

Supporting Information (i): High Salt systems with M^{III} as background ions.

Addition of 1M Al^{3+} or Fe^{3+} completely dissolves $FePP_i$, see Figure A1. This might be caused by the lowered pH, see Table A2.

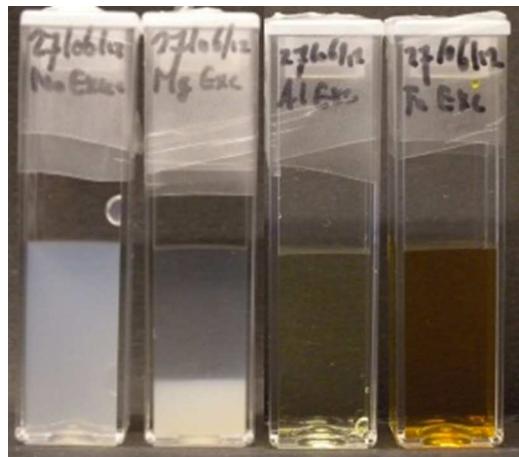


Figure A1. M^{II}/M^{III} excess dispersions: (a) Freshly prepared $FePP_i$ (b) 1M $MgCl_2$, (c) 1M $AlCl_3$, (d) 1M $FeCl_3$.

Table A2. pH of $FePP_i$ dispersions.

Added cation	Added concentration (M)	pH
No salt*	0	3.30
Li^+	2	2.84
K^+	2	2.69
Mg^{2+}	1	2.68
Al^{3+}	1	1.79
Fe^{3+}	1	0.81
Fe^{3+} 10%	0.0002	2.80

*No additional salt: freshly prepared $FePP_i$ system.

Supporting Information (ii): Fe or PP_i deficient systems

Fe/PP_i deficient samples were prepared by reducing the amount of the respective salt by 10%. The PP_i deficient systems had a positive zeta-potential insufficient to keep the system dispersed, see Table A1. The Fe deficient system remained stable but did not have an increased zeta-potential that might be expected for an electrocratic system.

Table A1. 10% Fe or PP_i deficient systems

	Size (nm)	ζ -potential (mV)	Conductivity (mS/cm)
Stoichiometry	210	-37	2.2
Fe deficit	250	-40	2.1
PP_i deficit	*20000	*4.2	2.5

*Indicative values, size and measurement polydispersity too large for accurate analysis.

Supporting Information (iii) Solubility of iron(III) pyrophosphate

The solubility of iron pyrophosphate at pH 3.8 and a solid content of 25 g/L (0.14 M iron, 0.11 M pyrophosphate assuming pure FePP_i) was determined by ICP-AES analysis to be 0.61 mM iron and 0.67 mM pyrophosphate. Solubility of similar materials have been shown to vary with experimental conditions such as solid content[1].

References

[1] Leach, S. A. Electrophoresis of Synthetic Hydroxyapatite. *Arch. Oral Biol.* **1960**, 3, 48-56.