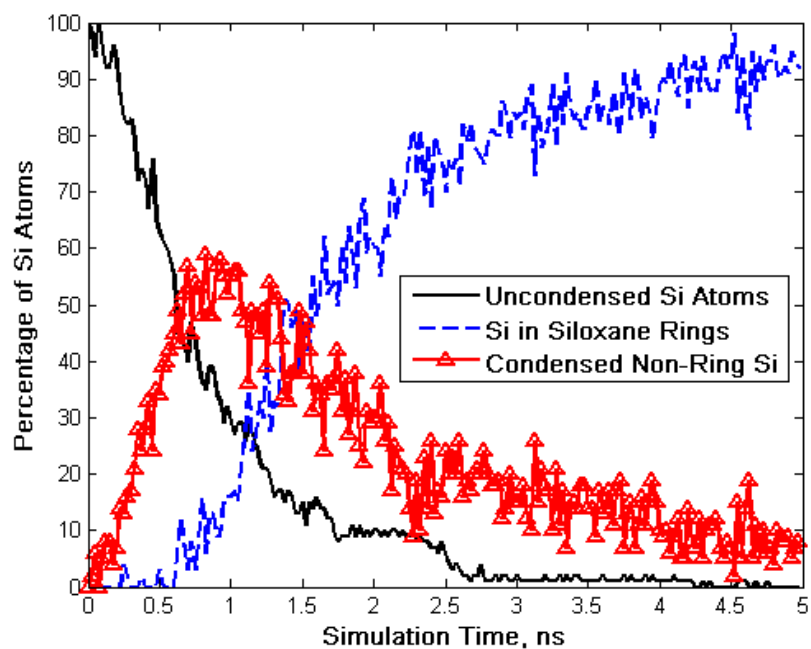


Figure S42: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:4:8 molar ratio of TetraMS, methanol, and water.

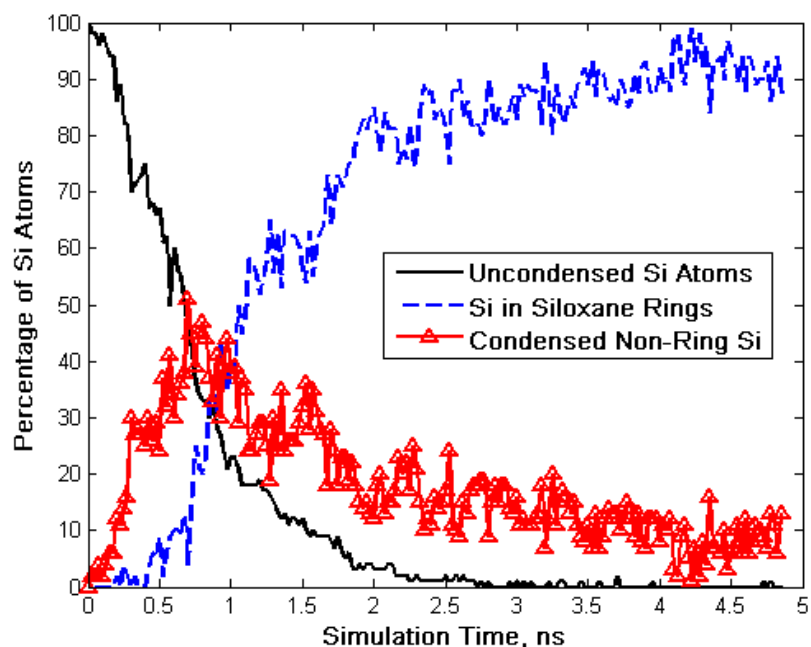


Figure S43: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:4:12 molar ratio of TetraMS, methanol, and water.

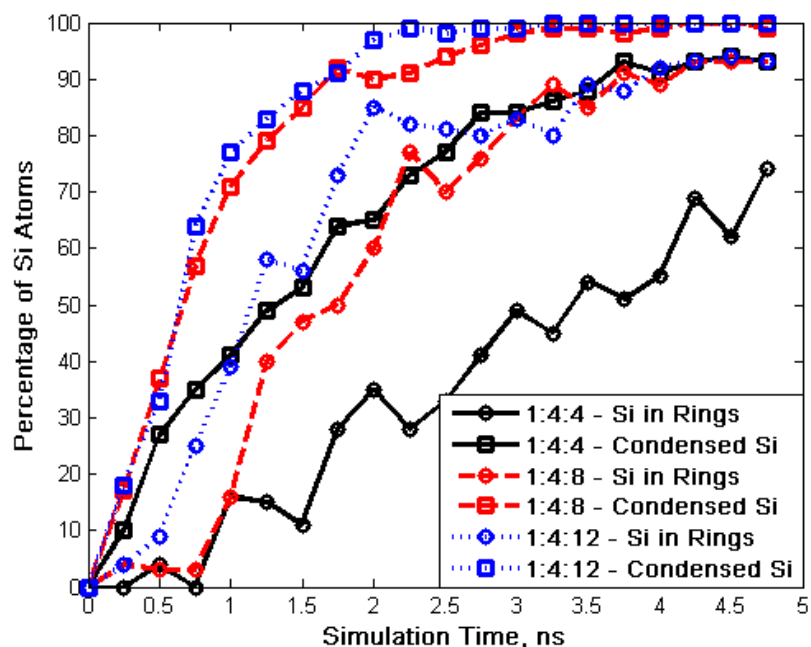


Figure S44: Percent of silicon atoms condensed compared with the percent in siloxane rings. Three systems are modeled with different ratios of TetraMS: methanol: water. The three systems compared here have different methanol concentrations at 1:4:4, 1:4:8, and 1:4:12.

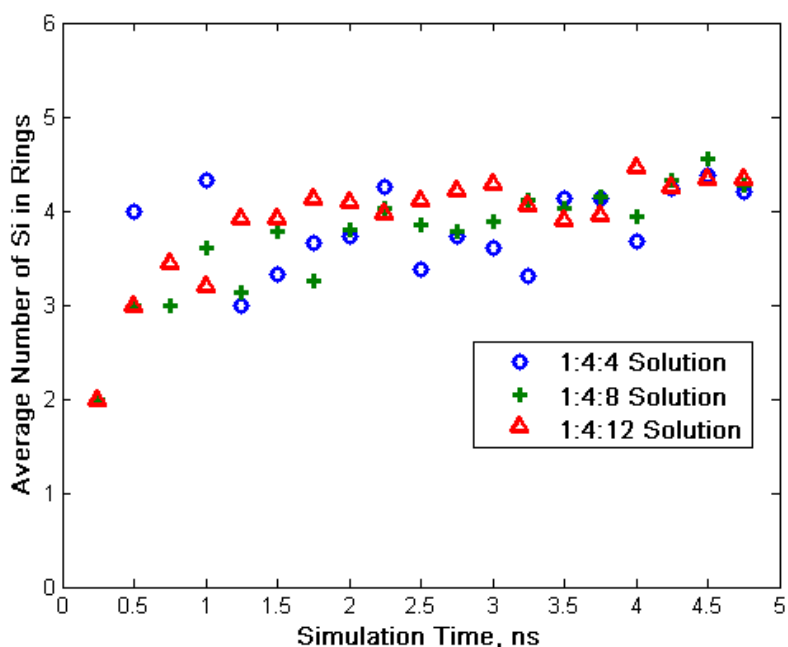


Figure S45: Average number of silicon atoms in siloxane rings. Two systems are modeled with different ratios of TetraMS:methanol:water at 1:4:4, 1:4:8, and 1:4:12.

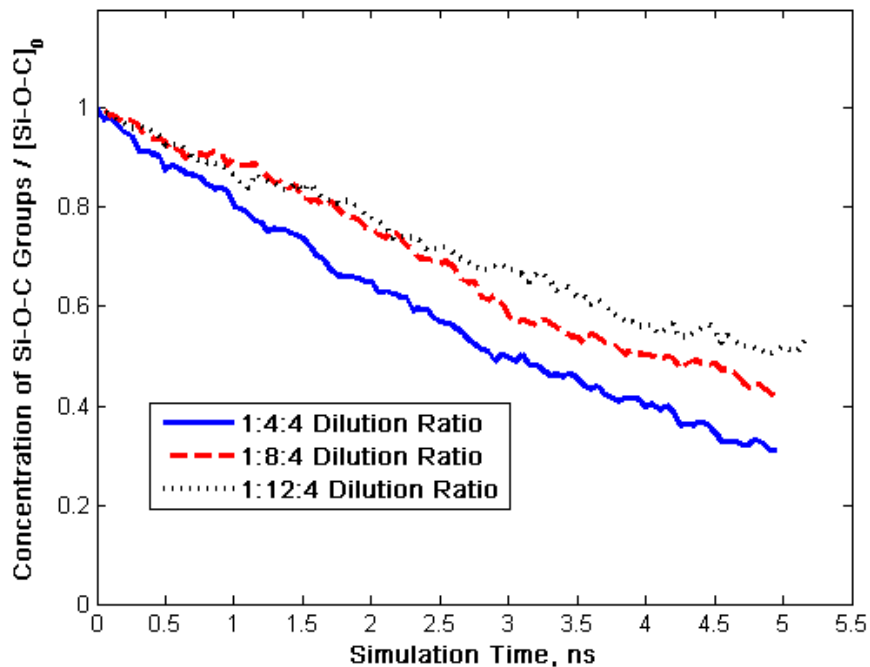


Figure S46: Normalized concentration of alkoxy groups (Si-O-C) in system. Three molar ratios of 1:4:4, 1:8:4, and 1:12:4 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

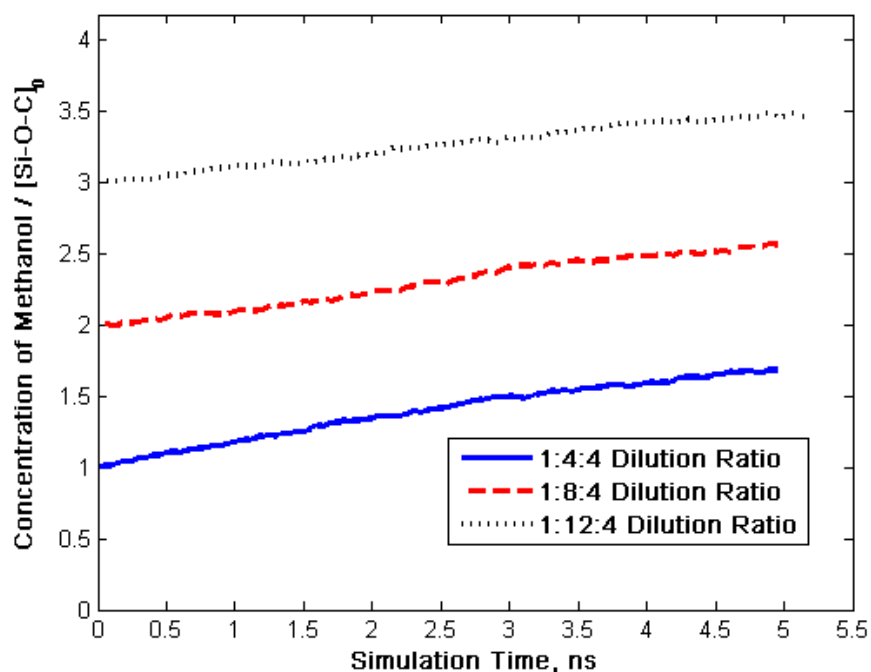


Figure S47: Normalized concentration of methanol in system. Three molar ratios of 1:4:4, 1:8:4, and 1:12:4 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

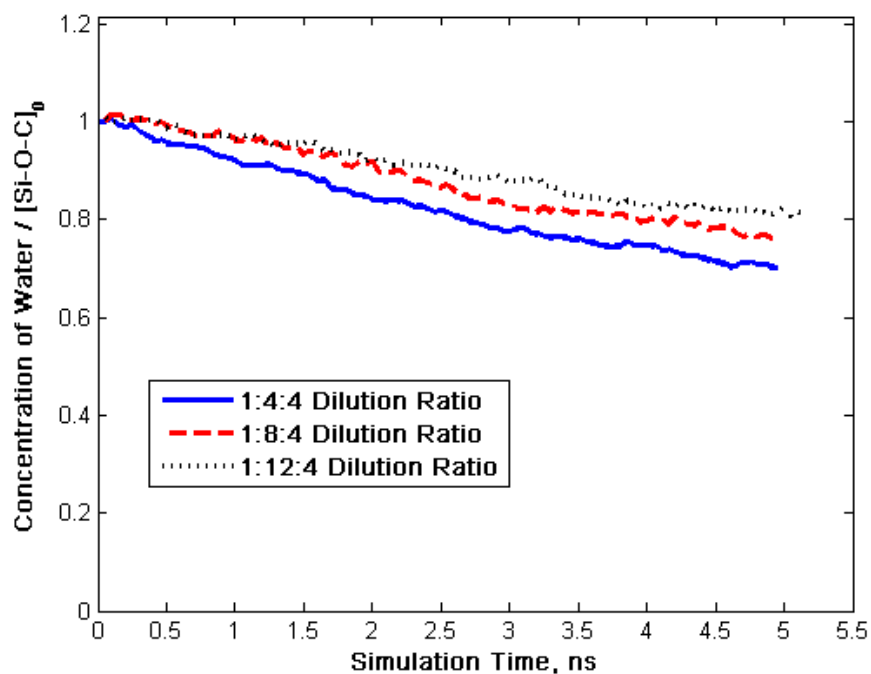


Figure S48: Normalized concentration of water in system. Three molar ratios of 1:4:4, 1:8:4, and 1:12:4 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

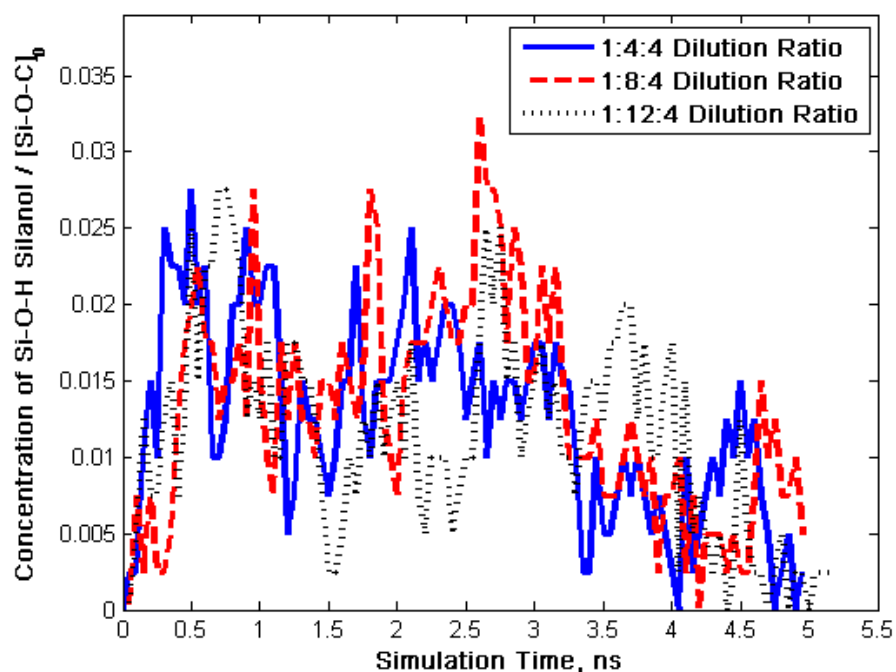


Figure S49: Normalized concentration of silanol (Si-O-H) in system. Three molar ratios of 1:4:4, 1:8:4, and 1:12:4 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

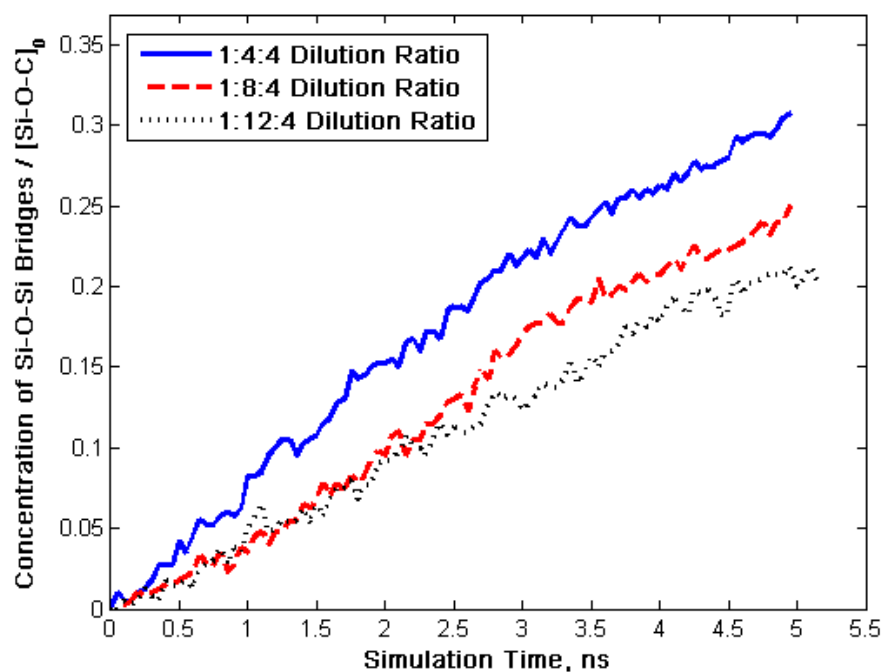


Figure S50: Normalized concentration of siloxane bridges (Si-O-Si) in system. Three molar ratios of 1:4:4, 1:8:4, and 1:12:4 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

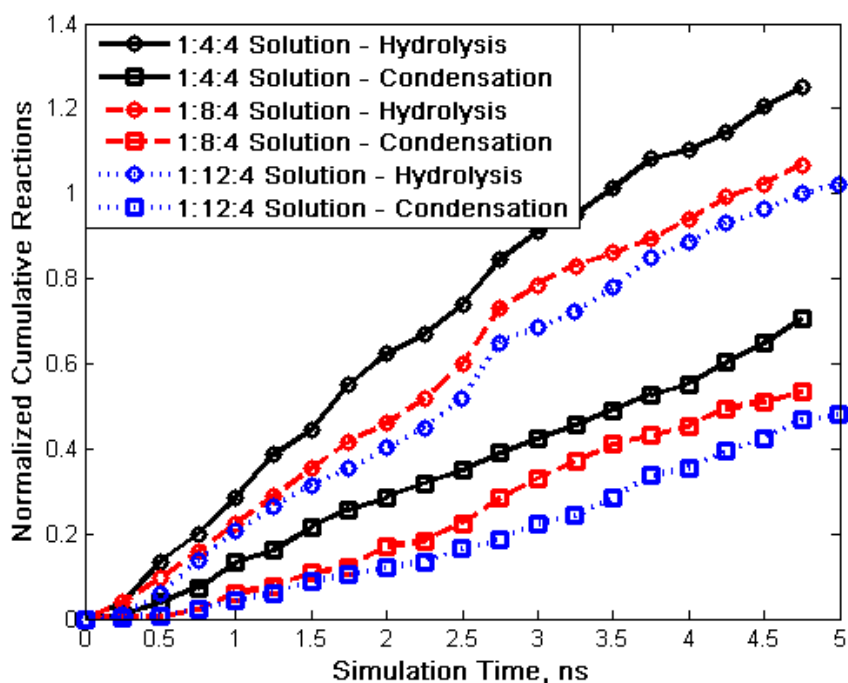


Figure S51: Normalized cumulative hydrolysis and condensation reactions. Three molar ratios of 1:4:4, 1:8:4, and 1:12:4 tetramethoxysilane:methanol:water were modeled. Number of reactions are normalized by initial number of alkoxy silane groups in the simulation.

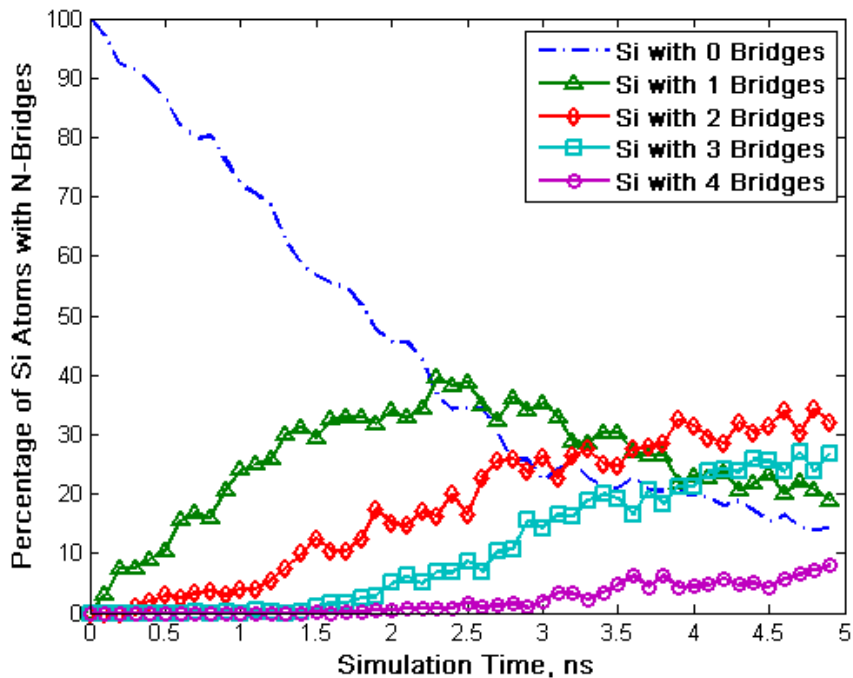


Figure S52: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:8:4 molar ratio of TetraMS, methanol, and water.

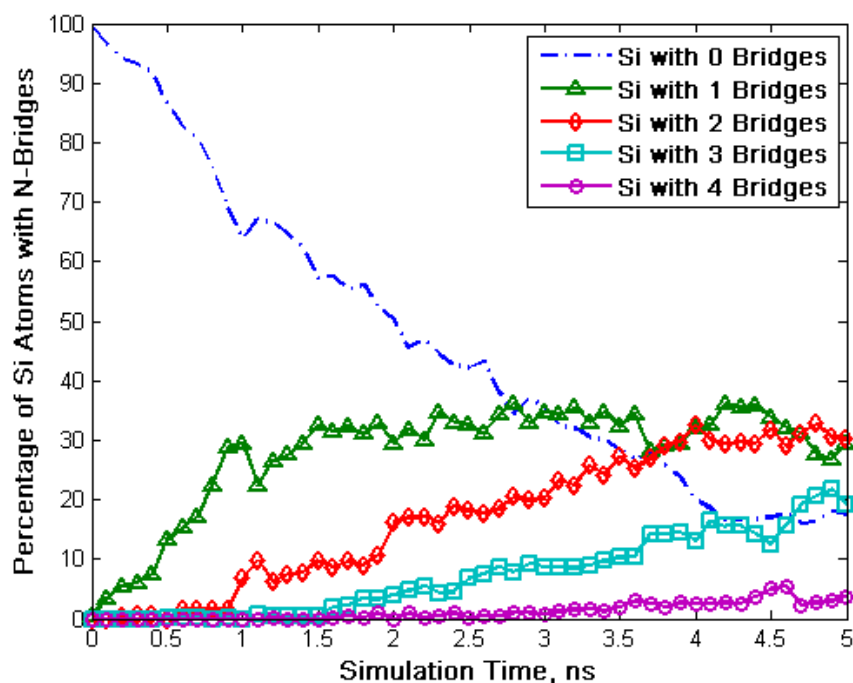


Figure S53: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:12:4 molar ratio of TetraMS, methanol, and water.

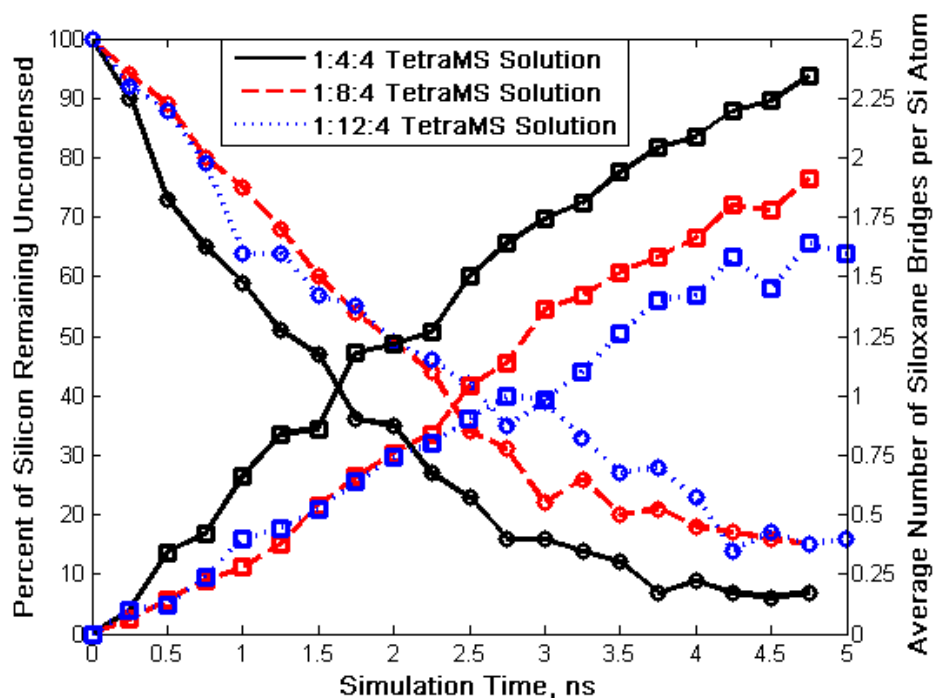


Figure S54: Percent of silicon atoms remaining uncondensed compared with the average number of siloxane Si-O-Si bridges per silicon atom. Three systems are modeled with different ratios of TetraMS:methanol:water. The three systems compared here have different methanol concentrations at 1:4:4, 1:8:4, and 1:12:4.

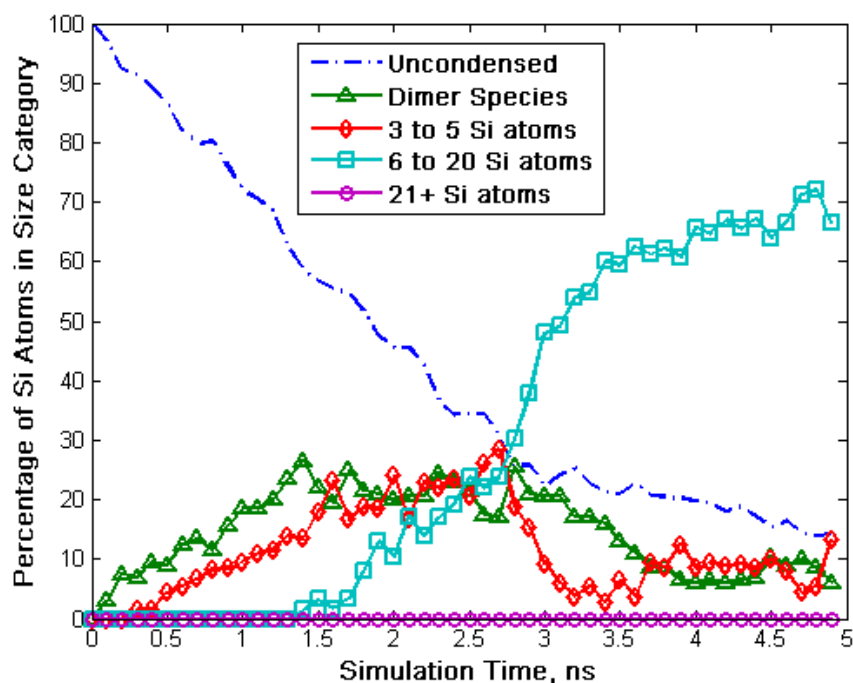


Figure S55: Siloxane cluster size distribution for a solution with an initial composition of a 1:8:4 molar ratio of TetraMS, methanol, and water.

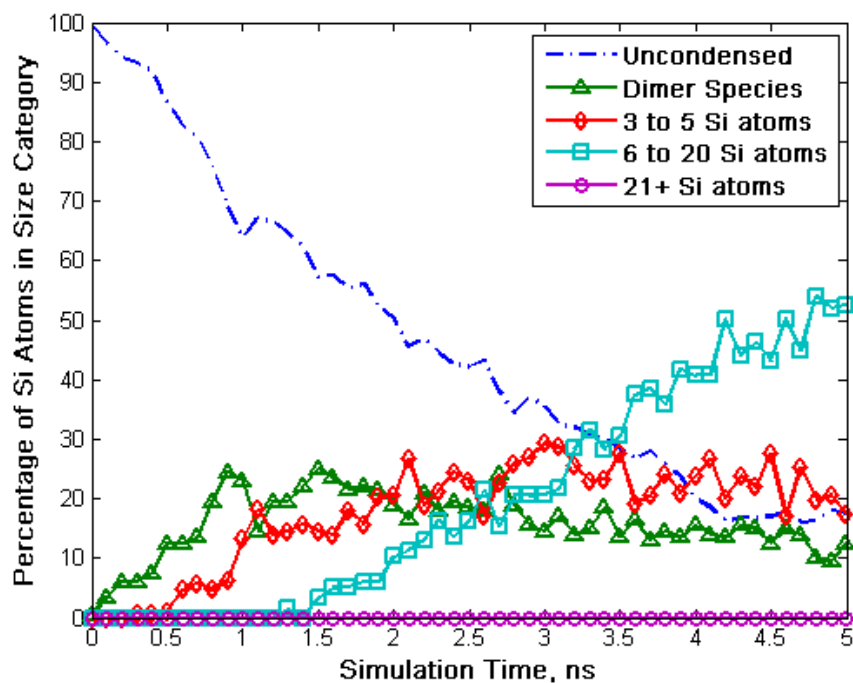


Figure S56: Siloxane cluster size distribution for a solution with an initial composition of a 1:12:4 molar ratio of TetraMS, methanol, and water.

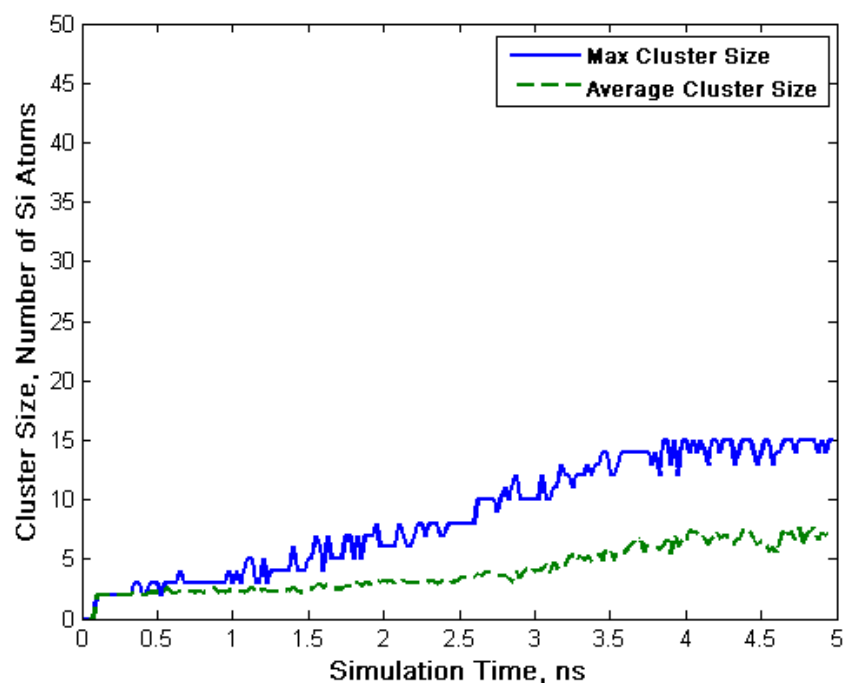


Figure S57: Maximum and average siloxane cluster size for a system containing one hundred tetramethoxysilane molecules. The initial composition of the solution is a 1:8:4 molar ratio of TetraMS, methanol, and water.

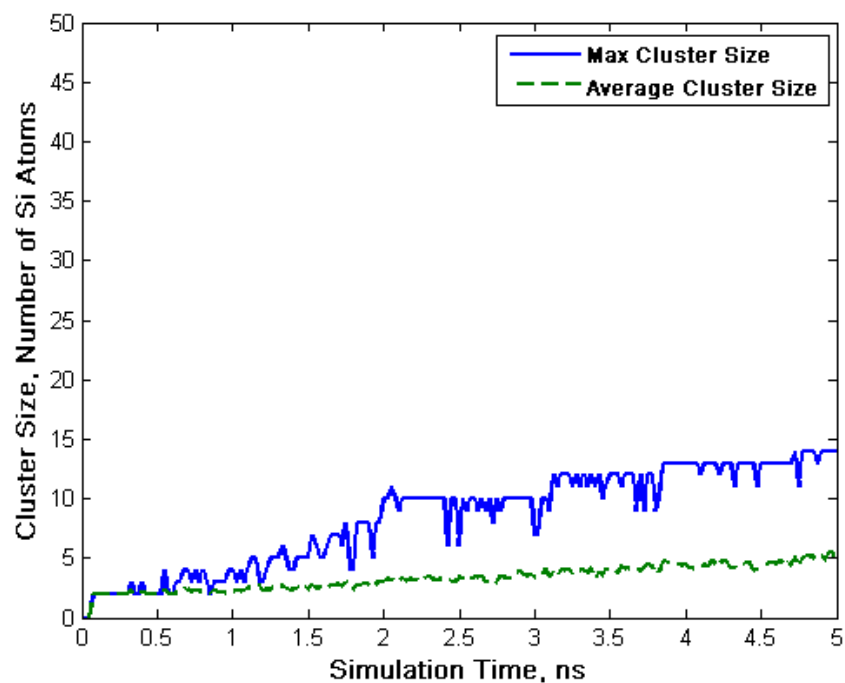


Figure S58: Maximum and average siloxane cluster size for a system containing one hundred tetramethoxysilane molecules. The initial composition of the solution is a 1:12:4 molar ratio of TetraMS, methanol, and water.

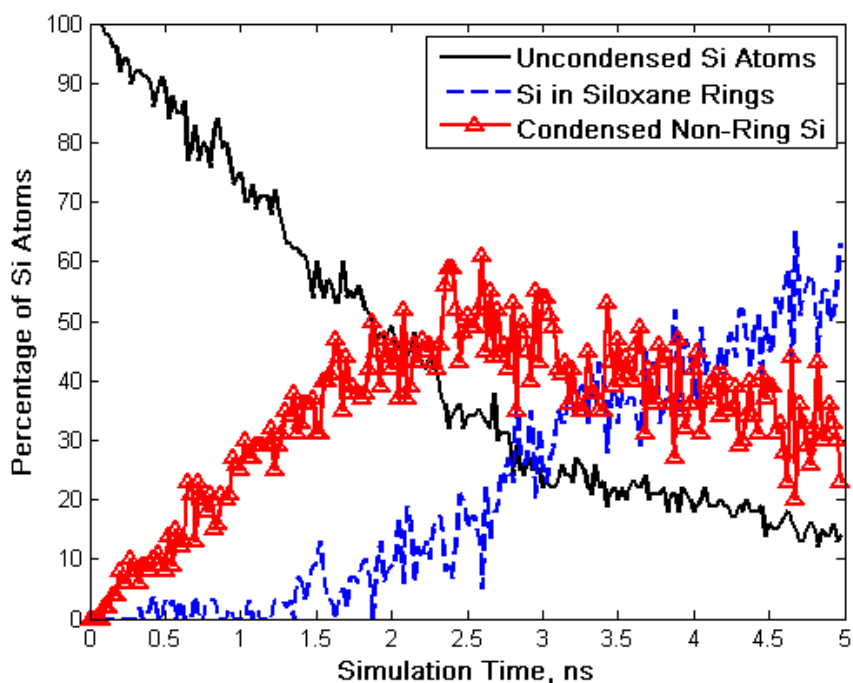


Figure S59: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:8:4 molar ratio of TetraMS, methanol, and water.

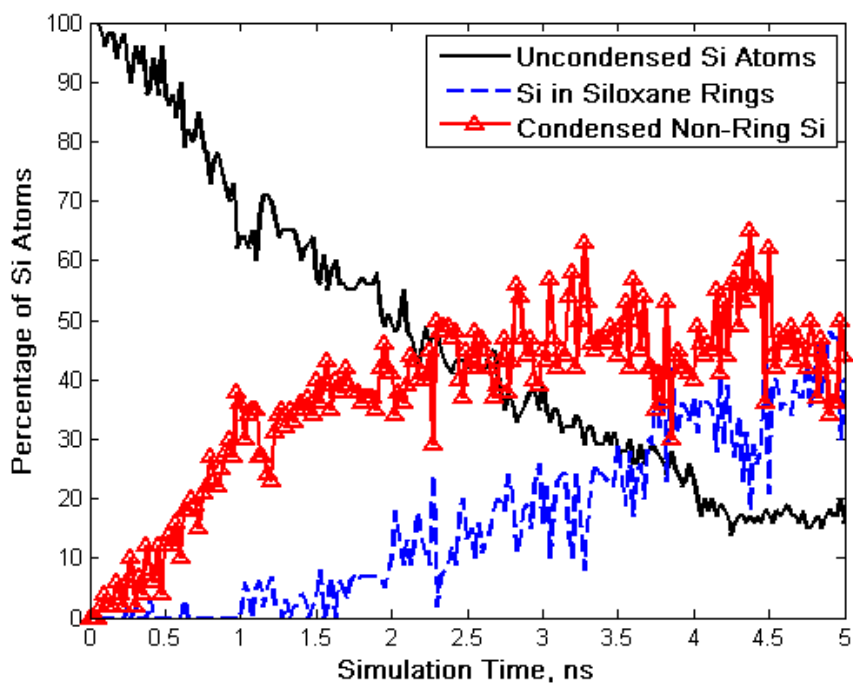


Figure S60: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:12:4 molar ratio of TetraMS, methanol, and water.

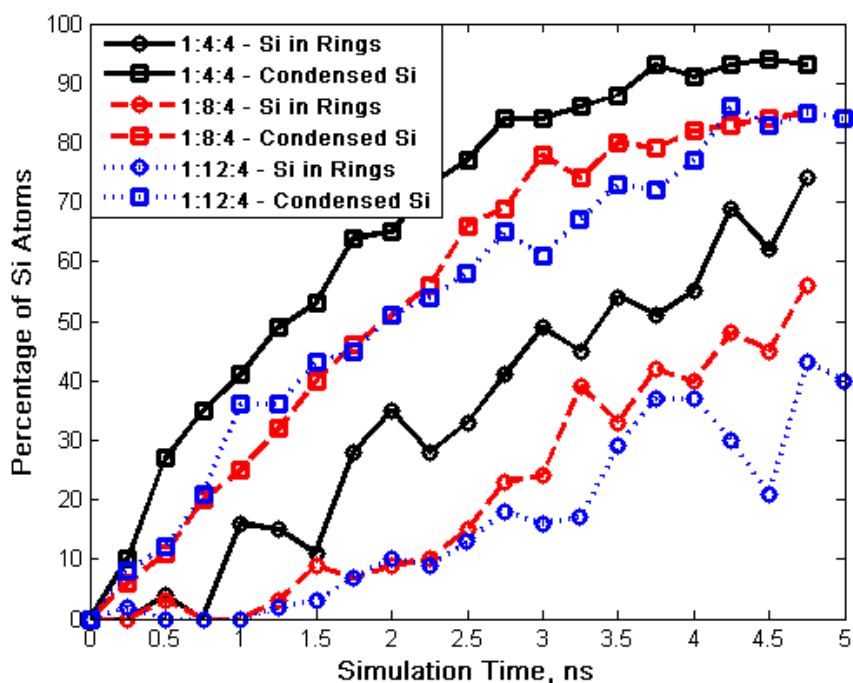


Figure S61: Percent of silicon atoms condensed compared with the percent in siloxane rings. Three systems are modeled with different ratios of TetraMS:methanol:water. The three systems compared here have different methanol concentrations at 1:4:4, 1:8:4, and 1:12:4.

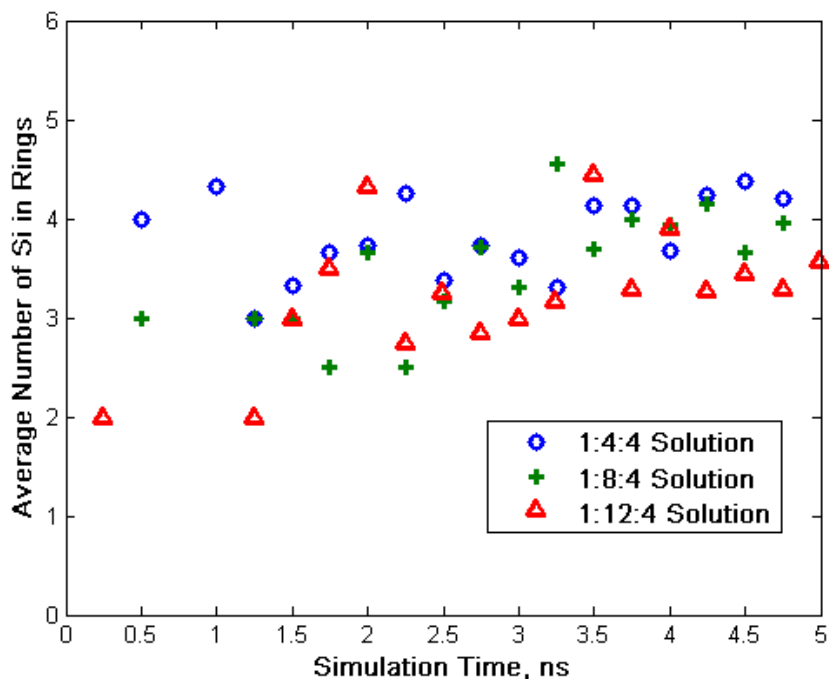


Figure S62: Average number of silicon atoms in siloxane rings. Two systems are modeled with different ratios of TetraMS:methanol:water at 1:4:4, 1:8:4, and 1:12:4.

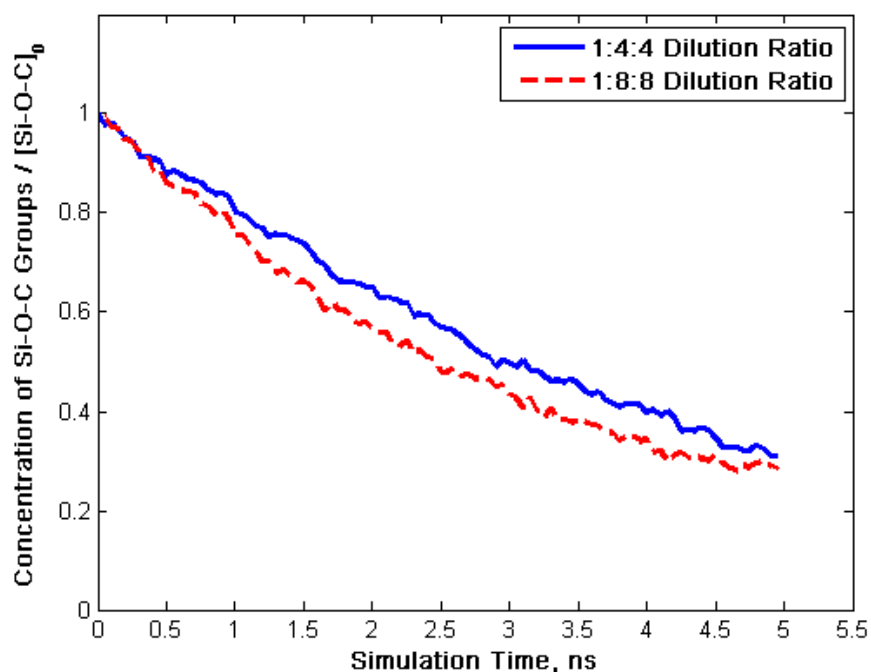


Figure S63: Normalized concentration of alkoxy groups (Si-O-C) in system. Two molar ratios of 1:4:4, and 1:8:8 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

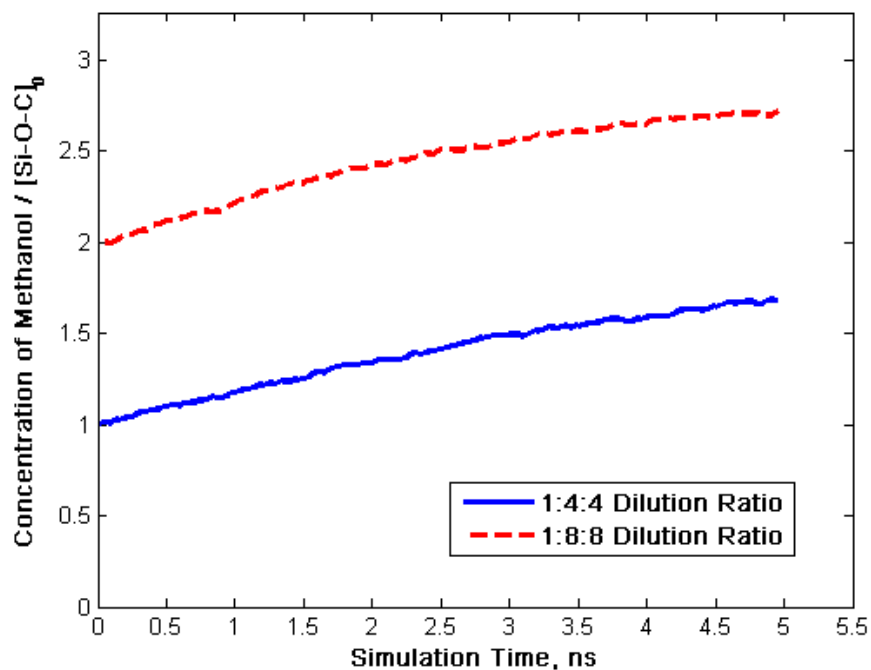


Figure S64: Normalized concentration of methanol in system. Two molar ratios of 1:4:4, and 1:8:8 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

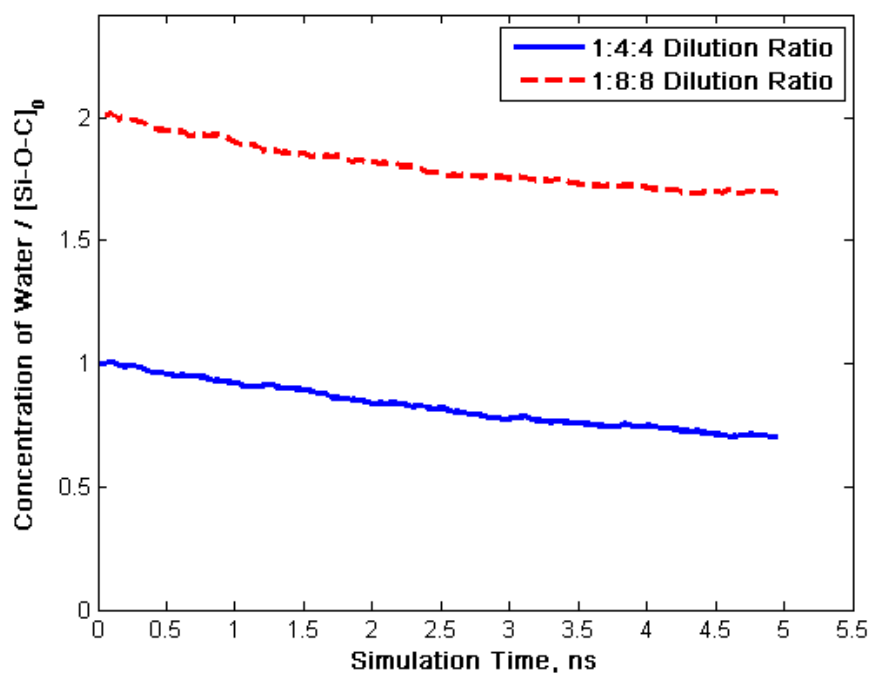


Figure S65: Normalized concentration of water in system. Two molar ratios of 1:4:4, and 1:8:8 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

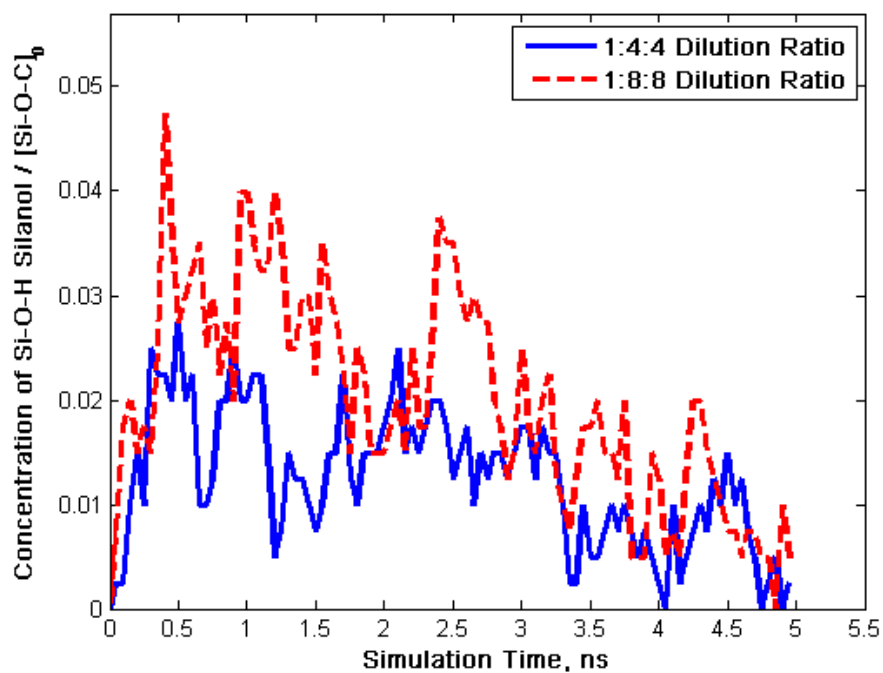


Figure S66: Normalized concentration of silanol (Si-O-H) in system. Two molar ratios of 1:4:4, and 1:8:8 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy groups in each simulation.

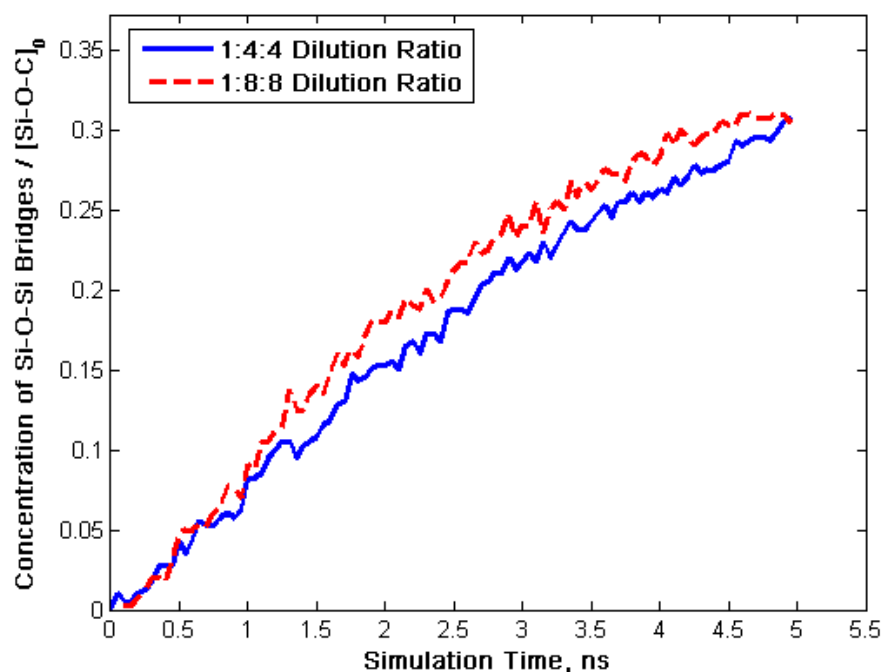


Figure S67: Normalized concentration of siloxane bridges (Si-O-Si) in system. Two molar ratios of 1:4:4, and 1:8:8 tetramethoxysilane:methanol:water were modeled. Concentration is normalized by the initial number of alkoxy silane groups in each simulation.

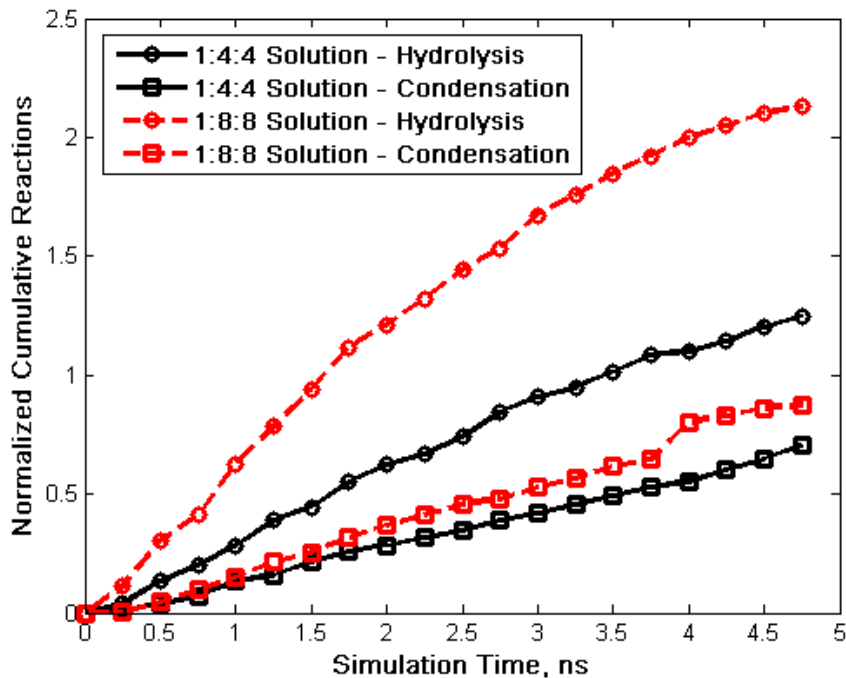


Figure S70: Normalized cumulative hydrolysis and condensation reactions. Two molar ratios of 1:4:4, and 1:8:8 tetramethoxysilane:methanol:water were modeled. Number of reactions are normalized by initial number of alkoxy silane groups in the simulation.

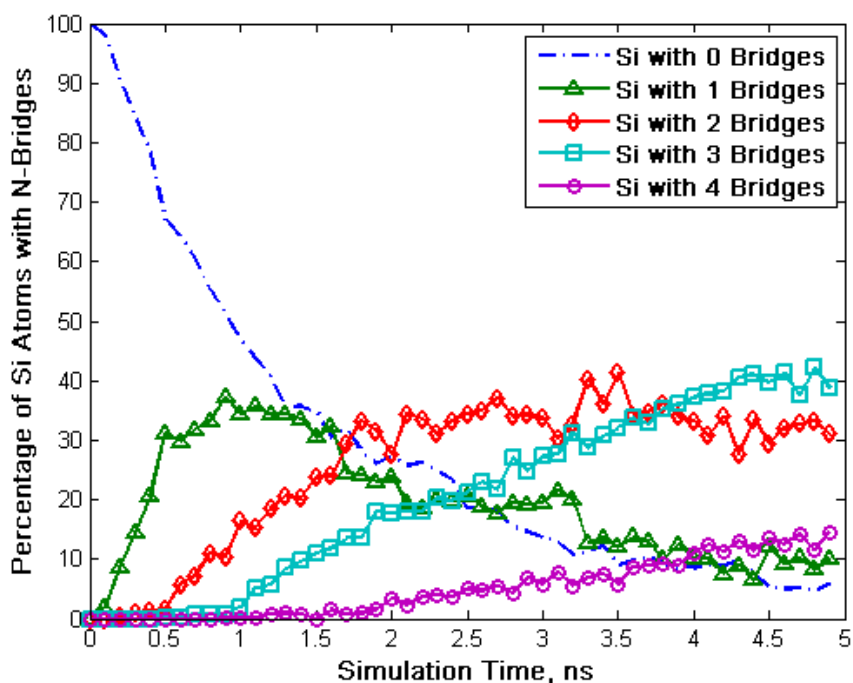


Figure S71: Distribution of silicon atoms with different numbers of condensed siloxane Si-O-Si bridges. The initial composition of the solution is a 1:8:8 molar ratio of TetraMS, methanol, and water.

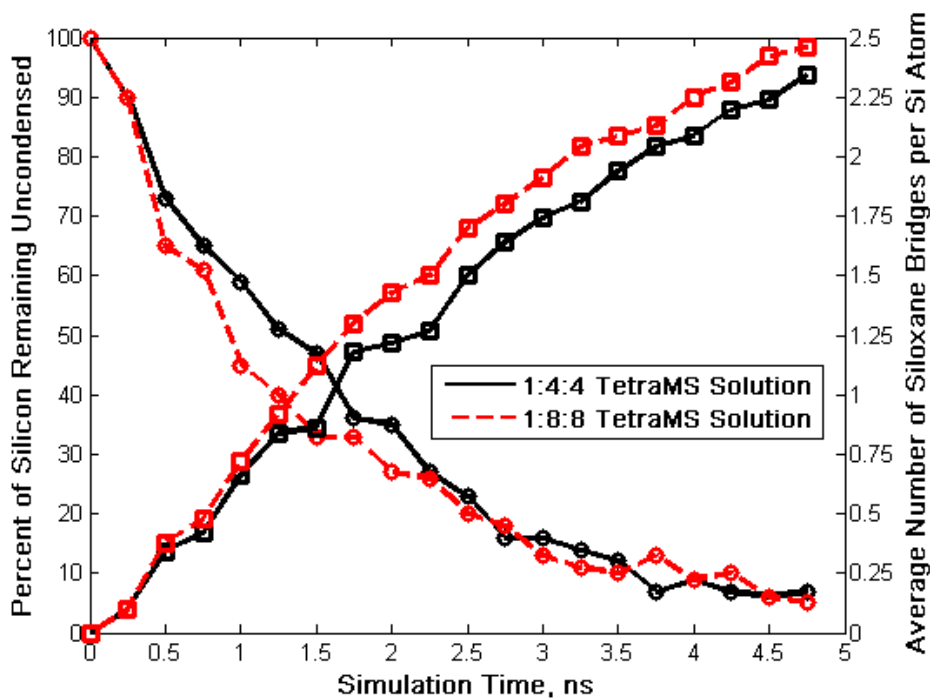


Figure S72: Percent of silicon atoms remaining uncondensed compared with the average number of siloxane Si-O-Si bridges per silicon atom. Two systems are modeled with different ratios of TetraMS:methanol:water. The two systems compared here have different TetraMS concentrations at 1:4:4, and 1:8:8.

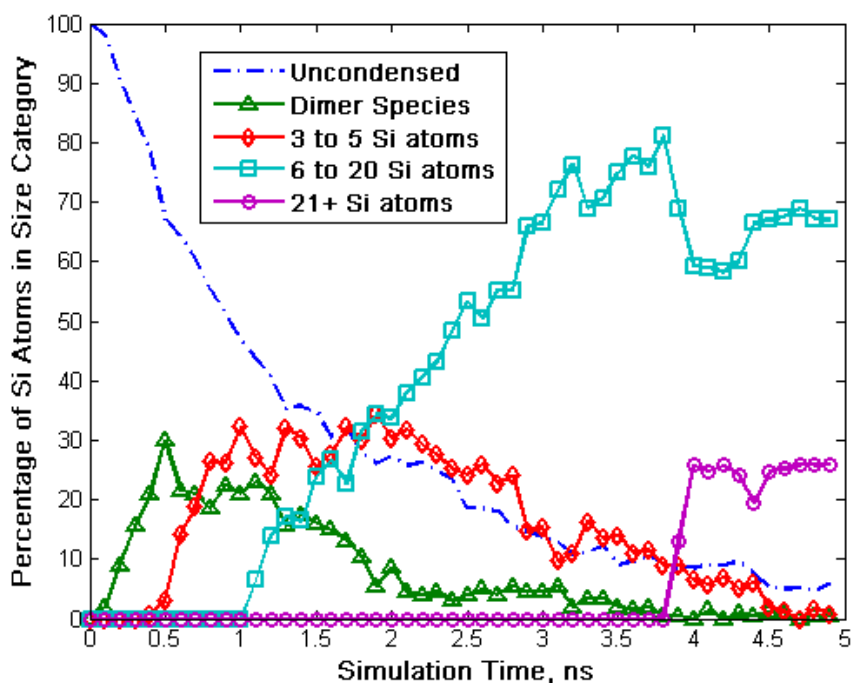


Figure S73: Siloxane cluster size distribution for a solution with an initial composition of a 1:8:8 molar ratio of TetraMS, methanol, and water.

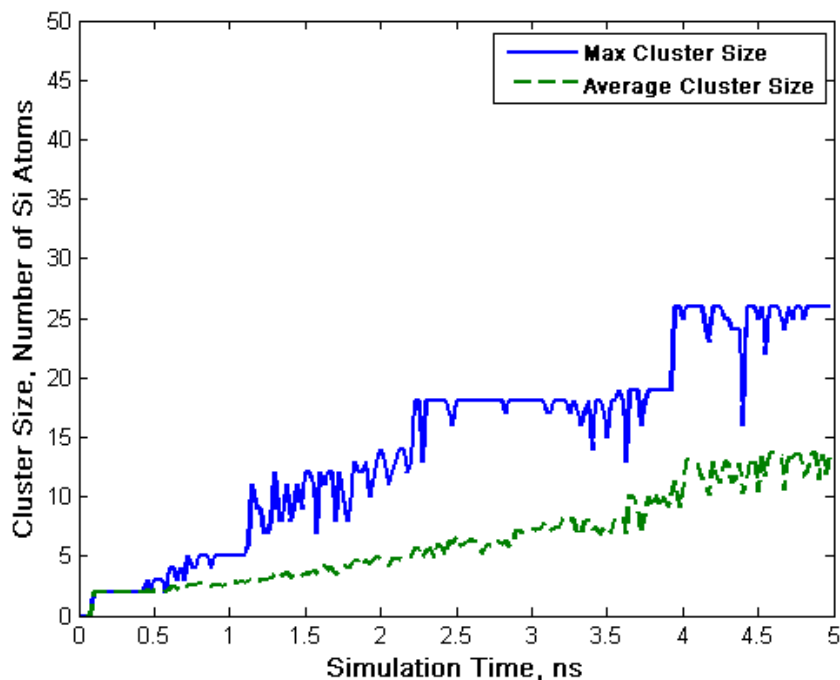


Figure S74: Maximum and average siloxane cluster size for a system containing one hundred tetramethoxysilane molecules. The initial composition of the solution is a 1:8:8 molar ratio of TetraMS, methanol, and water.

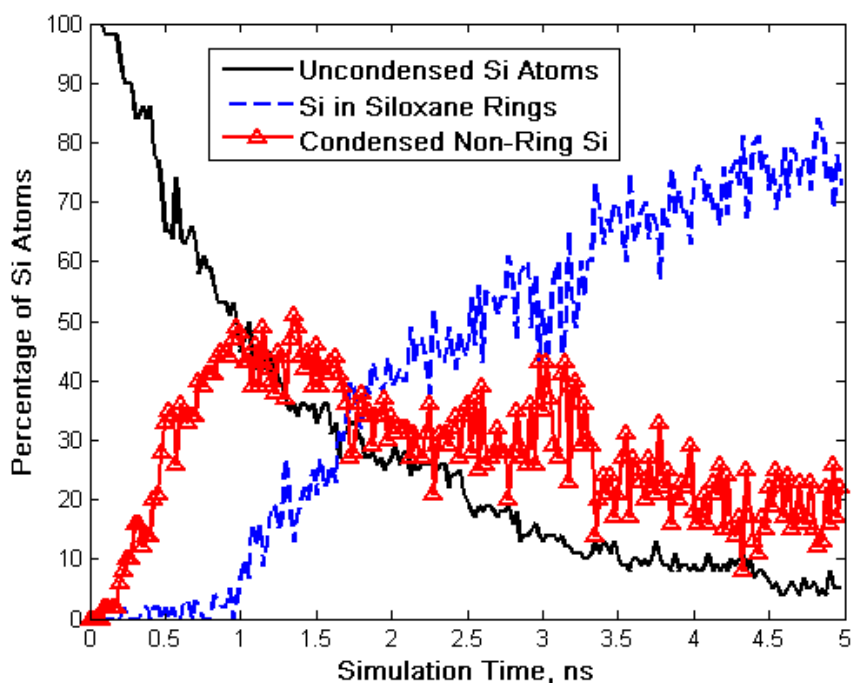


Figure S75: Percentage of silicon atoms which are uncondensed, condensed in siloxane rings, or condensed but not in rings. The initial composition of the solution is a 1:8:8 molar ratio of TetraMS, methanol, and water.

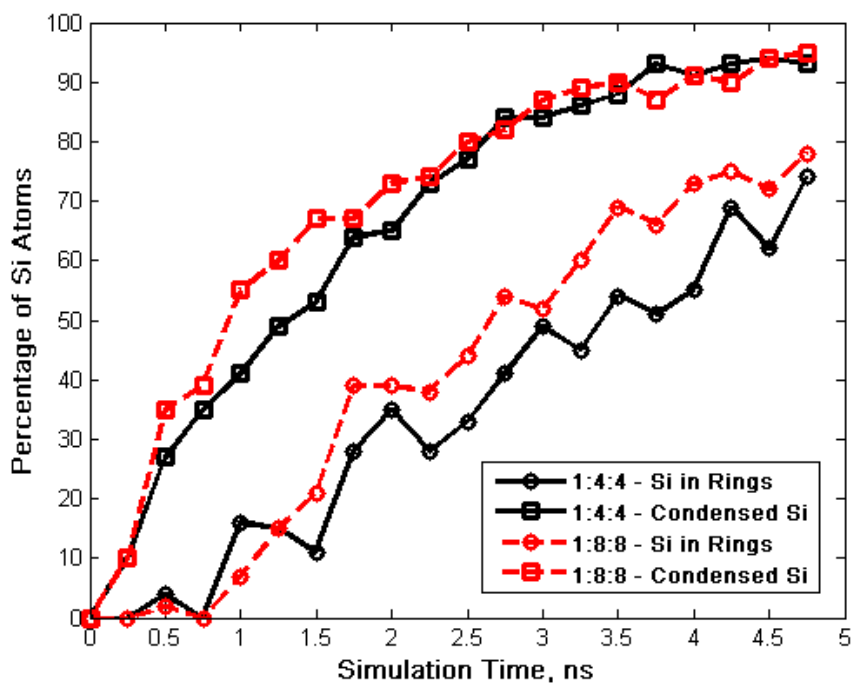


Figure S76: Percent of silicon atoms condensed compared with the percent in siloxane rings. Two systems are modeled with different ratios of TetraMS:methanol:water. The two systems compared here have different tetramethoxysilane concentrations at 1:4:4, 1:8:8.

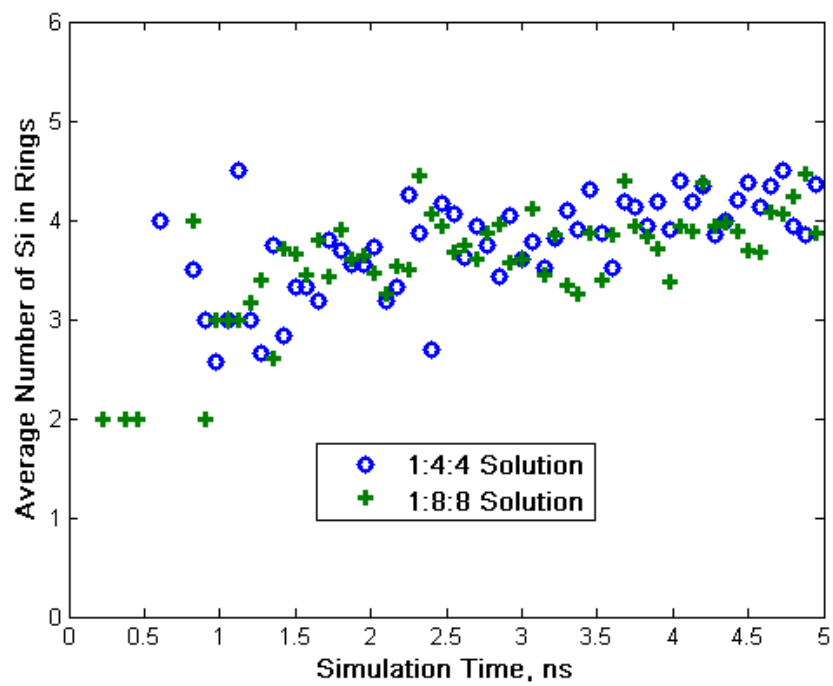


Figure S77: Average number of silicon atoms in siloxane rings. Two systems are modeled with different ratios of TetraMS:methanol:water at 1:4:4, and 1:8:8.