Electronic Supplementary Material (ESI) for RSC Advances.


# Removal of microplastics via tannic acid-mediated coagulation and *in vitro* impact assessment

## 1. Formation of coordination complex of tannic acid and $FeCl_3$

0.033 – 0.001 mM of tannic acid and 3 mM of $FeCl_3$ solution was dissolved in DI water using vortex (DH.WVM00010, DAIHAN) and sonication (NXP 1002, NEXUL). The two solutions were mixed at 1 : 1 volume ratio. Formation of coordination complex was checked using an optical microscopy (Eclipse TS100, Nikon) and ultraviolet–visible spectrophotometer (Biochrom Libra S50).

## 2. Coagulation by Fe-based salts on PS beads

2.5 wt% bead solution was diluted to 0.5 wt% using distilled water. The 0.5 wt% bead solution and 3 mM FeCl3 solution were mixed in a 2 : 1 volume ratio. After 2 h, the mixed solution was checked for precipitation using a microscopy.

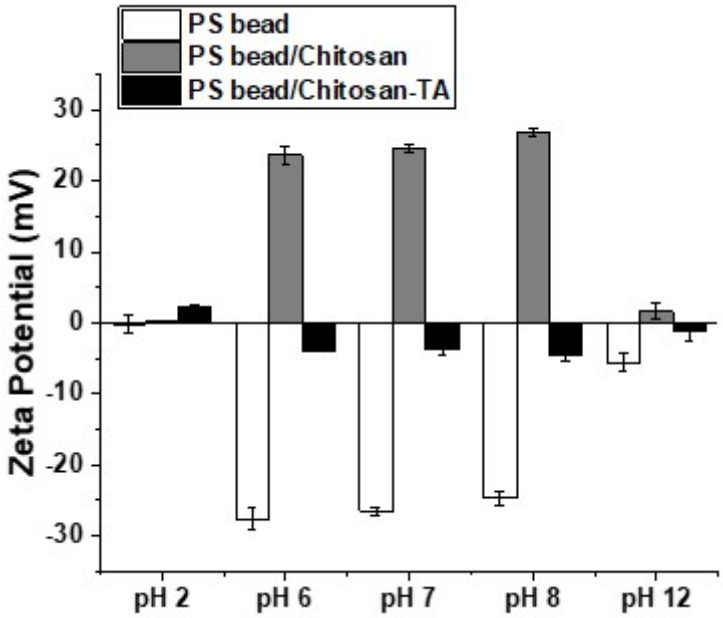
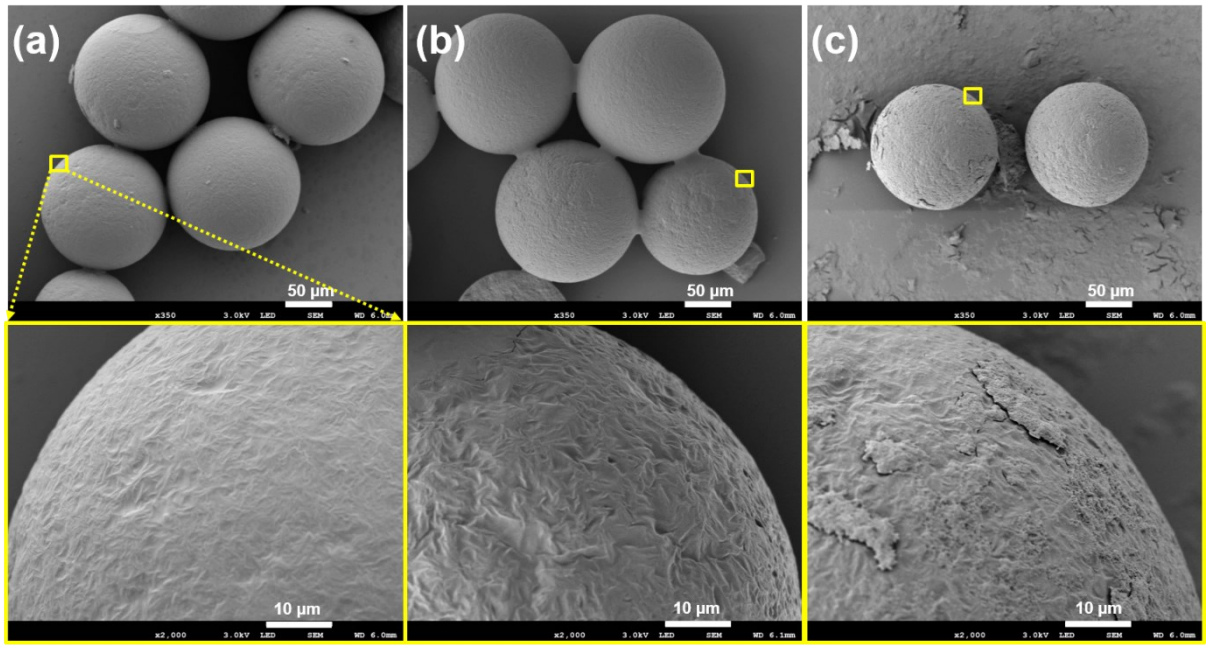



**Figure S1.** pH-dependent zeta potential of PS beads (90 μm) according to the modification.




(d)

| Atomic % / Sample | C | N | O |
|---|---|---|---|
| PE beads | 88.44 | 3.64 | 7.86 |
| PE beads /Chitosan | 95.67 | 2.58 | 1.73 |
| PE beads /Chitosan-TA | 83.09 | 3.53 | 13.35 |

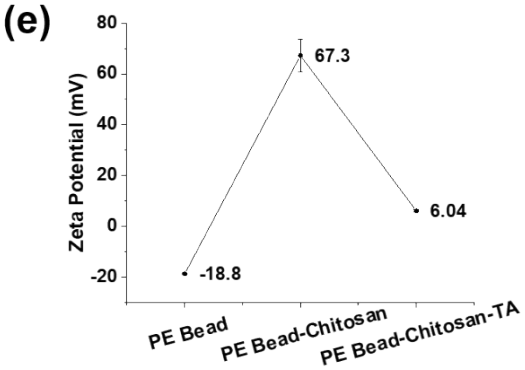
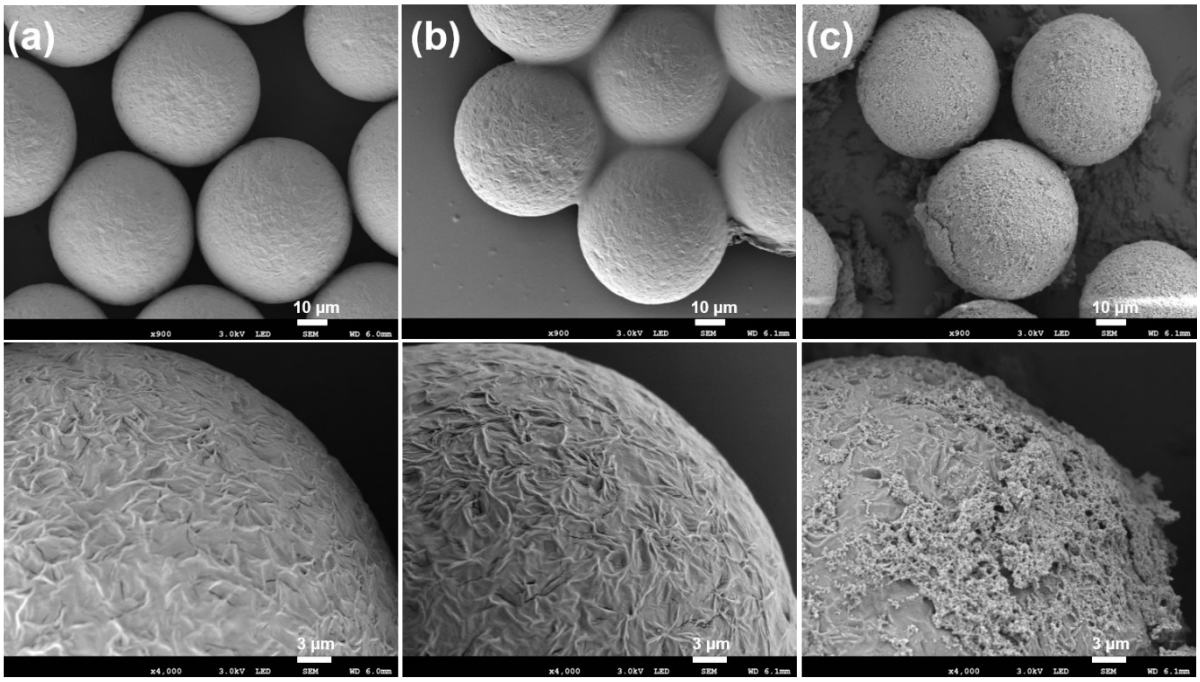



**Figure S2.** Scanning electron microscopy images of (a) PE beads (106–125 μm) and (b) modification with chitosan and (c) with chitosan and tannic acid. (d) Surface element analysis of the PE beads through scanning electron microscopy/energy-dispersive X-ray spectroscopy according to the modification of 106–125 μm PE bead. (e) Zeta potetential of the PE beads according to the modification.




(d)

| Atomic % / Sample | C | N | O |
|---|---|---|---|
| PE beads | 95.27 | 3.41 | 2.65 |
| PE beads /Chitosan | 95.38 | 2.82 | 1.76 |
| PE beads /Chitosan-TA | 85.11 | 2.53 | 12.30 |

(e)

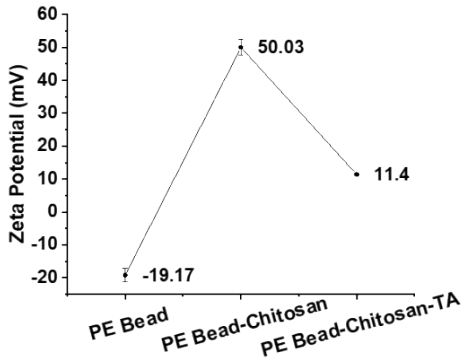



**Figure S3.** Scanning electron microscopy images of (a) PE beads (45–53 μm) and (b) modification with chitosan and (c) with chitosan and tannic acid. (d) Surface element analysis of the PE beads through scanning electron microscopy/energy-dispersive X-ray spectroscopy according to the modification of 45–53 μm PE bead. (e) Zeta potetential of the PE beads according to the modification.

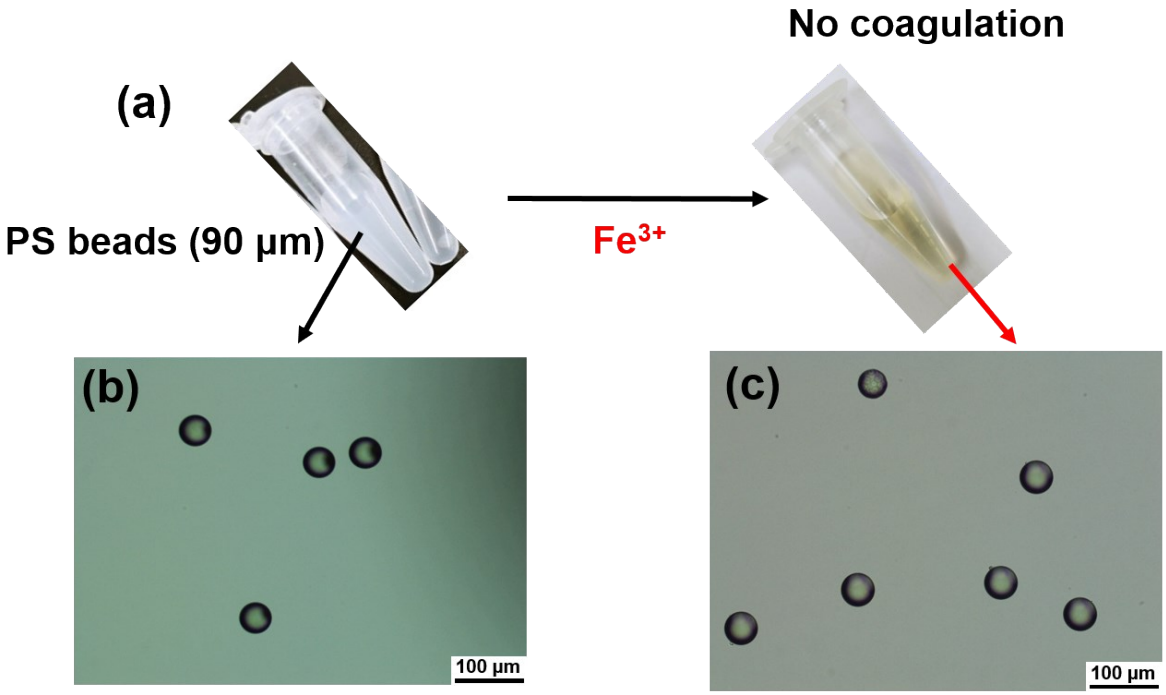



**Figure S4.** Image of $Fe^{3+}$ added to untreated 90 μm PS bead.


0.033 mM, 0.016 mM, 0.008 mM, 0.004 mM, 0.002 mM, 0.001 mM


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**Figure S5.** Image of reaction with $Fe^{3+}$ by diluting tannic acid 0.5 times at 0.033 mM. (a) Image of tannic acid reacted with $Fe^{3+}$ up to 0.016 mM. (b) Image of reaction $Fe^{3+}$ up to 0.004 mM when chitosan and tannic acid were treated with beads.

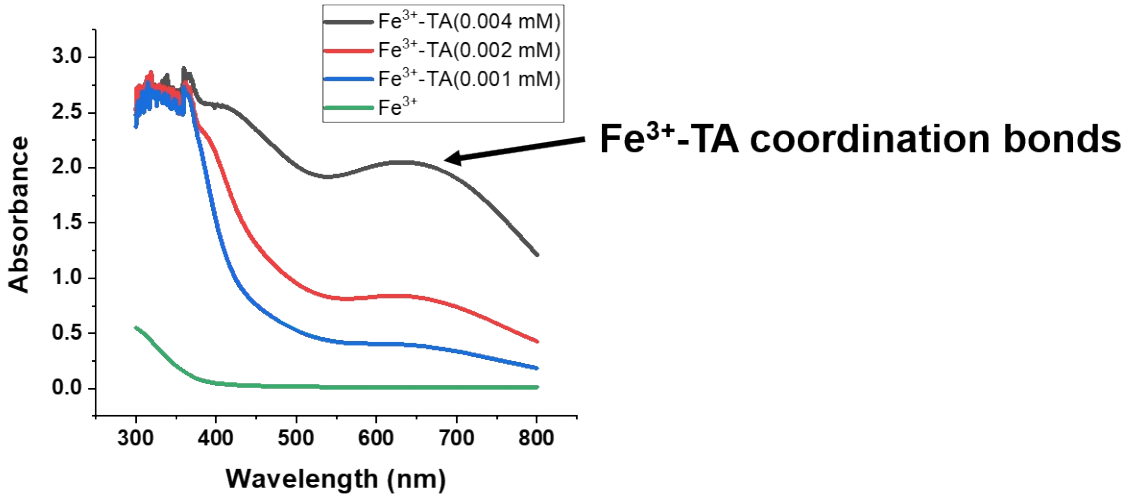


**Figure S6**. UV-Vis spectra of coagulated PS beads according to various concentrations of tannic acid

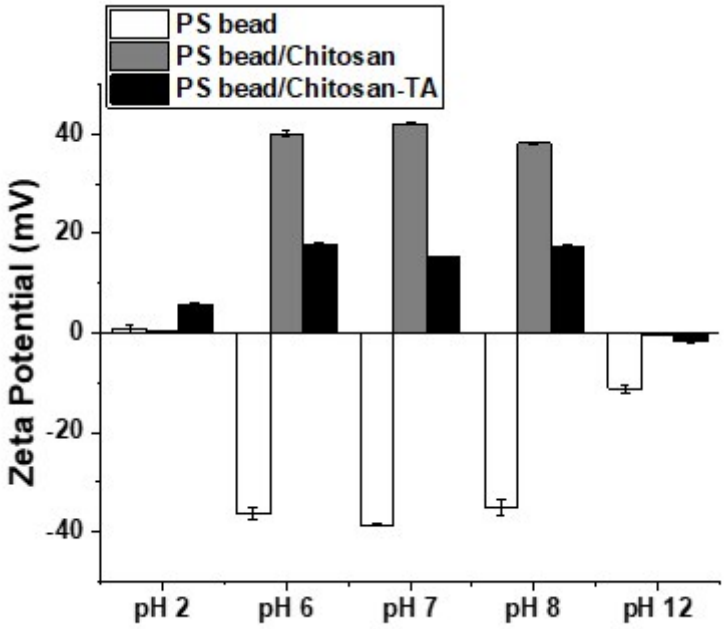


**Figure S7.** pH-dependent zeta potential of PS beads (0.5 μm) according to the modification.

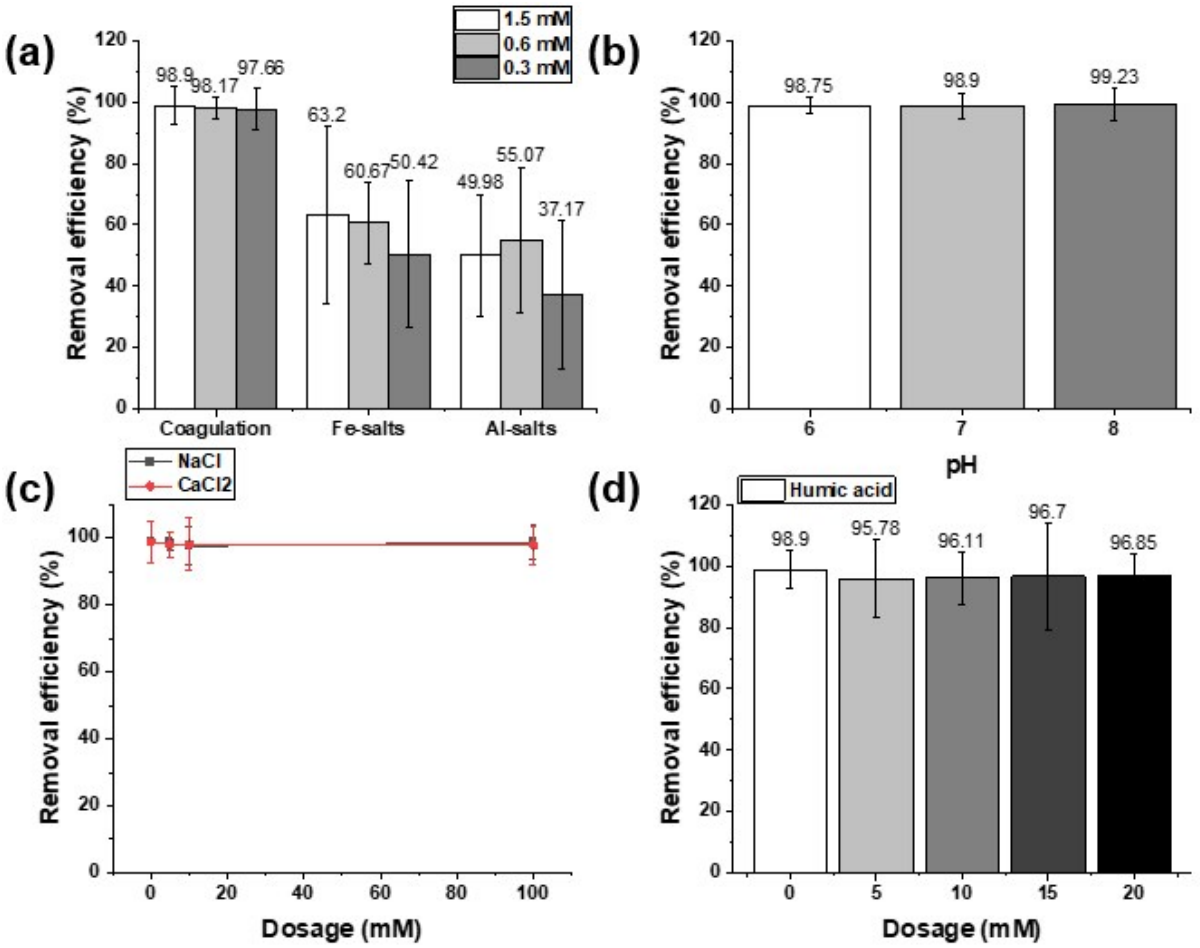


**Figure S8**. (a) Concentration of metal ion (1.5, 0.6, 0.3 mM) in coagulant-dependent removal efficiency of 0.5 μm PS beads (0.1 mg/mL). (b) Sample pH-dependent removal efficiency of 0.5 μm PS beads. (c) Actual water conditions, including various kinds of ions, removal efficiency of 0.5 μm PS beads. (d) Actual water conditions, including humic acid in NOM, removal efficiency of 0.5 μm PS beads.

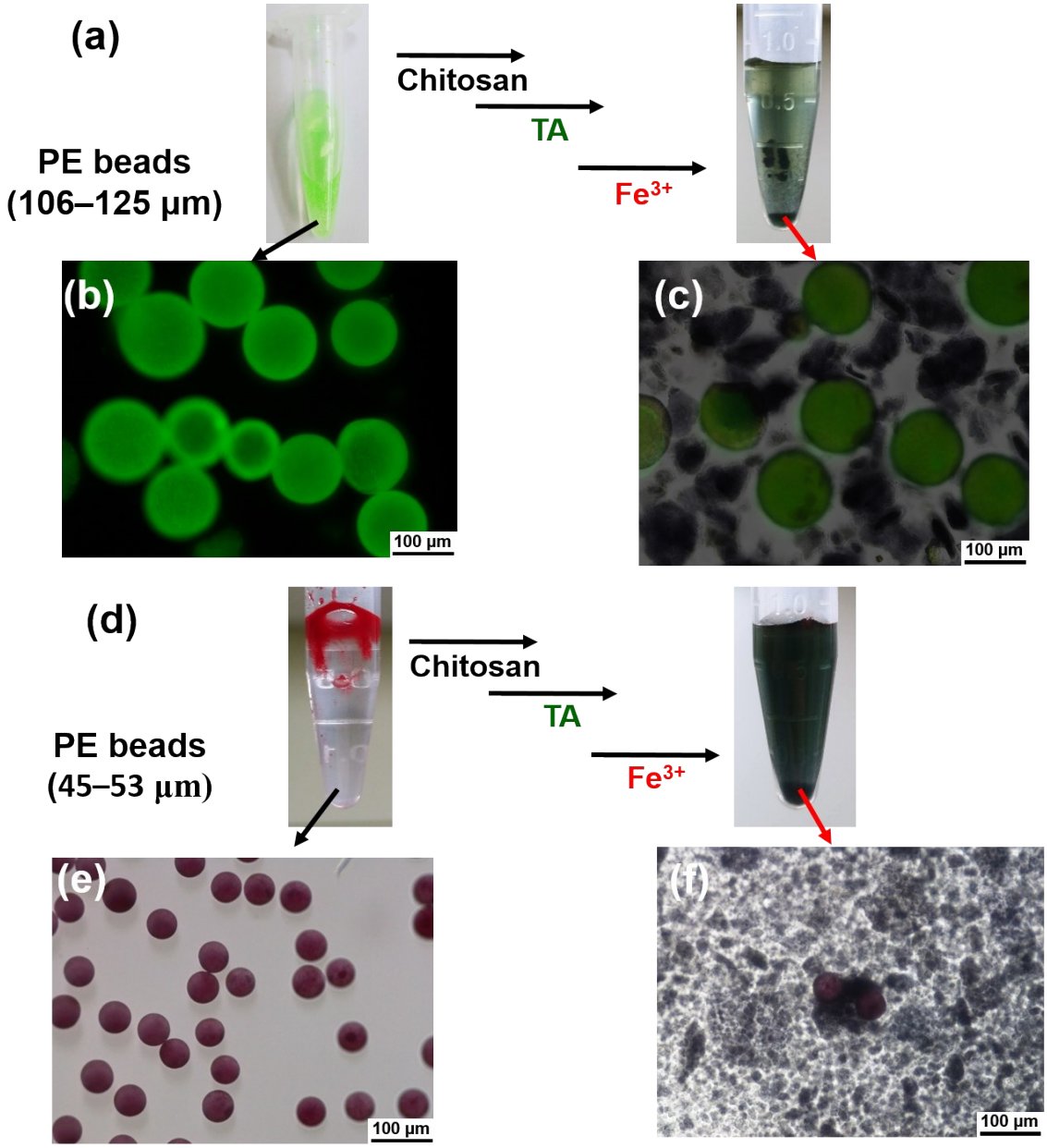


**Figure S9.** (a) Photographs of coagulation process of PE beads (106–125 µm). Fluorescent microscopy image of (b) non-treated PE beads and (c) coagulated beads. (d) Photographs of coagulation process of PE beads (45–53 µm). Fluorescent microscopy image of (e) non-treated PE bead and (f) coagulated beads.

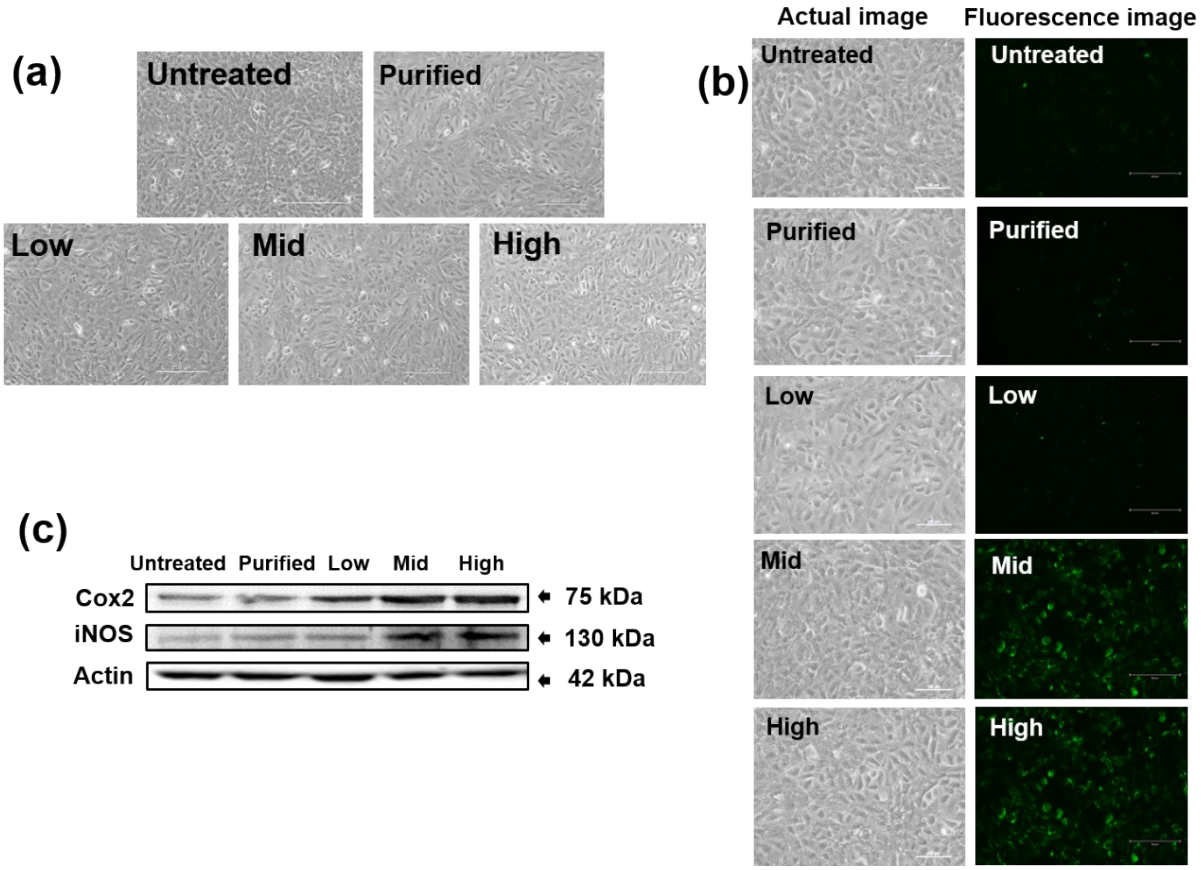


**Figure S10.** Optical microscopy image from (a) IEC18 Cell viability and (b) ROS tests. Optical microscopic images of Western blot from (c) *in vitro* inflammation tests of PS beads (0.5 μm). Sample of purified PS beads was compared with low (0.01 mg/mL), middle (0.05 mg/mL) and high (0.1 mg/mL) concentration of non-treated PS beads for these tests.

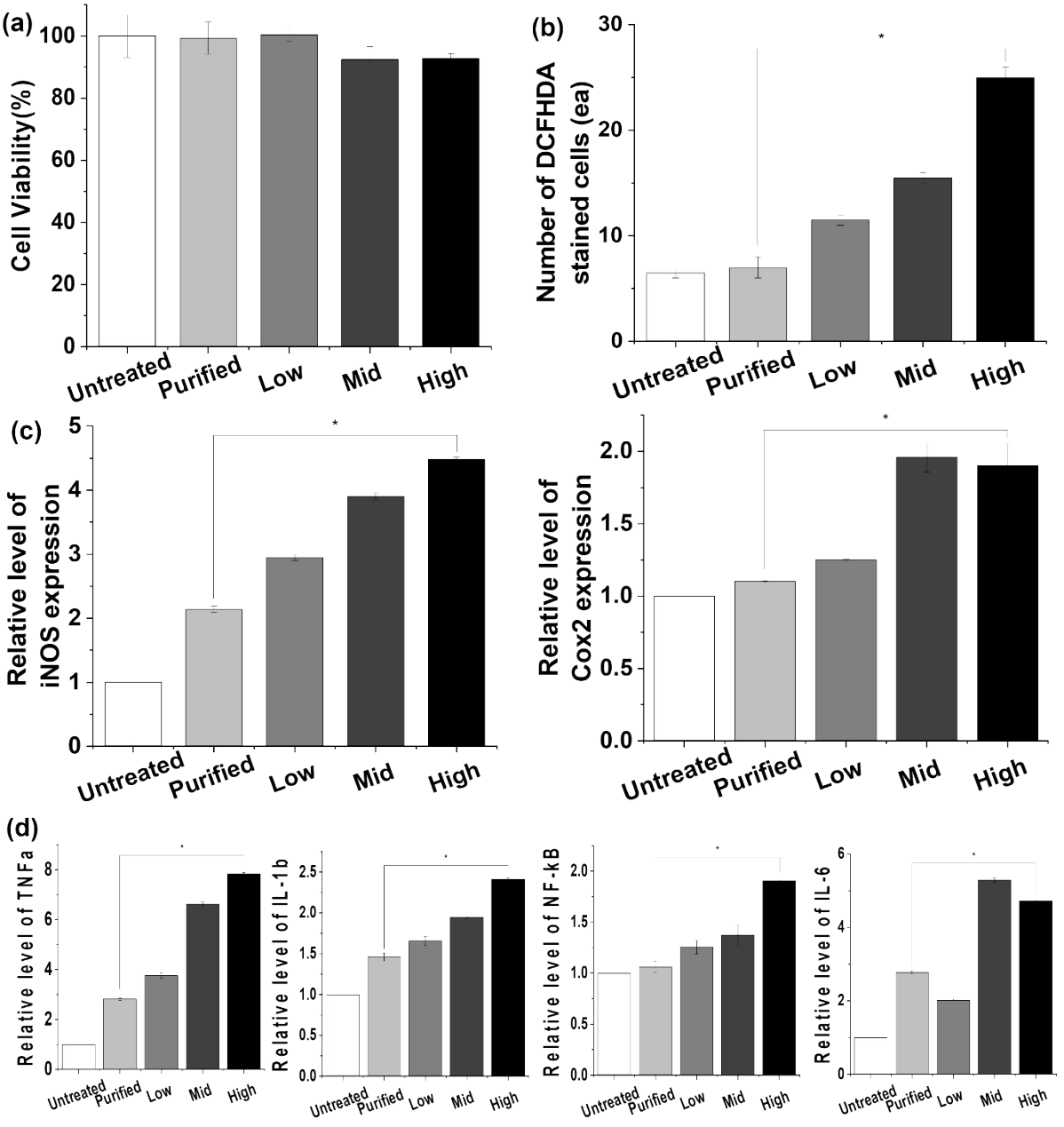


**Figure S11.** (a) HepG2 Cell viability, (b) ROS tests, (c) *in vitro* inflammation, and (d) cytokine tests of PS beads (0.5 μm). Sample of purified PS beads was compared with low (0.01 mg/mL), middle (0.05 mg/mL) and high (0.1 mg/mL) concentration of non-treated PS beads for these tests.

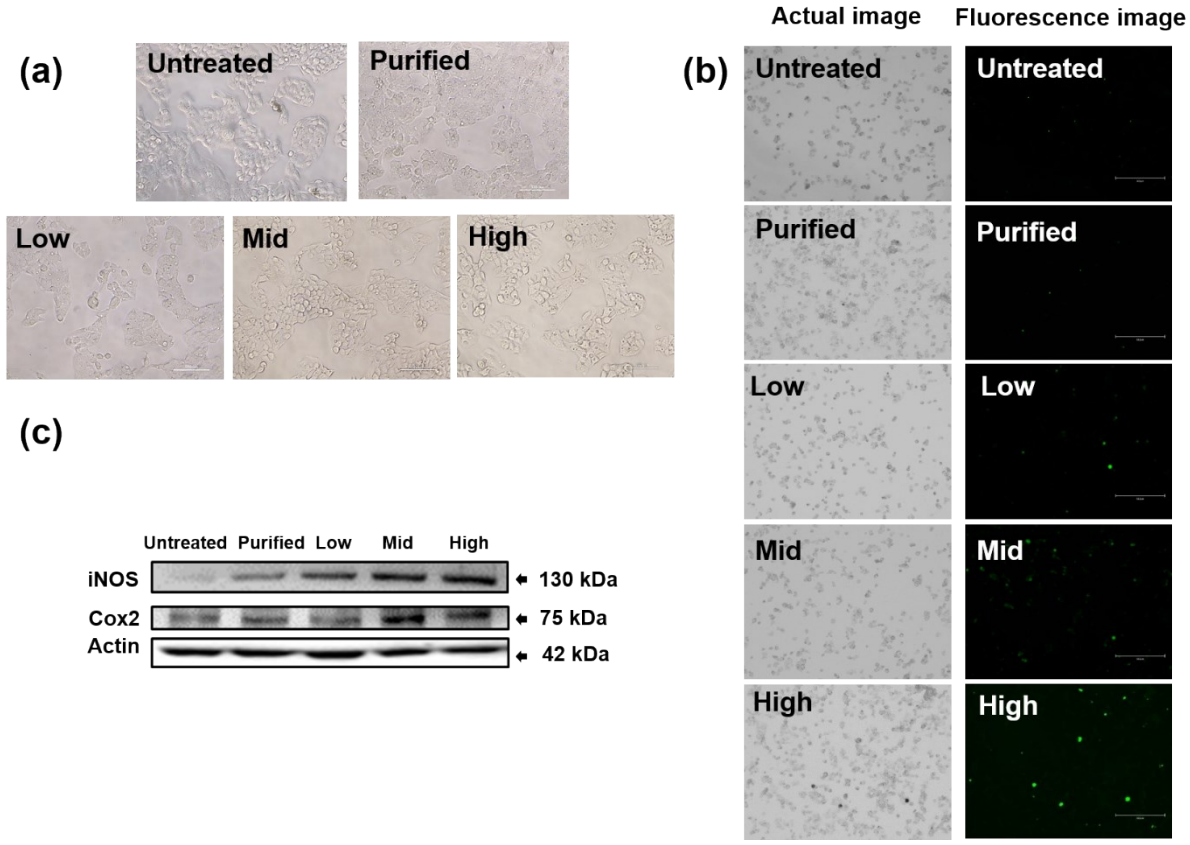


**Figure S12.** Optical microscopy image from (a) HEpG2 Cell viability and (b) ROS tests. Optical microscopic images of Western blot from (c) *in vitro* inflammation tests of PS beads (0.5 μm). Sample of purified PS beads was compared with low (0.01 mg/mL), middle (0.05 mg/mL) and high (0.1 mg/mL) concentration of non-treated PS beads for these tests.