

## Supporting Information

**Table SI-1.** Compositions of PGPs and PGFs expressed in atomic % measured via EDX.

Sample	P	Ca	Na	Mn	O
PGP-Mn0	20.5 ± 0.4	8.1 ± 0.2	4.1 ± 0.2	-	67.3 ± 0.9
PGP-Mn1	21.9 ± 0.4	6.2 ± 0.5	3.5 ± 0.3	0.2 ± 0.3	68.2 ± 0.4
PGP-Mn3	20.0 ± 0.3	7.4 ± 0.2	3.9 ± 0.4	0.6 ± 0.5	68.1 ± 1.1
PGP-Mn5	20.9 ± 0.4	6.9 ± 0.5	3.6 ± 0.5	1.0 ± 0.4	67.6 ± 0.5
PGP-Mn10	20.5 ± 0.5	6.6 ± 0.4	2.1 ± 0.2	2.2 ± 0.4	68.6 ± 1.0
PGF-Mn0	20.5 ± 0.8	6.5 ± 0.3	4.2 ± 0.5	-	68.8 ± 1.2
PGF-Mn1	20.7 ± 0.5	5.4 ± 0.4	4.2 ± 0.2	0.2 ± 0.4	69.5 ± 0.4
PGF-Mn3	20.8 ± 0.4	5.0 ± 0.5	3.2 ± 0.4	0.6 ± 0.4	70.4 ± 0.8
PGF-Mn5	21.8 ± 0.7	5.2 ± 0.2	1.7 ± 0.4	1.1 ± 0.5	70.3 ± 1.5
PGF-Mn10	22.3 ± 0.9	4.0 ± 0.4	1.2 ± 0.8	2.2 ± 0.3	70.3 ± 0.6

Table SI-2. Summary of key references on PGs.

<i>Synthesis</i>	<i>Morphology</i>	<i>TMI</i>	<i>Mol %</i>	<i>Cytocompatibility</i>	<i>Antimicrobial properties</i>	<i>Properties</i>	<i>Ref</i>
SG	Mesoporous PGP	Sr <sup>2+</sup>	1-3-5			Pore size 11.8-18.6 nm. Specific Surface Area (SSA) 123-73 m <sup>2</sup> /g. Sr <sup>2+</sup> acts as a cross-linker. Good drug delivery system (DDS).	43
SG	Mesoporous PGP	Cu <sup>2+</sup>	1-3-5		Antibacterial effect against <i>S. aureus</i> from day 1 (3 and 5 mol% of Cu <sup>2+</sup> ). Activity against <i>E. coli</i> was for all compositions after day 2.	Pore size 8-20 nm. SSA 124-67 m <sup>2</sup> /g. The role of Cu <sup>2+</sup> changes from network modifying to network forming with increasing Cu <sup>2+</sup> content. Good DDS.	44
SG	Mesoporous PGP	Zn <sup>2+</sup>	1-3-5			Average pore size ~12 nm. SSA 124-76 m <sup>2</sup> /g. Good DDS.	45
COA	PGP	Zn <sup>2+</sup>	2-10-15	MTT assay on MG-63 and HaCaTs cells. Good cell viability of all PGs, with slightly reduced cell viability at higher loadings. ~ 100% for MG-63 ≥ 69% for HaCaTs		Zn <sup>2+</sup> adopts a 6-coordinate geometry. Good DDS.	42
COA	PGP	Cu <sup>2+</sup>	2-10-15	MTT assay on MG-63 and HaCaTs cells. Good cell viability of all PGs, with slightly reduced cell viability at higher loadings. ≥ 79% for MG-63 ≥ 73% for HaCaTs		Cu <sup>2+</sup> adopts a 4-coordinate geometry. Good DDS.	42
ES-COA	PGF	Cu <sup>2+</sup>	1-3-5	Cu <sup>2+</sup> enhancement of MG-63 viability and proliferation.	Cu <sup>2+</sup> has antibacterial activity against <i>S. aureus</i> and <i>E. coli</i> .	PGFs average diameter 1-3 μm. Cu <sup>2+</sup> acts as cross-linker.	13
ES-COA	PGF	Ag <sup>+</sup>	1-2-4-6-10	Dissolution products non-toxic against HaCaTs.  Enhancement of wound closure in <i>ex vivo</i> models, (≥ 4 mol % Ag <sup>+</sup> )  HaCaT migration/proliferation demonstrated by <i>in vitro</i> scratch assays starting from PGF-Ag6, reaching 72% closure with 10 mol % Ag <sup>+</sup> .	PGF-Ag (Ag <sub>2</sub> O ≥ 4 mol%) antibacterial activity against <i>S. aureus</i> and <i>E. coli</i> .	PGFs average diameter 4.2 μm. Good DDS.	9
ES-COA	PGF	Zn <sup>2+</sup>	1-2-4-6-10	Dissolution products non-toxic against HaCaTs.	PGF-Zn10 antibacterial activity against <i>S. aureus</i>	PGFs average diameter: 1.4 μm. Good DDS.	9
ES-COA	PGF	Fe <sup>3+</sup>	1-2-4	Dissolution products are non-toxic against HaCaTs. Acceleration of healing in chronic wounded skin via <i>ex vivo</i> assay (> 30%) on 1 mol% Fe.		PGFs average diameter 5.2 μm. Good DDS.	9
ES-COA-SUP	Porous PGF	Ga <sup>3+</sup>	0.2-0.5-1	All PGF dissolution products are non-toxic against HaCaTs.	PGFs have antibacterial activity against <i>E. coli</i> . 1 mol% Ga <sup>3+</sup> has antibacterial activity against <i>S. aureus</i> .	Pores diameters from 100 nm to 2 μm. Ga <sup>3+</sup> acts as cross-linker. All PGFs are able to encapsulate clove oil (99.99 %). Clove oil is mostly released within the first 6 hours. The presence of Ga <sup>3+</sup> promotes the release of clove oil. Clove oil and Ga <sup>3+</sup> enhanced the antioxidant properties of PGFs.	14

COA = coacervation; ES-COA = ES of the coacervate gel; ES-COA-SUP = ES of the coacervate gel combined with the supramolecular templating technique

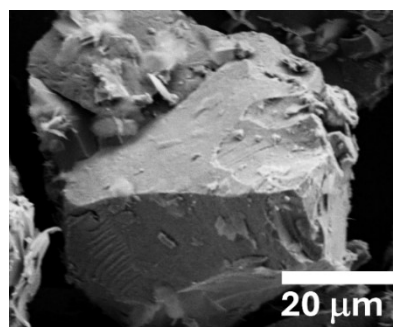


Figure SI-1. SEM image of PGP-Mn1

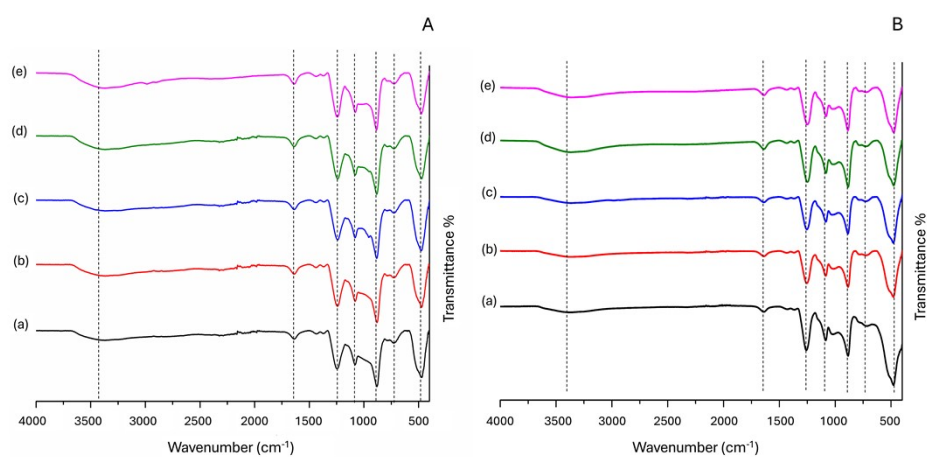


Figure SI-2. FT-IR spectra of A): (a) PGP-Mn0; (b) PGP-Mn1; (c) PGP-Mn3; (d) PGP-Mn5; (e) PGP-Mn10 and B): (a) PGF-Mn0; (b) PGF-Mn1; (c) PGF-Mn3; (d) PGF-Mn5; (e) PGF-Mn10.

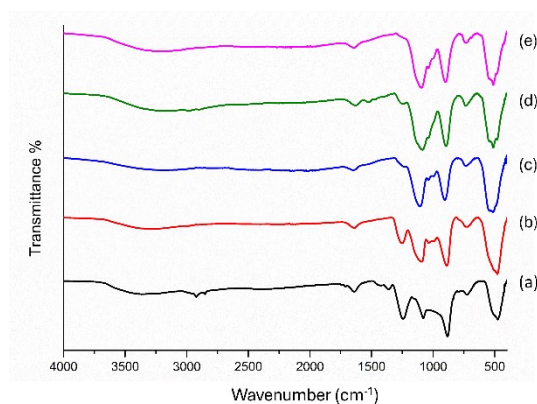


Figure SI-3. FT-IR spectra of PGP-Mn1 before (a) and after immersion in CM for 24 h (b), 72 h (c) and after immersion in Tris-B for 24 h (d), 72 h (e).