

Supporting Information

Folding of Electrostatically Charged Beads-on-a-String: An Experimental Realization of a Theoretical Model in Polymer Science

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Construction of a random motion generator

To verify the randomness of the motion induced by the motion generator, we analyzed the two-dimensional motion of a dot on the surface. Our analysis included two calculations: First, we measured the displacement of the dot on arbitrary Cartesian coordinates and calculated the correlation function for incremental displacements along each axis. We collected 1000 images of the dot at a frame rate of 17 frames per second, and tabulated the x- and y-coordinates for the dot on each frame. We calculated the correlation function using equation 1:

$$\rho = \frac{\sum (x - \bar{x})(y - \bar{y})}{\sqrt{\sum (x - \bar{x})^2 (y - \bar{y})^2}}, \quad [1]$$

where ρ is the correlation function, x and y are the incremental positions of the dot on the x - and y -axes, and \bar{x} and \bar{y} are the average positions of the dot. The value of -0.2 for the correlation function indicated that the motion of the surface along one axis was not correlated with motion along the orthogonal axis.

Second, we plotted the frequency distribution for incremental changes in angle for the dot on polar coordinates (**Fig. S1**). The normal Gaussian distribution we observed for this plot indicated that the motion of the surface was random (1).

Selection of PMMA for the Spacer Beads

We selected PMMA for two reasons: (i) PMMA is located between Teflon and Nylon in the triboelectric series. We assumed that upon collision of the beads, similar amounts of positive (from Teflon) and negative (from Nylon) charge would transfer to the PMMA beads, and thus collision among the beads would result in PMMA beads that were approximately electrically neutral, (ii) these beads do not contact the surface on which we agitate the chains; therefore these beads did not tribocharge against the surface. Given these two assumptions, the approximately electrically neutral spacer beads should not interact appreciably with the larger electrostatically charged beads in this model. We verified these assumptions.

Characterization of Electrical Charging: Individual Spherical Beads

We first measured the charge accumulated on the surfaces of individual beads. We agitated a set of 20 Teflon (T), 20 Nylon (N) and 40 PMMA (P) beads in the stainless steel bowl (orbital motion), and measured the charge on ten of each bead. The values of charges on the beads were: $q_T = -1400 \pm 200$ pC, $q_P = +400 \pm 25$ pC, and $q_N = +600 \pm 200$ pC.

In order to confirm that the dye did not significantly change the magnitude of the charge accumulated on the beads, we measured the charge on the surface of the dyed beads and compared this charge to that of the undyed beads. We agitated mixtures of dyed or undyed Nylon beads with equal numbers of Teflon beads (7–10 of each bead) on a steel surface. The

charge measured on the undyed Nylon beads, $q_N^{\text{undyed}} = +1100 \pm 200$ pC, with $q_T^{\text{undyed}} = -1000 \pm 300$ pC. The charge on the dyed Nylon beads, $q_N = +890 \pm 150$ pC, with $q_T = -1200 \pm 300$ pC. We concluded that the difference in charge between the dyed and undyed Nylon beads was not statistically significant.

Characterization of Electrical Charging: Individual Cylindrical Beads

We first measured the charge accumulated on the surfaces of individual beads at RH \sim 20%. We agitated a set of 20 Teflon (T), 20 Nylon (N) cylindrical beads and 20 PMMA (P) spherical beads in a stainless steel bowl covered with paper (orbital motion), and measured the charge on ten of each bead. The values of charges on the beads were: $q_T = -3850 \pm 1350$ pC, $q_P = +180 \pm 40$ pC, and $q_N = +3330 \pm 1300$ pC.

We increased the humidity to \sim 70%, agitated the beads till there were no evident interactions among the beads and measured the charge on ten of each bead. The values of charges on the beads were significantly lower than the values measured in RH \sim 15-30%: $q_T = -390 \pm 130$ pC, $q_N = +125 \pm 60$ pC and the charge on the PMMA beads (q_P) was in the noise range.

Characterization of Electrical Charging: Beads on a String

We measured the charge accumulated on the surfaces of the beads on a string in low humidity (RH \sim 20%). We agitated a sequence of 7 Teflon (T), 7 Nylon (N) and 39 PMMA (P) beads on the surface of paper, and measured the charge on each bead. The values of charges on the spherical beads were: $q_T = -2200 \pm 430$ pC, $q_P = +56 \pm 25$ pC, and $q_N = +1833 \pm 230$ pC. The values of charges on the cylindrical beads were: $q_T = -4400 \pm 300$ pC, $q_P = +90 \pm 30$ pC, and

$q_N = +3600 \pm 300$ pC. Without the crimp beads, the values of charges on the cylindrical beads were: $q_T = -4700 \pm 300$ pC, $q_P = +90 \pm 50$ pC, and $q_N = +3700 \pm 600$ pC.

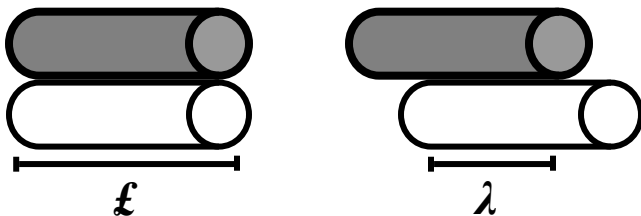
Agitating the beads on a flat surface

We agitated the beads on surfaces made of either an insulating material such as paper or ceramic, or of a conductive material such as aluminum or gold.

In initial experiments, we charged the beads by agitating them on a flat aluminum surface surrounded by an aluminum frame. We noticed that in 90% of the experiments the sequence of beads reached the aluminum frame that surrounded the surface and remained close to the frame and interacted with the aluminum frame in addition to interacting with each other. Covering the inside of the frame with materials that are located in the middle of the triboelectric series, like paper or cotton did not prevent this phenomenon. Changing the geometry of the surface from a flat surface to a surface with a slight curvature (radius ~ 3 cm) at the perimeter (**Fig. 1C**) prevented this interaction.

Definition of a Pair for Cylindrical beads

We define beads to be paired when the length of overlap, λ , between them is 60% of their length, ℓ .



If $\lambda \geq 0.6\ell$, then pair

References

1. Nelson P in *Biological Physics: Energy, Information, Life* (W. H. Freeman, New York, 2008), pp 109-129.

Figure Captions

Fig. S1. Analysis of the radial motion of the surface. The histogram describes the angular displacement in time for the motion of the surface. The Cartesian coordinates of a point on the surface were collected for each frame and the angular displacements were converted to polar coordinates by calculating the arctangent of the ratio of $(y_i - y_{i+1})/(x_i - x_{i+1})$. The solid line represents a Gaussian fit of the counts in each bin for the 999 values of angular displacement observed.

Fig. S2. Images of the stable conformations (conformation that persisted for more than an hour) observed with sequences of 10 cylindrical beads. **(A)** The image represents a final conformation of a sequence of beads without spacer beads. **(B)** Each image represents a final conformation of a sequence with four PMMA beads. **(C)** Each image represents a final conformation of a sequence with five spacer beads.

Figure S3. Time-lapse photographs capture the motion of a sequence of 40 Nylon cylindrical beads on agitation. With time, the string of beads became more extended, rather than more condensed.

Figure S4. The plots represent the number of correct pairs of beads formed when folding a sequence of 40 cylindrical beads with alternating 5 Nylon and 5 Teflon beads and three spacer beads. Each plot corresponds to an individual experiment.