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Supporting Information

Switchable Self-assembly of Prussian Blue Analogs Nano-tiles Triggered by Salt Stimulus

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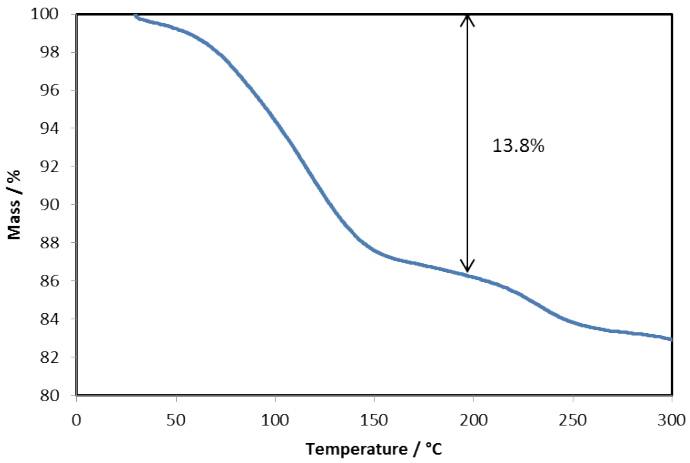

Fig. S1. Thermogravimetry data obtained by sample heating with the speed of 5°C/min in nitrogen atmosphere. Mass lost up to 200°C was attributed to the water evaporation and further to decomposition of cyano bonds.

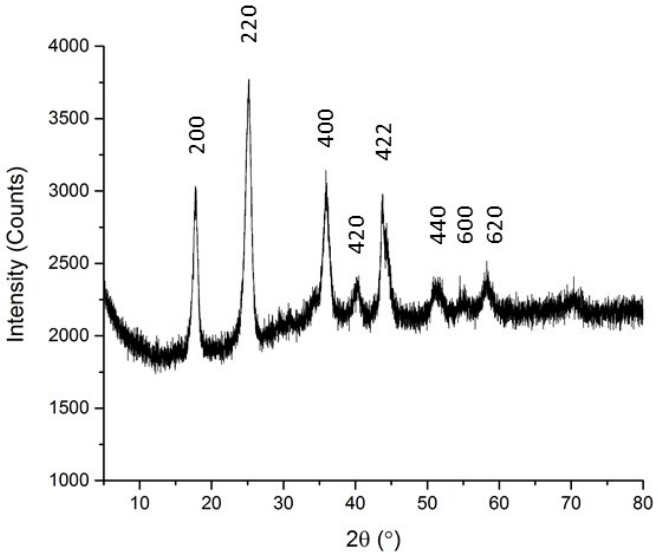

Fig. S2. X-Ray powder diffraction pattern of a dried Cu-PBA powder

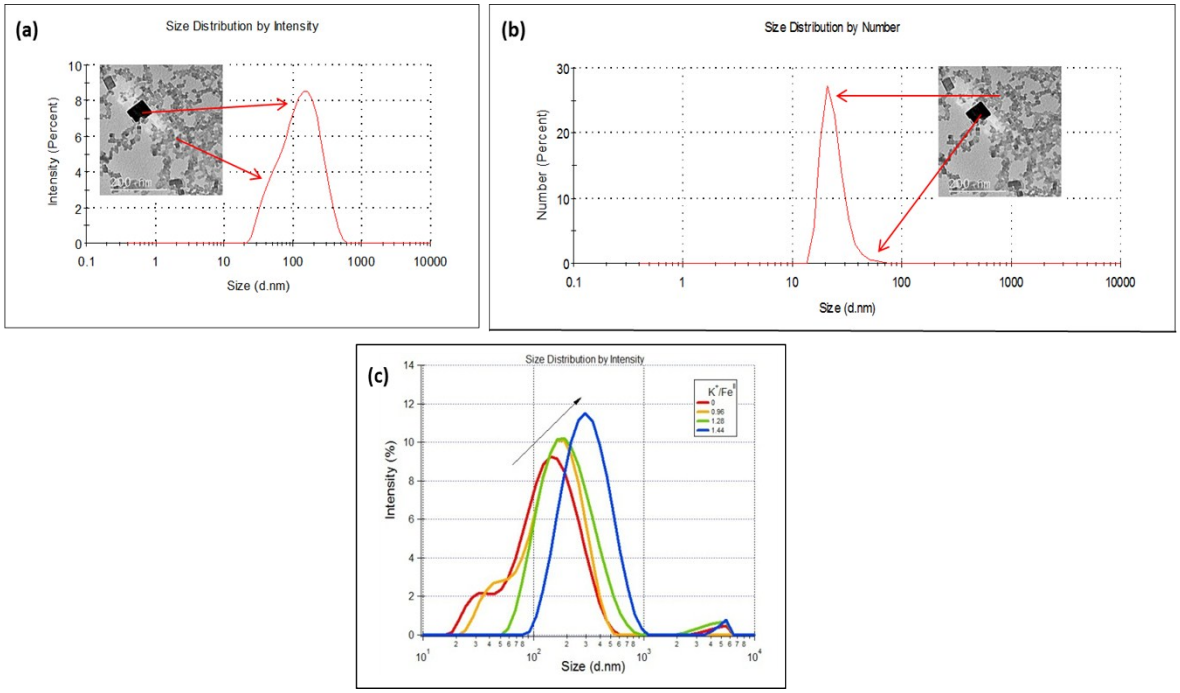

Fig. S3. (a) Size distribution by intensity and (b) size distribution by number of 20 mM Cu-PBA suspension (0.85 wt%). Insert shows corresponding TEM image with the domains where nanocrystals aggregate to form platelets. (c) Evolution of the size distribution in intensity by increasing the K^+/Fe^{II} ratio, i.e. the ionic strength (IS = [KNO_3]), for 20 mM of Cu-BPA showing the equilibrium shift between the small and large populations of nanoparticles towards the larger ones.

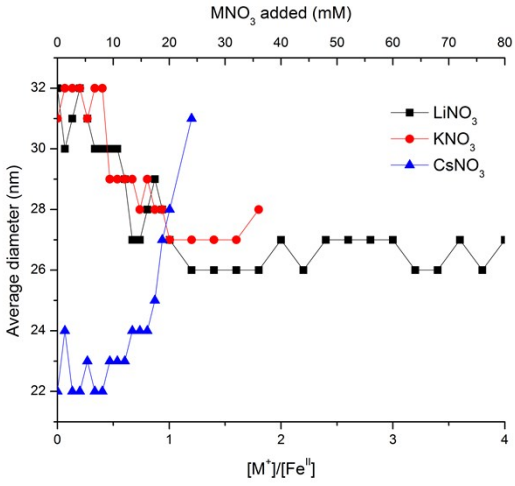

Fig. S4. Average diameter determined by acoustic attenuation measurement of Cu-PBA dispersion (20mM) as a function of salt concentration and M^{+}/Fe^{II} ratio.

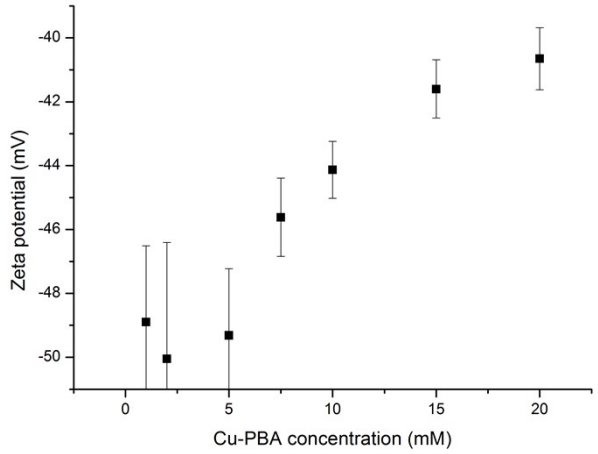

Fig. S5. Zeta potential of Cu-PBA nanoparticles as a function of suspension concentration.

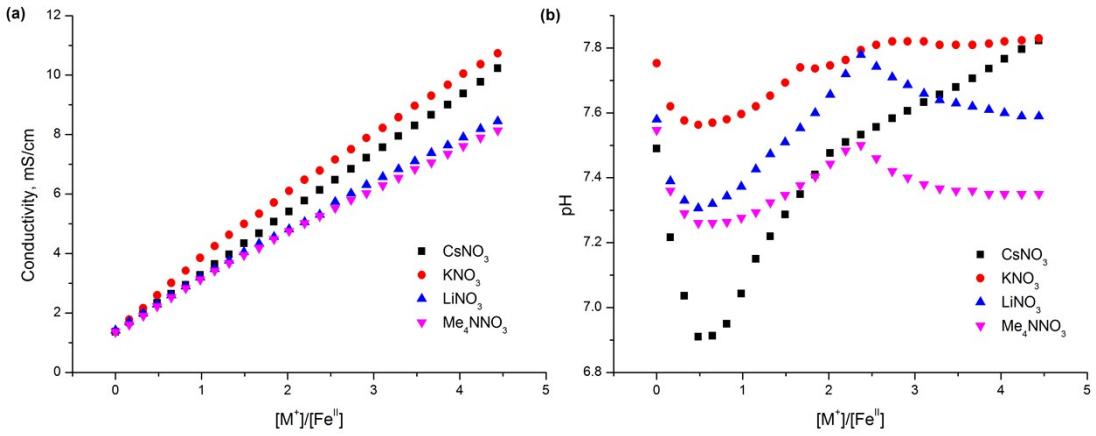

Fig. S6. (a) Conductivity and (b) pH of the colloidal Cu-PBA suspension as a function of M^{+}/Fe^{II} ratio for different salts.

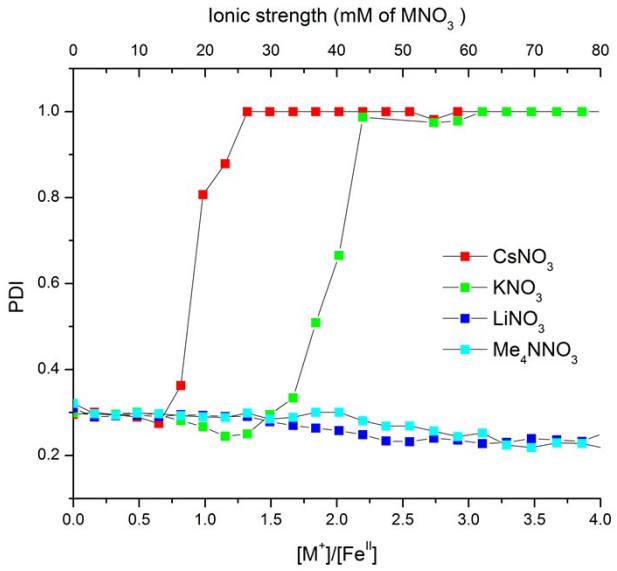

Fig. S7. Polydispersity index (PDI) of 20 mM Cu-PBA suspension as a function of ionic strength and M^+/Fe^{II} ratio as determined from DLS measurements. PDI above 0.3 indicates the NPs flocculation.

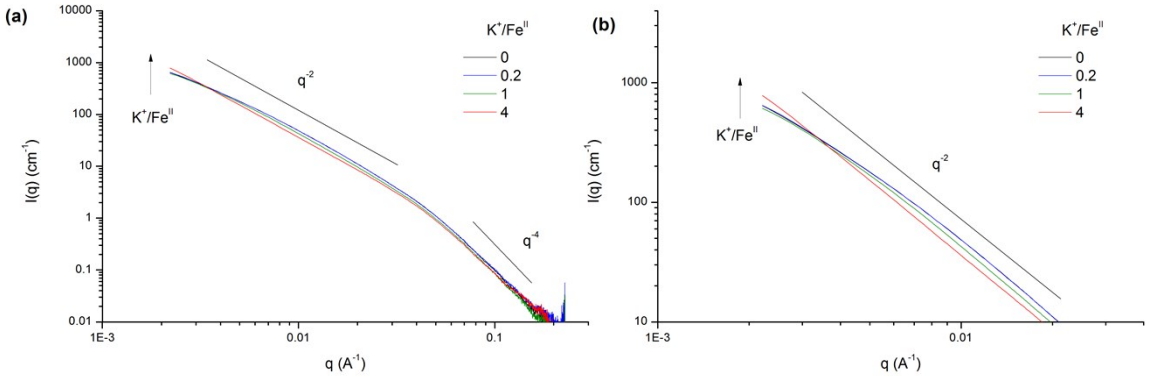

Fig. S8. (a) SAXS spectra of 20 mM Cu-PBA suspension upon titration with KNO_3. (b) Zoom of the region where curves show q^{-2} dependence.

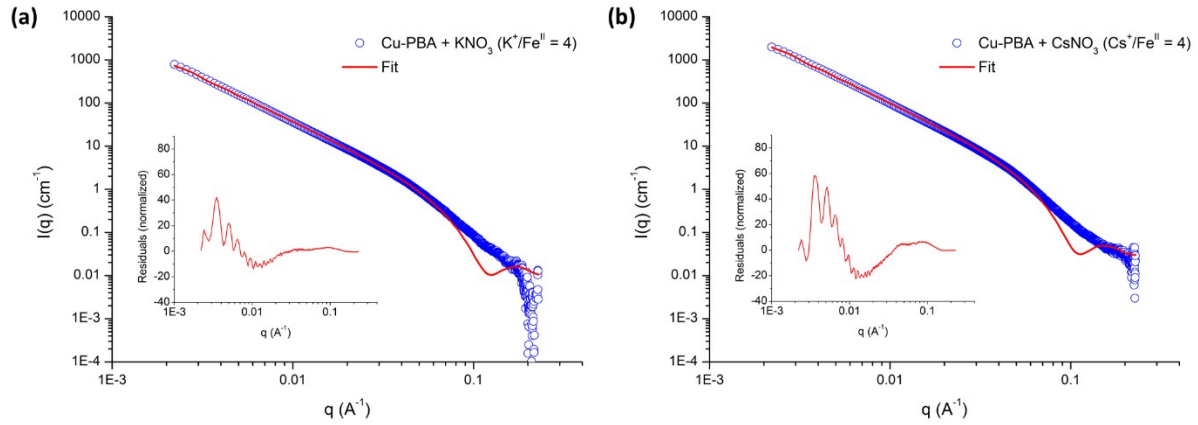

Fig. S9. Fitting of synchrotron SAXS data with the Disc model for the 20mM Cu-PBA suspensions in the presence of 80 mM (a) KNO_3 (disc diameter = 210 nm, thickness = 5.0 nm) and (b) $CsNO_3$ (disc diameter = 207 nm, thickness = 5.6 nm). No polydispersity was taken into account for the thickness; this explains the difference between the scattering data and the model at high q-values, above 0.1 $Å^{-1}$. Inserts on each plot show normalized residuals of fitting. The structure factor S(q) characterizing the interactions between NPs was taken to 1 since inter-particles repulsion was suppressed by salt addition.

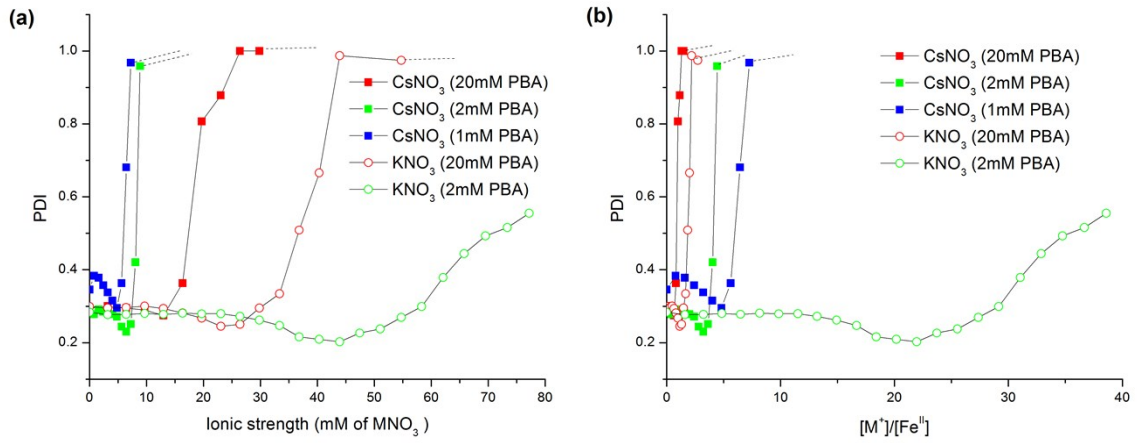

Fig. S10. Results of DLS measurements showing for different concentrations of Cu-PBA the evolution of aggregates Polydispersity index (PDI) as a function of the electrolyte

concentration (a) and as a function of the added ion (M^+) to Fe^{II} molar ratio (b). Dashed lines represent conditions under which fast flocculation occurs.

Expression for hydrodynamic radius of disk particles

$$R_h = \frac{3}{2} r \left(\left[1 + \left(\frac{t}{2r} \right)^2 \right]^{\frac{1}{2}} + \frac{2r}{t} ln \left[\frac{t}{2r} + \left[1 + \left(\frac{t}{2r} \right)^2 \right]^{\frac{1}{2}} \right] - \frac{t}{2r} \right)^{-1} \quad (1)$$

Equation of the hydrodynamic radius for a disk-shaped nanoparticle as a function of the disk radius r and its thickness t, equation extracted from appendix A in ref [1]

References

1 N. A. Mazer, G. B. Benedek and M. C. Carey, *Biochemistry*, 1980, **19**, 601.