Supporting information.

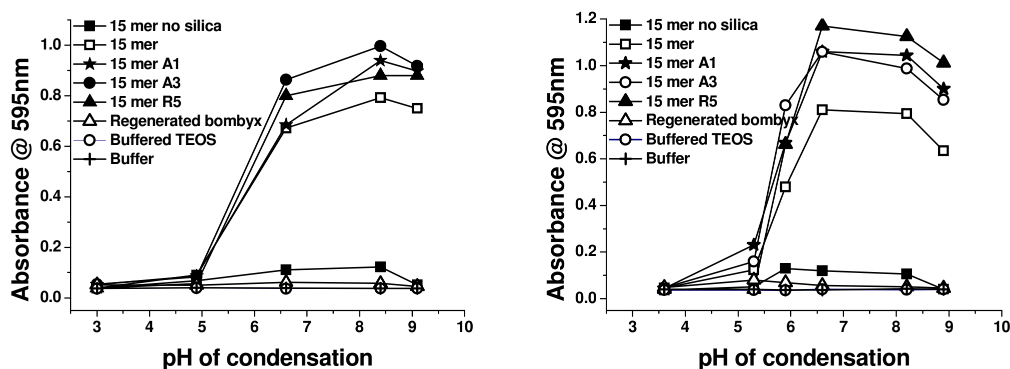

Figure 1. Buffer comparison for scattering data. LHS, phosphate buffer; RHS, bis propane/ citric acid buffer.

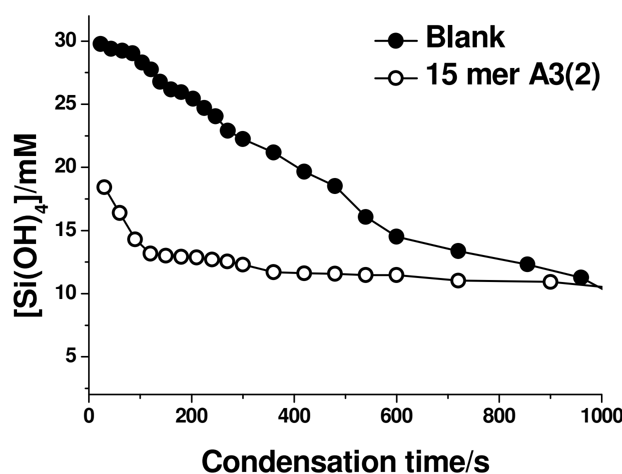

Figure 2. Disappearance of molybdenum active silicate species after initiation of condensation by pH adjustment to 6.9

Equilibrium monosilicic acid concentrations:

For each reaction, the equilibrium point of the silica condensation reaction was determined as the time after which the level of monosilicic acid in the silicifying system was stabilised. Twenty µl of supernatant from each sample was added to 180 µl of molybdic acid reagent (ammonium molybdate (20 g) dissolved in 500 cm^3 ddH_2O, hydrochloric acid added (60 cm^3 concentrated) and diluted to 1000 cm^3 with ddH_2O). Reducing reagent (oxalic acid (20 g), 4 methylaminophenolsulphate (6.67g) and sodium sulphite (4 g) dissolved in ddH_2O (800 cm^3), Sulphuric acid added (98%, 100 cm^3) and diluted to 1,000 cm^3 with ddH_2O) was added after 15 minutes and the absorbance at 810 nm measured after a minimum of 2 hours and maximum of 48 hours (corresponding to the completion of reduction and known stability of

the silicomolybdous acid complex formed). Measurements were compared with standards prepared by dilution of a 1,000 ppm SiO_2 stock standard treated in the same way.

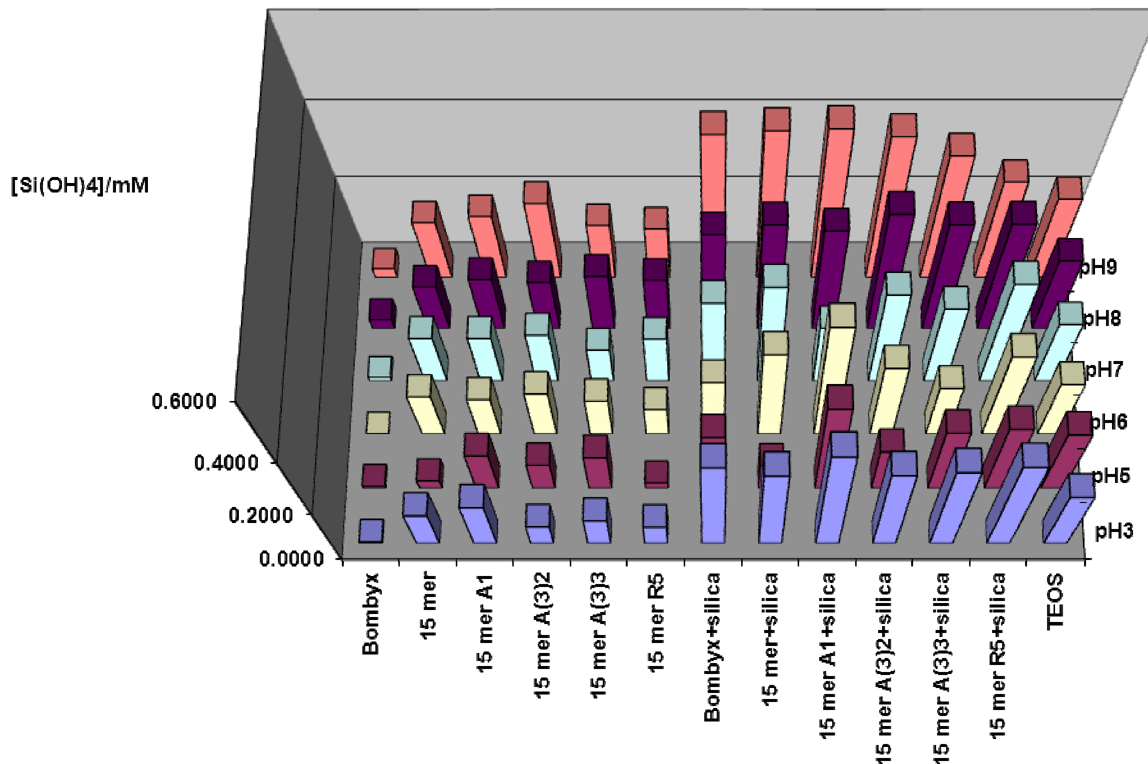

Figure 3 [Si(OH)$_4$] at equilibrium/mM

Residual soluble protein:

Determination of remaining soluble protein after silica condensation was carried out by a modified Bradford protein assay method. Coomassie blue stock reagent – 60µl (100 mg Coomassie blue in 50 cm^3 methanol with 100 cm^3 85% orthophosphoric acid diluted to 200 cm^3 with water) was diluted with 130 µl distilled water and 10 µl of supernatant from each sample added (after centrifugation at 3000 rpm for 5 minutes). Absorbance at 595 and 450 nm measured using a labtech LT-5000 plate reader with manta software (build 0460). Protein determined as the ratio of the absorbances at 595 nm and 450 nm and measured against 0.1 – 1.0 $mgcm^{-3}$ aqueous solutions of non chimeric 15 mer and regenerated *B. mori* fibroin as standards for the residual 15 mer chimeras and *B. mori* proteins respectively.

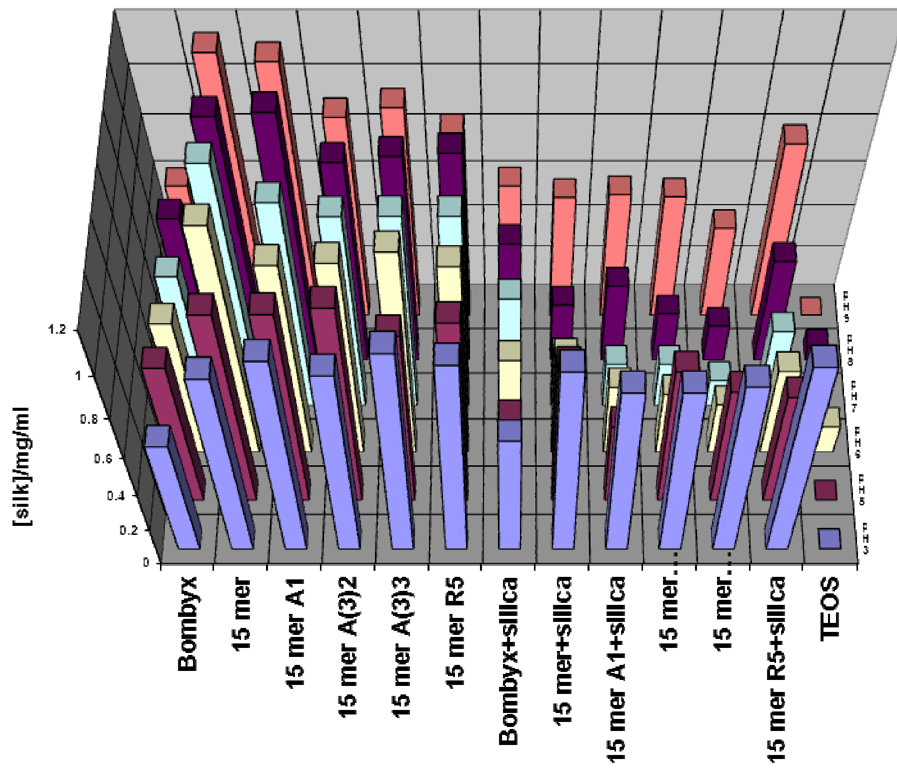

Figure 4. Aqueous protein at equilibrium time/mgcm^{-3}

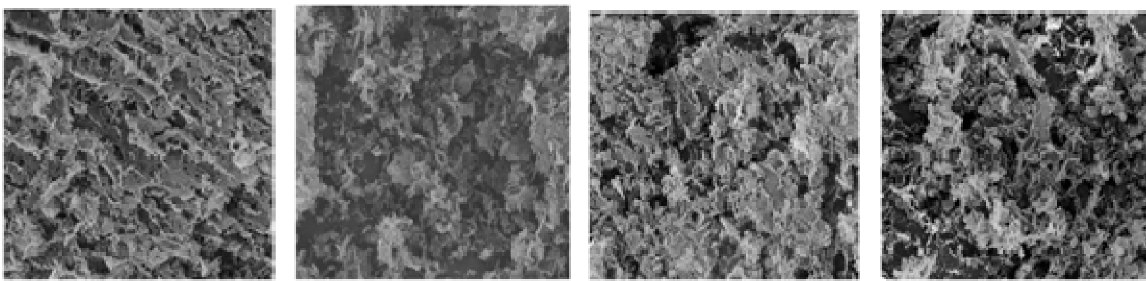

Figure 5. Precipitated silica blanks. Left to right pH 4.7, 6.7, 7.3 and 7.9. Scale bar represents 40µm

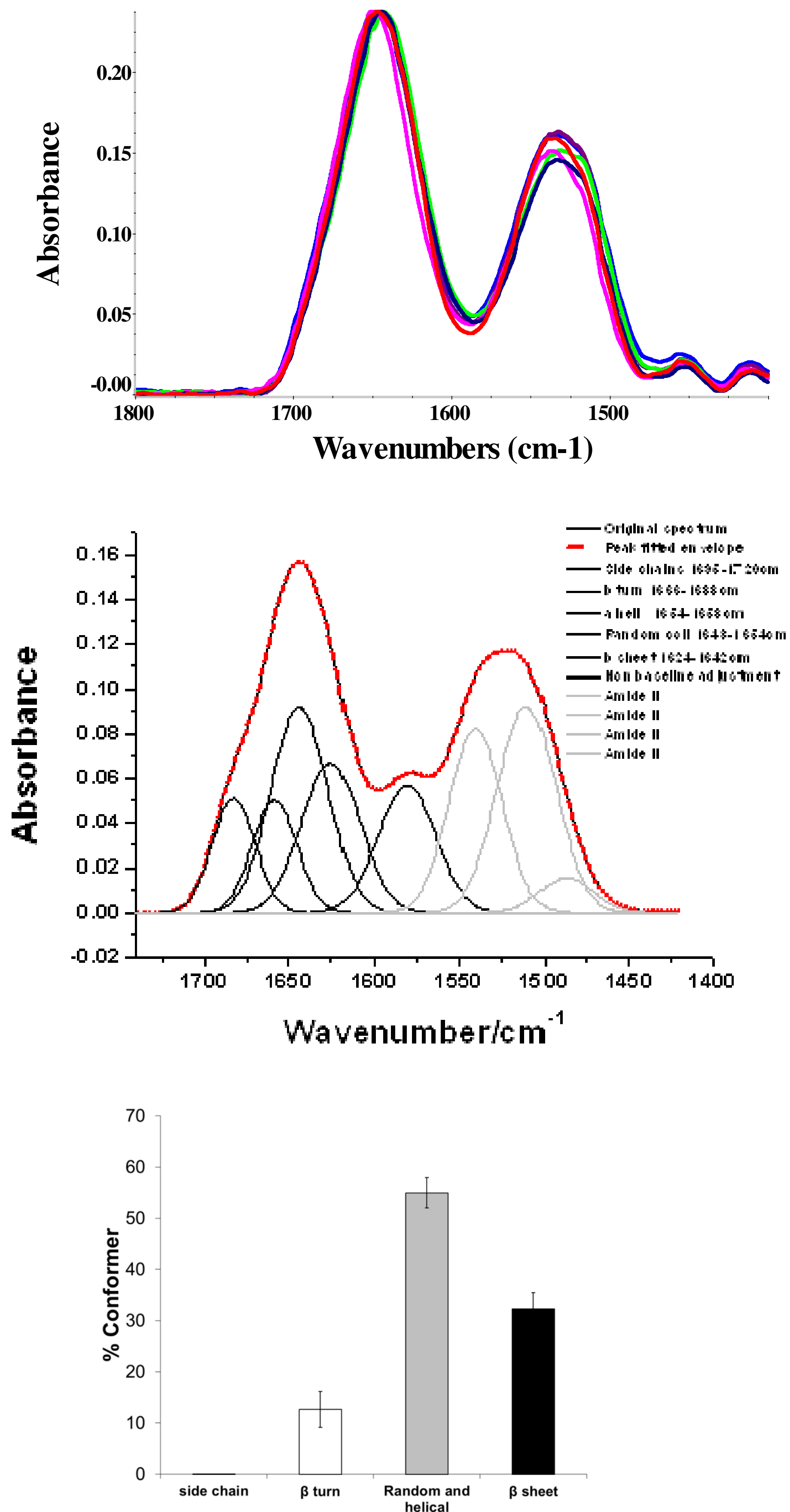

Figure 6. Top- representative FTIR of 15 mers. - Silk consensus repeats only, - A1 chimera, - A3 chimera, - R5 chimera. Middle- Example of amide I band curve fitting of standard α lactalbumin. **Bottom-** Statistical analysis of 5 repeats (sampling, data collection and analysis) of α lactalbumin.

Silk/ chimera	pH 3.1	pH 4.7	pH 6.7	pH 7.4	pH 7.8	pH 8.7
Silk fibroin		X	X	X	X	
15mer		X s	X s	X s	X	
15mer-A1	X	X s	X s	X s	X s	X
15mer-A3		X s	X s	X s	X s	X
15mer-$(A3)_2$	X	X s	X s	X s	X s	X
15mer-$(A3)_3$	X	X s	X s	X s	X s	X
15mer-R5	X	X s	X s	X s	X s	X

Table 1. pH at which isolable material was recovered from silicifying systems