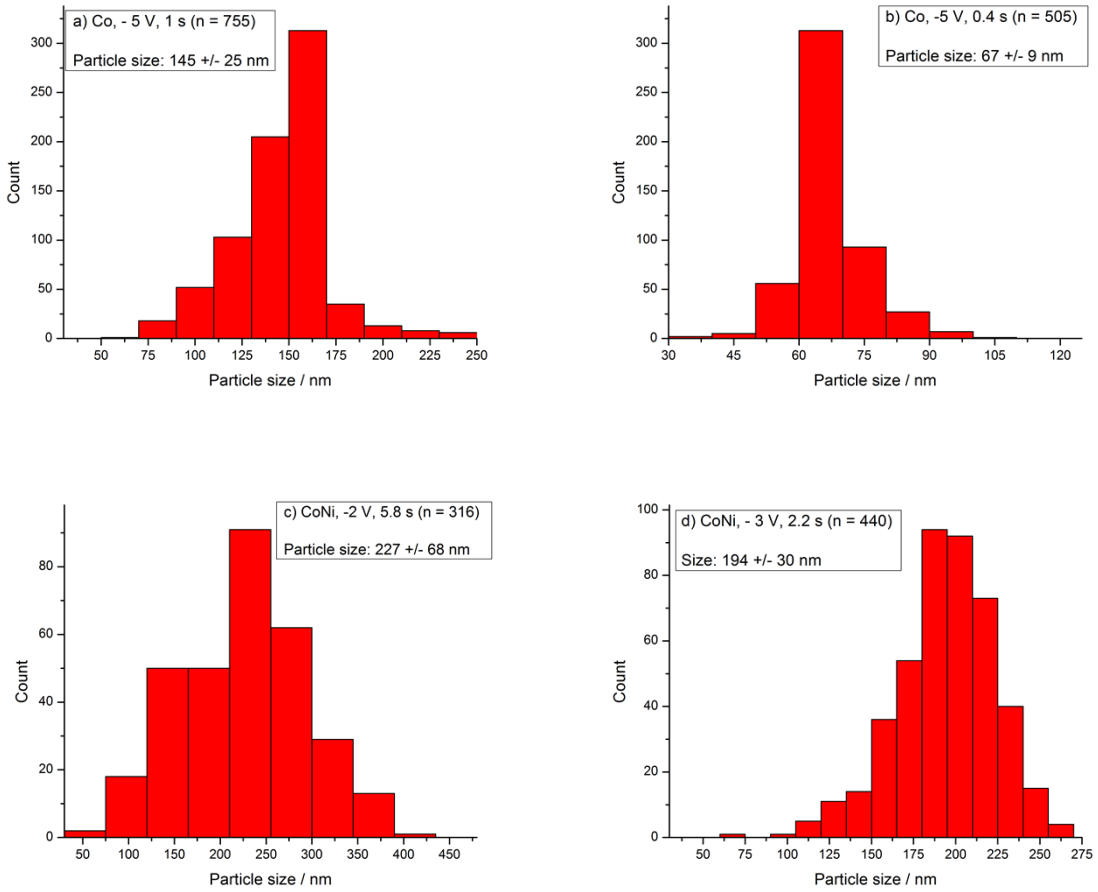
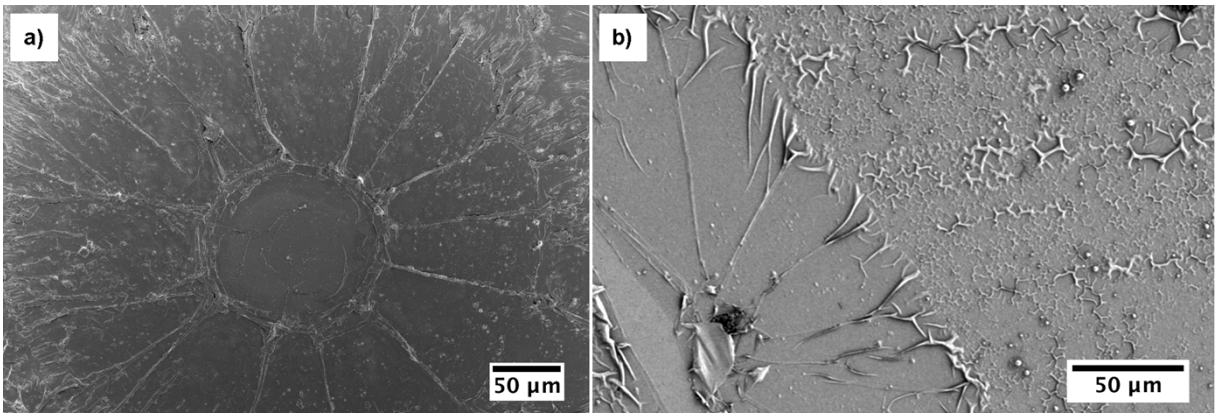
Supporting Information

Figure S1. Particle size distribution corresponding to the SEM images of Figure 3.



The particle size distribution has been determined by counting more than 300 NPs per condition using Image J software. In all cases, moderate size distributions are observed, with most of the particles having sizes close to the average value.

Figure S2. Top view SEM image of sandwich type nanocomposite fabricated with a secondary layer of Ppy at a) 1V, 30 seconds and b) 1V, 60 seconds without a rest phase. The H_2 bubbles which were formed during the CoNi NPs cycle were trapped in between during the secondary Ppy layer growth due to a lack of rest phase. The bubbles afterwards burst creating the wrinkled Ppy film (a, b) and tears (b).



The rest phase conditions were optimized to prevent film detachment. As shown in Figure S3 when the rest phase is conducted potentiostatically the nanocomposite film gets detached from the gold surface. This is probably due to the H_2 evolution taking place which does not get released completely because the current is not 0 A as opposed to 0 V.

Figure S3. Images showing cross-section of nanocomposite fabricated using a potentiostatic rest phase which was set to 0 V.

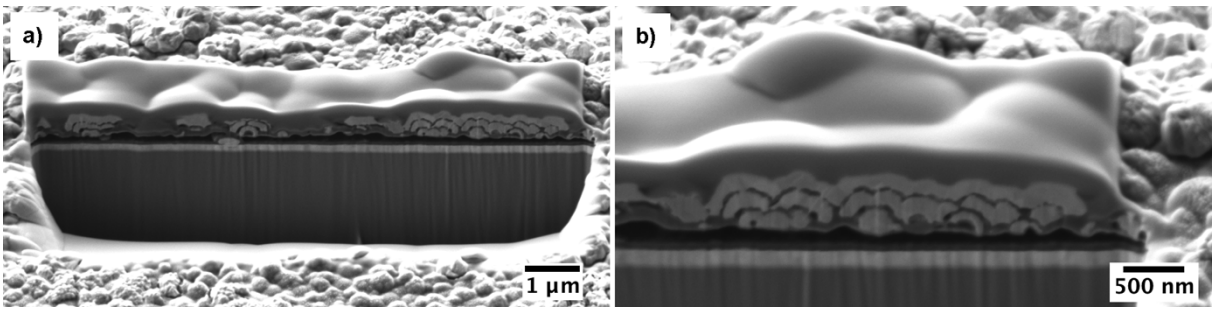


Figure S4. SEM top view image of Ppy – CoNi NPs nanocomposite (image was taken at an angle of 54° with SE2 detector). The second layer of Ppy was grown at 1V for 60 seconds. CoNi NPs are not completely covered by the second layer of Ppy, since they are still visible from a top view image.

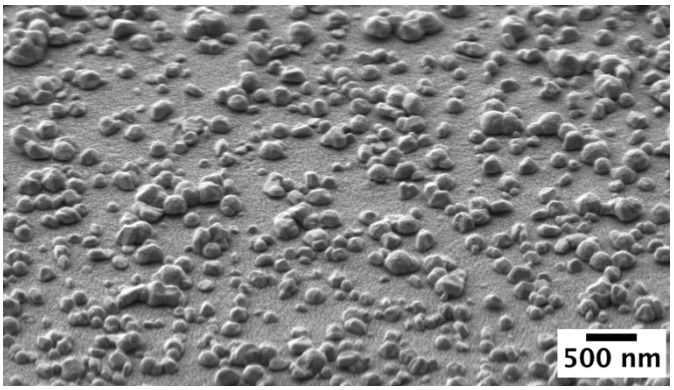


Table S1. Average particle size (ϕ), saturation magnetization (M_S) and coercivity (H_C) values for the samples shown in Figure 8. The values have been extracted from the room-temperature hysteresis loops recorded by VSM.

Sample	ϕ (nm)	M_S (emu/cm^3)	H_C (Oe)
Co – Ppy bilayer	147 ± 25	850	290 (in-plane)
CoNi – Ppy bilayer	227 ± 68	380	177 (in-plane)
CoNi – Ppy multilayer (5 layers)	227 ± 68*	380	110 (in-plane) 195 (out-of-plane)

*Average particle size of the initial bilayer