Electronic Supplementary Material (ESI) for Materials Advances.

Electronic Supporting Information

Formation of Liquid Marbles & Aggregates: Rolling and Electrostatic Formation Using Conductive Hexagonal Plates

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Micrographs of PET plates

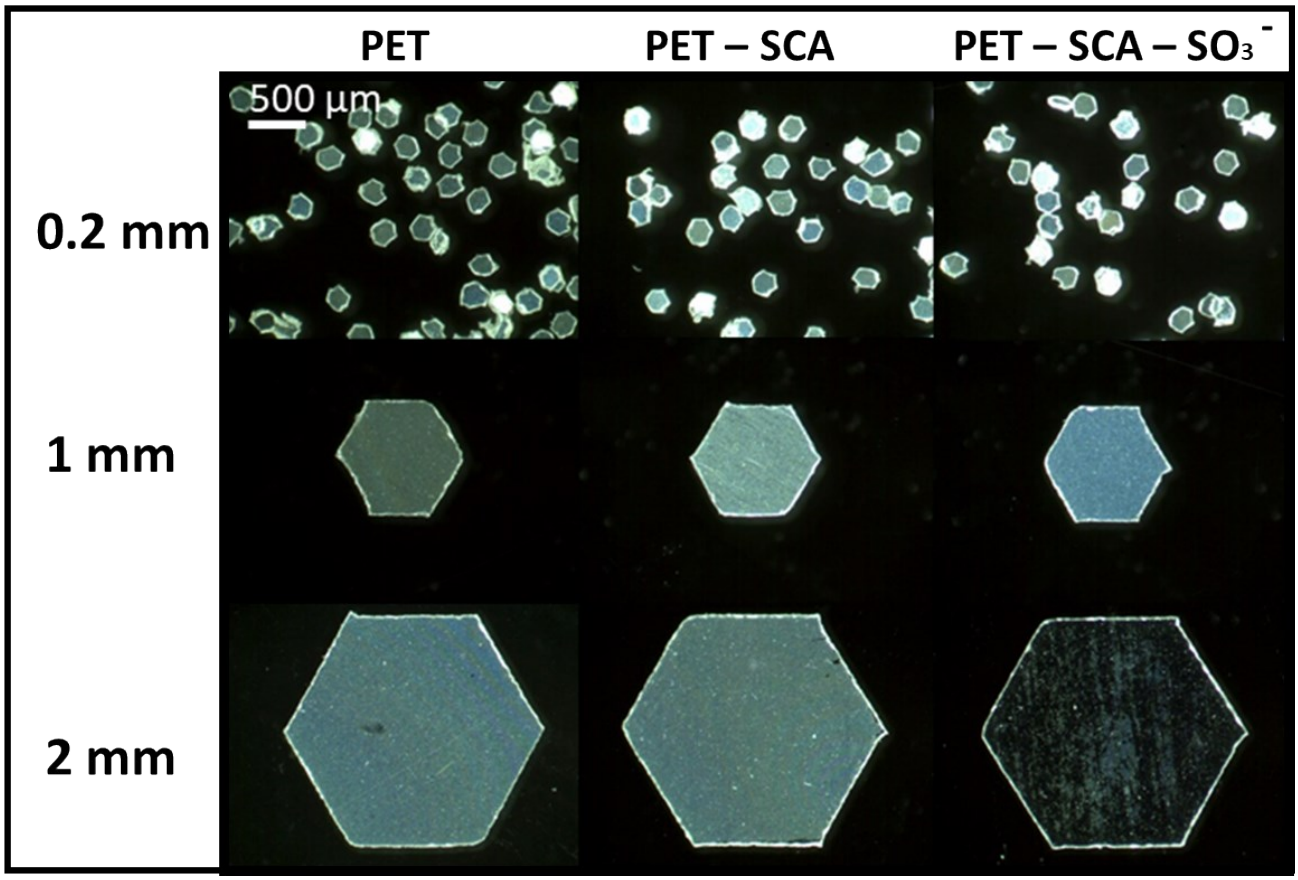

Figure S1. Optical microscopy images of uncoated PET plates.

SEM images used to determine coating thickness of polymer coating

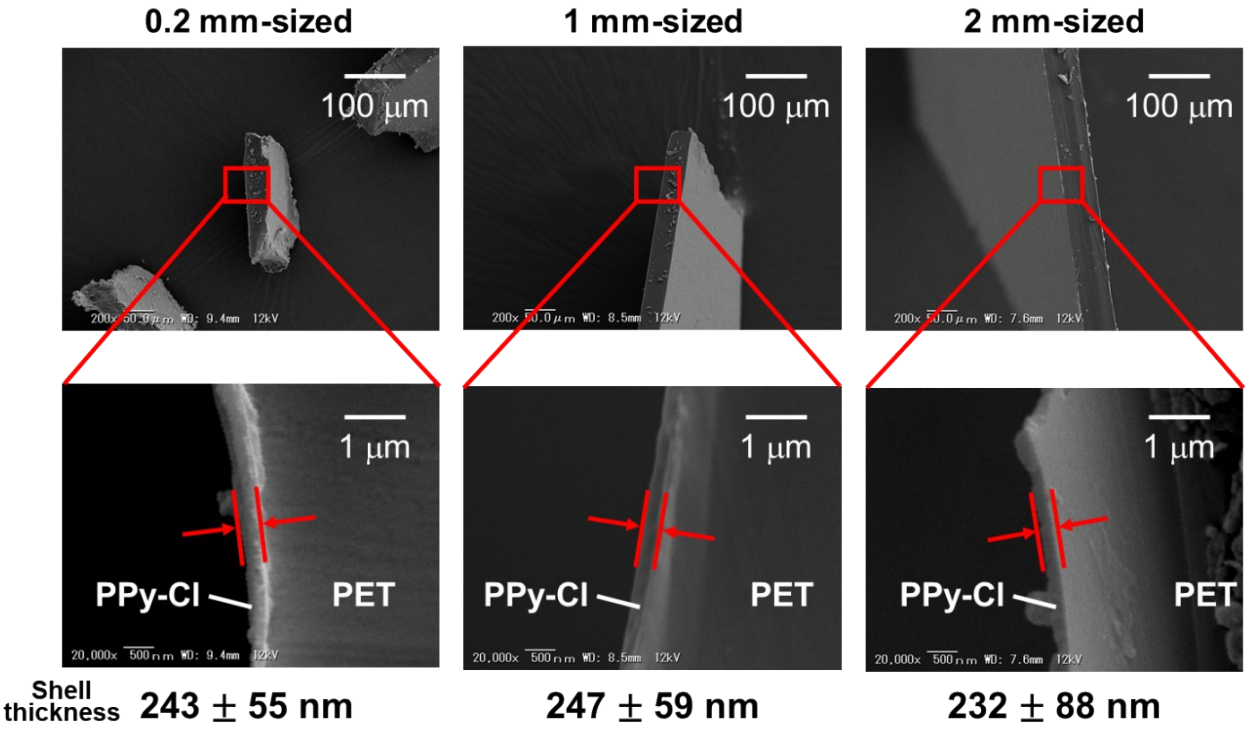

Figure S2. Annotated SEM images of edge coating thickness P of PPy-Cl on PET substrate.

SEM images used to determine coating thickness of polymer coating

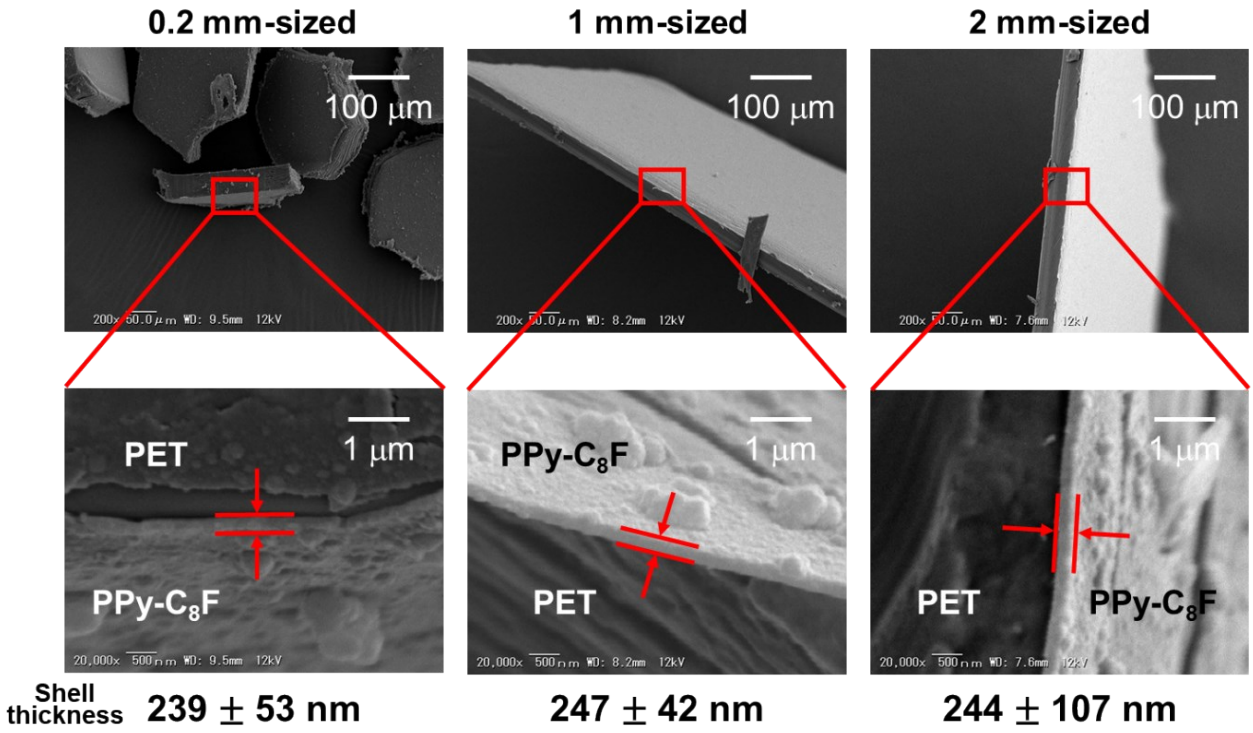

Figure S3. Annotated SEM images of edge coating thickness P of PPy-C_8F on PET substrate.

Photos of particle prepared particle bed showing orientation of platelet particles before experiments

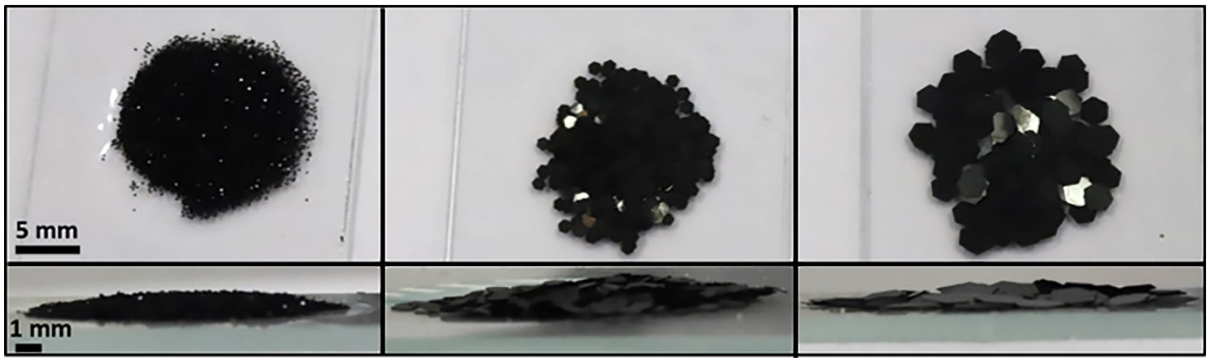

Figure S4. Representative particle beds (left to right: 0.2, 1 and 2 mm platelets) after being gently dropped on to metal plate from approximately 5 mm ten times. Top row: top-down image. Bottom row: side image. The photos demonstrate that the bed thickness is less than 2.5 mm in each case

Mass and volume calculations

Volume and mass % of coating were determined using the thickness of the coating observed in Figures S2 & S3. Particles were treated as if hexagonal prisms in which case volume may be calculated via the following formula:

$$V_{PET\ Plate} = \frac{3\sqrt{3}}{2} S^2 T$$

where S and T are the side length and platelet width (Figure S5). The volume of the coating may be determined by treating the edges of the platelet as rectangular prisms of height equal to coating thickness and each face as a hexagonal prism of thickness equal to P (Figure S2 & S3).

$$V_{Coating} = \left(2 \times \frac{3\sqrt{3}}{2} S^2 P\right) + (6SP[T + 2P])$$

$$Volume\% = \frac{V_{Coating}}{V_{PET\ Plate}} \times 100\%$$

Similarly,

$$Mass\% = \frac{V_{Coating}\rho_1}{V_{PET\ Plate}\rho_2} \times 100\%$$

Where ρ_1 and ρ_2 are the density of PET and PPy respectively.

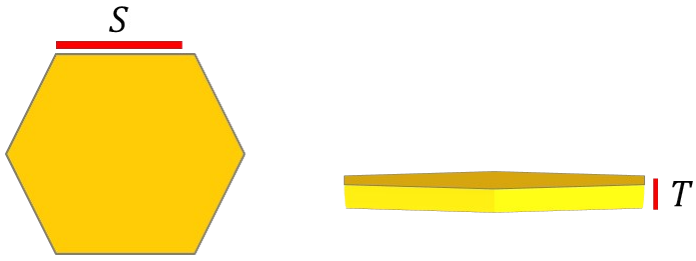

Figure S5. Schematic detailing hexagon dimensions

Rolled marbles formed from 0.2 mm platelets before coating with PPy

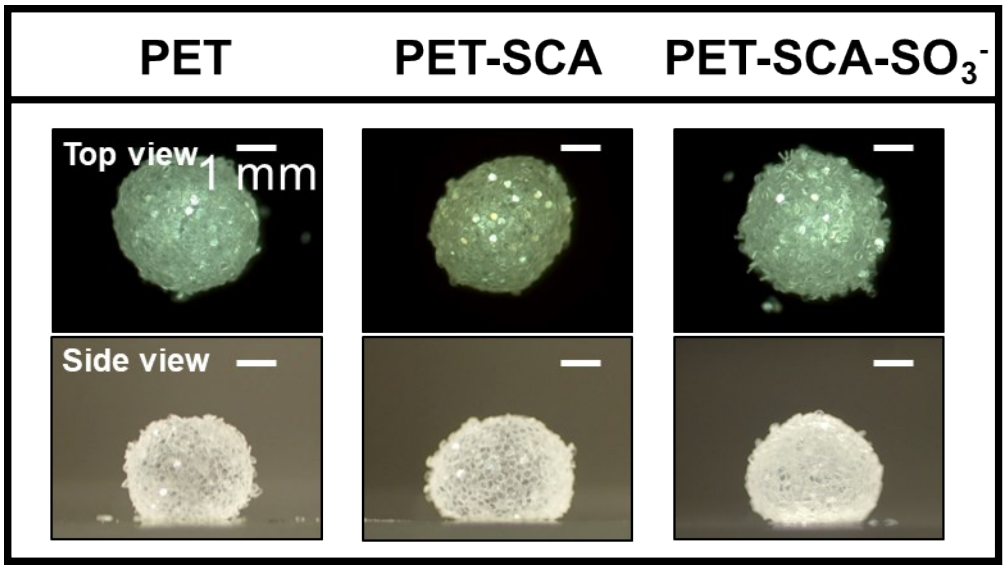

Figure S6. Attempts at rolled liquid marbles with 0.2 mm platelets before coating with PPy. Internal liquid is 15 μL. PET plates were unable to maintain stability after compression. The PET-SCA demonstrated some stability due to the lower interfacial energy of the particles after coating. Finally the PET-SCA-SO_3^- only showed limited stability.

Rolled liquid marbles being subjected to compressive force

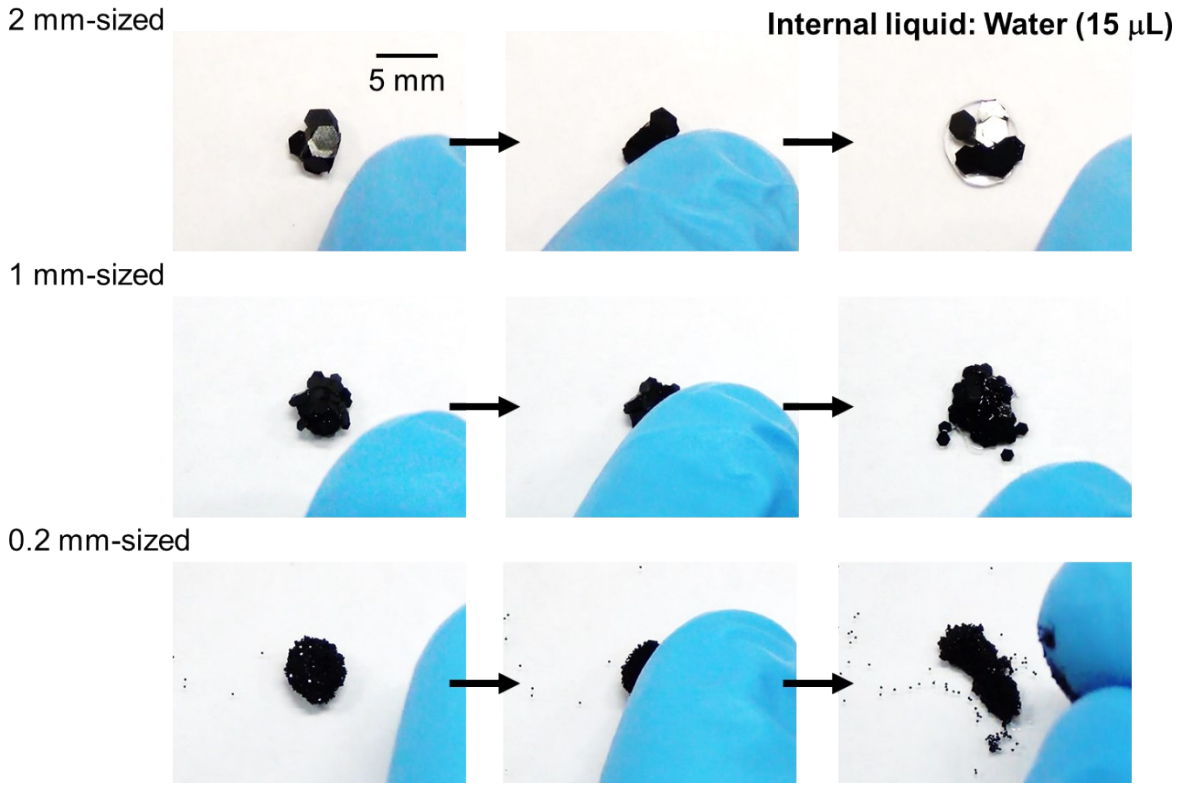

Figure S7. PET-PPy-Cl stabilised droplets of all tested sizes before and after compressive pressure applied. This demonstrates the tendency of the water to adhere to the platelets due to their hydrophilic nature; this is especially prominent in the 0.2 mm sized plates.

Rolled liquid marbles being subjected to compressive force

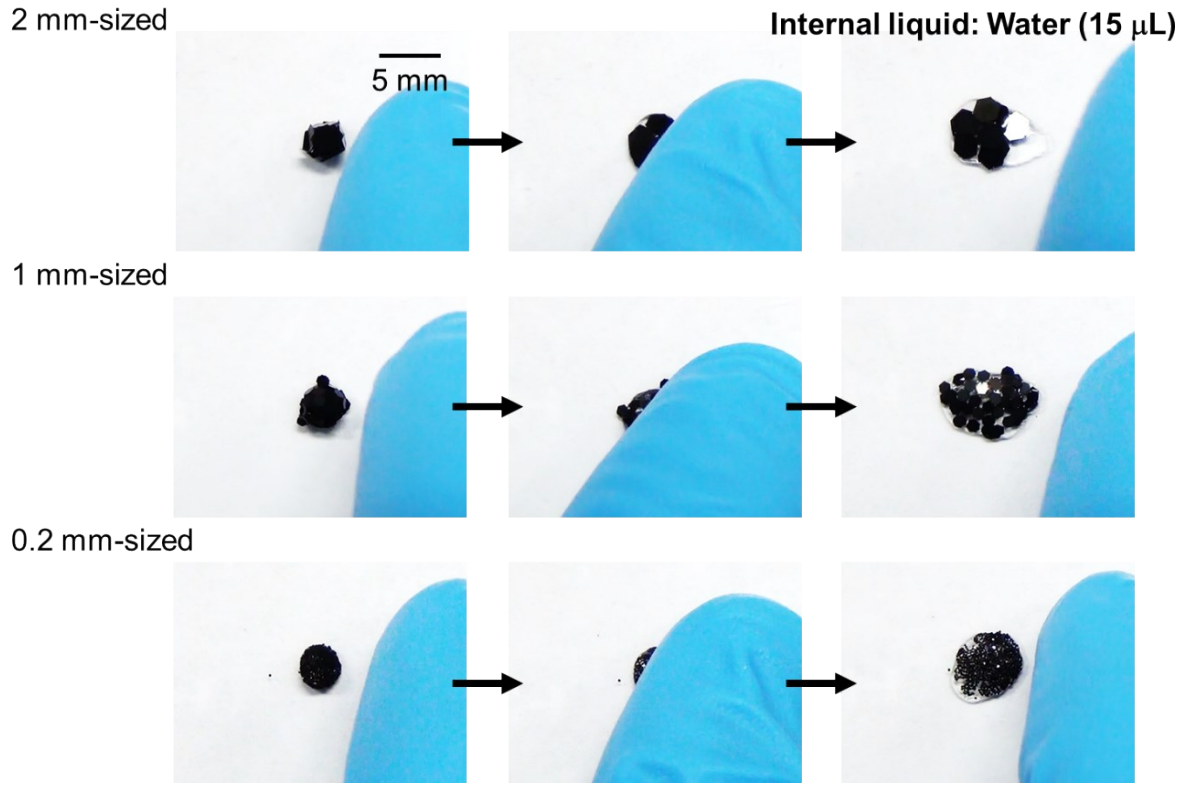

Figure S8 PET-PPy-C_8F stabilised droplets of all tested sizes before and after compressive pressure applied. Demonstrating different behaviour of the chemically hydrophobic particles when compared to the hydrophilic Cl^- doped particles

Raw & processed electrometer data

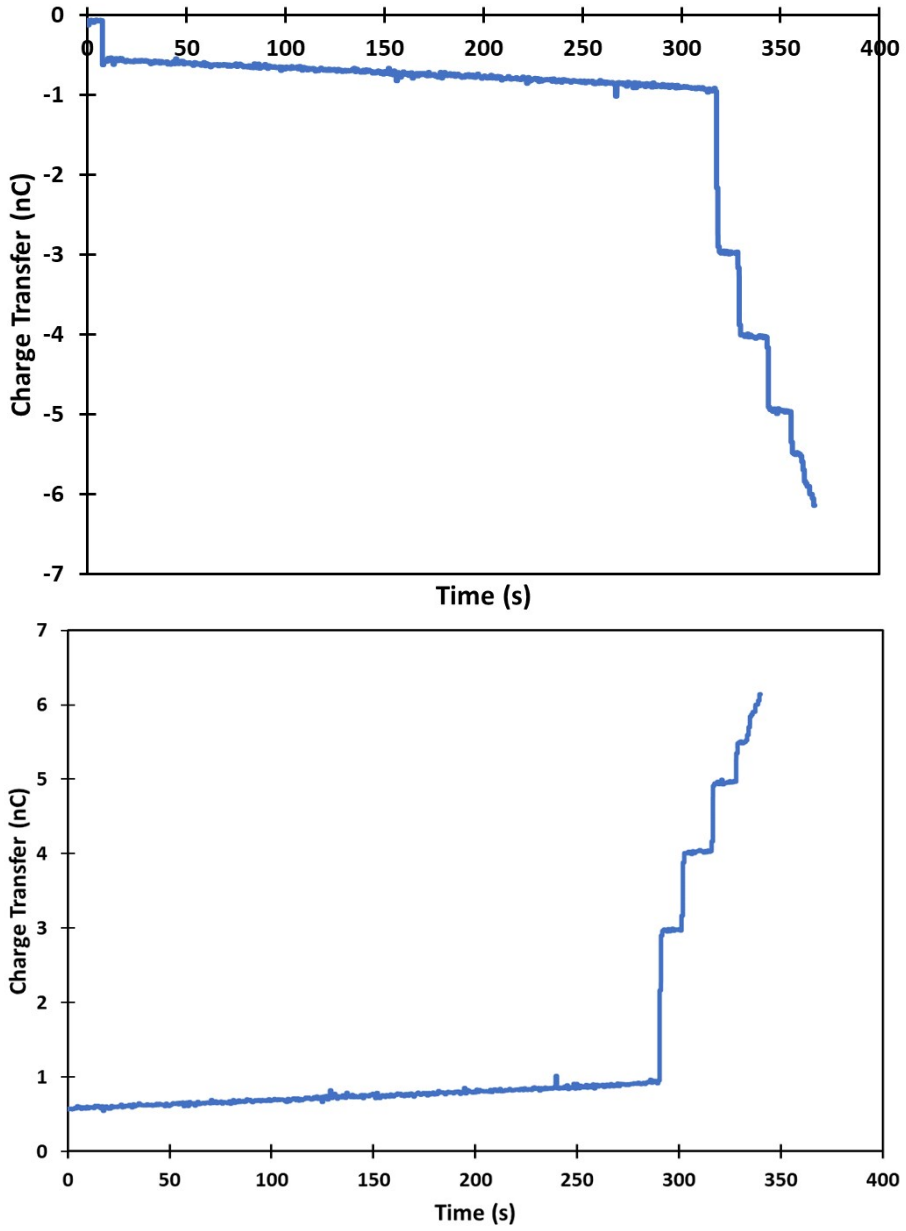

Figure S9. Electrometer data collected for 2 mm PET-PPy-C_8F platelets. Top: Raw data. The initial increase in charge corresponds to the time at which the voltage is applied to the charging plate. Bottom: Data modified to measure magnitude of charge and begin data reporting when the bed begins moving towards the droplet.

Gravimetric measurements of rolled liquid marbles

Table S1. Measured mass percentage of coated polymer plates 15 µL water droplet. Measured by weighing the marble or aggregate before and after evaporation of the internal liquid phase. Used in calculation of number of particles in Figure 3 in manuscript.

Nominal PET Plate Size (mm)	Mass % PET-PPy-Cl/ H_2O in aggregate	Mass % PET-PPy-C_8F/ H_2O in liquid marble
0.2	42 ± 11	16 ± 5
1	53 ± 6	20 ± 1
2	89 ± 6	9 ± 1

Table S2. Calculated surface area of adsorbed particles based on gravimetric analysis in Table S1.

Nominal PET Plate Size (mm)	Surface Area of Plate (m^2)	Number of PET-PPy-Cl plates Measured	PET-PPy-Cl Platelet Surface Area (m^2)	Coverage Fraction PET-PPy-Cl plates	Number of PET-PPy-C_8F plates Measured	PET-PPy-C_8F Platelet Surface Area (m^2)	Coverage Fraction PET-PPy-C_8F plates
0.2	1.04×10^{-7}	5693	5.92×10^{-4}	10	629	5.47×10^{-5}	1
1	1.83×10^{-6}	157	2.87×10^{-4}	5	63	1.20×10^{-4}	2
2	7.77×10^{-6}	28	2.18×10^{-4}	4	11	8.58×10^{-5}	2

surface area of a 15 µL droplet is 2.94×10^{-5} m^2.

Determination of theoretical field strength

To determine the force of extraction in Table 3 we must first know E. For this we use the approach of Morrison.[1] The original work determined the electric field produced by a charged conducting sphere sitting a distance h above a charge conducting plate at a given applied potential. In our application of this model the conducting sphere is the droplet, the plate is the particle bed and h represents the drop-bed separation distance. The Morrison model allows determination of the electric vector field components at any point in space close to the droplet. We calculate the total electrostatic field strength as a vector sum of both the radial E_r and vertical E_z component of the electric field as close as possible to directly beneath the droplet at a given separation distance

$$E_{Total} = \sqrt{E_r^{\,2} + E_z^{\,2}}$$

Consider the data for "*1.0 kV*" in Figure S9. At a constant potential, as the drop-bed separation distance decreases, the electric field strength increases and so does the extracting force that is acting upon it. Furthermore, as the applied potential increases at a given separation h so too does the electric field. Conversely, as we decrease potential and/or increase the separation distance the field also decreases. Using this data a value for E could be determined for drop-bed separation distances experimentally determined from extracted stills at known potentials and used for calculations in Table 3.

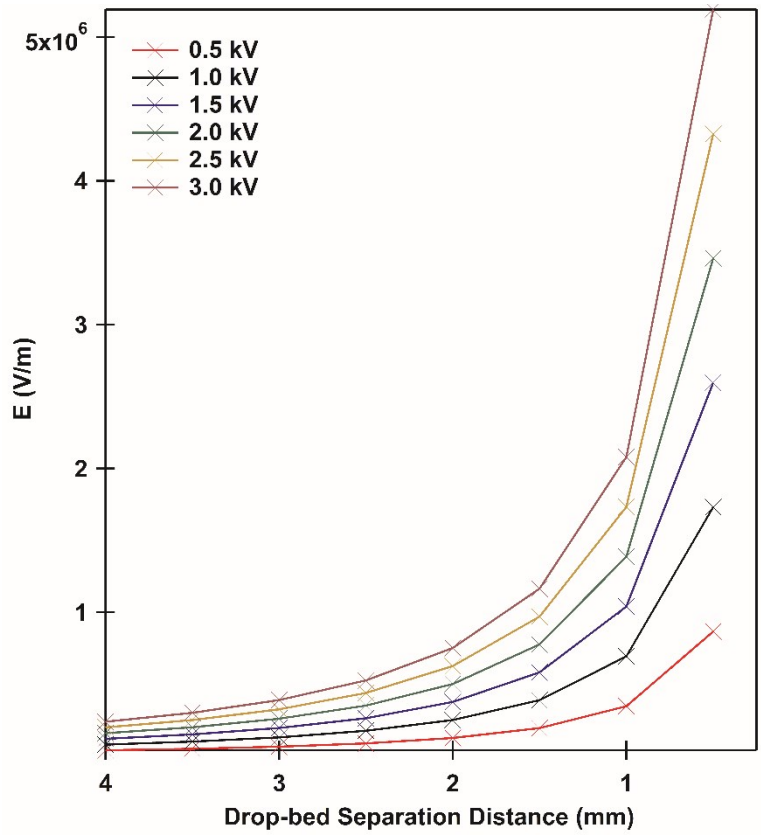

Figure S10. Total electrostatic field strength at increasing voltages as a function of decreasing drop-bed separation.

Table S3. Calculated electrostatic field values at given separation distanced and applied potentials and measured charge per particle used to determine extraction force on particles in Table 3.

Voltage (kV)	Drop-bed Separation (mm)	Number of Particles	Charge Before Particle Extraction (nC)	Charge After Particle Extraction (nC)	Charge Per Particle (nC)	E (V/m) $\times 10^5$	F (mN)
0.5	0.560	1	0.80	4.58	3.78	7.66	2.89
1.0	1.59	1	1.53	9.89	8.36	3.56	2.97
1.0	1.35	1	1.55	10.4	8.80	4.53	3.99
1.0	1.29	1	1.55	8.20	6.65	4.84	3.22
1.5	2.28	1	2.30	12.1	9.80	3.06	3.00
1.5	1.89	1	2.30	11.1	8.80	4.10	3.61
1.5	2.10	1	2.45	17.1	14.63	3.48	5.08
2.0	2.02	2	3.20	11.6	4.20	4.93	2.07
2.0	2.49	2	3.20	18.2	7.50	3.53	2.65
2.0	2.78	1	3.18	15.0	11.82	2.95	3.48
2.5	3.50	2	4.00	19.8	7.90	2.50	1.98
2.5	3.06	2	4.00	17.3	6.65	3.14	2.09
2.5	2.98	2	4.60	27.2	11.3	3.28	3.70
3.0	5.12	1	4.70	20.0	15.3	1.54	2.36
3.0	3.37	3	4.65	21.0	5.45	3.20	1.74

References

1. C. A. Morrison, *The potential and electric fields of a conducting sphere in the presence of a charged conducting plane*, Adelphi MD, 1989.