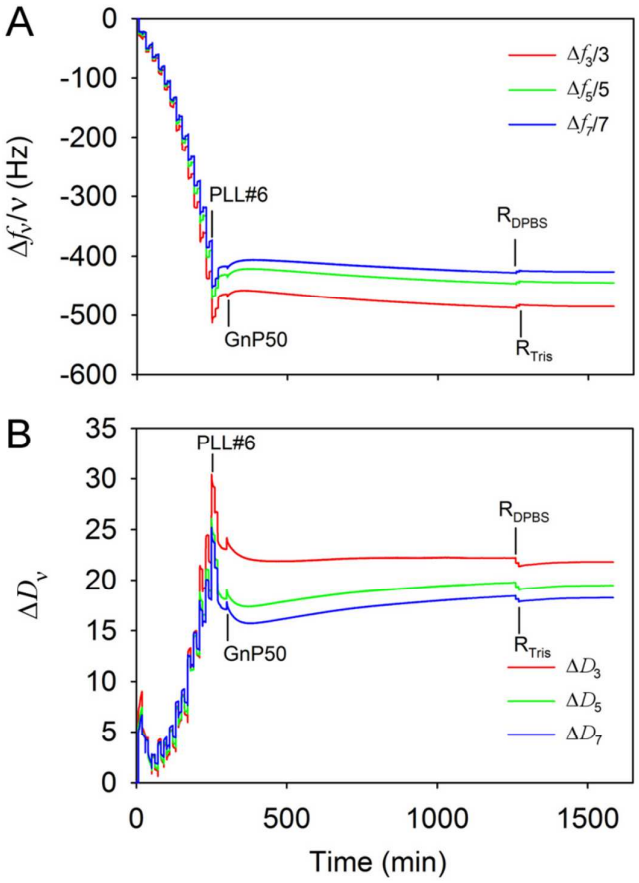

Figure S1. Evolutions of the QCM-D parameters during the buildup and GnP50 treatment of a PEI/(CSA/PLL)$_6$ architecture: (A) Normalized resonance frequencies $\Delta f_v/v$ and, (B) Dissipation factors ΔD_v.

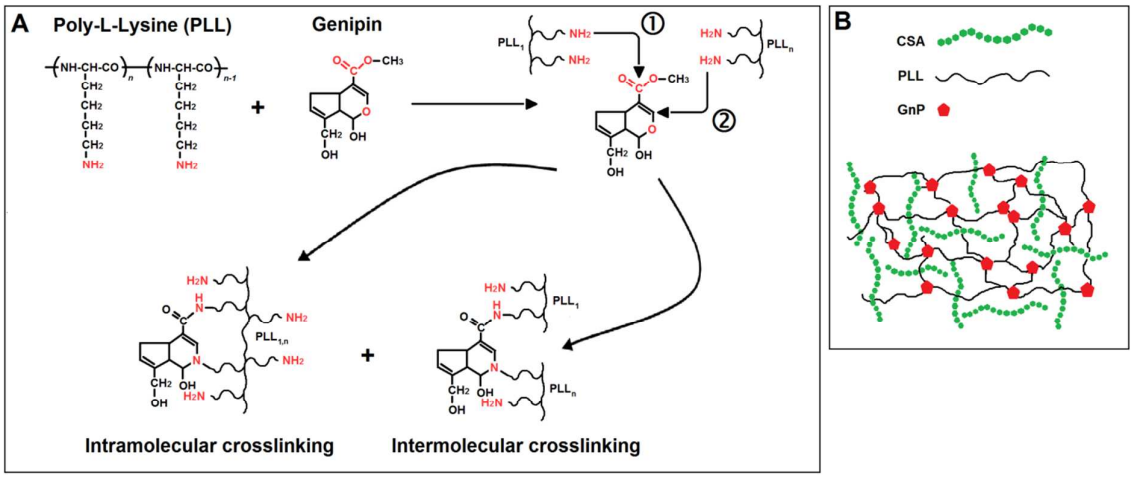

Figure S2. (A) Possible crosslinking reactions between genipin and PLL, involving ① the carboxymethyl group, and ② the C-3 atom of genipin leading to intra- and/or intermolecular crosslinking. (B) Proposal of a scheme describing the likely semi-IPN structure obtained after the genipin post-treatment of $(CSA/PLL)_n$.

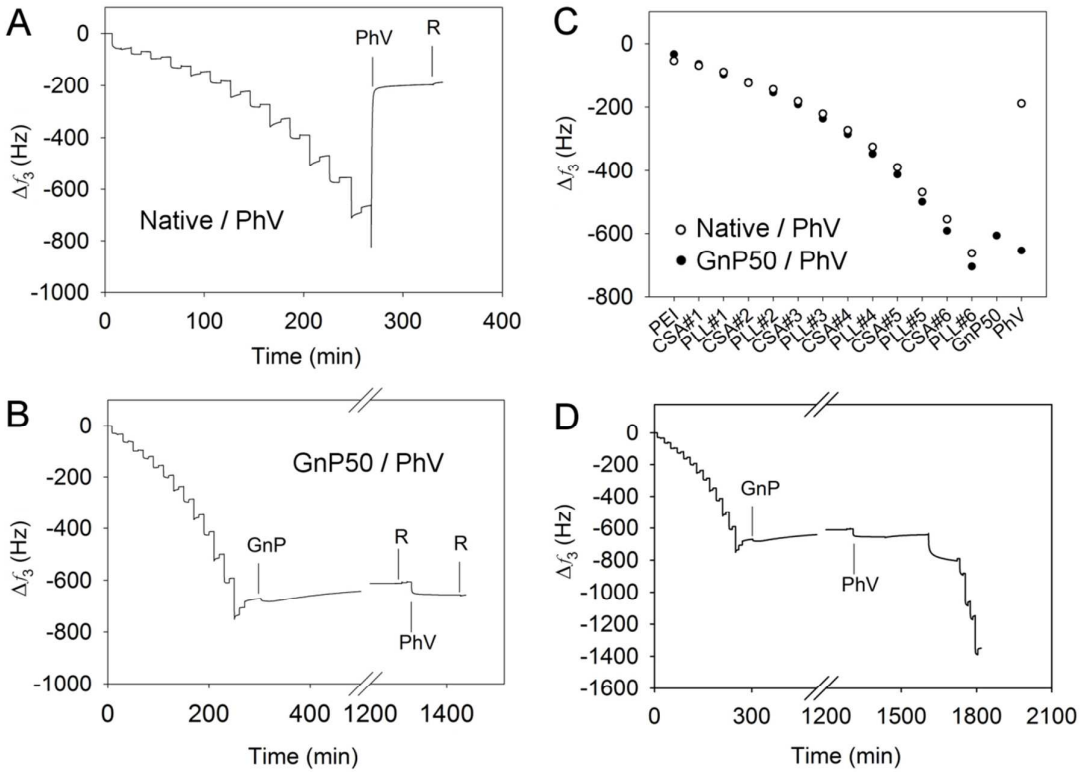

Figure S3. Evolutions of the normalized resonance frequency $\Delta f_3/3$, during the buildup and the treatment with a 1 mg mL^{-1} PhV solution, of (A) a PEI/$(CSA/PLL)_6$ film and, (B) a PEI/$(CSA/PLL)_6$-GnP50 film, with (C) the corresponding step-by-step evolutions. (D) Continuation of the buildup beyond the treatment with PhV, until reaching a PEI/$(CSA/PLL)_6$-GnP/PhV/$(PLL/CSA)_2$/PLL architecture.

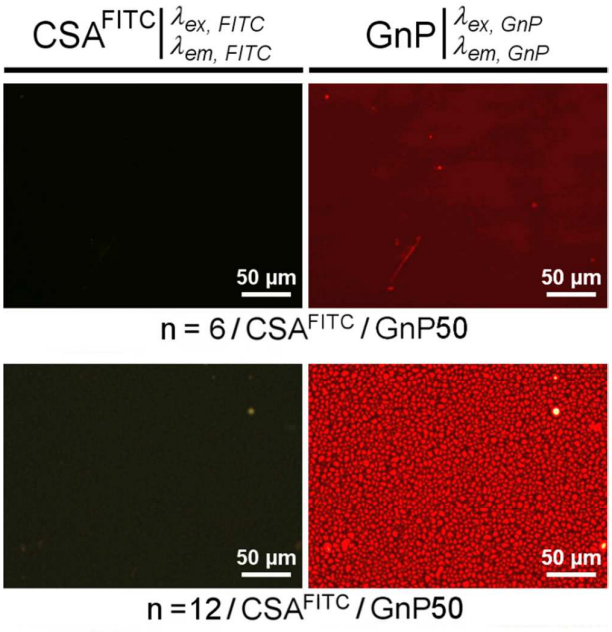

Figure S4. Epifluorescence micrographs at characteristic excitation and emission wavelengths of FITC (λ_{ex} = 490 nm; λ_{em} = 525 nm) and amino-bound GnP (λ_{ex} = 550 nm; λ_{em} = 575 nm) for PEI/$(CSA^{FITC}/PLL)_n$-GnP50 films.

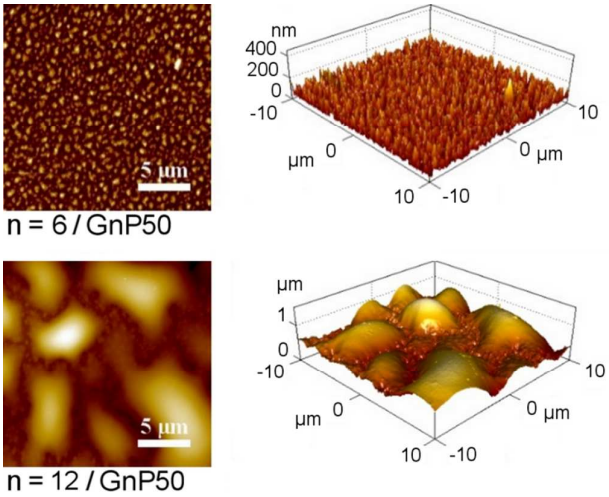

Figure S5. 2D and 3D AFM images representing the topographies in buffer medium of the PEI/(CSA/PLL)$_n$-GnP50 (n = 6 and 12) films.

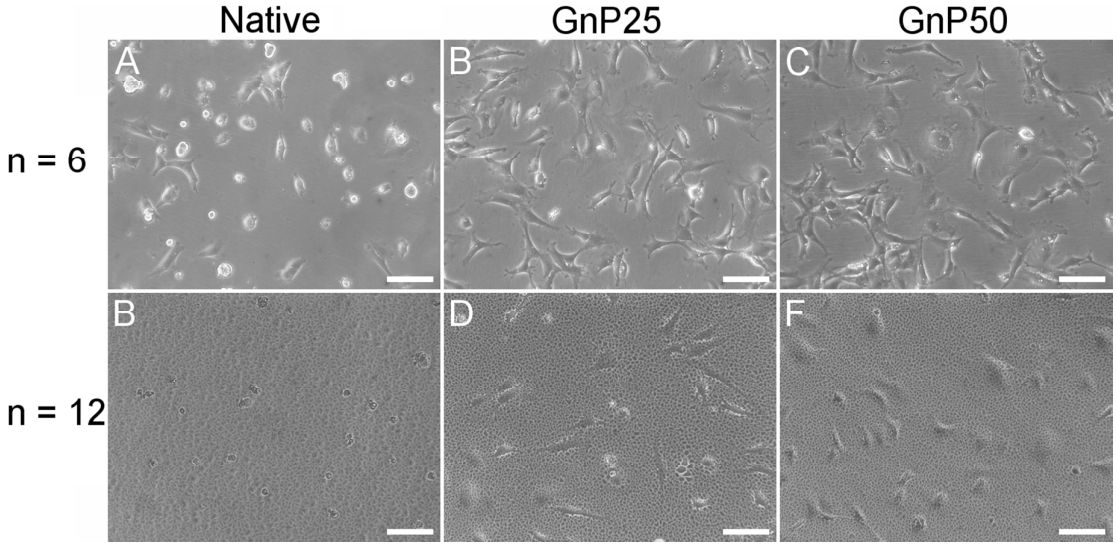

Figure S6. Optical micrographs of MC3T3-E1 pre-osteoblasts 24 h after seeding atop (A,B) native, (C,D) GnP25-crosslinked and, (E,F) GnP50-crosslinked PEI/(CSA/PLL)$_n$ (n = 6 and 12, respectively) films. Bar: 100 μm.