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## **Supplementary information**

## Cathodic electrodeposition of zinc-zinc phosphate-calcium phosphate composite coatings on pure iron for biodegradable implant applications

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Table S1: Assignment of FT-IR and Raman bands of zinc - zinc phosphate - calcium phosphate composite coatings deposited on pure iron by CED under varying current densities at 27 °C for 30 min

FT-IR bands (cm <sup>-1</sup> )	Assignment	Raman bands (cm <sup>-1</sup> )	Assignment
3521	(O-H) stretching of H <sub>2</sub> O	302	M-O bending mode
3189	(O-H) stretching of H <sub>2</sub> O	442	$v_2$ bending mode of $HPO_4^{2-}$ and $PO_4^{3-}$
1635	H-O-H bending of H <sub>2</sub> O	572	$v_4$ bending mode of $HPO_4^{2-}$ and $PO_4^{3-}$
1100	v <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>	808	P-O(H) stretching of HPO <sub>4</sub> <sup>2</sup> -
1069	v <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>	986	ν <sub>1</sub> P-O symmetric stretching of PO <sub>4</sub> <sup>3-</sup>
1009	v <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>	1070	v <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>
929	v <sub>1</sub> P-O symmetric stretching of HPO <sub>4</sub> <sup>2-</sup>	1136	v <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>
627	ν <sub>L</sub> liberation of the OH <sup>-</sup> group of H <sub>2</sub> O	1220	P-O-H in plane bending of HPO <sub>4</sub> <sup>2</sup> -
570	O-P-O(H) bending	1380	P-O-H in plane bending of HPO <sub>4</sub> <sup>2</sup> -
		1626	H-O-H bending of H <sub>2</sub> O
		1756	H-O-H bending of H <sub>2</sub> O

Table S2: Assignment of FT-IR and Raman bands of zinc -zinc phosphate - calcium phosphate composite coatings deposited on pure iron by CED at 2 mA/cm<sup>2</sup> at 27 °C for 30 min, after immersion in HBSS at 37  $\pm$  1 °C for 168 h

IR	Assignment	Raman	Assignment
bands		bands	
(cm <sup>-1</sup> )		(cm <sup>-1</sup> )	
3346	(O-H) stretching of H <sub>2</sub> O	310	M-O bending mode
2921	(P)O-H stretching	597	v <sub>4</sub> bending mode of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>
2850	H-O-H bending and rotation of H <sub>2</sub> O	943	$v_1$ P-O symmetric stretching of $PO_4^{3-}$
1747	H-O-H bending of H <sub>2</sub> O	997	ν <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>
1624	H-O-H bending of H <sub>2</sub> O	1057	v <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>
1445	v <sub>3</sub> Asymmetric stretching of $CO_3^{2-}$	1154	ν <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>
1382	P-O-H in-plane bending		
1192	P-O-H in-plane bending		
1103	ν <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>		
1024	ν <sub>3</sub> P-O asymmetric stretching of HPO <sub>4</sub> <sup>2-</sup> and PO <sub>4</sub> <sup>3-</sup>		
946	ν <sub>1</sub> P-O symmetric stretching of HPO <sub>4</sub> <sup>2-</sup>		

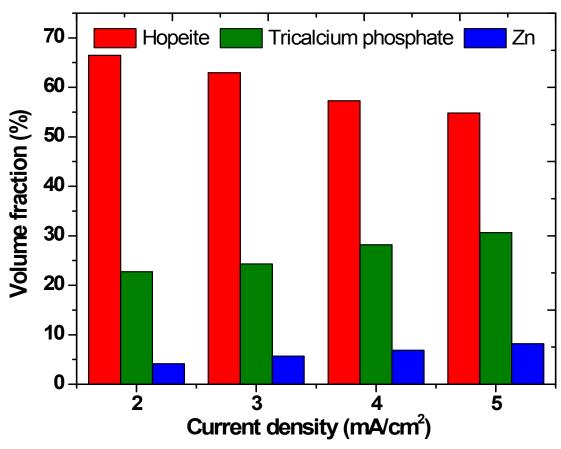


Fig. S1 Volume fraction of zinc phosphate (hopeite), calcium phosphate and zinc in the composite coatings formed on pure iron by cathodic electrodeposition under varying current densities (2 to 5 mA/cm²) at 27 °C for 30 min, estimated from the XRD patterns