Supporting Information

Experimental Section

Fabrication of 1D NAPPy/TCA on Implants: 1D NAPPy/TCA over a large area on the titanium was formed via template-free electrochemical polymerization. Briefly, the small electrochemical cell included a biomedical titanium sheet (effective area of 15 mm × 15 mm) as a working electrode, copper sheet as a counter electrode, saturated calomel electrode (SCE) as a reference electrode, and an electrolyte. A prenucleation film (PNF) was first formed on the titanium. To form PNF, the titanium sheet (i.e., the working electrode) was immersed into the electrolyte (0.2 M KCl solution containing 0.1 M Py). Then the PNF was formed on the working electrode at 0.8 V (*vs* SCE) for 20 s at room temperature under the control of electrochemical station (Zennium Zahner, Germany). Subsequently, the PNF was rinsed in deionized water and dried in a vacuum. To fabricate 1D NAPPy/TCA, a phosphate buffer solution (PBS, 0.1 M, physiological pH value) containing 0.2 M Py and 0.07 M TCA was used as an electrolyte. 1D NAPPy/TCA was formed on the PNF-coated biomedical titanium (as working electrode) galvanostatically at 0.9 mA/cm^2 for 30 min. The as-obtained products were rinsed in deionized water for several times, and dried in vacuum.

Potential-induced reversible switch in wettability: To determine the redox potentials of 1D NAPPy/TCA, cyclic voltammetry (CV) was measured in an electrochemical system made of an electrolyte (PBS, pH 7.4, containing 0.07 M TCA), biomedical titanium sheet (deposited with NAPPy/TCA) as a working electrode, platinum electrode as a counter electrode, and SCE as a reference electrode. The CV curves of 1D NAPPy/TCA were recorded by applying a scanning potential from 0.8 V to -1.0 V at a scan rate of 20 mV/s. According to the resultant CV curves of 1D NAPPy/TCA, we determined that the reduction and oxidation potential of NAPPy/TCA were -0.42 V and +0.21 V, respectively. Then we used the electrochemical station to apply -0.80 V (reduction potential) to trigger reduction reactions of the 1D NAPPy/TCA coated titanium (working electrode) for 10 min. The 1D NAPPy/TCA coated titanium was then taken out of the station and dried in vacuum, followed by the measurement of static contact angle (SCA) of a 1

μL water droplet on 1D NAPPy/TCA by surface contact angle analyzer (Filderstadt OCA15, Germany) at ambient temperature. Then the 1D NAPPy/TCA coated titanium was placed into the station again and an oxidation potential (+0.50 V) was applied to trigger the oxidation reactions of the 1D NAPPy/TCA coated titanium (working electrode) for 10 min. The 1D NAPPy/TCA coated titanium was dried in vacuum again and the SCA of 1 μL water droplet on 1D NAPPy/TCA by surface was measured. At least five measurements were taken on each specimen, and the average and standard deviation were calculated. The reduction and oxidation potential were repeatedly applied to the 1D NAPPy/TCA coated titanium in sequence along with the subsequent SCA measurements after applying each potential in order to characterize the potential-induced reversible wettability.

Potential-induced reversible switch in protein adsorption: The 1D NAPPy/TCA specimens in switch-on or switch-off states were taken out of the electrochemical station and immersed in PBS (pH 7.4) for 30 min prior to protein adsorption. BSA, Fn and PAS, with isoelectric point of approximately 4.8 (acidic), 5.5 (acidic) and 12.0 (basic), respectively, were chosen as model proteins. The protein solution (0.1 mg/mL for BSA and PAS; 10 μg/mL for Fn) was freshly prepared by dissolving the protein in PBS. To study reversible switch of the protein adsorption, the surface of 1D NAPPy/TCA with a diameter of 10 mm was exposed to 1 mL protein solution and incubated at 37°C for 8 h. Then the protein solution was removed, followed by shaking the 1D NAPPy/TCA in 0.2 mL of 1 wt % sodium dodecylsulfate (SDS) to elute the adsorbed protein. The amount of the adsorbed protein on 1D NAPPy/TCA surface was determined by bicinchoninic acid (BCA) assay. The absorbance of the eluent was measured at 562 nm by a microplate reader (Thermo 3001, USA) with at least three repetitions for each group.

Cell culture and seeding on 1D NAPPy/TCA: MC3T3-E1 osteoblasts were cultured in α-modified minimum essential medium (α-MEM) supplemented with 10% fetal bovine serum (FBS) in a humidified incubator at 37 ℃ and 5% CO_2. Antibiotics (penicillin/streptomycin, P/S) were added to culture media for all cell growth experiments on the polymers. The medium was changed every 2-3 days. Before seeding, PPy-based materials placed in 48-well plates were sterilized by immersion in 70 % ethanol for 5 min, followed by drying under a sterile condition

and exposure to ultraviolet (UV) light for 20 min, and then immersed in α-MEM overnight. The cells were seeded at the density of 2.0×10^5 cells/mL. The cell viability at 1, 3, 5 and 7 days was used to quantitatively evaluate the toxicity of NAPPy/TCA by Cell Counting Kit-8 (CCK-8) assay.

Morphological observation of cells on 1D NAPPy/TCA by fluorescence staining: To image MC3T3-E1 osteoblasts on PPy-based materials after being cultured for 8 h, cells were fixed in ice-cold 4% paraformaldehyde for 30 min and then washed twice in PBS. Nuclei were stained with DAPI (1 μg/mL) for 10 min at room temperature. F-actin of cells was stained with 50 μg/mL rhodamin-labeled phalloidin in PBS for 20 min at room temperature. Then the cells were washed three times with PBS to remove unbound phalloidin. Images were acquired under a laser scanning confocal microscope (Zeiss LSM 780, Germany).

Characterization: Field emission scanning electron microscopy (FE-SEM, ZEISS Ultra 55, Germany) with acceleration voltage of 5 kV, was employed to examine the 1D NAPPy/TCA coated with gold nanolayer. Attenuated total reflection Fourier-transform infrared (ATR-FTIR, Bruker Vector 33, Germany) and electron probe micro-analyzer (EPMA, Shimadzu 1720, Japan) were utilized to analyze the chemical composition of as-obtained products. The surface potentials of specimens were characterized using Kelvin probe force microscopy (KFM, Shimadzu SPM-9600, Japan). For investigating the effect of cell growth on the TCA conformation or orientation between α-face and β-face of TCA in 1D NAPPy/TCA, MC3T3-E1 osteoblasts were cultured on 1D NAPPy/TCA in switch-off state and switch-on state for 3 days. Prior to characterization by KFM, the cells were removed from 1D NAPPy/TCA by rinsing ultrasonically in deionized water for 10 min.

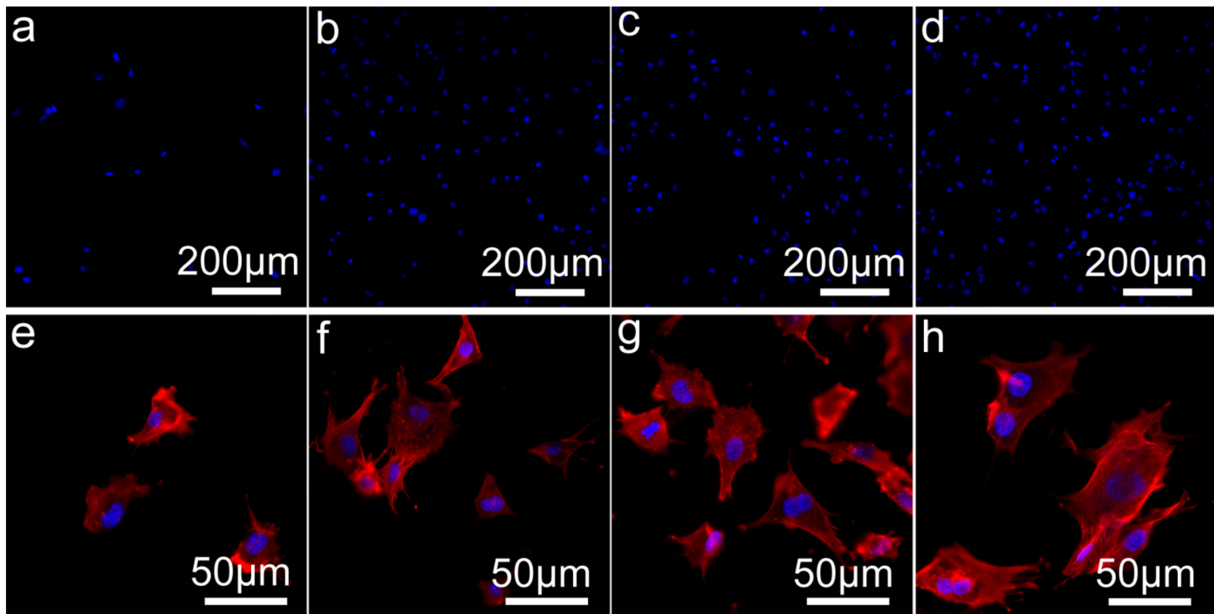

Figure S1. Fluorescence images of MC3T3-E1 osteoblasts seeding on PPy/TCA irregular films (a and e), 1D NAPPy/TCA (b and f), PPy/TCA tightly packed microparticles (c and g) and unmodified Ti substrates (d and h) for 8 h. Nuclei were stained to be blue and cytoskeletal actin was stained to be red. This result shows that 1D NAPPy/TCA could promote the adhesion when compared to other irregular PPy films and showed similar capability in cell adhesion as un-coated Ti implants and PPy/TCA microparticles on Ti.

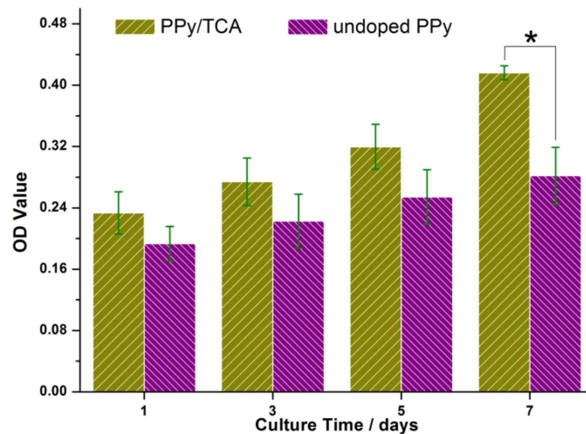

Figure S2. The plot of optical density (OD) values versus time of culturing MC3T3-E1 osteoblasts on PPy/TCA (i.e., 1D NAPPy/TCA) and undoped PPy determined by cell counting kit 8 (CCK-8) assay. The * indicated significant difference ($p<0.05$). It is well-known that PPy has excellent tissue compatibility, and we therefore used the undoped PPy as a control sample to test the cell toxicity of PPy/TCA by CCK-8

assay. This result showed that OD values of PPy/TCA were higher than that of undoped PPy, implying that the PPy/TCA is non-toxic for cells.

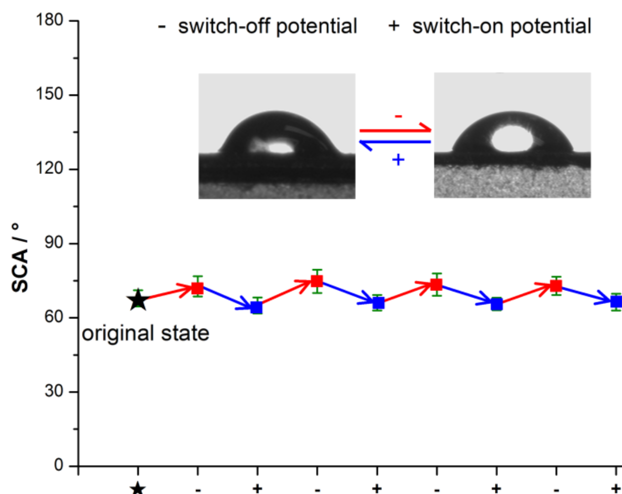

Figure S3. The plots of SCA of PPy/Cl versus switch-off potentials (-0.80 V) and switch-on potentials (+0.50 V) *in situ*. Inset: SCA images. This result showed that when the dopant of 1D NAPPy was changed from TCA to Cl^-, the reversible switch in wettability becomes trivial.

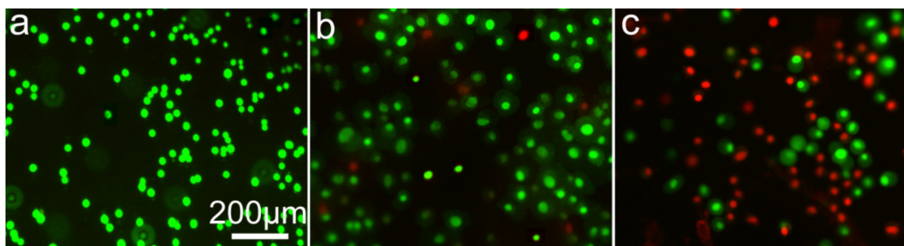

Figure S4. Acridine orange–propidium iodide (AO/PI) staining fluorescence images of MC3T3-E1 osteoblasts on 1D NAPPy/TCA after applying 50 cycles of periodic switching potentials with different magnitudes. (a) -0.80 V / +0.50 V, (b) -1.80 V / +1.50 V, (c) -2.80 V/+2.50 V. The MC3T3-E1 osteoblasts viability on 1D NAPPy/TCA after applying 50 cycles of different magnitudes of periodic switching potentials was assayed using AO/PI staining. MC3T3-E1 osteoblasts on 1D NAPPy/TCA were incubated with the AO/PI mixture for 10 min and observed under a fluorescence microscope. Live cells were stained green (AO) while dead cells were colored red (PI). Figure S4 showed that under the weak periodic switching potentials (-0.80 V / +0.50 V), which were applied to induce reversible switch in

wettability and protein adsorption in this work, all the cells remained alive during 50 cycles of switching potential. However, when the magnitudes of periodic switching potentials was increased (-1.80 V / +1.50 V, -2.80 V / +2.50 V), some cells became dead, suggesting that relatively high potentials were unacceptable for MC3T3-E1 osteoblasts.

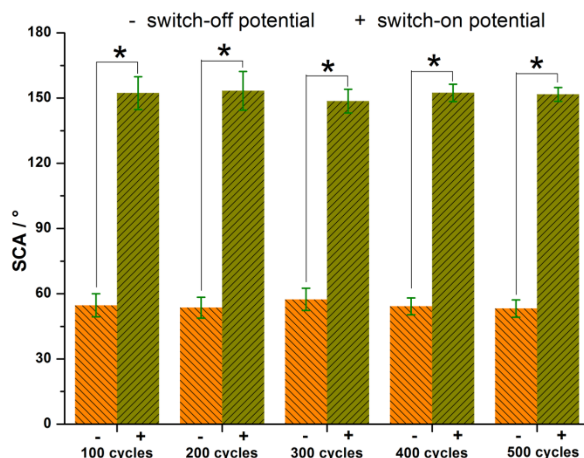

Figure S5. The plots of SCA of 1D NAPPy/TCA after applying various cycle numbers of periodic switching potentials. The * indicated significant difference ($p<0.05$). This result showed that the SCA of 1D NAPPy/TCA in both switch-on and switch-off states remained relatively stable during the 500 cycles of periodic switching potentials, indicating a good stability of potential-induced wettability of 1D NAPPy/TCA films on Ti substrate.

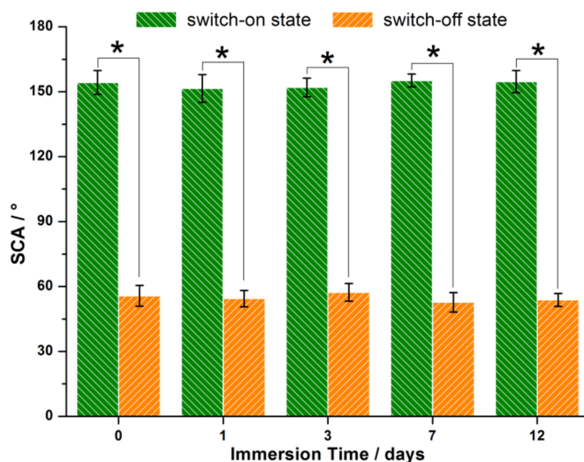

Figure S6. The plots of SCA of 1D NAPPy/TCA in switch-on (hydrophobic) state and switch-off (hydrophilic) state versus the time of immersion in PBS. The * indicated significant difference ($p<0.05$). This data showed that regardless of the time of immersion in PBS, both the switch-on state and switch-

off state of specimens exhibited a relatively stable static contact angle. Therefore, the 1D NAPPy/TCA was stable in physiological condition.

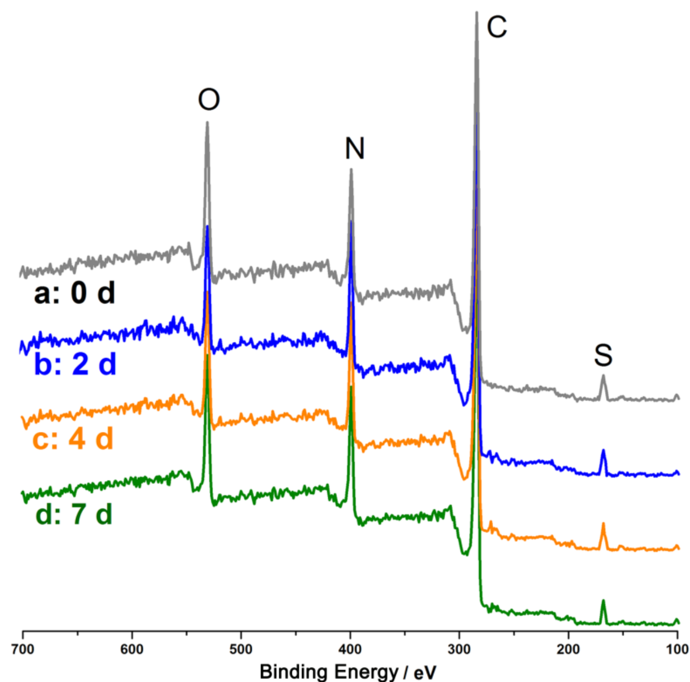

Figure S7. XPS of 1D NAPPy/TCA after immersion in PBS for 0 (a), 2 (b), 4(c) and 7 (d) days. To find out whether TCA would be released from 1D NAPPy/TCA in physiological medium, the specimens were placed in PBS (pH 7.4) for various times. After the 1D NAPPy/TCA was washed with deionized water, XPS was employed to analyze the TCA amount in specimens. This figure showed that no visible change in the peak intensity of S (sulphur, presence in TCA molecule) could be observed for up to 7 days, implying that almost no TCA was released from 1D NAPPy/TCA in physiological medium. In addition, the intensities of other peaks also remained nearly unchanged, further suggesting good chemical stability in physiological conditions.

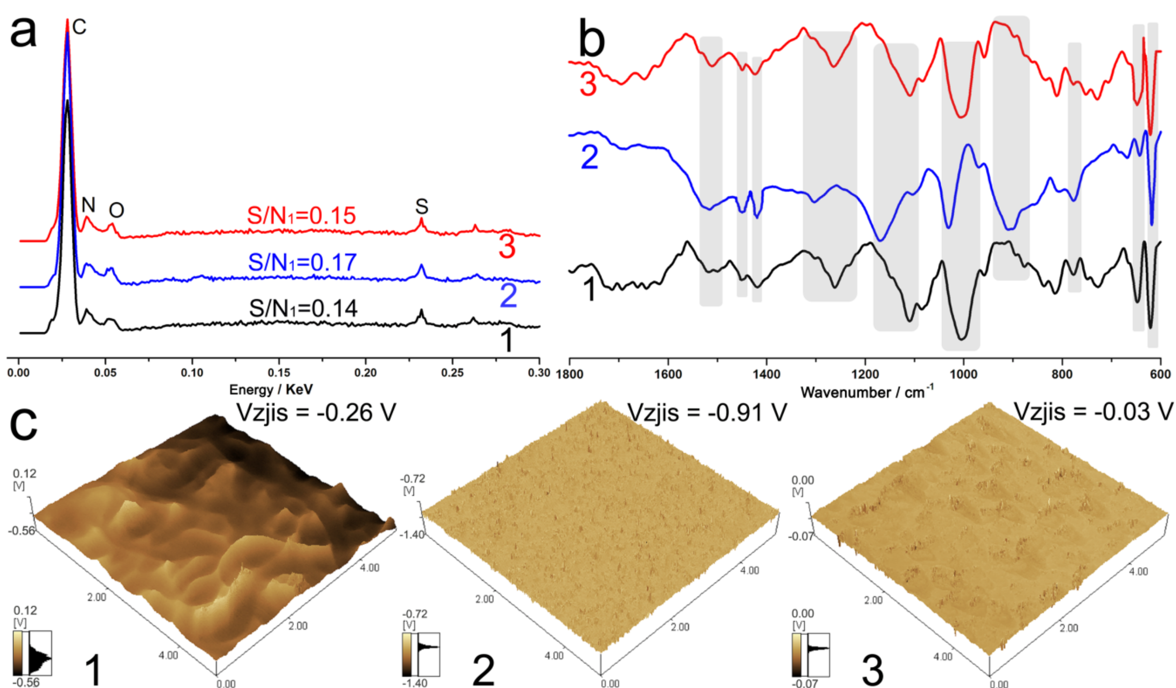

Figure S8. Electron probe micro-analysis (EPMA) profiles (a) and attenuated total reflection Fourier-transform infrared spectroscopy (ATR-FTIR) spectra (b) and Kelvin probe force microscopy (KFM) surface potential patterns of 1D NAPPy/TCA in original state (1) and switch-off state (2) and switch-on state (3). With S/N_1 representing the molar ratio of TCA to Py monomer, where S is the number of sulfur in TCA and N_1 is the number of nitrogen in TCA, original state, switch-off state and switch-on state of 1D NAPPy/TCA exhibited an S/N_1 value of 0.14, 0.17 and 0.15 (**Figure S8a**), respectively, suggesting no significant difference in TCA concentration between the two switching states. Interestingly, several changes emerged on ATR-FTIR spectra of specimens in two switch states (**Figure S8b**). Three peaks at 1520, 1451 and 777 cm^{-1} appeared in all spectra and could be assigned to the C=C stretching vibration, C–N stretching vibration, C-H out-plane ring deformation of PPy, respectively (Reference: G. Lu, C. Li, G. Shi, *Polymer* **2006**, *47*, 1778.). The peaks at 1426, 647 and 621 cm^{-1} corresponded to $-CH_3$ deformation vibration, -OH out-plane deformation vibration and O=C-NH deformation vibration of TCA, respectively. The peak regions of 1320 – 1250 cm^{-1}, 1180 – 1100 cm^{-1} and 1040 – 990 cm^{-1} were attributed to the -OH stretching vibration, asymmetric and symmetric O=S=O stretching vibration (strong) of TCA, respectively (Reference: Y. Zhu, D. Hu, M. Wan, L. Jiang, Y. Wei, *Adv. Mater.* **2007**, *19*, 2092.). It was obvious that, compared with **Figure S8b1** (original state) and **Figure S8b3** (switch-on state), the three peaks in **Figure S8b2** (switch-off state) exhibited a blueshift in those peak regions, implying that the chemical environment of TCA molecule in switch-off state was different from that in other states. In addition, a

strong peak at 905 cm^{-1} only observed in **Figure S8b2** might be assignable to -C-O^- generated due to the deprotonation of a part of -C-OH in forming reduction state of TCA, which was confirmed by the decreased ratio in the intensity of peaks at 647 cm^{-1} (-OH) and 621 cm^{-1} (O=C-NH).

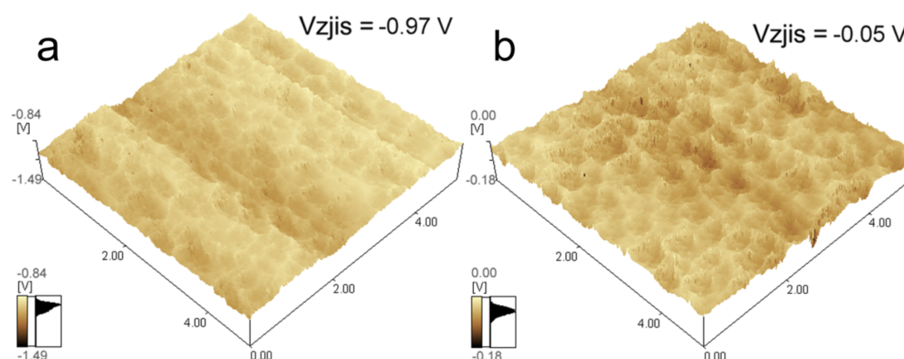

Figure S9. KFM surface potential patterns of 1D NAPPy/TCA in switch-off state (a) and switch-on state (b) after seeding MC3T3-E1 osteoblasts for 3 days. A comparison in the surface potential (Vzjis) shown in Figures S8c suggested that the cell growth on 1D NAPPy/TCA almost did not change the surface potential in switch-on or switch-off state, indicating that the cell growth did not alter the orientation (exposure of α-face or β-face on the surface) of TCA in 1D NAPPy/TCA.

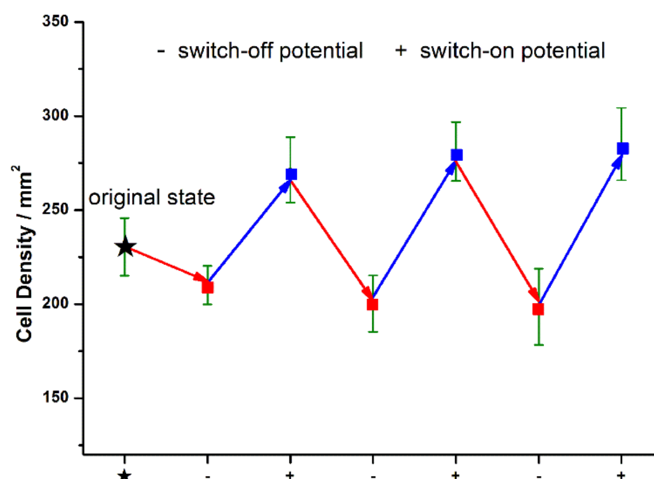

Figure S10. The reversible change in cell density (the number of cells per mm^2) on 1D NAPPy/TCA versus switch-off and switch-on states. The number of cells was counted from DAPI-stained fluorescence cell images.

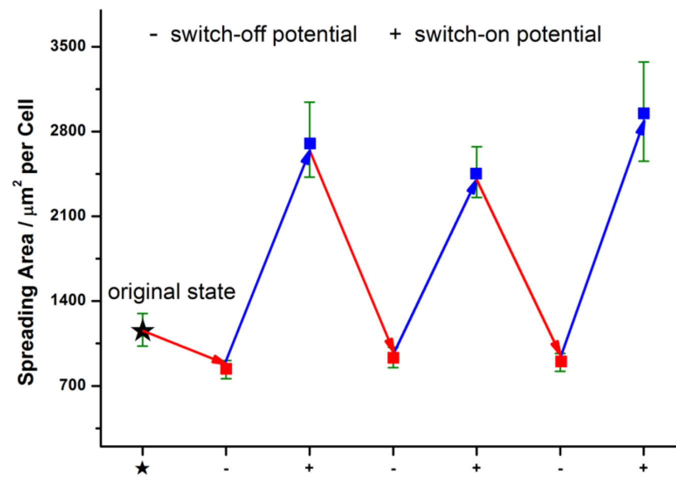

Figure S11. The plots of spreading area (average area per cell) of MC3T3-E1 osteoblasts on 1D NAPPy/TCA versus switch-off and switch-on states.