

1 **Supplementary data**

2

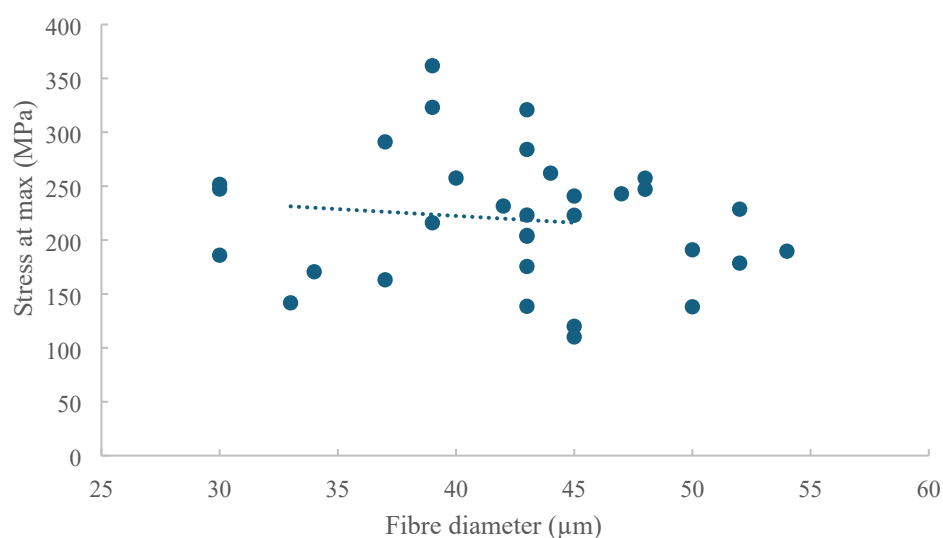
3 **Recapitulating native aculeate silk structure enables enhanced**
4 **mechanical performance in recombinant protein materials**

5

6

7

8 Lyndall J. Briggs¹, Michelle Michie¹, Jian-Wei Liu², Denise Thomas³, David Cantor³, Jacinta
9 Poole⁴, Tara D. Sutherland¹, Caitlin L. Johnston¹



10

11 **Supplementary Figure 1. Relationship between fibre diameter and stress at mx for the 33**
12 **recombinant silk fibres in this study, showing trend line and R² value.**

13

14

15 **Supplementary Table 1. The protein and amino acids identified as contributing to**
16 **spontaneous or thermally induced isopeptide cross-links between two proteins in F1-F4 silk**
17 **materials. ‘Same’ indicates the same lysine as in the row above. For simplicity, peptides with**
18 **more than one cross-link were excluded from the analysis.**

19

Protein hosting lysine (K) side of cross-link	Spontaneous cross-links (Controls)		~115 °C induced cross-links (SDS PAGE gel band)		~190 °C induced cross-links (Silk films)	
F1	F1:247[K]	F4:145[E]	F1:41[K]	F4:201[E]	F1:41[K]	F3:179[E]
			F1:66[K]	F3:129[T]	F1:83[K]	F2:113[T]
			same	F3:132[T]	same	F4:148[E]
			F1:118[K]	F4:126[T]	same	F4:149[S]
			same	F4:132[E]	F1:139[K]	F2:111[E]
			same	F4:242[E]	same	F2:113[T]
			same	F4:245[S]	same	F3:308[S]
			same	F4:249[E]	same	F3:309[E]
			F1:149[K]	F2:131[T]	F1:149[K]	F2:90[E]
			F1:151[K]	F3:116[S]	F1:174[K]	F4:317[T]
			same	F3:119[E]	same	F4:319[E]
			F1:247[K]	F4:145[E]	F1:187[K]	F2:111[E]
			same	F4:149[S]	same	F2:113[T]
			F1:295[K]	F3:181[T]	same	F3:308[S]
					same	F3:309[E]
	F2					F2:118[K]
					F2:156[K]	F4:126[T]
					F2:178[K]	F4:319[E]
					F2:178[K]	F4:316[T]
					F2:288[K]	F1:177[E]
F3			F3:163[K]	F4:126[T]	F3:52[K]	F1:177[E]
			same	F4:131[E]	same	F4:145[E]
			F3:307[K]	F4:170[E]	same	F4:175[E]
			same	F4:176[S]	F3:163[K]	F4:126[T]
					same	F4:127[E]
F4	F4:30[K]	F2:195[S]	F4:30[K]	F2:256[T]	F4:11[K]	F2:113[T]
			same	F2:258[E]	same	F2:134[S]
			F4:146[K]	F2:71[T]	same	F2:139[E]
			same	F2:75[E]	F4:146[K]	F3:27[D]
			same	F2:168[E]	same	F3:116[S]
			F4:152[K]	F1:84[E]	same	F3:119[E]
			same	F1:85[T]	same	F2:90[E]
			F4:303[K]	F3:233[E]	F4:152[K]	F2:134[S]
			same	F3:236[S]	same	F2:139[E]
					same	F3:303[E]
				F4:191[K]	F3:308[S]	

					same	F3:309[E]
					F4:307[K]	F2:113[T]

1

2