

## Mussel-inspired hydrogel for fast fabrication of flexible SERS tape for point-of-care testing of $\beta$ -blockers

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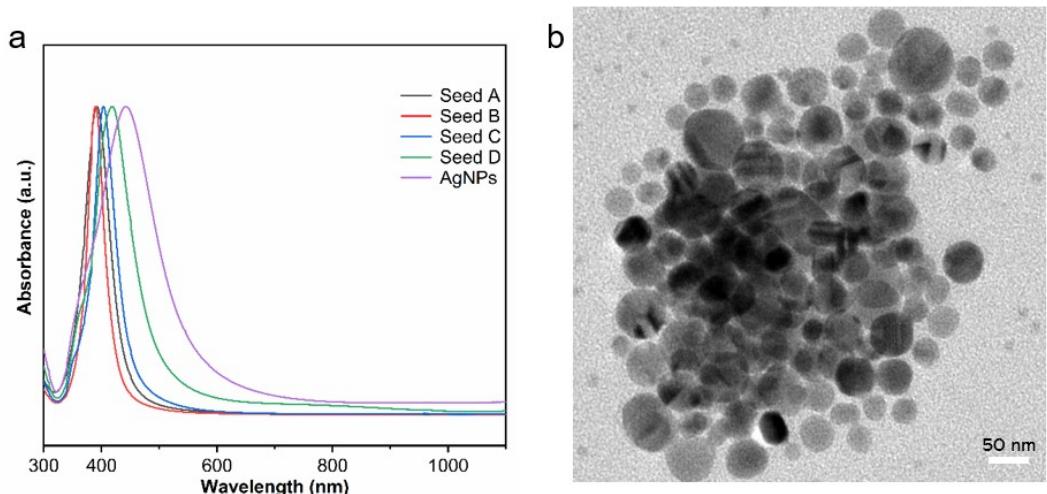
**Table S1. Composition of the hydrogels**

Samples	AA (mL)	Al(NO <sub>3</sub> ) <sub>3</sub> •9H <sub>2</sub> O (g)	CHI (g)	GA (g)	BIS (mg)	KPS (g)	Deionized water (g)
P	3	0	0	0	10	0.08	10
P-0.3C	3	0	0.3	0	10	0.08	10
P-0.08Al	3	0.08	0	0	10	0.08	10
P-0.3C-0.06GA	3	0	0.3	0.06	10	0.08	10
P-0.3C-0.06GA-0.02Al	3	0.02	0.3	0.06	10	0.08	10
P-0.3C-0.06GA-0.04Al	3	0.04	0.3	0.06	10	0.08	10
P-0.3C-0.06GA-0.06Al	3	0.06	0.3	0.06	10	0.08	10
P-0.3C-0.06GA-0.08Al	3	0.08	0.3	0.06	10	0.08	10
P-0.3C-0.06GA-0.10Al	3	0.10	0.3	0.06	10	0.08	10
P-0.08Al-0.06GA	3	0.08	0	0.06	10	0.08	10
P-0.15C-0.06GA-0.08Al	3	0.08	0.15	0.06	10	0.08	10
P-0.45C-0.06GA-0.08Al	3	0.08	0.45	0.06	10	0.08	10
P-0.3C-0.08Al	3	0.08	0.3	0	10	0.08	10
P-0.3C-0.01GA-0.08Al	3	0.08	0.3	0.01	10	0.08	10
P-0.3C-0.02GA-0.08Al	3	0.08	0.3	0.02	10	0.08	10
P-0.3C-0.04GA-0.08Al	3	0.08	0.3	0.04	10	0.08	10
P-0.3C-0.06GA-0.086Fe <sup>*</sup>	3	0.086	0.3	0.08	10	0.08	10
P-0.3C-0.06GA-0.051Cu <sup>*</sup>	3	0.055	0.3	0.08	10	0.08	10
P-0.3C-0.06GA-0.085Cr <sup>*</sup>	3	0.063	0.3	0.08	10	0.08	10

\*As for P-0.3C-0.06GA-0.086Fe, P-0.3C-0.06GA-0.055Cu and P-0.3C-0.06GA-0.063Cr, 0.086 g of Fe(NO<sub>3</sub>)<sub>3</sub>•9H<sub>2</sub>O, 0.055 g of Cu(NO<sub>3</sub>)<sub>2</sub>•3H<sub>2</sub>O or 0.063 g of Cr(NO<sub>3</sub>)<sub>3</sub>•9H<sub>2</sub>O was added instead of Al(NO<sub>3</sub>)<sub>3</sub>•9H<sub>2</sub>O. All the samples listed here were synthesized following the same methodology mentioned in the “Preparation of hydrogel” session.

**Table S2. Experimental vibrational band wavenumbers of BSE, MET, ESE and ACE.**

BSE	MET	ESE	ACE
363 $\delta(\text{O-CH}_2)$	357 $\delta(\text{OH})+\delta(\text{CH})+\delta(\text{CH}_2)+\delta(\text{CH}_3)$	336 $\delta(\text{O-CH}_2)$	
485 $\rho(\text{CH}_2)$		486 $\rho(\text{CH}_2)$	485 $\rho(\text{CH}_2)$
		575 $\tau(\text{CCC ring})+\delta(\text{OC-NH})$	571 $\tau(\text{CCC ring})+\delta(\text{OC-NH})$
644 $\delta(\text{CCC ring})+\rho(\text{CH}_2)$	643      in-plane ring deformation	644 $\delta(\text{CCC ring})+\rho(\text{CH}_2)$	644 $\delta(\text{CCC ring})$
749 $\delta(\text{NH})+\delta(\text{CC ring})$	749 $\delta(\text{CCC ring})+\rho(\text{CH}_2)$	748 $\delta(\text{CCC ring})+\delta(\text{NH})$	750 $\delta(\text{CCC ring})+\delta(\text{NH})$
821 $\omega(\text{CH})+\text{ring breathing}$	821 $\gamma(\text{CH ring})$	816 $\gamma(\text{CH ring})$	818 $\gamma(\text{CH ring})$
889 $\nu(\text{CO})+\nu(\text{NH})$	890      ring breathing	890 $\nu(\text{CO})+\nu(\text{NH})$	890 $\nu(\text{CO})+\nu(\text{NH})$
910 $\nu(\text{CO})+\rho(\text{CH}_2)$	911 $\nu_{\text{as}}(\text{CCN})+\delta(\text{CH}_2)$	911 $\nu(\text{CO})+\rho(\text{CH}_2)$	912 $\nu(\text{CO})+\rho(\text{CH}_2)$
953 $\delta(\text{CH}_3)+\delta(\text{CH}_2)$	952 $\delta(\text{CH}_3)+\delta(\text{CH}_2)$	955 $\delta(\text{CH ring})+\delta(\text{CH}_2)$	952 $\delta(\text{CH ring})+\delta(\text{CH}_2)$
999 $\nu(\text{CO})+\rho(\text{CH}_2)$	998 $\delta(\text{CH ring})+\text{in plane ring deformation}$	998 $\nu(\text{CO})+\rho(\text{CH}_2)$	999 $\nu(\text{CO})+\rho(\text{CH}_2)$
	1063 $\nu(\text{OC})$		
1073 $\nu(\text{OC})$	1075 $\nu(\text{CH-CH})+\delta(\text{CH}_3)+\delta(\text{CH}_2)$	1077 $\nu(\text{OC})$	1075 $\nu(\text{CCH})+\delta(\text{CCH})$
1102 $\nu(\text{CC})+\delta(\text{CCH})$	1103 $\nu_{\text{as}}(\text{OCC})$	1102 $\nu(\text{CC})$	1103 $\nu(\text{CC})$
1115 $\delta(\text{CH ring})$	1116 $\rho(\text{CH}_2)+\rho(\text{CH}_3)+\rho(\text{NH})$	1119 $\rho(\text{CH}_2)+\rho(\text{CH}_3)+\rho(\text{NH})$	1118 $\delta(\text{CH ring})$
1176 $\nu(\text{CC})+\delta(\text{CH ring})$	1176 $\rho(\text{CH}_2)+\rho(\text{CH}_3)$		1178 $\nu(\text{OCC})+\delta(\text{CH ring})$
	1208 $\delta(\text{OH})+\delta(\text{CH})$	1203 $\delta(\text{CC ring})$	1207 $\delta(\text{CC ring})$
1217 $\nu_{\text{as}}(\text{CC ring})+\tau(\text{CH}_2)$	1218 $\nu_{\text{as}}(\text{CC ring})+\tau(\text{CH}_2)$	1215 $\nu_{\text{as}}(\text{OC})$	1217 $\nu_{\text{as}}(\text{OC})$
1244 $\tau(\text{CH}_2)+\rho(\text{CH}_2)$	1244 $\nu_{\text{as}}(\text{OCC})+\delta(\text{CC})$	1244 $\tau(\text{CH}_2)+\rho(\text{CH}_2)$	1244 $\tau(\text{CH}_2)+\rho(\text{CH}_2)$
1256 $\tau(\text{CH}_2)+\rho(\text{CH}_2)$	1257 $\rho(\text{CH}_2)$	1252 $\nu_{\text{as}}(\text{OCC})+\delta(\text{CH})$	1257 $\nu_{\text{as}}(\text{OCC})+\delta(\text{CH})$
1286 $\delta(\text{CH}_2)+\delta(\text{OH})$	1285 $\delta(\text{CH}_2)+\delta(\text{OH})$	1270 $\delta(\text{CH}_2)+\delta(\text{OH})$	1285 $\delta(\text{CH}_2)+\delta(\text{OH})$
1323 $\delta(\text{NH})+\delta(\text{CH})$	1324 $\delta(\text{NH})+\delta(\text{CH})$	1315 $\delta(\text{NH})+\delta(\text{CH})$	1323 $\delta(\text{NH})+\delta(\text{CH})$
1336 $\omega(\text{CH}_2)+\delta(\text{CH})$	1335 $\omega(\text{CH}_2)+\delta(\text{CH})$		1337 $\omega(\text{CH}_2)+\delta(\text{CH})$
1357 $\omega(\text{CH}_2)+\tau(\text{CH}_2)$	1357 $\omega(\text{CH}_2)+\tau(\text{CH}_2)/\delta(\text{CH}_3)$	1355 $\omega(\text{CH}_2)+\tau(\text{CH}_2)$	1357 $\omega(\text{CH}_2)+\tau(\text{CH}_2)$
1458 $\delta(\text{CH}_2)$	1455 $\delta_{\text{as}}(\text{CH}_3)$	1458 $\delta_{\text{as}}(\text{CH}_3)$	1458 $\delta_{\text{as}}(\text{CH}_3)$
1570 $\nu_{\text{as}}(\text{CC ring})+\delta(\text{CH ring})$	1570 $\nu_{\text{as}}(\text{CC ring})+\delta(\text{CH ring})$	1569 $\nu_{\text{as}}(\text{CC ring})+\delta(\text{CH ring})$	1570 $\nu_{\text{as}}(\text{CC ring})+\delta(\text{CH ring})$
1597 $\nu(\text{CC ring})+\delta(\text{CH ring})$	1597 $\nu(\text{CC ring})+\delta(\text{CH ring})$		1597 $\nu(\text{CC ring})+\delta(\text{CH ring})$
1615	1615	1615 $\nu(\text{CC ring})+\delta(\text{CH ring})$	1615 $\nu(\text{CC ring})+\delta(\text{CH ring})$



**Fig. S1.** (a) UV-vis spectrum of the solution of synthesized Seed A, B, C, D and AgNPs. (b) TEM image of AgNPs.



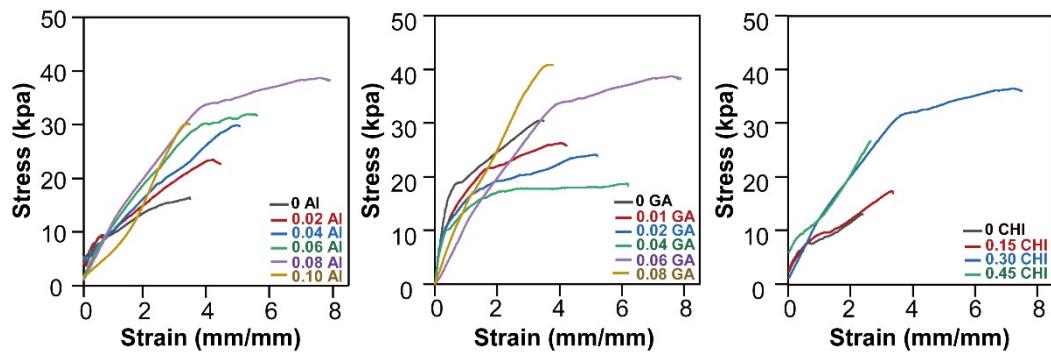
**Fig. S2.** Large amount of CHI (from left to right: 0.45, 0.5, 0.6g) failed to reach gelation. Gel with 0.6 g CHI was too soft to reach gelation.



**Fig. S3.** The impact of different  $\text{Al}^{3+}$  (from left to right: 0, 0.02, 0.04, 0.10g) contents on gelation of the GA-containing samples.



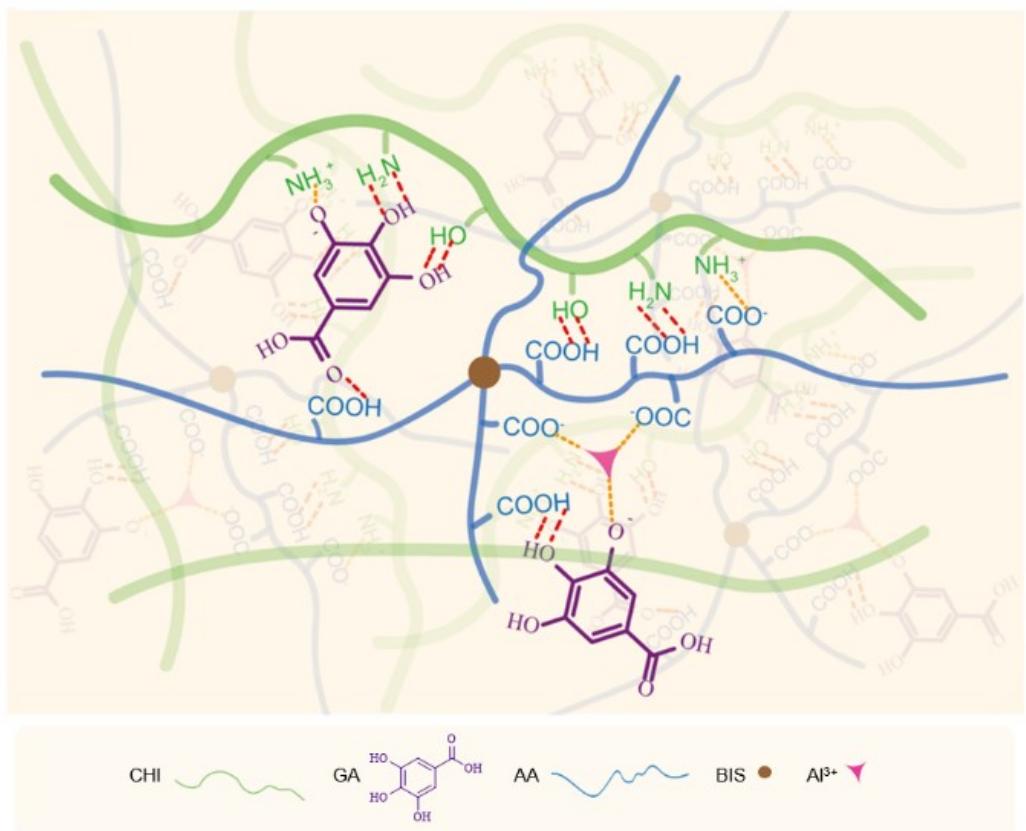
**Fig. S4.** The impact of different GA (from left to right: 0, 0.01, 0.02, 0.04, 0.06, 0.08g) contents on gelation.



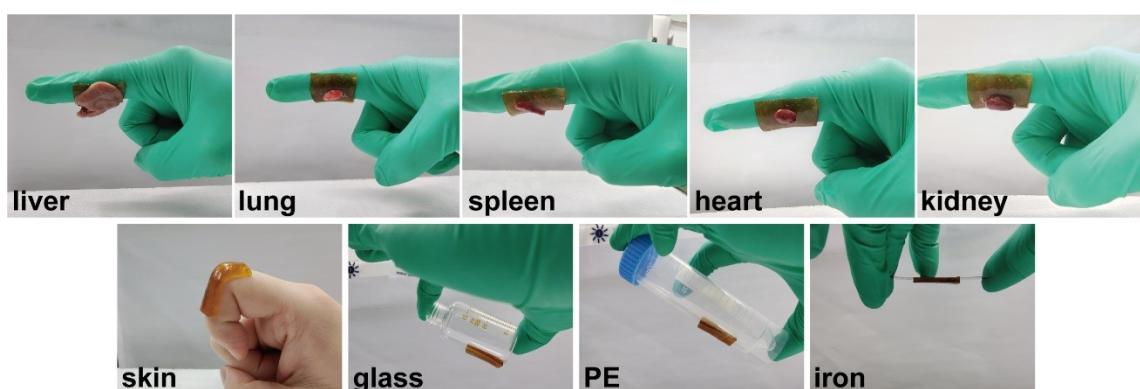
**Fig. S5.** The tensile curves of (a) P-0.3C-0.06GA-(0, 0.02, 0.04, 0.06, 0.08, 0.10)Al, (b) P-0.3C-(0, 0.01, 0.02, 0.04, 0.06, 0.08)GA-0.08Al and (c) P-(0, 0.15, 0.3, 0.45)C-0.06GA-0.08Al.



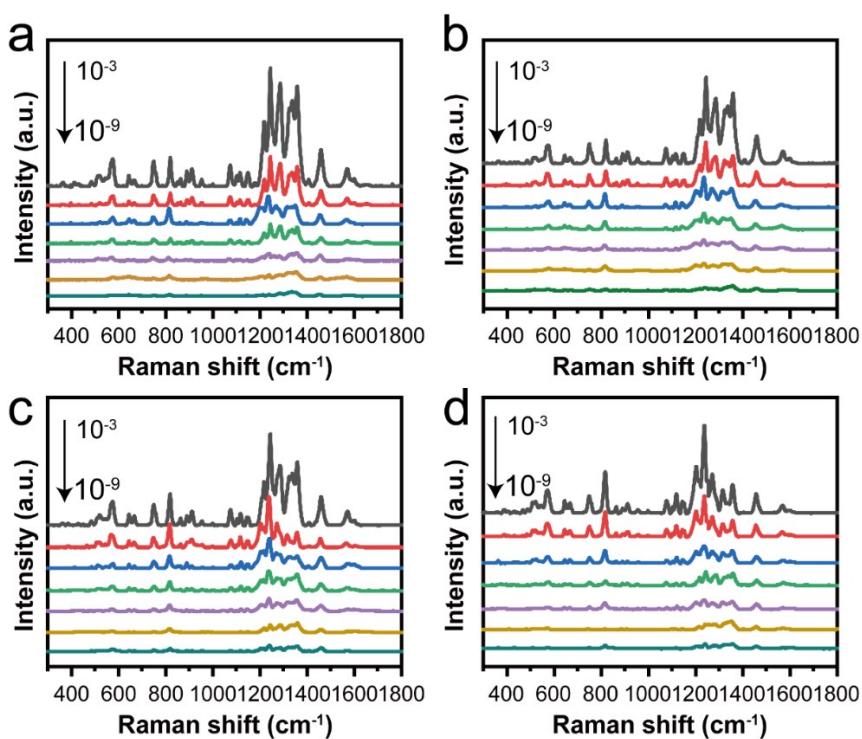
**Fig. S6.**  $\text{Al}^{3+}$  in P-0.3C-0.06GA-0.08Al was replaced by equal molar amount of  $\text{Fe}^{3+}$ ,  $\text{Cu}^{2+}$  and  $\text{Cr}^{3+}$  (from left to right), making P-0.3C-0.06GA-0.086Fe, P-0.3C-0.06GA-0.051Cu and P-0.3C-0.06GA-0.085Cr hydrogels. Only the gelation of P-0.3C-0.06GA-0.085Cr succeeded.



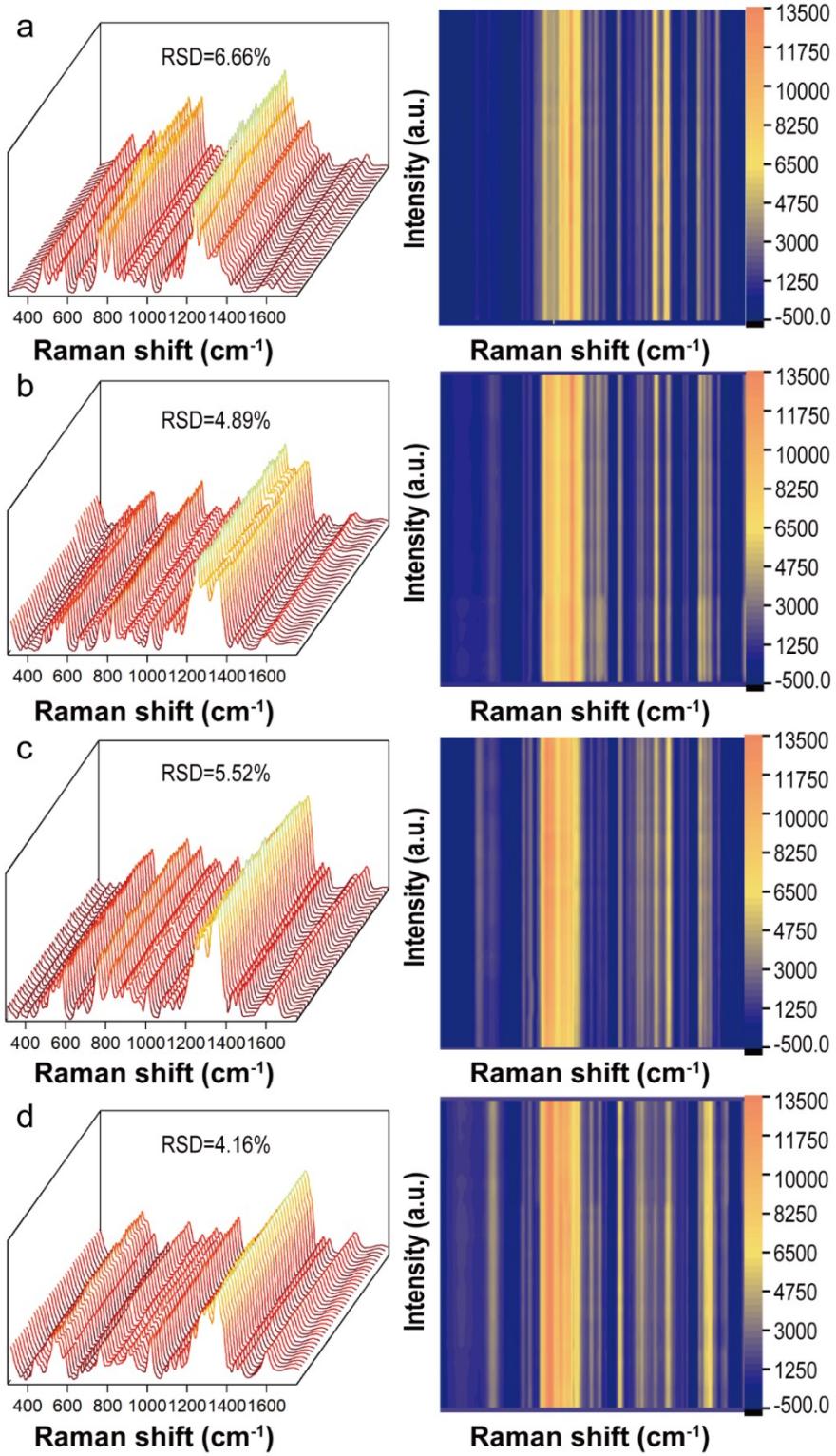
**Fig. S7.** Multiple hydrogen bonds and electrostatic interactions in P-0.3C-0.06GA-0.08Al hydrogel.



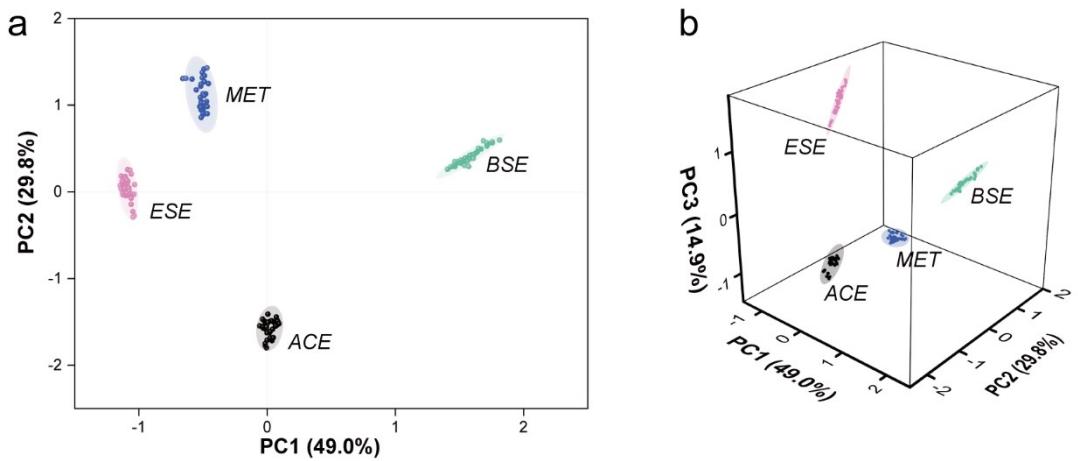
**Fig. S8.** Adhesiveness of P-0.3C-0.06GA-0.08Al to various surfaces including: liver, lung, spleen, heart, kidney, skin, glass, PE and iron.



**Fig. S9.** SERS spectra of (a) BSE, (b) MET, (c) ACE and (d) ESE with a concentration of  $10^{-3}\text{-}10^{-9}$  M. 5  $\mu\text{L}$  of BSE, MET, ACE or ESE simulated urine solution was dropped onto the substrate for each sample. Raman spectra were collected by a portable Raman spectrometer. Exposure time was 1500 ms, and laser wavelength was 785 nm.



**Fig. S10.** SERS spectra of (a) BSE, (b) MET, (c) ACE and (d) ESE with a concentration of  $10^{-5}$  M. 5  $\mu$ L of  $10^{-5}$  M BSE, MET, ACE or ESE simulated urine solution was dropped onto the substrate for each sample. Raman spectra were collected by a portable Raman spectrometer. Exposure time was 1500 ms, and laser wavelength was 785 nm.



**Fig. S11.** (a) 2D-PCA and (b) 3D-PCA of four  $\beta$ -blockers simulated urine solution samples are shown as scatter plots. Green dots represent BSE, blue dots represent MET, black dots represent ACE and pink dots represent ESE.