

## Straining flow spinning: Production of regenerated silk fibers under a wide range of mild coagulating chemistries

### Supplementary Information

Rodrigo Madurga,<sup>a,b</sup> Alfonso M. Gañán-Calvo,<sup>c</sup> Gustavo R. Plaza,<sup>a,b</sup> Gustavo V. Guinea,<sup>a,b,d</sup> Manuel Elices<sup>a,b</sup> and José Pérez-Rigueiro<sup>a,b,d,†</sup>

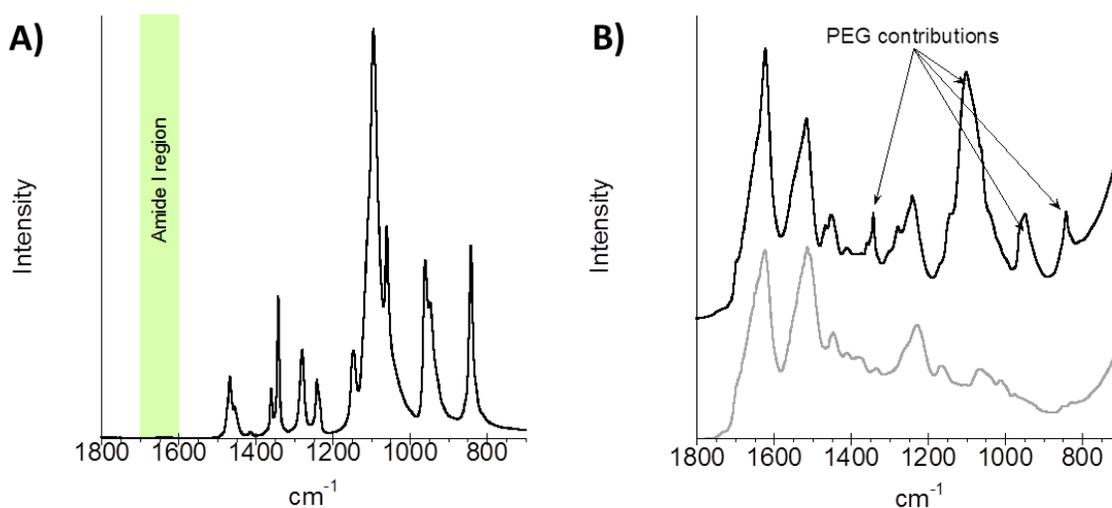
<sup>a</sup> Centro de Tecnología Biomédica. Universidad Politécnica de Madrid. 28223 Pozuelo de Alarcón (Madrid). Spain.

<sup>b</sup> Departamento de Ciencia de Materiales. ETSI Caminos, Canales y Puertos. Universidad Politécnica de Madrid. 28040. Madrid. Spain.

<sup>c</sup> Escuela Técnica Superior de Ingenieros. Universidad de Sevilla. 41092. Sevilla. Spain.

<sup>d</sup> Biomedical Research Networking Center in Bioengineering, Biomaterials and Nanomedicine (CIBER-BBN), Madrid, Spain

**Figure S1**



**Figure S1:** ATR-FTIR spectra of PEG (A) and fibers spun with PEG (B): black curve is for as spun fibers and gray curve is for the same spinning conditions but after washing the fibers in water to remove the PEG coating.

**Table S1**

Code	D ( $\mu\text{M}$ )	SD	SD/D	$\sigma_u$ (MPa)	$\epsilon_u$ (%)	$E_i$ (GPa)	$W_f \cdot 10^{-3}$ (MJ/m <sup>3</sup> )
AC-ET80-12-H	15	2	0.15	32	0.7	4.7	116
AC-ET80-12-L	12	2	0.17	24	0.4	6.1	52
AC-ET80-12-M	19	6	0.34	17	0.3	5.0	30
AC-ET80-3-H	20	2	0.12	21	0.4	5.5	45
AC-ET80-3-L	18	2	0.09	6	0.2	3.9	8
AC-ET80-3-M	16	1	0.08	21	0.4	5.5	46
AC-ET80-6-H	10	1	0.11	-	-	-	-
AC-ET80-6-L	16	2	0.14	31	0.5	5.6	84
AC-ET80-6-M	14	2	0.17	28	0.5	5.8	71
AC-ET80-9-H	14	3	0.19	20	0.4	5.0	52
AC-ET80-9-L	13	2	0.15	20	0.3	5.3	37
AC-ET80-9-M	10	1	0.12	20	0.4	4.7	45
AC-ISO80-12-H	9	2	0.24	19	0.7	2.8	75
AC-ISO80-12-L	11	1	0.11	30	0.8	3.4	167
AC-ISO80-12-M	10	2	0.16	24	0.9	2.6	117
AC-ISO80-3-H	18	3	0.17	12	0.6	1.9	40
AC-ISO80-3-L	11	2	0.15	21	0.9	2.2	108
AC-ISO80-3-M	18	2	0.14	15	0.5	2.6	48
AC-ISO80-6-H	13	3	0.24	16	0.6	2.6	56
AC-ISO80-6-L	13	1	0.09	15	0.5	2.8	41
AC-ISO80-6-M	13	2	0.19	15	0.7	2.0	64
AC-ISO80-9-H	11	3	0.24	13	0.5	2.4	40
AC-ISO80-9-L	12	2	0.19	33	0.9	3.5	175
AC-ISO80-9-M	9	3	0.28	17	0.5	2.9	55
AC-PEG10-3-H	7	1	0.17	22	2.4	2.9	520
AC-PEG10-3-L	11	1	0.11	24	3.3	2.8	763
AC-PEG10-3-M	10	3	0.27	20	1.1	3.0	148
AC-PEG30-3-H	21	6	0.26	13	11.4	1.3	1321
AC-PEG30-3-L	21	6	0.28	9	13.1	1.2	1188
AC-PEG30-3-M	15	5	0.31	12	6.4	1.4	616
AC-PEG30-6-H	14	3	0.19	12	1.7	1.8	175
AC-PEG30-6-L	11	2	0.14	18	0.7	2.6	93
AC-PEG30-6-M	16	3	0.17	9	1.9	1.7	105
ET-3-H	18	5	0.27	1	0.3	0.7	2
ET-3-L	23	4	0.18	3	0.2	1.9	4
ET-3-M	18	5	0.25	1	0.5	0.3	4
ET-6-H	16	5	0.30	1	0.3	0.3	2

ET-6-L	15	5	0.35	1	0.3	0.5	3
ET-6-M	16	5	0.30	1	0.4	0.5	3
ET80-3-H	18	2	0.13	-	-	-	-
ET80-3-L	17	3	0.18	21	1.0	2.6	94
ET80-3-M	17	1	0.05	25	0.5	4.2	70
ET80-6-H	11	2	0.20	0	0.0	0.0	0
ET80-6-L	13	1	0.09	11	0.5	2.9	45
ET80-6-M	15	2	0.17	-	-	-	-
ET90-3-H	25	4	0.17	22	0.6	3.4	68
ET90-3-L	17	4	0.25	9	0.2	4.7	12
ET90-3-M	25	5	0.19	11	0.3	3.2	18
ET90-6-H	13	2	0.13	-	-	-	-
ET90-6-L	14	3	0.18	21	0.6	3.7	65
ET90-6-M	12	1	0.13	-	-	-	-
ET-9-H	14	3	0.18	3	0.4	0.8	8
ET-9-L	13	3	0.22	2	0.4	0.5	5
ET-9-M	12	3	0.21	1	0.2	0.8	1
ISO-12-L	11	4	0.39	4	0.5	0.8	12
ISO-12-M	12	2	0.16	6	0.8	0.8	34
ISO-3-H	16	4	0.24	3	0.9	0.5	19
ISO-3-L	22	3	0.15	5	0.8	0.7	23
ISO-3-M	20	3	0.16	3	0.5	0.6	8
ISO-6-H	16	3	0.22	2	1.1	0.2	15
ISO-6-L	17	2	0.13	6	0.7	0.9	24
ISO-6-M	14	2	0.16	5	1.0	0.5	29
ISO80-12-H	10	2	0.25	12	0.5	2.2	34
ISO80-12-L	9	2	0.27	11	0.5	2.9	33
ISO80-12-M	11	3	0.26	24	0.8	3.1	108
ISO80-3-H	15	5	0.36	9	0.2	3.0	16
ISO80-3-L	15	4	0.26	8	0.3	2.6	13
ISO80-3-M	16	4	0.27	12	0.3	3.6	24
ISO80-6-L	11	4	0.32	15	0.9	4.2	143
ISO80-6-M	10	4	0.38	10	0.3	2.7	23
ISO80-9-H	9	2	0.26	16	0.9	2.1	94
ISO80-9-L	14	4	0.26	21	0.7	3.5	88
ISO80-9-M	13	3	0.20	28	0.8	3.6	136
ISO90-12-H	9	2	0.27	3	0.9	0.5	20
ISO90-12-L	11	4	0.33	14	0.5	2.9	38
ISO90-12-M	12	3	0.23	18	0.7	2.6	68
ISO90-3-H	17	4	0.21	10	0.4	2.6	23
ISO90-3-L	18	6	0.35	6	0.2	2.8	12

ISO90-3-M	16	2	0.13	8	0.3	4.1	13
ISO90-6-H	14	4	0.26	7	0.3	2.5	10
ISO90-6-L	11	3	0.23	12	0.4	3.4	41
ISO90-6-M	12	2	0.20	13	0.4	3.6	37
ISO90-9-H	13	3	0.27	6	0.3	2.1	11
ISO90-9-L	10	3	0.32	10	0.2	3.3	16
ISO90-9-M	11	2	0.17	27	1.1	3.3	188
ISO-9-H	13	3	0.24	5	1.0	0.6	34
ISO-9-L	15	3	0.22	4	0.2	1.6	5
ISO-9-M	13	4	0.33	2	0.6	0.3	9
PEG30-3-H	21	6	0.29	14	1.4	1.4	138
PEG30-3-L	20	3	0.14	15	11.6	1.7	1725
PEG30-3-M	22	5	0.22	16	0.7	2.2	58
PEG30-6-H	18	5	0.26	9	1.0	1.3	71
PEG30-6-L	29	12	0.43	12	1.5	1.5	115
PEG30-6-M	23	5	0.23	10	1.0	1.1	66

**Table S1:** Results obtained from the morphological and mechanical characterization of the as spun fibers. Fibers where no mechanical results are available correspond to fibers that were too brittle that it was not possible to perform the tensile tests. The code is defined with the code of the coagulant (see table 1) followed by the value of  $V_{R1}$  in m/min followed by a letter (L for low, M for medium and H for high) relative to the value of the flow rate of the focusing fluid.

**Table S2**

<b>Code</b>	<b><math>\sigma_u</math> (MPa)</b>	<b><math>\epsilon_u</math> (%)</b>	<b><math>W_f</math> (MJ/m<sup>3</sup>)</b>
AC-ET80-12-L	5.5	172	6.19
AC-ET80-3-H	7.1	213	9.60
AC-ET80-3-L	9.9	246	13.56
AC-ET80-3-M	3.2	92	2.26
AC-ET80-6-H	4.9	174	5.66
AC-ET80-6-L	9.5	184	10.13
AC-ET80-6-M	4.1	74	2.21
AC-ET80-9-M	4.2	117	3.23
AC-ISO80-12-H	1.5	28	0.30
AC-ISO80-12-L	5.8	27	0.63
AC-ISO80-12-M	4.3	82	2.53
AC-ISO80-3-H	4.0	154	4.28
AC-ISO80-3-L	8.7	137	6.84
AC-ISO80-3-M	6.2	165	6.21
AC-ISO80-6-H	4.9	135	4.14
AC-ISO80-6-L	8.3	94	4.78
AC-ISO80-6-M	16.3	185	16.16
AC-ISO80-9-H	1.6	61	0.55
AC-ISO80-9-L	10.0	87	5.28
AC-ISO80-9-M	2.9	48	0.98
AC-PEG10-3-H	14.7	72	6.03
AC-PEG10-3-L	13.3	184	12.64
AC-PEG10-3-M	9.1	133	6.39
AC-PEG30-3-H	11.1	120	7.37
AC-PEG30-3-L	5.6	130	4.31
AC-PEG30-3-M	12.5	106	7.36
AC-PEG30-6-H	2.0	76	0.91
AC-PEG30-6-L	5.5	88	3.28
AC-PEG30-6-M	2.2	102	1.29
ET-3-H	0.3	16	0.03
ET-3-L	0.4	33	0.09
ET-3-M	0.2	16	0.02
ET-6-L	0.5	17	0.05
ET-6-M	0.5	33	0.10
ET80-3-H	3.5	66	1.69
ET80-3-L	5.8	139	5.15
ET80-3-M	6.1	227	8.04

ET80-6-L	8.4	242	10.42
ET80-6-M	5.6	142	5.30
ET90-3-L	2.0	17	0.25
ET90-6-L	1.2	38	0.37
ET-9-H	0.4	39	0.10
ET-9-M	0.5	75	0.26
ISO80-12-H	3.5	69	1.59
ISO80-12-L	2.6	41	0.63
ISO80-12-M	1.0	58	0.28
ISO80-3-H	6.5	90	3.77
ISO80-3-L	4.0	71	1.85
ISO80-3-M	4.0	63	1.68
ISO80-6-L	6.1	64	1.93
ISO80-6-M	3.9	54	1.37
ISO80-9-H	1.8	57	0.79
ISO80-9-L	2.2	28	0.34
ISO80-9-M	2.6	66	0.99
ISO90-12-L	3.2	47	0.94
ISO90-3-L	1.1	48	0.32
ISO90-3-M	3.0	56	1.12
ISO90-6-H	0.9	24	0.13
ISO90-6-L	2.6	67	1.03
ISO90-6-M	3.6	41	1.02
ISO90-9-H	2.1	43	0.54
ISO90-9-L	2.3	37	0.41
ISO90-9-M	3.7	48	1.11
PEG30-3-H	1.1	26	0.16
PEG30-3-L	2.2	136	1.59
PEG30-3-M	1.7	129	1.25
PEG30-6-H	8.9	177	8.80
PEG30-6-L	0.8	78	0.33
PEG30-6-M	5.0	265	5.15

**Table S2:** Mechanical properties of the regenerated fibers tested in water at 25°C. The fibers that do not appear in this table are those that were no possible to test either due to their brittleness or because they were water soluble. The code is defined with the code of the coagulant (see table 1) followed by the value of  $V_{R1}$  in m/min followed by a letter (L for low, M for medium and H for high) relative to the value of the flow rate of the focusing fluid.

**Table S3**

<b>Code</b>	<b><math>\sigma_u</math> (MPa)</b>	<b><math>\epsilon_u</math> (%)</b>	<b><math>E_i</math> (Gpa)</b>	<b><math>W_f</math> (MJ/m<sup>3</sup>)</b>
AC-ET80-12-L	93	4.0	6.9	3.0
AC-ET80-3-L	66	34.9	4.7	20.2
AC-ET80-6-H	77	1.8	5.1	0.7
AC-ET80-9-M	73	1.4	5.5	0.6
AC-ISO80-3-M	42	0.8	4.5	0.2
AC-ISO80-6-M	78	10.5	2.8	6.8
AC-ISO80-9-L	102	1.4	8.3	0.9
AC-PEG10-3-L	41	3.8	2.6	1.3
AC-PEG30-3-L	35	2.6	2.9	0.7
AC-PEG30-6-M	38	4.0	2.6	1.3
ET80-3-M	46	4.4	3.6	1.5
ET80-6-L	59	13.7	3.1	6.9
ET90-6-L	10	2.5	0.3	0.1
ISO80-12-H	44	1.8	2.8	0.5
ISO80-3-L	43	1.3	3.9	0.3
ISO80-9-M	39	1.5	3.6	0.3
ISO90-6-M	13	1.0	1.5	0.1
ISO90-9-M	14	11.0	0.4	1.1
PEG30-3-L	134	32.8	6.5	37.5
PEG30-6-M	51	9.4	2.4	3.9

**Table S3:** Mechanical properties of the fibers after being subjected to wet-stretching in water.

Samples in bold letters correspond to those that are presented in figure 4.