

SUPPLEMENTARY INFORMATION

Trigger sequence can influence final morphology in the self-assembly of asymmetric telechelic polymers

Aatish Kumar,^a Christopher P. Lowe,^a Martien A. Cohen Stuart,^b and Peter G. Bolhuis^{*a}

In the supplementary information section we present several additional plots which are referred to in the main text. Fig. S1 shows the entire table of cluster sizes for the $N_b = 3$ and $N_b = 9$ collagen-silk systems for several volume fractions. In Fig. S2 we present the analog of Fig. 9 in the main text for larger systems, to investigate the influence of the system size. Fig. S3 and S4 show the results for the $N_b = 3$ and $N_b = 9$ collagen-silk systems at several volume fractions, while Fig. S5 does the same for the elastin-silk system. Fig. S6 depicts the initial silk node size distributions for silk-only activated $N_b = 3$ and $N_b = 9$ collagen-silk systems at several densities. In Fig. S7 we show the intra- and intercluster CC and SS radial distribution functions. Fig. S8 indicates that the silk node size distribution at maximum sticker attraction strength for the $N_b = 3$ and $N_b = 9$ collagen-silk systems. Finally, Fig. S9 presents the cluster network for $N_b = 3$ in an alternative way, namely by viewing each node as a single point in the graph. This highlights the concept of bridging collagen nodes, which connect two silk nodes. The trigger sequence dependence of this metric is shown in the table. Note that while this is a different network metric, the conclusions remain the same.

^a Amsterdam Center for Multiscale Modeling, van 't Hoff Institute for Molecular Sciences, University of Amsterdam, PO Box 94157 1090 GD Amsterdam, The Netherlands

^b Laboratory of Physical Chemistry and Colloid Science, Wageningen University, Dreijenplein 6, 6703 HB Wageningen, The Netherlands

* E-mail: p.g.bolhuis@uva.nl

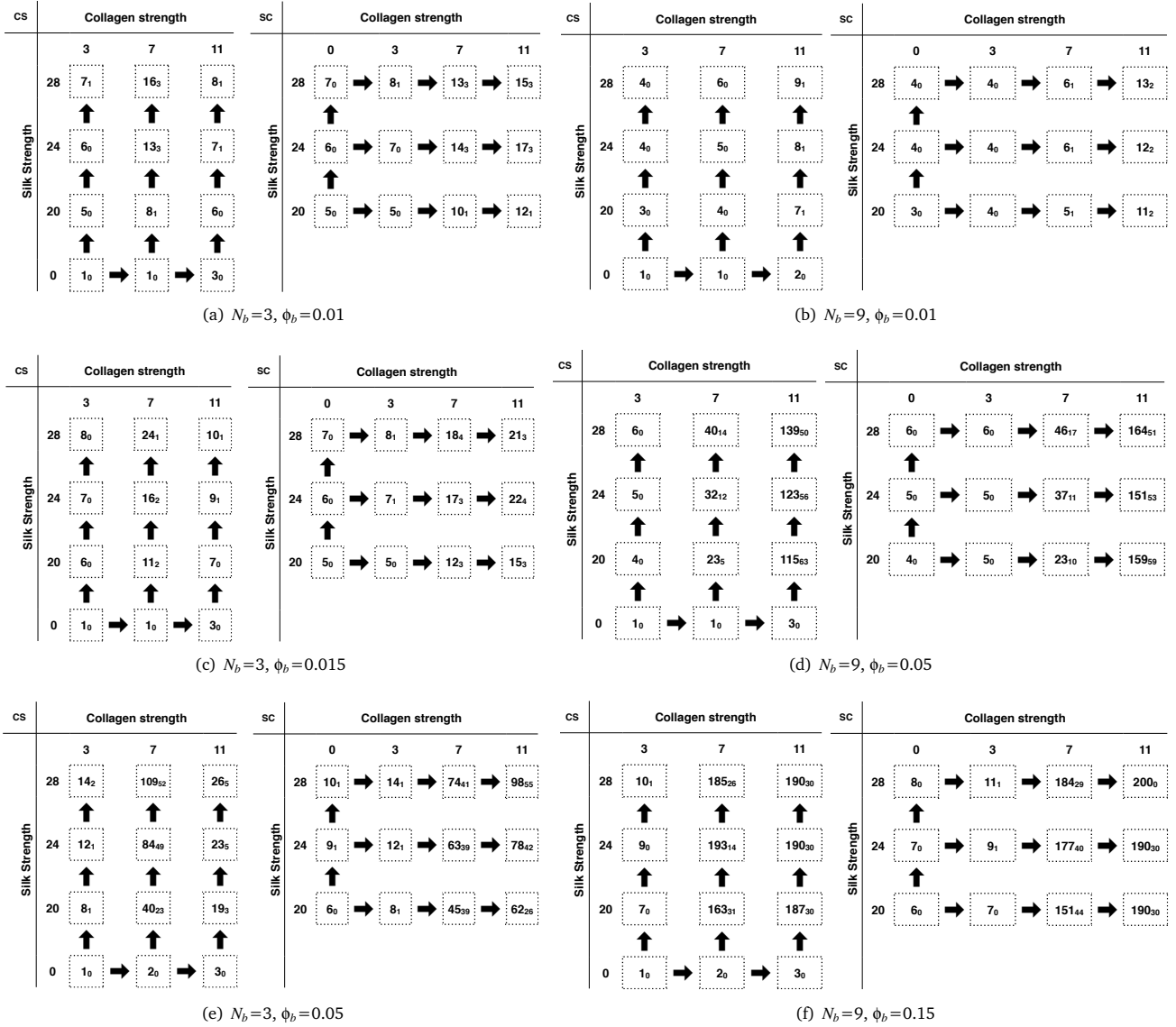


Fig. S1 Average cluster sizes for the collagen-silk TP systems, for $N_b=3$ (left column) and $N_b=9$ (right column) at several blob volume fractions. For each setting, the left graph shows the average cluster size for collagen-firs trigger sequence. The right graph are for a silk-first trigger sequence. The arrows denote the order in which the simulations have been performed, using the output of one simulation as input for the next.

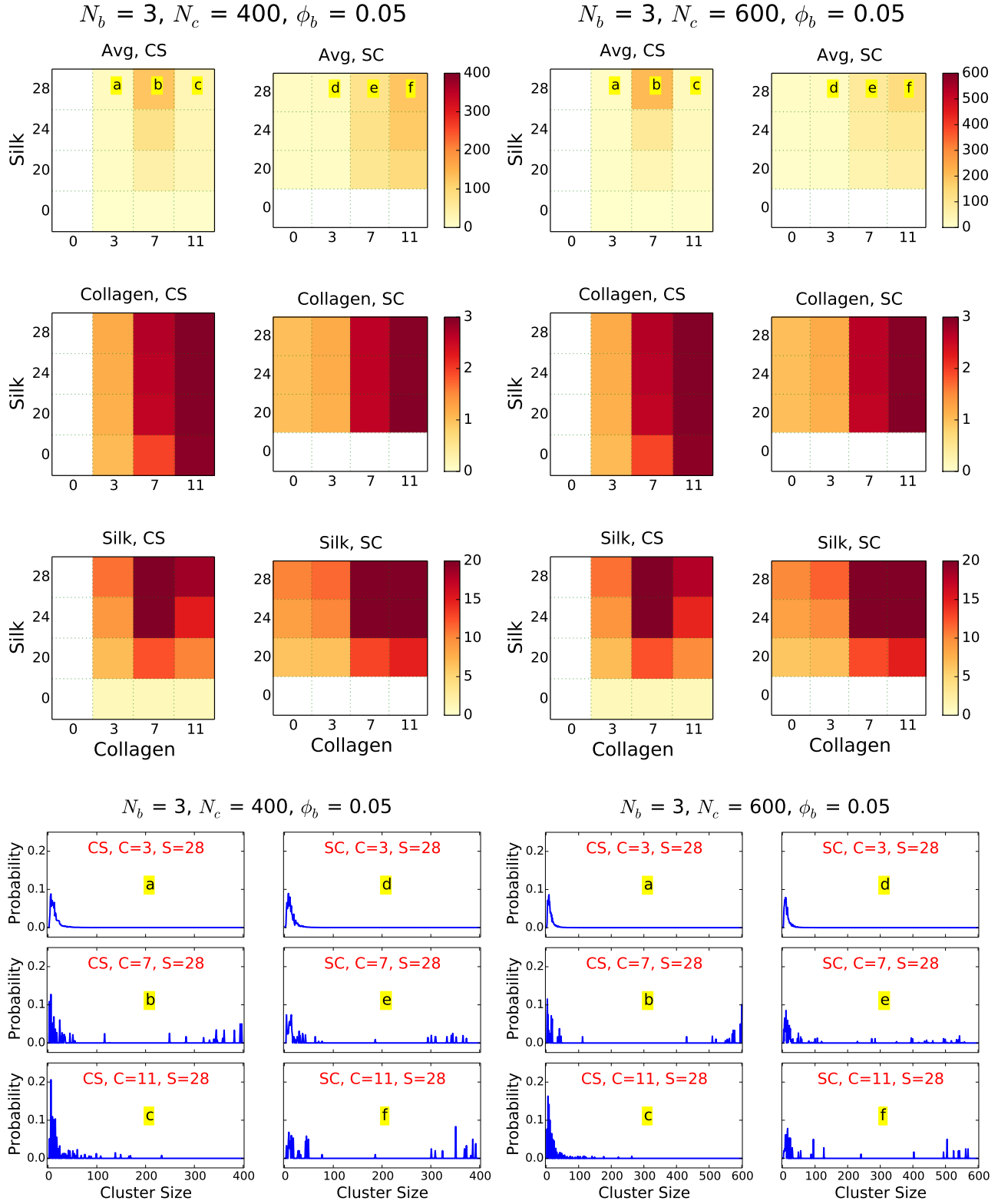


Fig. S2 Top: Average cluster and node sizes for collagen-silk TP for $N_b = 3$ for blob volume fractions, $\phi_b = 0.05$ and number of chains, $N_c = 400$ and 600 (left to right). Bottom: cluster size distributions corresponding to settings indicated by labels a-f in top row.

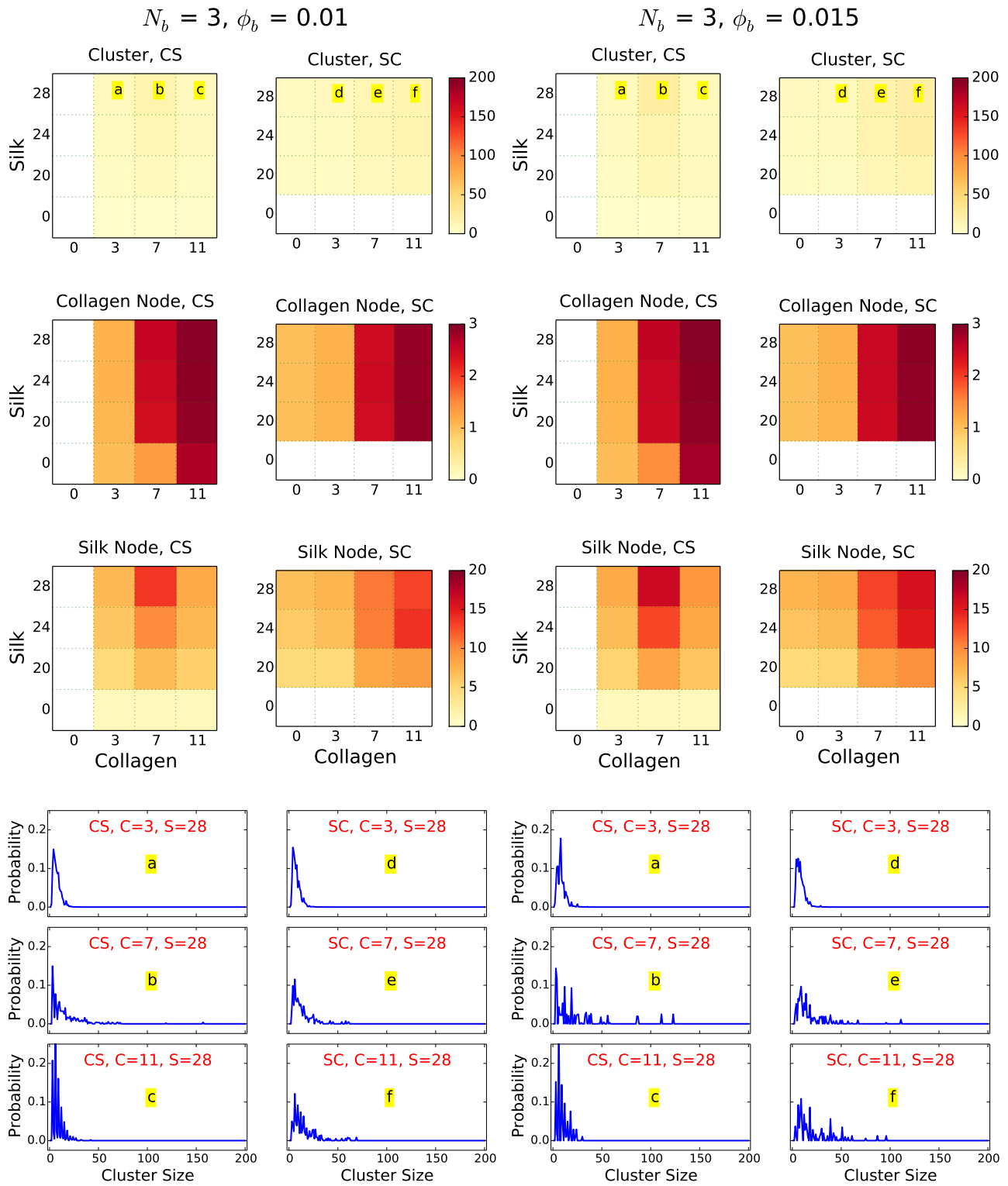


Fig. S3 Top: Average cluster and node sizes for collagen-silk TP for $N_b = 3$ for blob volume fractions ϕ_b of 0.01 and 0.015 (left to right). Bottom: cluster size distributions corresponding to settings indicated by labels a-f in top row.

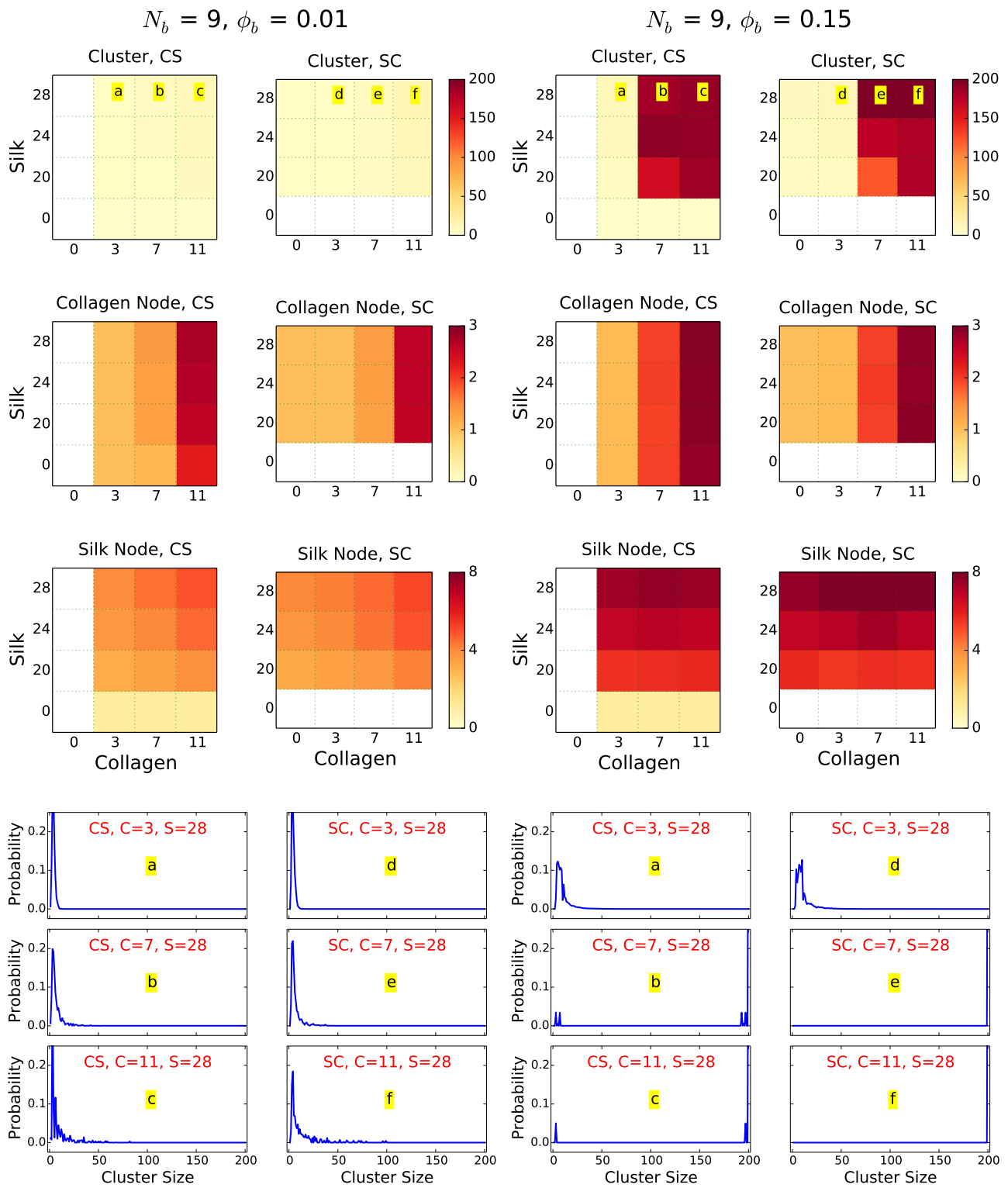


Fig. S4 Top: Average cluster and node sizes for collagen-silk TP for $N_b = 9$ for blob volume fractions ϕ_b of 0.01 and 0.15 (left to right). Bottom: cluster size distributions corresponding to settings indicated by labels a-f in top row.

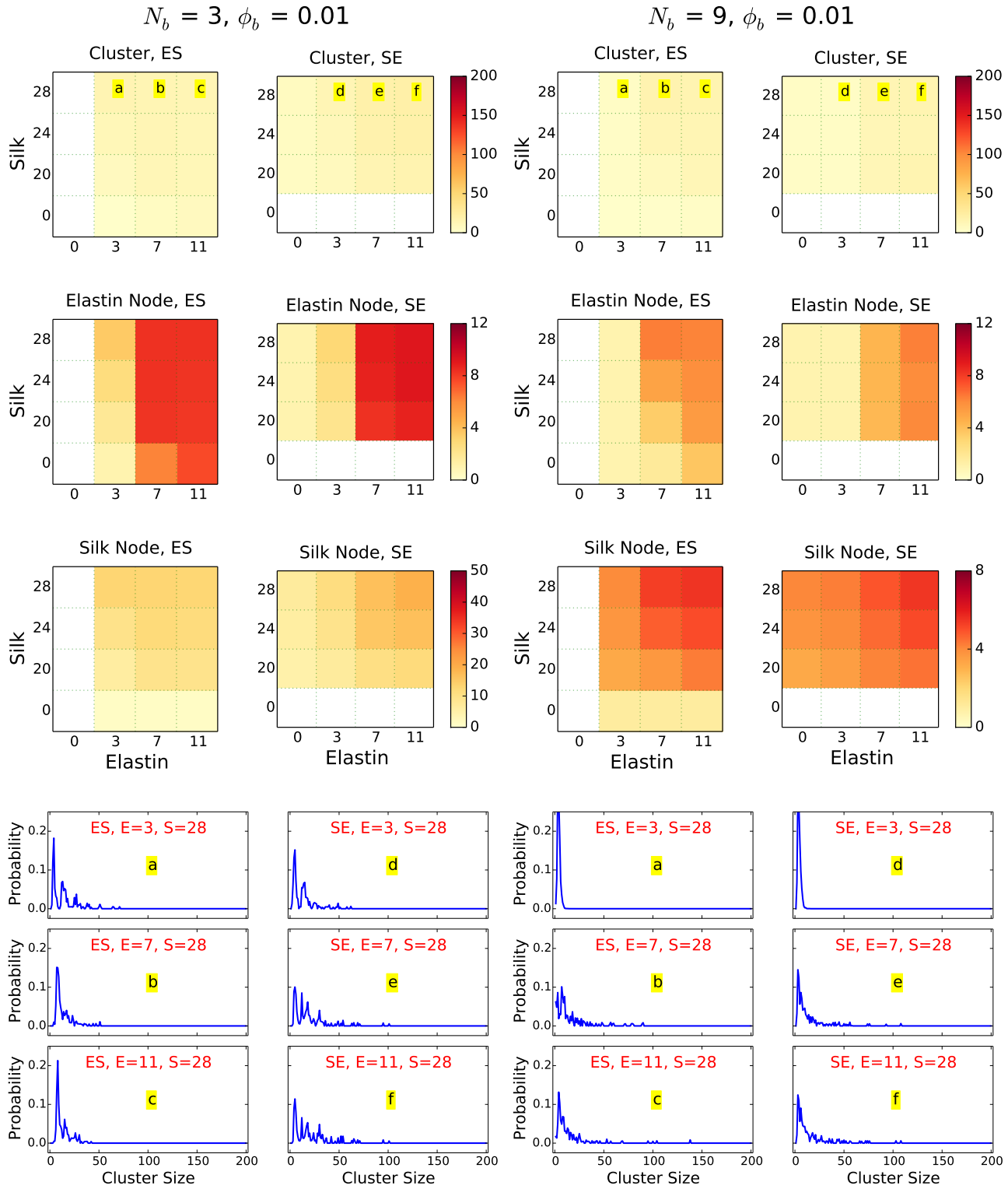


Fig. S5 Top: Average cluster and node sizes for elastin-silk TP for $N_b = 3$ (left) and $N_b = 9$ (right) for blob volume fraction $\phi_b = 0.01$. Bottom: cluster size distribution corresponding to settings indicated by labels a-f in top row.

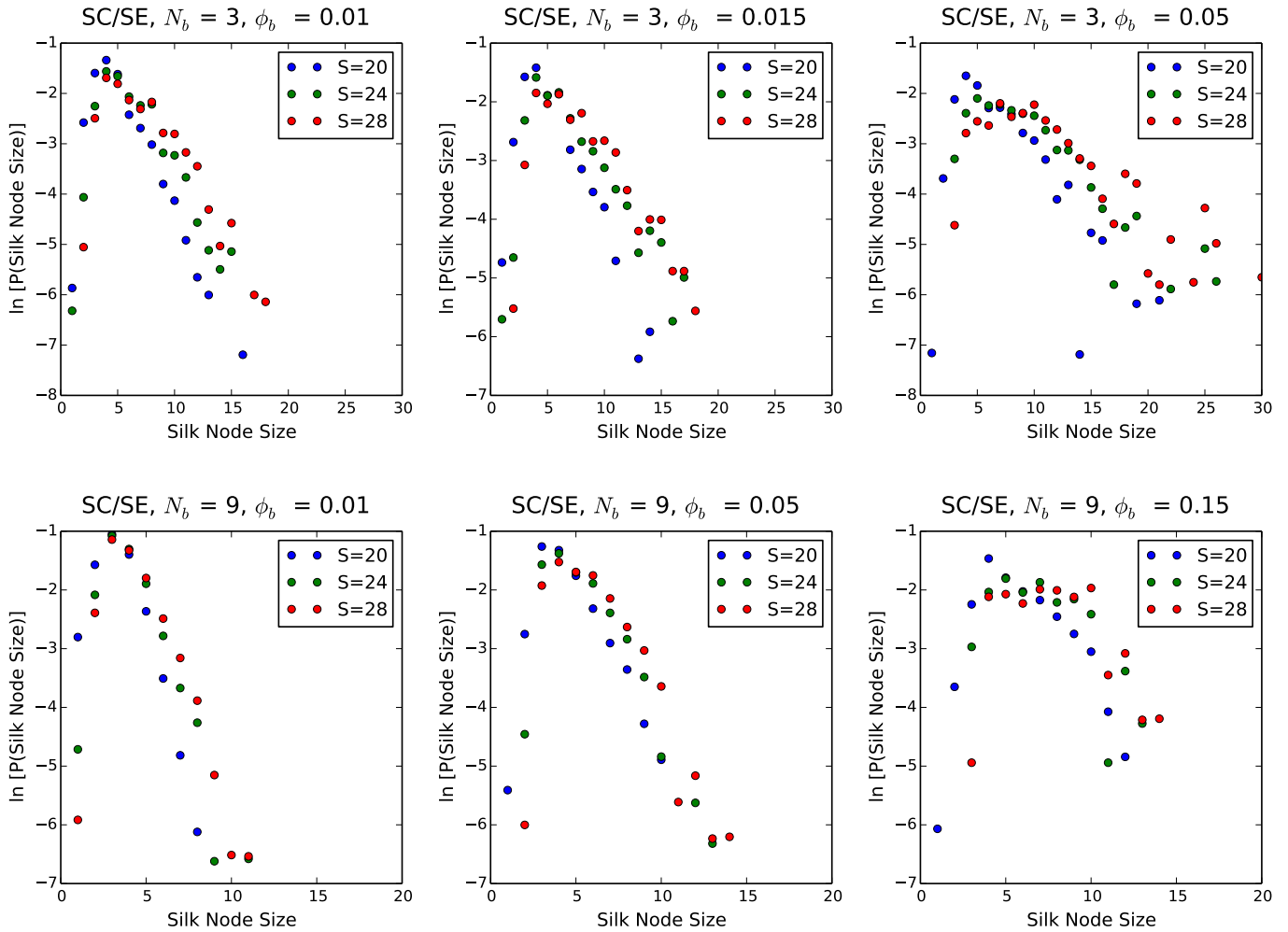


Fig. S6 Top: Initial silk node size distributions for $N_b = 3$ for blob volume fractions ϕ_b of 0.01, 0.015 and 0.05 (left to right). Bottom: Initial silk node size distributions for $N_b = 9$ for blob volume fractions ϕ_b of 0.01, 0.05 and 0.15 (left to right).

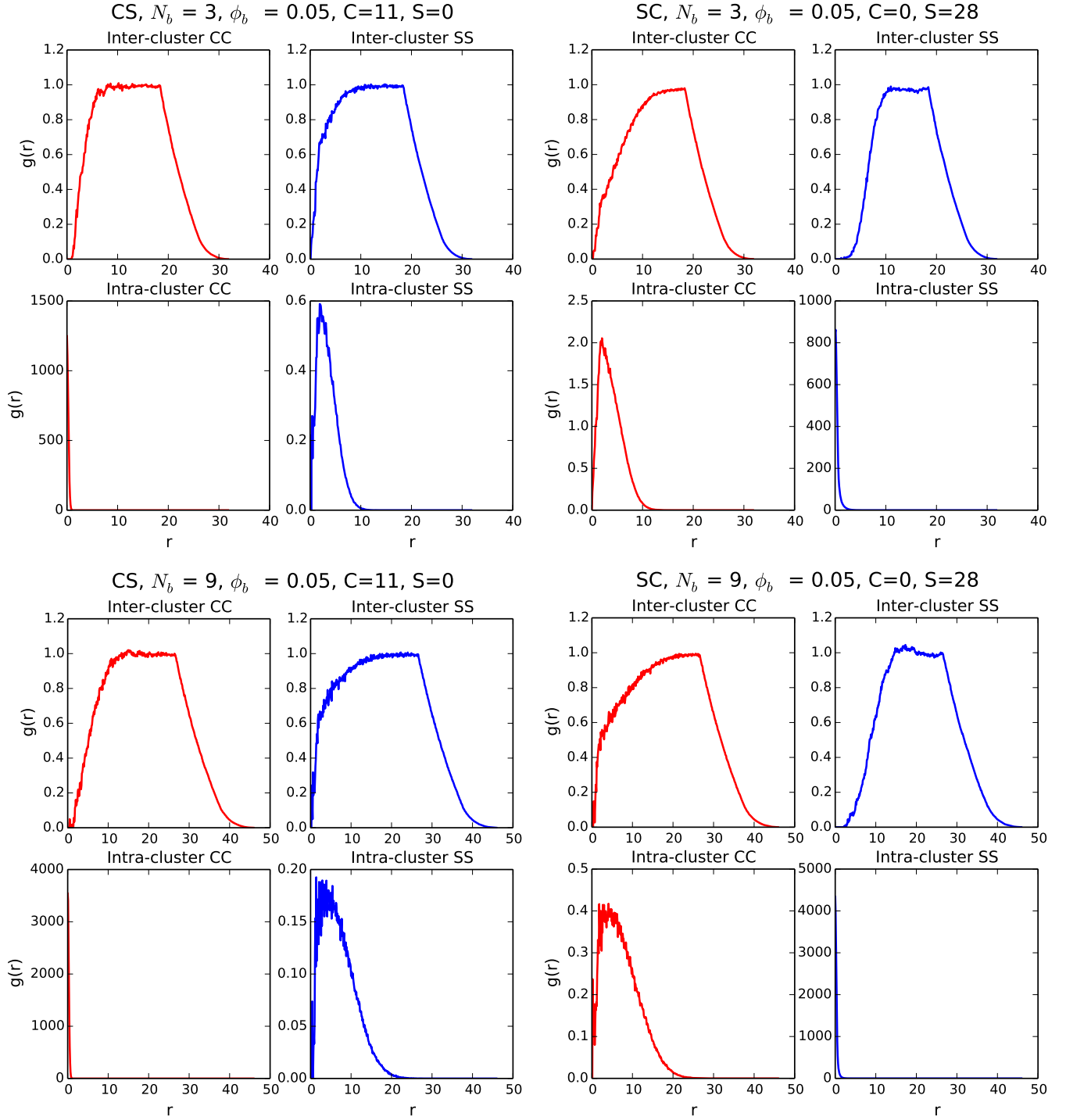


Fig. S7 Top: Inter- and Intra-cluster sticker radial distribution functions for $N_b = 3$ for blob volume fraction ϕ_b of 0.05 for CS and SC routes (left to right). Bottom: Inter- and Intra-cluster sticker radial distribution functions for $N_b = 9$ for blob volume fraction ϕ_b of 0.05 for CS and SC routes (left to right).

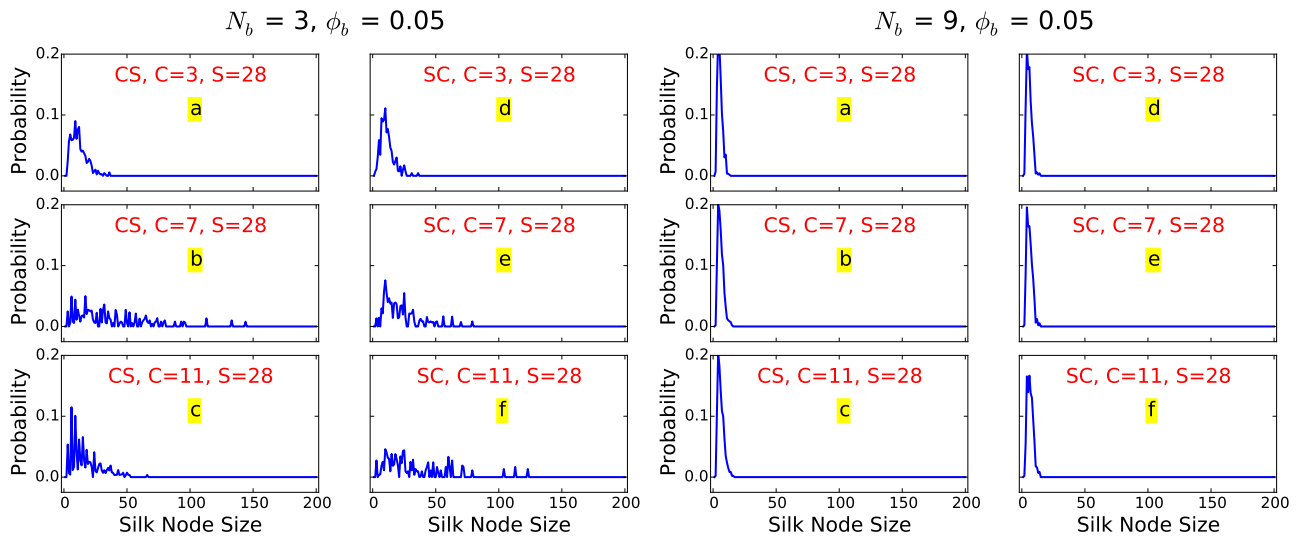


Fig. S8 Silk node size histograms for $N_b = 3$ (left) and $N_b = 9$ (right) for blob volume fraction ϕ_b of 0.05 for CS and SC routes. a-f as indicated in Fig. S3.

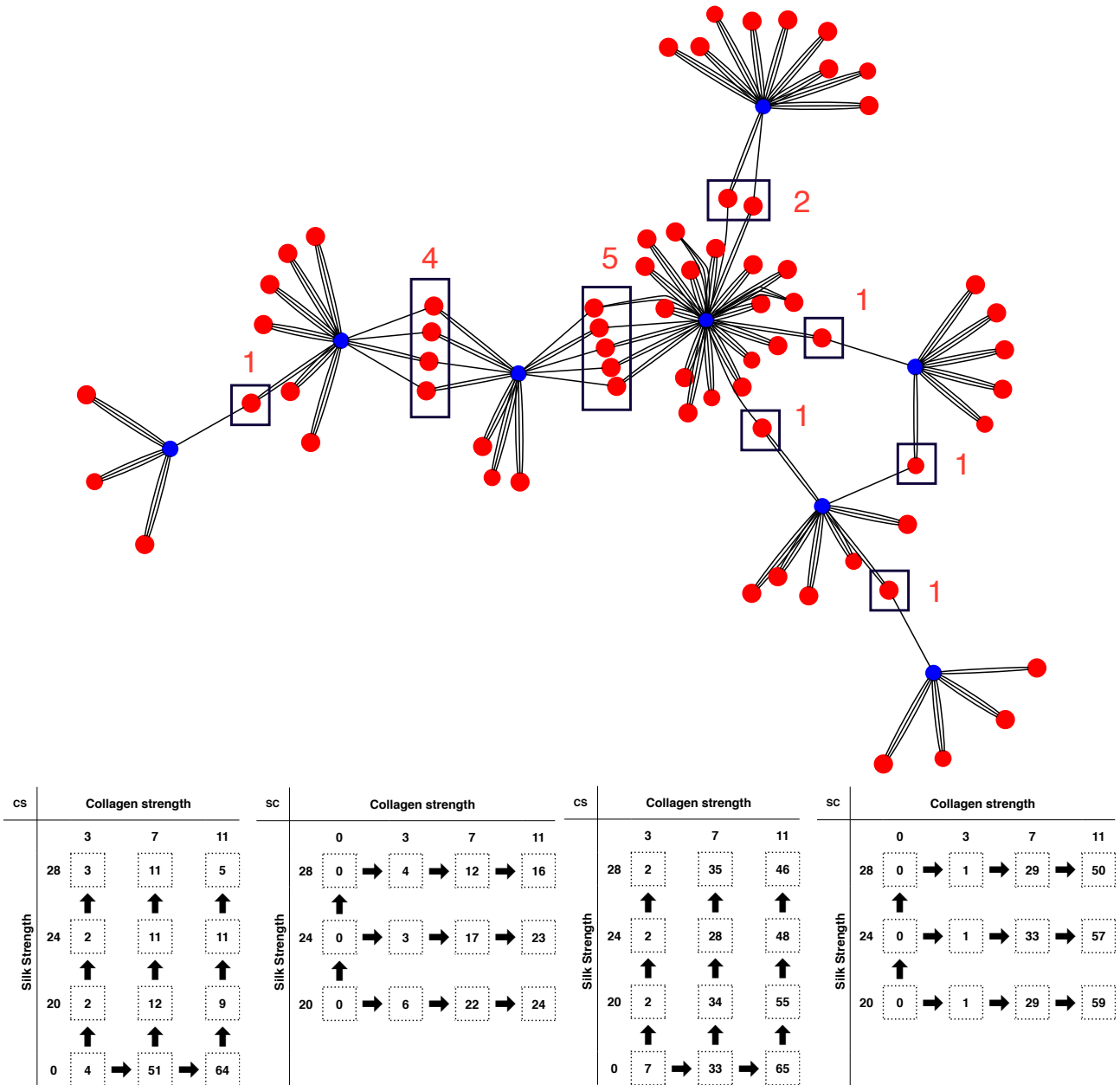


Fig. S9 Top: Alternative representation of graph network of Fig 7 bottom right ($N_b = 3$, $\phi_b = 0.05$, SC route, $C = 11$, $S = 28$). Red and blue circles indicate collagen and silk nodes whereas black lines indicate the polymer connections. Bridging collagen (indicated by black boxes) are collagen nodes which connect two silk nodes. Bottom: Bridging collagen for $N_b = 3$ (left) and $N_b = 9$ (right) for blob volume fraction ϕ_b of 0.05 for CS and SC routes for a single trajectory.