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## Supplemental information

## RNA chain length and stoichiometry govern surface

## tension and stability of protein-RNA condensates

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## Supplementary information

Energy Term	Parameter Values	Meaning
$k_b^P (r - r_0^P)^2$	$r_0^P = 0.38, \ k_b^P = 10$	Protein chain bonding
$k_b^R (r - r_0^R)^2$	$r_0^R = 0.34, \ k_b^P = 10$	RNA chain bonding
$k_{ heta}^R( heta- heta_0)^2$	$\theta_0 = 1.34, \ k_\theta^R = 1$	RNA chain stiffness
$\frac{q_i q_j}{4\pi Dr} exp(-\kappa r)$	$D = 80,  k = [1, \dots 3]$	Electrostatic interactions
$\boxed{\epsilon_{ij} \left[ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^6 \right]}$	$\epsilon_{ij}=1$ Values of $\sigma_{ij}$ come from Dignon et al. Plos Comp Bio 2018	Short-range attraction and excluded volume terms acting between all RNA and protein units

Table S1: Listing of all energy terms describing interactions of coarse-grained molecular models of RNA and RGG along with parameters and their physical meaning; Related to the main figure 1.



**Figure S1. Experimental state diagrams of RGG-PolyU mixtures; Related to the main figure 6.** Shown are phase diagrams at 150 mM NaCI (left) and 325 mM NaCI (right). Blue-filled circles indicate a homogenous mixed state. Red-filled circles indicate LLPS. The solid black line is drawn as a guide to the eye. Samples were prepared at the desired RGG and PolyU concentration in a buffer containing 25 mM Tris-HCI (pH 7.5), 20 mM DTT, and the indicated salt concentration.



**Figure S2.** Snapshots of simulations done under NPT ensemble; Related to the main figure 4. Showing snapshots of NPT simulations with (A) P=1 and (B) P=10 conditions demonstrate the potential changes in condensate density that may occur as a function of external crowding. Each panel shows a slice which is taken across the diagonal of a simulation box (top) and a 2nm slab is taken from a simulation box to better show the difference in packing of chains (bottom)



Figure S3. Relative entropy and energies of condensation; Related to the main figure 5 (A) Shannon entropy measure computed by image segmentation of simulation snapshots. Entropy is

measured relative to uniformly dissolved state (B) Average inter-molecular energy accounting for excluded volume, dispersion, and electrostatic forces.